Organisation de Coopération et de Développement Économiques
Organisation for Economic Co-operation and Development

NUCLEAR ENERGY AGENCY
NUCLEAR SCIENCE COMMITTEE

Actinide and Fission Product Partitioning and Transmutation

Workshop Proceedings of the Fourteenth Information Exchange Meeting
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17-20 October 2016

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Foreword

The benefits of partitioning and transmutation (P&T) have now been established worldwide and, as a result, many countries are pursuing research and development (R&D) programmes to advance the technologies associated with P&T. In this context, the OECD Nuclear Energy Agency (NEA) has organised a series of biennial information exchange meetings to provide experts with a forum to present and discuss state-of-the-art developments in the field of partitioning and transmutation since 1990. Previous meetings were held in Mito (Japan) in 1990, at Argonne National Laboratory (United States) in 1992, in Cadarache (France) in 1994, in Mito (Japan) in 1996, in Mol (Belgium) in 1998, in Madrid (Spain) in 2000, in Jeju (Korea) in 2002, in Las Vegas (United States) in 2004, in Nîmes (France) in 2006, in Mito (Japan) in 2008, in San Francisco (United States) in 2010, in Prague (Czech Republic) in 2012 and in Seoul (Korea) in 2014. They were co-sponsored by the European Commission (EC) and the International Atomic Energy Agency (IAEA). The 14th Information Exchange Meeting was held in San Diego (United States) on 17-21 October 2016, hosted by Idaho National Laboratory (INL). The workshop comprised a plenary session on national and international programmes followed by technical sessions and a poster session covering various aspects of P&T. The information exchange meetings on P&T form a part of the NEA programme of work in the field of advanced nuclear fuel cycles. These proceedings include papers presented at the 14th Information Exchange Meeting. The opinions expressed are those of the authors only and do not necessarily reflect the NEA views or those of its member countries.
Acknowledgements

The NEA wishes to thank the Idaho National Laboratory for hosting the 14th Information Exchange Meeting on Actinide and Fission Product Partitioning and Transmutation. The NEA would also like to express its gratitude to all the scientists who participated in this workshop and contributed to the improvement of international co-operation in the field of partitioning and transmutation. Special thanks are due to T. Todd, J. Law and M. Oliveira for their invaluable assistance and support in organising this conference. We would also like to thank members of the Scientific Advisory Committee for devoting time to organising this workshop and in particular to reviewing the abstracts. The NEA also extends its gratitude to the IAEA representatives for their co-operation.
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<th>Description</th>
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<tr>
<td>AAS</td>
<td>Atomic absorption spectrophotometry</td>
</tr>
<tr>
<td>ABR</td>
<td>Advanced burner reactor</td>
</tr>
<tr>
<td>ACPF</td>
<td>Advanced spent fuel conditioning process demonstration facility</td>
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<tr>
<td>ACSEPT</td>
<td>Actinide reCycling by SEParation and Transmutation</td>
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<tr>
<td>ACTOFF</td>
<td>Analysis of options and experimental examination of fuels for water-cooled reactors with increased accident tolerance</td>
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<tr>
<td>ADS</td>
<td>Accelerator-driven system</td>
</tr>
<tr>
<td>ADUN</td>
<td>Acid deficient uranyle nitrate</td>
</tr>
<tr>
<td>AFC</td>
<td>Advanced fuel cycle</td>
</tr>
<tr>
<td>AGF</td>
<td>Alpha-gamma facility</td>
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<tr>
<td>AGR</td>
<td>Advanced gas reactor</td>
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<td>AGS</td>
<td>Alpha-gamma section</td>
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<tr>
<td>ALARA</td>
<td>As low as reasonably attainable</td>
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<tr>
<td>ALE</td>
<td>Alkaline earths</td>
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<tr>
<td>ALFUS</td>
<td>ALloyed fuel unified simulator</td>
</tr>
<tr>
<td>ALG</td>
<td>Alginate</td>
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<tr>
<td>ALLEGRO</td>
<td>Experimental low-power, non-electricity generating, gas-cooled fast reactor (GFR) project</td>
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<tr>
<td>ALSEP</td>
<td>Actinide-lanthanide separation process</td>
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<tr>
<td>AmBB</td>
<td>Americium bearing blanket</td>
</tr>
<tr>
<td>AMBER</td>
<td>Assisted model building with energy refinement</td>
</tr>
<tr>
<td>AmN</td>
<td>Americium nitride</td>
</tr>
<tr>
<td>AMP</td>
<td>Ammonium molybdroposphate</td>
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<tr>
<td>AMUSE</td>
<td>Astrophysical multipurpose software environment</td>
</tr>
<tr>
<td>ANL</td>
<td>Argonne National Laboratory</td>
</tr>
<tr>
<td>ANS</td>
<td>American Nuclear Society</td>
</tr>
<tr>
<td>AOSTA</td>
<td>Activation of OSMOSE sample in TAPIRO</td>
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<tr>
<td>APS</td>
<td>Advanced photon source</td>
</tr>
<tr>
<td>APT</td>
<td>Atom probe tomography</td>
</tr>
<tr>
<td>AREVA</td>
<td>French multinational group specialising in nuclear power and renewable energy (headquartered in Paris La Défense)</td>
</tr>
<tr>
<td>Abbreviation</td>
<td>Description</td>
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<tr>
<td>ARIS</td>
<td>Advanced reactors information system</td>
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<td>AFSR</td>
<td>Argonne fast source reactor – Idaho Falls</td>
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<td>ASTRID</td>
<td>Advanced sodium technological reactor for industrial demonstration</td>
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<tr>
<td>ATR</td>
<td>Advanced test reactor</td>
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<tr>
<td>ATW</td>
<td>Accelerator-driven transmutation of waste</td>
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<tr>
<td>AWP</td>
<td>Ammonium tungstophosphate</td>
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<tr>
<td>B&amp;B</td>
<td>Breed and burn</td>
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<tr>
<td>BigT</td>
<td>Burnable absorber-integrated Guide Thimble</td>
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<tr>
<td>BNFL</td>
<td>British Nuclear Fuels Limited</td>
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<tr>
<td>BOC</td>
<td>Beginning of cycle</td>
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<tr>
<td>BU</td>
<td>Burnup</td>
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<tr>
<td>BWR</td>
<td>Boiling water reactor</td>
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<tr>
<td>CANDU</td>
<td>Canadian deuterium uranium reactor</td>
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<tr>
<td>CBP</td>
<td>Chaîne blindée procédé</td>
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<tr>
<td>CEA</td>
<td>Cost-effectiveness analysis</td>
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<tr>
<td>CERCER</td>
<td>Ceramic ceramic</td>
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<tr>
<td>CFD</td>
<td>Computational fluid dynamics</td>
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<tr>
<td>CHON</td>
<td>Carbon, hydrogen, oxygen and nitrogen</td>
</tr>
<tr>
<td>CIEMAT</td>
<td>Centro de Investigaciones Energéticas, Medioambientales y Tecnológicas</td>
</tr>
<tr>
<td>CMPO</td>
<td>Octyl(phenyl)-N,N-diisobutylcarbonylmethylphosphine oxide</td>
</tr>
<tr>
<td>CN</td>
<td>Co-ordination numbers</td>
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<td>CNL</td>
<td>Canada National Laboratories</td>
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<tr>
<td>COEX™</td>
<td>Co-extraction of actinides</td>
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<td>COGEMA</td>
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<td>CR</td>
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<td>CRIEPI</td>
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<td>CRMP</td>
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<td>CSA</td>
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<td>DDR</td>
<td>Declining discount rate</td>
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<td>Description</td>
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<tr>
<td>DF</td>
<td>Decontamination factor</td>
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<td>DUPIC fuel development facility</td>
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<td>DGA</td>
<td>Diglycolamide</td>
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<td>DGR</td>
<td>Deep geological repository</td>
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<td>DIAMINO</td>
<td>Dispositif d'Irradiaion d'Actinides MINeurs dans Osiris</td>
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<td>DPA</td>
<td>Displacement per atom</td>
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<td>DOE</td>
<td>Department of Energy, United States</td>
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<td>DRUP</td>
<td>Direct reuse of used pressurised water reactor</td>
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<td>Diethylenetriaminepentaacetic acid</td>
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<td>DU</td>
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<td>Direct use of spent pressurised water reactor fuel into CANDU reactor</td>
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<td>EFPD</td>
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<td>FCCI</td>
<td>Fuel-cladding chemical interaction</td>
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<td>Fuel-cladding mechanical interaction</td>
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<td>FCRD</td>
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<td>Field emission gun</td>
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<td>FEM</td>
<td>Finite element method</td>
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<td>Fusion-fission hybrid reactor</td>
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<td>FFTF</td>
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<td>FMF</td>
<td>Fuel monitoring facility</td>
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<td>First of a kind</td>
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<td>FRKP</td>
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<tr>
<td>HCl</td>
<td>Hydrochloric acid</td>
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<td>HDEHP</td>
<td>Di-2-ethylhexylphosphoric acid</td>
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<td>N-(2-hydroxyethyl)-ethylenediamine-N,N',N'-triacetic acid</td>
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<td>HEH[EHP]</td>
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<td>HEU</td>
<td>High enriched uranium</td>
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<td>Hydrofluoric acid</td>
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<td>HFIR</td>
<td>High flux isotope reactor</td>
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<td>High-level liquid radio-waste</td>
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<td>HLM</td>
<td>Heavy liquid metal</td>
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<td>HLW</td>
<td>High-level waste</td>
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<td>HMVF</td>
<td>Heavy metal volume fraction</td>
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<td>HONTA</td>
<td>HexaoctylNitrilotriacetamide</td>
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<td>HTGR</td>
<td>High-temperature gas-cooled reactor</td>
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<td>High temperature reactor</td>
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<td>HZDR</td>
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<td>IAEA</td>
<td>International Atomic Energy Agency</td>
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<td>IBG</td>
<td>Internal breeding gain</td>
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<td>LCAE</td>
<td>Levelised cost of electricity at equilibrium</td>
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<tr>
<td>ICAPP</td>
<td>International Congress on Advances in Nuclear Power Plants</td>
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<td>IDC</td>
<td>Interest during construction</td>
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<td>IE</td>
<td>Initial enrichment</td>
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<td>INPRO</td>
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<tr>
<td>ILW</td>
<td>Intermediate-level waste</td>
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<td>IOZ</td>
<td>Inner oxidation zone</td>
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<tr>
<td>IR</td>
<td>Infrared</td>
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<td>IRMM</td>
<td>Institute for Reference Materials and Measurements (European Commission Joint Research Centre)</td>
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<td>ITU</td>
<td>Institute for Transuranium Elements (European Commission Joint Research Centre)</td>
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<td>JAEA</td>
<td>Japon Atomic Energy Agency</td>
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<td>JB</td>
<td>Jarque-Bera</td>
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<td>Acronym</td>
<td>Description</td>
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<td>JNC</td>
<td>Japan Nuclear Cycle Development Institute</td>
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<td>JNFL</td>
<td>Japan Nuclear Fuel Limited</td>
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<td>JOPRAD</td>
<td>Joint Programme on Radioactive Waste Disposal</td>
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<td>JRC</td>
<td>Joint Research Centre (European Commission)</td>
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<td>KAERI</td>
<td>Korea Atomic Energy Research Institute (Korea)</td>
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<td>KAPF</td>
<td>Korea Advanced Pyroprocess Facility (Korea)</td>
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<tr>
<td>KCuFC</td>
<td>Potassium copper hexacyanoferrate</td>
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<td>KUCA</td>
<td>Kyoto University Critical Assembly</td>
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<td>LBE</td>
<td>Lead-bismuth eutectic</td>
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<td>LCC</td>
<td>Liquid Cd cathode</td>
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<td>LEU</td>
<td>Low-enriched uranium</td>
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<td>LFR</td>
<td>Lead fast reactor</td>
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<td>LHR</td>
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<td>LINAC</td>
<td>Linear acceleraror</td>
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<td>LKE</td>
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<td>Loss-of-coolant accident</td>
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<td>LSC</td>
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<td>Light water reactor</td>
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<td>MA</td>
<td>Minor actinide</td>
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<td>MABB</td>
<td>Minor-actinide bearing blankets</td>
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<tr>
<td>MCNP</td>
<td>Monte Carlo N-Particle</td>
</tr>
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<td>MADB</td>
<td>Minor actinide database</td>
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<td>MATADOR</td>
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<td>MC</td>
<td>Monte Carlo</td>
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<td>MCT</td>
<td>Medium-chain triglycerides</td>
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<td>MDD</td>
<td>Modified direct denitration</td>
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<td>MEXICO</td>
<td>Mass exchanger in continuous operation</td>
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<td>Ministry of Economy, Trade and Industry (Japan)</td>
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<td>Ministry of Education, Culture, Sports, Science and Technology (Japan)</td>
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<td>MgO</td>
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<td>MRCAT</td>
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<td>MS</td>
<td>Member states</td>
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<td>Description</td>
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<td>MSDR</td>
<td>Molten salts dynamics rig</td>
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<td>MSFR</td>
<td>Molten salt fast reactor</td>
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<td>MOX</td>
<td>Mixed oxide</td>
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<td>MSR</td>
<td>Molten salt reactor</td>
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<td>MTZ</td>
<td>Mass transfer zone</td>
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<td>MYRRHA</td>
<td>Multipurpose hybrid research reactor for high-tech applications</td>
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<td>Nuclear Energy Agency</td>
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<td>Nuclear Energy University Programme (US Department of Energy)</td>
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<td>NDT</td>
<td>Non-destructive tests</td>
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<td>NEAMS</td>
<td>Nuclear energy advanced modelling and simulation</td>
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<td>NES</td>
<td>Nuclear energy systems</td>
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<td>NFCIS</td>
<td>Nuclear fuel cycle information system</td>
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<td>Nuclear fuel cycle and materials section</td>
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<td>National Nuclear Security Administration (US Department of Energy)</td>
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<td>Nth of a kind</td>
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<td>Nitrogen oxide</td>
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<td>Nuclear power plant</td>
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<td>Nuclear Power Technology Development Section (International Atomic Energy Agency)</td>
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<td>Nuclear Science Committee (Nuclear Energy Agency)</td>
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<td>Natural uranium</td>
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<td>O&amp;M</td>
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<td>Oxide dispersion strengthened</td>
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<td>Optical microscopy</td>
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<td>Public Engagement Commission on Spent Nuclear Fuels (Korea)</td>
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<td>PERSENT</td>
<td>Perturbation and sensitivity for transport</td>
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<td>Pentaerythritol</td>
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<td>PFPF</td>
<td>Plutonium fuel production facility</td>
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<td>PGSFR</td>
<td>Prototype of generation IV sodium-cooled fast reactor</td>
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<td>PGM</td>
<td>Platinum group metals</td>
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<td>PHITS</td>
<td>Particle and Heavy Ion Transport Code System</td>
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<td>Physics of reactor</td>
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<td>PPP</td>
<td>Purchase power parity</td>
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<td>Quantum and radiological science and technology</td>
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<td>Research co-ordination meeting</td>
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<td>R&amp;D</td>
<td>Research and development</td>
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<tr>
<td>RD&amp;D</td>
<td>Research, development and demonstration</td>
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<td>RE</td>
<td>Rare earth</td>
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<td>REFINRE</td>
<td>Researching Fracking in Europe</td>
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<td>RF</td>
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<td>Reverse flow diverter</td>
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<td>Remote handling parts test rig</td>
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<td>Safety of actinide separation processes</td>
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<td>SAXS</td>
<td>Small angle X-ray</td>
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<td>Scanning and transmission electron microscopy</td>
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<td>Sustainable Nuclear Energy Technology Platform (European Commission)</td>
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<td>SmART</td>
<td>Small amount of reuse fuel test cycle</td>
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<td>Spent nuclear fuel</td>
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<td>Scientific readiness levels</td>
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<td>STAAR</td>
<td>Sigma team for advanced actinide recycle</td>
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<tr>
<td>STH</td>
<td>System thermal hydraulic</td>
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<td>SWU</td>
<td>Separative work unit</td>
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<tr>
<td>SYNERGIES</td>
<td>Synergistic nuclear energy regional group interactions evaluated for sustainability</td>
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<tr>
<td>T2EHDGA</td>
<td>N,N,N',N'-tetra-2-ethylhexyl diglycolamide</td>
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<td>TAPIRO</td>
<td>Taratura pila rapida a potenza 0</td>
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<td>TBZ</td>
<td>Tritium breeding zone</td>
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<td>TBP</td>
<td>Tributylphosphate</td>
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<td>TBR</td>
<td>Tritium breeding ration</td>
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<td>TD</td>
<td>Theoretical density</td>
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<td>Transmutation experimental facility</td>
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<td>Transmission electron microscopy</td>
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<td>TEPCO</td>
<td>Tokyo Electric Power Company (Japan)</td>
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<td>THORP</td>
<td>Thermal oxide reprocessing plant</td>
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<td>TIARA</td>
<td>Takasaki ion accelerators for advanced radiation application</td>
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<td>Tungsten inert gas</td>
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<td>TIMS</td>
<td>Thermal ionisation mass spectrometry</td>
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<td>TOA</td>
<td>Triocetyl amine</td>
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<td>TOF</td>
<td>Time of flight</td>
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<td>TPDN</td>
<td>N,N,N',N'-tetrakis(pyridin-2-ylmethyl)- decane-1,2-diamine</td>
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<tr>
<td>TPH</td>
<td>Total petroleum hydrocarbons</td>
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</table>
TRL    Technology readiness level
TRU    Transuranic
TRUEX  Transuranic element extraction
TTB    Transport-to-biosphere
TWG    Technical working groups
UDV    Ultrasonic doppler velocimetry
ULOF   Unprotected loss of flow
UMACS  Uranium-minor actinide conventional sintering
UNF    Used nuclear fuel
UOX    Uranium oxide
UREX   Uranium extraction
US DOE  United States Department of Energy
UTOP   Unprotected loss of power
VHLW   Vitrified high-level waste
VIPAC  Vibratory compaction
VVF    Void volume fraction
WAR    Weak acid resin
WIRAF  Waste from innovative types of reactors and fuel
WPFC   Working Party on Scientific Issues of the Fuel Cycle (Nuclear Energy Agency)
WT     Waste transmutation
WTR    Waste transmutation ratio
XAFS   X-ray absorption fine structure
XRD    X-ray diffraction
ZrN    Zirconium nitride
Executive summary

The Nuclear Energy Agency (NEA) Information Exchange Meeting on Actinides and Fission Products Partitioning and Transmutation (IEMPT) has been organised since 1990 and acts as a forum for experts to present and discuss the state-of-the-art development in the field of partitioning and transmutation (P&T). Thirteen meetings have been organised so far and were held in Japan, the United States, France, Belgium, Spain, Korea and the Czech Republic. This 14th meeting was hosted by Idaho National Laboratory, held in San Diego (United States) and was organised in co-operation with the International Atomic Energy Agency (IAEA).

The meeting covered strategic and scientific developments in the field of P&T such as: fuel cycle strategies and transition scenarios, the role of P&T in the potential evolution of nuclear energy as part of the future energy mix; radioactive waste management strategies; transmutation fuels and targets; advances in pyro and aqueous separation processes; P&T specific technology requirements (materials, spallation targets, coolants, etc.); transmutation systems: design, performance and safety; impact of P&T on the fuel cycle; fabrication, handling and transportation of transmutation fuels.

A total of 89 presentations (41 oral and 48 posters) were discussed among the 94 participants from 15 countries and 3 international organisations. The meeting consisted of one plenary session where national and international programmes were presented followed by five technical sessions on:

- Fuel cycle strategies and transition scenarios;
- Transmutation fuels and targets;
- Transmutation systems and infrastructures;
- Waste management for P&T;
- Advanced nuclear fuel recycling.

Welcome address and opening lecture

The welcome address was given by J. Law (US) and S. Cornet (NEA).

Session 1: National and International Programmes

S. Cornet (NEA) summarised the ongoing activities of the NEA related to P&T. Most work is carried out within the Nuclear Science programme of work and the Working Party of Scientific Issues of the Fuel Cycle (WPFC). Current activities involve MA-bearing fuel performance benchmark studies, an international review of separation processes, minor actinides management and recycling.

An overview of the IAEA activities on P&T was given by A. Gonzales-Espartero (IAEA). The framework of the IAEA activities is carried out by several Nuclear Energy sections and technical working groups (TWG) and is implemented through co-ordinated research projects (CRP), the organisation of international conferences and workshops, the publication of technical documents and reports and the management of specific databases. A report on the status and trends in pyroprocessing should be published soon. Other ongoing activities include the preparation of a
technical document on the challenges of reprocessing fuels from fast reactors and advanced fuel cycle for waste burden minimisation. Transmutation activities focus on fast neutron systems such as fast reactors (FR) and accelerator-driven systems (ADS). In addition, the IAEA will organise an international conference on fast reactors and related fuel cycles (FR17) in 2017 in Yekaterinburg, Russia.

**P. Paviet** (US) gave an overview of the US research and development (R&D) activities. The US is evaluating advanced (closed) fuel cycles for a potential deployment around mid-century. A comprehensive nuclear fuel cycle evaluation and screening study has been completed and indicated that continuous recycle of U/Pu or U/TRU are the most promising fuel cycles. The US fuel cycles technologies programmes are performing R&D on advanced partitioning technologies and advanced transmutation fuels for potential fuel cycles, investigating both homogeneous and heterogeneous approaches and focusing on more robust and simpler processes. Aqueous (for oxide fuels) and electrochemical (for metal fast reactor fuels) technologies are being developed.

**F. Rayment** (UK) gave an update on nuclear energy and P&T programmes in the United Kingdom. Current programmes involve collaboration between industries, universities and national laboratories. The foreseen programmes for the next decades will mostly focus on fuel manufacturing, reactor design, advanced spent fuel recycling and strategic toolkit. Spent fuel recycling programmes primarily focus on aqueous technologies but investigations on pyroprocessing as a complementary technology are also being carried out. The UK is involved in various European and international projects.

The recent advances on P&T R&D in France were presented by **E. Touron** (France). The Commissariat à l’énergie atomique et aux énergies alternatives (CEA) co-ordinates research work carried out by public research bodies (ANDRA, CEA, CNRS, Universities) and their industrial partners (AREVA, EDF) on plutonium multirecycling and on long-lived radioactive elements partitioning and transmutation, in close connection with the work carried out on the new generation of nuclear reactors. Scenario studies have been carried out with industrial partners and confirmed the benefits of a progressive deployment of SFR (sodium fast reactors). The French SFR programme focuses on the ASTRID reactor. The main interest of transmutation is the significant reduction of the residual heat of wastes through the separation of Am. Separation of Am is achieved with the ExAm process. Recovered Am can be reused in fuel and some experimental irradiations on UAmO₂ fuels have been performed.

**J-P. Glatz** (EU) presented the development of safe and responsible P&T options in the European Union (Eu). The status of nuclear energy in EU was summarised and the different European R&D projects related to P&T were highlighted. Projects related to fuels, materials, reactors are being carried out. Most projects support the development of ASTRID, ALFRED, MYRRHA and ALLEGRO in relation with the European Sustainable Nuclear Industrial Initiative (ESNII).

**K. Tsujimoto** (Japan) reviewed the research activities on partitioning and transmutation in Japan. The Japanese government has issued a new strategic energy plan in April 2014 where the importance of R&D on P&T was stated. Since the accident of Fukushima Daiichi, the 54 reactors have been shut down and only 3 are currently operating. The Japan Atomic Energy Agency (JAEA) R&D activities on P&T technology is based on two concepts: fast reactors (FR) and accelerator-driven system (ADS). Two fuel cycle options are considered for P&T: homogeneous recycle in fast breeder reactor (FBR) and transmutation in ADS. Ongoing R&D programmes focus on transmutation of minor actinides (MA). The importance of international collaboration was mentioned.

In Belgium, fuel cycle activities are based on the MYRRHA project. The progress of the project was presented **H.A. Abderrahim** (Belgium). The history of lead-bismuth eutectic (LBE)-cooled ADS in
Europe was given. MYRRHA emerged as EU collaborative project for burning minor actinides through the ADOPT and EUROTRANS projects. The main outcomes of these projects were described. The MYRRHA programme was detailed along with the new implementation approach established from the period 2016-2030.

**J-H Baek** (Korea) summarised the progress of activities for recycling spent nuclear fuel in Korea. The government launched the Public Engagement Commission on Spent Nuclear Fuels (PECOS) in 2013 which made recommendations to the government for a spent fuel management policy in June 2015. For the moment, spent fuel policy is based on a “wait and see” option and final decision will be made following the results of studies performed on different options. Sodium-cooled fast reactor (SFR) coupled with pyroprocessing is expected to be a promising option for long-term spent fuel management in Korea. The prototype SFR (PGSFR) is under development for licence application planned in 2017, and its construction is planned for 2023. R&D on pyroprocessing has made significant progresses with U-only process facility (PRIDE). Hot process facilities for head-end (DFDF) and direct oxide reduction (ACPF) are completed. Prototype of Generation IV sodium-cooled fast reactor (PGSFR) combined with the Korea Advanced Pyroprocess Facility (KAPF) should be operational in the next 20 years, according to national R&D plan.

**Session 2: Fuel Cycle Strategies and Transition Scenarios**

**Y. Sagayama** (Japan) presented the results of fuel cycle scenarios studies. In Japan, a lot of effort is being made for the management of spent nuclear fuel. Fast reactor cycle technology is promoted for an effective utilisation of uranium resources and environmental impact reduction. Minor actinides (MA) partitioning, fuel fabrication and irradiation of MOX pellets containing MA are being investigated for a potential implementation of a FR fuel cycle. In particular, the SmART (Small Amount of Reuse Fuel Test Cycle) cycle is being developed. Transition scenarios from light water reactor (LWRs) to FRs with reprocessing have been studied with the code SCM developed by JAEA. Results of the study showed that fast reactor fuel cycle allow a better utilisation of resources and waste management in the medium and long term.

Fuel cycle analysis in the UK was described by **D. Mathers** (UK). The UK is committed to reduce its greenhouse gas emissions by 80% by 2020. To achieve this goal, an increase in its nuclear capacity with new build and the potential introduction of fast reactor fleet is envisaged. Scenario studies on a UK 40GWe fuel cycle were carried out with the ORION code. Results show that the transition to FR fleet is possible over the timescales considered in the scenarios. Volumes of spent fuel can be managed and reduced through recycle.

Fast reactors deployment scenarios were studied in France and the results were presented by **E. Touron** (France). The current strategy is the recycle Pu in current reactors to reduce the amount of natural uranium needs and the quantity of spent fuels in storage. Future fast reactor fuel cycles include actinide multirecycling. Scenarios studies were performed and results showed that the progressive introduction of fast reactors with multirecycling helps reduce the net production of Pu, the Pu inventory, natural-U needs, the amount of waste and the high-level waste (HLW) footprint.

**R. Wigeland** (US) summarised the US fuel cycle strategies and scenarios studies. The nuclear fuel cycle evaluation and screening study performed between 2011 and 2014 identified fuel cycles that have the potential for significant improvement in performance compared to the current US fuel cycle. Subsequent studies on minor actinide recycling indicated that the implementation of both U/Pu and U/TRU recycling will be challenging: for U/Pu recycle, the challenge is in waste form development.
and for U/TRU recycle, the challenge is recycle fuel and U/TRU reprocessing development. Transition scenarios studies have shown the potential for an option using LEU to start-up fast reactors.

The economics of fuel cycle was outlined by E. Hoffman (US) who explained that the transition to an alternative fuel cycle system introduces unique economic challenges not present in the ultimate system once fully deployed and operating as intended. A study was carried out to identify the economics issues that must be overcome for the deployment of an alternative fuel cycle option and to propose approaches and methodologies to evaluate these issues for different deployment pathways and fuel cycle options.

M. Tiphine presented the results of a benchmark study on the effects of uncertainties analysis of input parameters on nuclear fuel cycle scenario carried out within the NEA Expert Group on Advanced Fuel Cycle Scenarios (EGAFCS). The objectives of this international study were to provide guidance on which input parameters are important to be investigated thoroughly, and which components can be less well known, given the objectives of a particular study. A total of 17 key input parameters and 22 scenario outputs were identified and sensitivity analyses were conducted. This sensitivity study showed the relative impact of each parameter on each output metric. In order to compare the large amount of results, two methods of summarisation were adopted. The results and conclusions are included in an NEA report expected to be published in 2017.

Session 3: Transmutation Systems and Infrastructures

K Arie (Japan) gave a presentation on the development of innovative TRU (transuranic) burning fast reactor cycle with Uranium-free fuel. A 4-year research programme was initiated to clarify the feasibility of a core design with uranium-free TRU fuel, by evaluating the fundamental properties of the fuel, countermeasures against FCCI (fuel-cladding chemical interaction), the feasibility of electrochemical reprocessing, reactivity coefficient enhancements, etc. Preliminary results were summarised.

A. Aerts (Belgium) discussed the R&D programme on LBE technology to support the licensing and engineering design of MYRRHA facilities. Small and large scale LBE facilities have been constructed in the past years and several of these installations have already generated results while others are in the final phase of commissioning. Investigations on heavy metal coolant chemistry (MEXICO, HELIOS-3, CRAFT), thermal hydraulics (E-SCAPE & COMPLOT), ultrasonic instrumentation, and remote handling concepts (RHAPTER) are being carried out.

L. Buiron (France) gave an overview of the minor actinide transmutation advanced design for optimised reprocessing (MATADOR) concept, an innovative Am-bearing assembly designed to address decay heat limitation for transportation. This promising concept needs to be fully characterised to ensure its viability for heterogeneous minor actinide management.

F. Heidet (US) discussed the new analysis tools developed at Argonne National Laboratory (ANL) to assess the feasibility of burning TRU in a chloride-based MSR. This tool can easily model molten salt reactors (MSR) and determine the evolution of the fuel composition. The conclusions of the study show that by using salts having a low fuel molar concentration, a high TRU burning rate can be achieved. However, other issues should be addressed such as for example, determining the solubility limits of the actinides, assessing the feasibility of the process, etc.

T. Sugarawa (Japan) presented the results of using the subcriticality adjustment rod (SAR) to obtain a small burnup reactivity, enabling the reduction of the maximum proton beam current and enabling a more feasible beam window design.
The analysis of different americium bearing blanket (AmBB) transmutation strategies to support the characterisation of irradiation experiments was highlighted by N. Stauff (US). Transmutation performance of AmBB was investigated for oxide and metal fuels. AmBB assembly will be irradiated in advanced test reactor (ATR).

**Session 4: Transmutation Fuels and Targets**

Advances in metallic fuels were described by S. Hayes (US). Past irradiations of metallic fuels have demonstrated interesting properties. Irradiation of these types of fuels in fast reactors is considered as a key option for actinide management. Innovative metallic fuel technology is under investigation, in particular looking at Mo-based metallic fuel alloy system to eliminate fuel redistribution, low smeared density to achieve high burnup, and addition of Pd to fuel alloy to avoid fuel-cladding chemical interactions (FCCI).

On behalf of members of the NEA Expert Group on Innovative Fuels, N. Chauvin (NEA) summarised the state-of-the-art report on innovative fuels for advanced nuclear systems published in 2014. The report reviews the studies performed in member countries on fuels containing minor actinides. Different types of fuels are considered (oxide, metal, nitride, dispersion fuels, etc). An overview of the development and level of qualification of each fuel was given.

A. Aitkaliyeva (US) summarised the results of TEM (transmission electron microscopy) examination of metallic Zr alloys fuels designed for transmutation. TEM revealed the formation of subsurface phases which were not observed using other techniques.

D. Freis (EU) described the results of irradiation experiments carried out within the frame of the international collaborations FUTURIX-FTA, EUROTRANS and FAIRFUELS. The irradiation and PIE of two CerCer and two CerMet fuels for transmutation of Am in the Phénix fast reactors were highlighted (CEA and JRC). Promising results were achieved regarding CerCer fuels based of MgO matrix and on CerMet fuels based on Mo matrix.

A comparison between the AFC-1 and FUTURIX-FTA irradiations was outlined by L. Capriotti (US). Both irradiation experiments were designed to study fuel performance for candidate transmutation fuels. PIE results indicated that the fuel performed well during the irradiation. The metallic fuel pins behaviour is very similar in both irradiation campaigns and are comparable with EBR-II experiments.

T. Ogata (Japan) reported the results of PIE of the METAPHIX irradiation programme on MA-bearing metal fuels. Non-destructive PIE is completed and destructive PIE is still in progress. Results were compared to calculations performed with the fuel performance code ALFUS.

L. Ramond (France) described the process of fabrication of (U,Am)O₂ pellets with controlled porosity from oxide microspheres. This process presents the advantage of being dustless.

**Session 5: Waste Management for P&T Scenarios**

A. Salazar (US) presented an evaluation of criticality safety of geological repository. The goal of the work presented is to assess whether P&T can provide any benefits in avoiding critical mass formation. Models and simulation codes were developed for this purpose.

B. Merk (UK) discussed an innovative P&T system based on a reactor operating with direct spent nuclear fuel (SNF) from LWRs.
**M. Gobien** (Canada) described a high-level analysis of an advanced nuclear fuel cycle scenario in which TRU elements from the used CANDU fuel are burnt in fast reactors. The analysis assumed deployment of FRs only for the purpose of waste management and energy supply. The analysis showed that there are a number of benefits in burning TRU in fast reactors.

**Session 6: Advanced Nuclear Fuel Recycling**

**B. Moyer** (US) highlighted the achievements and progress made by the Sigma team for advanced actinide recycle (STAAR), in particular with the ALSEP process. STAAR focuses on R&D on MA actinides separation. The process flowsheet was designed and is ready for demonstration. Investigations on centrifugal contractor, new aqueous complexants, new N, O extractants have helped improving the performance of the ALSEP process.

**M.-C. Charbonnel** (France) described the ExAm process developed for the selective recovery of Am from the PUREX raffinate. ExAm was demonstrated in the ATALANTE facility in 2010 followed by spiked tests in 2014 and hot tests in 2015. The process was modelled with the PAREX code and calculations results were compared with experimental data.

On behalf of members of the NEA Expert Group on Fuel Recycling Chemistry (EGFRC), **E. Collins** (US) summarised the technology readiness levels (TRL) assessment of separation processes for current and advanced fuel cycles which constitutes one chapter of a state-of-the-art report on progress in separation chemistry prepared by members of the expert group.

**R. Taylor** (UK) described the actinide separation processes investigated at the UK National Nuclear Laboratory (NNL). Although the current policy in the UK is stop reprocessing SNF around 2020, the option to return to a closed fuel cycle at a later stage is kept open. The development of potential flowsheets for thermal and fast reactor reprocessing following both heterogeneous and homogeneous routes for actinide recycle is being investigated through international collaboration.

**M. Matsumura** (Japan) presented the MA separation process being developed at JAEA which consists of three solvent extraction processes. Suitable extractants were selected for each solvent extraction process and hot tests using genuine HLLW are conducted since 2015. New extractants were tested for An(III)/RE (Rare Earth) separation and Am/Cm separation processes. The basic design of process flow was demonstrated.

**S. Bourg** (France) gave an overview of the results of the European project SACSESS (2013-2016). This project has provided a structured framework to improve existing fuel cycles by addressing the safety issues of actinide separation processes. These processes involve new chemical systems. An important experimental work was dedicated to the improvement of the process flowsheets, focusing not only on the key performances of the process but also to numerous side parameters linked to kinetics, radiolytic stability, downstream effects. Methodologies have been developed and applied to these processes in order to identify safety issues and to optimise these processes.

**P. Soucek** (EU) presented the results of R&D on pyrochemical processes achieved within the European project SACSESS in which four work packages were devoted to the safety issues of pyrochemical processes. One part focused on safety aspects of pyrochemical systems. The physicochemical behaviour of actinides and fission products that impacts the chemical safety, e.g. solubility, volatility, influence of oxygen ingress, viscosity, both in chloride and in fluoride melts was studied. The second work package was dedicated to safety related to metallic fuel treatment, where electrochemical techniques were investigated. The third section dealt with safety related to refractory CerMet and CerCer fuels treatment, where the specific safety factors arising from treating
these fuels by the liquid-liquid reductive extraction were addressed. The last work package investigated the conditioning of used chloride and fluoride molten salts into waste streams suitable for a safe storage or disposal in a deep geological repository.

The engineering and materials considerations for the industrialisation of LKE molten salt systems were described by D. Graham (UK). Results of investigations showed that 316 stainless steel is a suitable material under an inert atmosphere.

T. Marakami (Japan) highlighted the recent activities for pyroprocessing of SNF carried out at CRIEPI. Electrochemical behaviours of the irradiated metallic fuels in molten chloride salts were investigated in detail and actinides recovery from the irradiated metallic fuels were successfully demonstrated. A novel pyroprocessing concept using liquid Ga as an electrode was mentioned.

**Poster Session**

A total of 48 posters were presented covering all areas of P&T: ADS, fuels, waste management and reprocessing
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<th>Time</th>
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<tr>
<td>18:00</td>
<td>Registration</td>
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<tr>
<td></td>
<td>Welcome reception</td>
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<td></td>
<td><strong>Monday 17 October 2016</strong></td>
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<tr>
<td>08:30</td>
<td>Welcome address from INL</td>
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<td></td>
<td>J. Law (INL, US)</td>
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<td>08:40</td>
<td>Welcome address from NEA</td>
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<td>S. Cornet (NEA)</td>
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<td><strong>Session I : International and National Programmes</strong></td>
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<td><strong>Chair: T. Todd</strong></td>
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<tr>
<td>08:50</td>
<td>NEA activities related to P&amp;T</td>
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<td></td>
<td>S. Cornet/N. Chauvin (NEA)</td>
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<td>09:05</td>
<td>Ongoing activities on P&amp;T at IAEA</td>
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<td>A. Gonzalez-Espartero (IAEA)</td>
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<td>09:20</td>
<td>US Department of Energy, fuel cycle technologies R&amp;D outlook</td>
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<td>P. Paviet (DOE, US)</td>
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<td>09:35</td>
<td>An update on nuclear energy and P&amp;T programmes within the UK</td>
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<td>F. Rayment (NNL, UK)</td>
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<td>09:50</td>
<td>COFFEE</td>
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<td></td>
<td><strong>Session II: Fuel Cycle Strategies and Transition Scenarios</strong></td>
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<td></td>
<td><strong>Chairs: E. Touron &amp; D. Mathers</strong></td>
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<tr>
<td>10:20</td>
<td>Fuel cycle development and scenario studies in Japan</td>
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<td>M. Takeuchi (IAEA, Japan)</td>
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<td>10:45</td>
<td>Fuel Cycle analysis of a long-term nuclear power scenario for the UK</td>
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<td>D. Mathers (NNL, UK)</td>
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<td>11:10</td>
<td>Analysis of fast reactor deployment scenarios in the French nuclear power fleet</td>
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<td>E. Touron (CEA, France)</td>
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<td>11:35</td>
<td>Analysis of fuel cycle strategies and US transition scenarios</td>
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<td>R. Wigeland (ANL, US)</td>
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<td>12:00</td>
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<td>13:15</td>
<td>Economics of fuel cycle deployment</td>
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<td>E. Hoffman (ANL, US)</td>
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<td>13:40</td>
<td>The effects of the uncertainty of input parameters on nuclear fuel cycle scenario studies</td>
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<td>M. Tiphine (EGAFCS, NEA)</td>
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<tr>
<td>14:05</td>
<td>Presentations of posters for Session II</td>
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<td>3 min. * 6 posters</td>
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<td>14:30</td>
<td>COFFEE</td>
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### Session III: Transmutation Systems and Infrastructures

**Chair:** T. Taiwo & N. Stauff

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<th>Time</th>
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<th>Speaker</th>
<th>Location</th>
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<tr>
<td>14:50</td>
<td>Programme overview on development of innovative TRU burning fast reactor cycle with uranium-free fuel</td>
<td>K. Arie (Toshiba Corporation, Japan)</td>
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<tr>
<td>15:15</td>
<td>R&amp;D on heavy liquid metal technology in support of MYRRHA: State of the art</td>
<td>A. Aerts (SCK•CEN, Belgium)</td>
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<td>15:40</td>
<td>Proposal for a first ADS of significant power (&gt; 1MW) at the Institute for Nuclear Research of the Russian Academy of Sciences</td>
<td>S. Sidorkin (INR RAS, Russia)</td>
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<td>16:05</td>
<td>MATADOR: An innovative sub-assembly design for heterogeneous minor actinides transmutation</td>
<td>L. Buiron (CEA, France)</td>
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<tr>
<td>16:30</td>
<td>Transmutation scoping studies for a chloride molten salt reactor</td>
<td>F. Heidet/B. Feng (ANL, US)</td>
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<tr>
<td>16:55</td>
<td>Investigation of beam window structure for accelerator-driven system with subcriticality adjustment</td>
<td>T. Sugawara (JAEA, Japan)</td>
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<tr>
<td>17:20</td>
<td>Am-bearing blanket transmutation strategies in sodium-cooled fast reactors</td>
<td>N. Stauff (ANL, US)</td>
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<tr>
<td>17:45</td>
<td>Presentations of posters for Session III</td>
<td>3 mins*8 posters</td>
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**Wednesday 19 October 2016**

### Session I: International and National Programmes

**Chair:** Y-J. Choi

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<tr>
<td>08:30</td>
<td>P&amp;T activities in France</td>
<td>E. Touron (CEA, France)</td>
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<tr>
<td>08:45</td>
<td>Development of safe and responsible concepts for advanced nuclear fuel cycles in the EU</td>
<td>JP. Glatz (JRC-ITU Karlsruhe, EU)</td>
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<tr>
<td>09:00</td>
<td>Review of research activities on P&amp;T in Japan</td>
<td>K. Tsujimoto, (JAEA, Japan)</td>
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### Session IV: Transmutation Fuels and Targets

**Chair:** N. Chauvin & L. Ramond

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<th>Time</th>
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<th>Speaker</th>
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<tr>
<td>09:15</td>
<td>Advances in metallic fuels for high burnup and actinide transmutation</td>
<td>S. Hayes (INL, US)</td>
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<td>09:40</td>
<td>State-of-the-art report on innovative fuels</td>
<td>N. Chauvin (EGIF, NEA)</td>
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<td>10:05</td>
<td>COFFEE</td>
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<td>10:35</td>
<td>Microstructural characterisation of metallic transmutation fuels</td>
<td>A. Aitkaliyeva (INL, US)</td>
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<td>11:00</td>
<td>Overview of PIE results on minor actinide bearing fuels in FUTURIX-FTA</td>
<td>D. Freis (EU)</td>
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<td>11:25</td>
<td>Preliminary post-irradiation examination comparison between AFC-1 and</td>
<td>L. Capriotti on behalf of J. Harp (INL, US)</td>
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<td>FUTURIX-FTA</td>
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<td>11:50</td>
<td>Post-irradiation examination of fast reactor metal fuel for minor</td>
<td>T. Ogata (CRIEPI, Japan)</td>
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<td>actinide transmutation</td>
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<td>12:15</td>
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<td>13:30</td>
<td>Fabrication of (U,Ce)O₂ and (U,Am)O₂ pellets with controlled porosity</td>
<td>L. Ramond (CEA, France)</td>
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<td>from oxides microspheres</td>
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<td>13:55</td>
<td>Presentations of posters for Sessions IV &amp; VI</td>
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<td>3 mins*(5+13) posters</td>
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<td>COFFEE</td>
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<td>15:25</td>
<td>Session V: Waste Management for P&amp;T</td>
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<td>Chair: T. Matsumura</td>
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<td>15:50</td>
<td>An innovative way of thinking partitioning and transmutation –</td>
<td>B. Merk (Uni. Liverpool, UK)</td>
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<td>neutron physics of a reactor directly operating on spent nuclear fuel</td>
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<td>16:15</td>
<td>Some implications of recycling CANDU used fuel in fast reactors</td>
<td>M. Gobien (Nuclear Waste Management</td>
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<td>Organisation, Canada)</td>
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<td>16:40</td>
<td>Presentations of posters for Sessions V &amp; VI</td>
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<td>17:45</td>
<td>Poster Session</td>
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<td>Chair: Y. Choi</td>
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**Thursday 20 October 2016**

**Session I: International and National Programmes**

*Chair: I-S. Hwang*

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<th>Time</th>
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<tr>
<td>09:00</td>
<td>Fuel cycle activities in Belgium: MYRRHA project status and new</td>
<td>H. Ait Abderrahim, SCK•CEN</td>
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<td>implementation plan</td>
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<td>09:15</td>
<td>Development activities for recycling spent nuclear fuel in Korea</td>
<td>JH. Baek, KAERI</td>
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Session VI: Advanced Nuclear Fuel Recycling
Chair: C. Ekberg & G. Lumetta

09:30  09:55  Sigma team for advanced actinide recycle: Highlights and directions
        B. Moyer (ORNL, US)

09:55  10:20  The EXAm process: towards the reprocessing of a concentrated PUREX raffinate
        M-C. Charbonnel (CEA, France)

10:20  10:50  COFFEE

10:50  11:15  Technology readiness levels assessment of separation processes for current and
        advanced fuel cycles
        E. Collins (EGFRC, NEA)

11:15  11:40  Actinide separation process development at the UK National Nuclear Laboratory
        R. Taylor (NNL, UK)

11:40  12:05  Recent progress on research and development of separation process for minor
        actinides using new extractants
        T. Matsumura (JAEA, Japan)

12:05  12:30  SACSESS: three years of optimisation of safe actinide separation processes
        S. Bourg (CEA, France)

12:30  14:00  LUNCH

Session VI: Advanced Nuclear Fuel Recycling
Chair: T. Murakami & S. Li

14:00  14:25  Status of research and development on pyrochemical processes for spent
        nuclear fuel treatment in Europe
        P. Soucek (JRC, EU)

14:25  14:50  Engineering and materials considerations of an industrialised LKE molten salt
        system
        D. Graham (NNL, UK)

14:50  15:15  Recent activities on pyroprocessing of spent nuclear fuel in CRIEPI
        T. Murakami (CRIEPI, Japan)

15:15  15:45  COFFEE

15:45  16:15  Summary Session (Session chairs)

16:15  17:00  Discussion and closing
Plenary Session I: International and National Programmes

Chair: T. Todd
NEA activities related to partitioning and transmutation

S. Cornet¹, N. Chauvin²
¹Nuclear Energy Agency, France
²CEA, France

Abstract

Partitioning and transmutation of minor actinides and fission products is investigated worldwide to address issues related to waste management. The elements contained in spent nuclear fuel (SNF) can be separated and transmuted in dedicated reactors to help reduce the amount of radioactive waste.

International collaboration is a key asset to develop this technology and the Nuclear Energy Agency (NEA) has been involved in P&T activities through the work carried out within its different committees and related expert groups. In particular, the Nuclear Science Committee (NSC) and the Working Party on Scientific Issues of the Fuel Cycle (WPFC) have initiated several studies on P&T and current activities on scenarios studies, minor actinides (MA) bearing fuels and separation chemistry are currently ongoing. This work is carried out by members of various expert groups.

The Expert Group on Advanced Fuel Cycle Scenarios (EGAFCS) focuses its activities on scenarios studies. A benchmark study on uncertainty analysis was recently completed and the group is currently working on a benchmark exercise on the management of transuranic (TRU).

Transmutation of MA in fuels is investigated through the activities of the Expert Group on Innovative Fuels (EGIF) which has recently published a state-of-the-art report on innovative fuels covering the fabrication, characterisation and behaviour of different type of fuels containing minor actinides and an assessment of current technology readiness level (TRL).

Finally, the separation of minor actinides is covered in the work performed by the Expert Group on Fuel Recycling Chemistry (EGFRC). A state-of-the-art report on separation chemistry is being edited and should be published soon. The report reviews the different processes being developed in member states. An assessment of the TRLs was also undertaken.

Introduction

The management of spent fuel from nuclear power plants varies according to the perspectives and scenarios in different countries. However, strong common features exist such as the need of a deep geological repository, the requirement to reduce the burden of the radiotoxic elements to be disposed of, non-proliferation issues, etc.

Partitioning and transmutation (P&T) is considered as valuable option to reduce the burden on a geological disposal by minimising the amount of nuclear waste, reducing the heat load and ultimately the quantity of potential radiotoxic isotopes.
To prove these potential benefits, large R&D initiatives on P&T were launched worldwide. The NEA is involved in a number of activities related to P&T covering both strategic and scientific issues. These are mainly conducted under the guidance of the Nuclear Science Committee (NSC). The work is carried out by expert groups of the NSC Working Party on Scientific Issues of the Fuel Cycle (WPFC) who deal with issues arising from various existing and advanced nuclear fuel cycles, including fuel cycle scenarios, separation chemistry and flowsheets, innovative fuels and materials, and waste management. The WPFC also interacts with other relevant NSC Working Parties and NEA Standing Technical Committees, to ensure that the respective work programmes are complementary and to support joint activities where appropriate. A series of reports covering these topics have been published or are in the process of being finalised.

This paper describes the latest NEA activities, developments and findings in the field of partitioning and transmutation.

**Minor actinides separation chemistry**

Recycling is recognised as an important step to minimise the quantity and residual heat of long-lived nuclear waste and alleviate the burden on future deep geological repositories. The first step in recycling technology considers the separation of actinides through chemical separation methods based on either solvent extraction or pyroprocessing. An efficient and selective recovery of key elements from spent nuclear fuel (SNF) is essential for a successful sustainable fuel cycle concept. The Expert Group on Fuel Recycling Chemistry has prepared a state-of-the-art report [1] to assess the status of current technologies for the separation of minor actinides (MA). The report reviews current and advanced processes using or pyroprocessing technologies to achieve either recovery of U and Pu alone or co-processing of U, Pu, Np. Processes for separation of Am and Cu are also reviewed. Comparison of chemical process and technical maturity is achieved through technology readiness levels (TRL). The PUREX technology is currently the only well-established process used at commercial scale. Other aqueous processes technologies are established but need to be industrially demonstrated. Pyroprocessing, however, is still at an early stage of development but appears as a promising technology to treat advanced spent fuels such as metal, nitride, etc.

**Minor actinides bearing fuels**

Over the last decade, minor actinides bearing fuels have been subject to numerous studies through different national and international R&D programmes. The Expert Group on Innovative Fuels (EGIF) has initiated a review of those programmes to assess the most recent advancements in the field in the form of a state-of-the-art report on innovative fuels [2] focusing, in particular, on fuel containing minor actinides (MA) to be irradiated in advanced nuclear systems. Different types of fuels were reviewed (metal, oxide, nitrides, dispersions fuels and special mechanical fuel forms) mainly for fast neutron reactors, examining different technical issues associated with their fabrication, characterisation, irradiation performance, design and safety criteria. The technical maturity of each fuel was also assessed. Metal and oxide fuels containing minor actinides have been widely investigated and irradiations tests and PIE have been performed. Additional results, however, are needed to gain further insights on He-gas release and fuel-cladding chemical interactions (FCCI). Nitrides fuels have numerous advantages for use in ADS compared to other types of fuels due to their higher density. However, more fundamental research is needed to determine their safety performance. On the other hand, dispersion fuels and special mechanical fuel forms (particles, namely VIPAC and SPHEREPAC) are new concepts. The first irradiation tests on inert matrix fuel containing MA started in the mid-90s. Three types of dispersion fuels have now been subjected to
safety tests. Some irradiation experiments have marked important milestones in the R&D of transmutation fuels. However, the examination of the behaviour and safety issues of transuranic bearing fuels are limited to laboratory scale (grams quantities of TRU) and irradiation tests have been limited to small samples or rodlets. More experimental data coupled to modelling are needed to gain better understanding of fuel behaviour under operating conditions.

Members of the expert group are currently working on a benchmark study on MA containing fuel performance codes focusing on minor actinide bearing oxide and metal fuels. Selected oxide and metal fuels irradiations test are being considered. Phase 1 of the benchmark is currently being finalised. Phase 2 will cover the study transient behaviour for the current range of innovative fuels.

Advanced fuel cycle scenarios

Through the development of computer codes, many countries are currently evaluating potential future nuclear energy, employing fuel cycle scenario studies as a mechanism to inform decisions on future reactor types, fuel types, and other fuel cycle facilities. To meet the demand from member countries, the NEA is conducting various projects related to fuel cycle transition scenarios in particular through the Expert Group on Advanced Fuel Cycle Scenarios (EGAFCS). The Expert Group performed a benchmark study on different selected scenarios to compare existing codes in term of capabilities, modelling and results [3,4].

A study to evaluate the effects of the uncertainties of input parameters on the outcomes of fuel cycle scenario studies is finalised and provides guidance on the importance of each input parameters. A reference fuel cycle scenario (transition from a fleet of UOX fuelled PWRs to a fleet of MOX fuelled FRs under a constant nuclear energy demand) was employed to ensure that all codes performed comparable analyses of the scenario, and to identify any differences attributable to the modelling approaches of the different codes. The results showed that the differences between codes can be considered as acceptable. The sensitivities of output metrics such as resource consumption, enrichment requirements, plutonium and spent fuel inventories, and reprocessing capacity to the various parameters were evaluated. Varied input parameters included: nuclear energy demand and energy demand profile, spent nuclear fuel cooling time, fresh fuel fabrication time, date and rate of introduction of the fast reactors, fuel burnup, fuel composition (including the minor actinide content of the fast reactor fuel in case of MA transmutation), fast reactor breeding ratio, reactor lifetime, and introduction date, capacity, and losses of the reprocessing plant. The results showed the relative impact of each parameter on each output metric.

Minor actinide management

Due to the difficulties in material handling, sample preparation and post-treatment, integral experiments for MA have been limited resulting in a lack of accuracy of nuclear data. In this context, an Expert Group on Integral Experiment for Minor Actinide Management (EGIEMAM) was established to review integral experiments for validating MA nuclear data, as well as to recommend additional integral experiments needed for validating MA nuclear data and to investigate the possibility to establish an international framework to facilitate integral experiments for MA management. The report “Review on existing integral data” [5] reviews the state of the art of nuclear data, relevant activities in international organisations, and existing integral data for MA. It concluded that further integral measurements are needed for $^{237}$Np, $^{241}$Am, $^{242m}$Am, $^{243}$Am, $^{243}$Cm, $^{244}$Cm, $^{244}$Cm.
Conclusions

Through the work conducted within its numerous expert groups, the NEA supports various activities related to partitioning and transmutation covering all areas of the fuel cycle (fuel fabrications, systems and infrastructures, reprocessing and waste disposal) and continues to help its member countries consolidating international knowledge. In addition, the information exchange meetings on actinide and fission product partitioning and transmutation give experts the opportunity to present, share and discuss state-of-the-art developments in the field of P&T.

References

Ongoing activities on partitioning and transmutation at the IAEA

Department of Nuclear Energy,
International Atomic Energy Agency (IAEA)

Abstract
The International Atomic Energy Agency (IAEA) actively supports its member states to improve their capabilities to develop and deploy advanced reactors and innovative related fuel cycle technologies with the aim to reduce the burden of high-level waste (HLW) and to enhance the sustainability of nuclear power.

To perform its scientific and technical activities, the IAEA defines and implements multiannual programmes considering the recommendations/requests expressed by its MSs through different means such as the yearly adopted resolutions, its Technical Working Groups (TWGs) and its Standing Advisory Groups (SAGs).

One of the major objectives of the IAEA activities is to foster and enhance the exchange of scientific and technical information among its MSs. In this regard and after the two successful IAEA Conferences on Fast Reactors and Related Fuel Cycles held in 2009, in Kyoto, and in 2013, in Paris, the next conference, to be held in June 2017, in Yekaterinburg, is under preparation. Three Technical Meetings were furthermore organised on “Challenges in Reprocessing Used Fast Reactor Fuels” in June 2015; on “Status of the IAEA Fast Reactor Knowledge Preservation Initiative” in December 2015 and on “Advanced Fuel Cycles for Waste Burden Minimisation” in July 2016.

In addition to these efforts, certain collaborative projects among MSs, organised within the frame of the International Project on Innovative Nuclear Reactors and Fuel Cycles (INPRO), carry out complex studies of regional and global multinational co-operation to enhance the sustainable development of nuclear energy systems including open and closed cycles. Specialised modelling tools and a technical sustainability metric have been developed to assist MSs to pursue studies in these areas and to enhance their national long-term planning and decision making. Principal among these are the “INPRO methodology” for sustainability assessment and MESSAGE-NES which allows sophisticated whole system analysis of nuclear reactors and fuel cycles with international trade relations.

Another mechanism the IAEA uses for collaborative work is the Co-ordinated Research Projects (CRPs). Currently IAEA has one CRP entitled “Accelerator-Driven System (ADS) Applications and the Use of Low-Enriched Uranium (LEU) in ADSs”. For their contributions to this CRP, some MSs are investigating the potential use of the LEU-driven system to perform transmutation of minor actinides. Another CRP on “Analysis of Options and Experimental Examination of Fuels for Water-Cooled Reactors with Increased Accident Tolerance” explores the potential to design and operate advanced fuel types that would be more resistant to fuel failure and hydrogen production to enhance the safety of current and advanced reactor fleets.
The IAEA also develops and establishes databases as the “Integrated Nuclear Fuel Cycle Information System (INFCIS)” that includes among others: updated and relevant information around the world on civilian nuclear fuel cycle facilities (NFCIS), a catalogue of post-irradiation examination facilities (PIE) and minor actinides properties database (MADB). An upgrade of the “advanced reactors information system (ARIS)” database has been recently released, which provides member states with comprehensive and balanced information about all advanced reactor designs and concepts, including transmutation systems.

Introduction

The International Atomic Energy Agency (IAEA) actively supports its member states to improve their capabilities to develop and deploy advanced reactors and innovative related fuel cycle technologies with the aim to reduce the burden of HLW and to enhance the sustainability of nuclear power.

To perform its scientific and technical activities, the IAEA defines and implements multiannual programmes considering the recommendations/requests expressed by its MSs through different means such as the yearly adopted resolutions, its Technical Working Groups (TWGs) and its Standing Advisory Groups (SAGs). For example the resolution 12 from the 59th General Conference (GC(59)/RES/12) encourages, among others, “the IAEA’s activities on advanced nuclear fuel cycle relating to fast reactors for potential waste burden minimisation” and “to pursue the investigation of new reactor and fuel cycle technologies with improved utilisation of natural resources and enhanced proliferation resistance, including those needed for the recycling of spent fuel and its use in advanced reactors under appropriate controls and for the long-term disposition of remaining waste materials, taking into account economic, safety and security factors” as well as “to consider jointly the improving of innovations in nuclear reactors, fuel cycles and institutional approaches, such as in the framework of the International Project on Innovative Nuclear Reactors and Fuel Cycles (INPRO)”.

The TWGs are groups of experts from MSs that mainly support the IAEA in the definition and implementation of its programmatic activities, and propose new initiatives and CRPs in the field. The TWG on Nuclear Fuel Cycle Options and Spent Fuel Management (TWG-NFCO) focuses its work on nuclear fuel cycle options with an emphasis on spent fuel management (storage and reprocessing and recycling), innovative fuel cycles and nuclear materials management. The TWG on Fuel Performance and Technology (TWG-FPT) focuses its work on status and trends in nuclear power reactor fuel performance and technology. It will cover nuclear core materials R&D, fuel design, manufacturing and utilisation, coolant chemistry, fuel performance analysis and quality assurance issues. TWG on fast reactors (TWG-FR) assists the IAEA in formulating an international vision applicable to fast spectrum transmutation systems, both critical and subcritical, for energy production and transmutation of long-lived radioactive nuclides.

One of the main objectives of the Agency is to foster and enhance the exchange of scientific and technical information among its MSs. The mechanisms that the Agency uses to achieve that are mainly the organisation of international conferences and workshops, the publication of technical documents and reports, the co-ordination of research activities through Co-ordinated Research Projects (CRPs) and the management of specific databases.
Main ongoing activities on partitioning and transmutation at the IAEA

Activities on advanced reactors and innovative related fuel cycles

After the two successful IAEA Conferences on Fast Reactors and Related Fuel Cycles held in 2009, in Kyoto [1], and in 2013, in Paris [2], the next conference, to be held in June 2017 in Yekaterinburg (Russian Federation), is under preparation. One of the main reasons for this proposed venue is that the sodium-cooled fast reactor BN-800 was connected to the grid in December 2015 at the Beloyarsk NPP, where the BN-600 reactor has been in operation since 1980.

The purpose of the conference is to provide a forum to exchange information on national and international programmes, and more generally new developments and experience, in the field of fast reactors and related fuel cycle technologies. A first goal is to identify and discuss strategic and technical options that may have been proposed by individual countries or companies. Another goal is to promote the development of fast reactors and related fuel cycle technologies in a safe, proliferation-resistant and cost-effective manner. A third goal is to identify gaps and key issues that need to be addressed in relation to the industrial deployment of fast reactors with a closed fuel cycle. A fourth goal is to engage young scientists and engineers in this field, in particular with regard to the development of innovative fast reactor concepts.

The collection of relevant information allows the IAEA to publish and update technical reports such as the one on “Status and Trends in Pyroprocessing of Spent Fuels”, currently under preparation, that includes an overview of national programmes and recent technical developments on pyrochemical processes and outlines the main issues for the industrial application of pyroprocessing technologies, especially for fast reactor fuels. This publication has been prepared using the information provided by MSs during two Technical Meetings held in 2011 and 2013 and it complements the IAEA publication on “Status and Developments in the Back-end of the Fast Reactor Fuel Cycle” [3] drafted with the perspective of a “nuclear renaissance” that prevailed at that time, before the Fukushima Daiichi accident.

In this regard two Technical Meetings were organised on “Challenges in Reprocessing Used Fast Reactor Fuels” in June 2015, and on “Advanced Fuel Cycles for Waste Burden Minimisation” in July 2016, with active participation of representatives from France, China, Hungary, India, Japan, Korea, Russia Federation and USA. Papers were submitted in the following areas: national approaches and present status of fast reactor fuel cycles; state of the art of advanced separation technologies; feasibility of transmutation of TRU nuclides, MAs and LLFPs; recovery of platinum group metals and useful fission products; and strategies and technologies aimed at waste burden minimisation. The drafting of a technical document on the different strategies and advanced technologies for waste burden minimisation in order to enhance nuclear power sustainability has been initiated. A third Technical Meeting was held in December 2015 on “Status of the IAEA Fast Reactor Knowledge Preservation Initiative (FRKP)” in response to the needs expressed by MSs and within a broader IAEA-wide effort in nuclear knowledge preservation. The main objectives of the FRKP were to halt the ongoing loss of information related to fast reactors and to collect, retrieve, preserve and make accessible already existing data and information on this topic. Recently, at the end of 2015, the first version of a new password-protected FRKP Portal has been released by the Agency, which provides information, data and documents in particular on ADS and transmutation systems.

International project on innovative nuclear reactors and fuel cycles (INPRO)

INPRO methodology is the IAEA tool for sustainability assessment of nuclear energy systems (NES). It covers all areas relevant to the sustainable development as it was defined in the United Nations
World Commission on Environment and Development Report entitled Our Common Future (often known as the Brundtland Report) [4]. These areas include economics, infrastructure, waste management, proliferation resistance, environmental impact from stressors and depletion of resources, and safety of reactors and fuel cycle facilities [5]. In every area a set of basic principles, user requirements and criteria has been formulated to define the objectives for sustainable development, actions which have to be taken to achieve these objectives and technical metrics for the assessment.

MSs applying the INPRO methodology to a specific planned national nuclear energy system may identify gaps in their programmes and define compensatory follow-up measures to close these gaps before they can create any potential problems.

The INPRO Task “Global Scenarios” develops, on the basis of scientific and technical analysis, nuclear energy scenarios that lead to a global vision of sustainable nuclear energy in the 21st century. The analysis framework was originally developed under the “GAINS” collaborative project [6]. Within this task the collaborative projects “Synergistic nuclear energy regional group interactions evaluated for sustainability” (SYNERGIES) and “Roadmaps for a Transition to Globally Sustainable Nuclear Energy Systems” (ROADMAPS) address the scenarios for enhancing global NES sustainability, specifically, through co-operation in nuclear fuel cycle among technology holder countries and technology user countries, including newcomer countries [7].

The INPRO collaborative project SYNERGIES performed multinational nuclear energy system scenario analyses to examine how the whole NES can achieve more than its parts. If one partner in a synergistic collaboration is achieving enhanced sustainability, then the other partner(s) may achieve similar enhancement without the requisite large national investments in technology R&D and related infrastructure development. Within SYNERGIES, 28 case studies were conducted by MSs to identify and evaluate mutually beneficial patterns of co-operation in the nuclear fuel cycle and the driving forces and impediments involved in such co-operation.

The INPRO collaborative project ROADMAPS, started in November 2014, has the objective of developing a structured approach for achieving globally sustainable nuclear energy, providing models for international co-operation and a template for documenting actions, scope of work, and time frames for specific collaborative efforts by particular stakeholders.

The INPRO Task “Innovations” designs and conducts collaborative projects on topics crucial to future nuclear energy sustainability and technological innovations. The utilisation of innovative fuels and fuel cycles can contribute directly to the development of sustainable NES, for technical and economic reasons as well as in terms of enhancing public acceptance. At present three collaborative projects in this field are carried out: “Nuclear fuel and fuel cycle analysis for future NES” (FANES); “Waste from innovative types of reactors and fuel cycles” (WIRAF) and “Study on co-operative approaches to the back-end of the nuclear fuel cycle”.

FANES is conducting feasibility analyses of selected advanced and innovative fuels and fuel cycles for different reactor systems in order to identify potential future NES using selected fuel and fuel cycles with the aim to enhance sustainability of NES.

WIRAF identifies any potential or known show-stopper issues for implementing advanced and innovative reactors and their fuel cycles that result from new or existing waste types.

Study on co-operative approaches to the back-end of the nuclear fuel cycle will examine how international co-operation in the nuclear fuel cycle back-end could be successfully built into national co-operation.
strategies and programmes, with the aim to enhance sustainability of NES. This includes all aspects of the back-end of the nuclear fuel cycle.

Co-ordinated research projects on research and technology development of ADS

Long-term nuclear fuel cycle can be enhanced by including accelerator-driven systems (ADSs), which have been developed in different countries for more than 40 years and offer new prospects and advantages for the transmutation of high-level radioactive waste.

In 2004, the IAEA published a report on energy generation and transmutation of nuclear waste with ADS [8]. The report demonstrated the state of the art of the ADS technology by reviewing the status and progress of national and international programmes in this field, as well as the most interesting projects which were conducted at that time in the MSs. Following the recommendation of the Technical Working Group on Fast Reactors (TWG-FR), the IAEA decided in 2006 to launch an initiative for updating the ADS status report, considering the relevant progress and advances made in many countries. In 2009, the IAEA published the TECDOC-1626 on “Advanced Reactor Technology Options for Utilisation and Transmutation of Actinides in Spent Nuclear Fuel” [9], which includes several studies concerning the use of ADS for long-lived radioactive waste utilisation and transmutation. In 2015, the IAEA published an updated “Status Report on ADS Research and Technology Development” [10], including the revision of the state of the art of this technology by discussing the different ADS concepts proposed worldwide since 2000, as well as the related R&D activities and demonstration initiatives carried out at national and international levels. The report contributes to establishing an international roadmap up to the full deployment of ADS, in the context of advanced nuclear fuel cycles, incorporating important achievements in the area, as well as identifying possible directions to stimulate and guide the national and international activities in the ADS technology.

In 2005, the IAEA started the CRP 32006 on “Analytical and Experimental Benchmark Analyses of Accelerator-Driven Systems” with the participation of 27 institutions from 18 MSs and two international organisations. Its specific objective was to improve the understanding of the coupling of an external neutron source with a multiplicative subcritical core. The participants performed computational and experimental benchmark analyses using integrated calculation schemes and simulation methods, addressing all major physics phenomena of the spallation source and its coupling with the subcritical core. To achieve the CRP’s objectives, experimental benchmarking was of paramount importance, with ADS concepts being the main thrust of the CRP. However, due to the fact that a series of experimental demonstration projects using non-spallation targets were also available, transmutation concepts based on subcritical cores driven by such neutron sources were also considered. During the CRP, the participants analysed eight benchmark problems, specifically: (i) YALINA-booster; (ii) spallation target parametric studies with experimental validation; (iii) spallation source efficiency and energy dependence; (iv) analytical and numerical benchmarking of methods and codes for ADS kinetics; (v) ADS concepts; (vi) subcritical experiments; (vii) photonuclear based transmutation benchmarks; and (viii) ADS performance. In addition, the scope of the CRP evolved to address other research areas that are relevant to external source driven nuclear systems, like nuclear data for both the proton target and the subcritical blanket. The results of this CRP will be published in a TECDOC, currently under publication.

As many of the ADS systems built in the past used high enriched uranium (HEU) for the subcritical assemblies, there has been a global effort to eliminate HEU from research facilities, and IAEA has assisted several MSs in converting the subcritical systems to low-enriched uranium (LEU). One concern on the part of facility owners and operators is the potential change in the performance
characteristics if a system is converted to LEU. In order to help alleviate this concern, IAEA has worked with MSs in analysing the ADS systems to facilitate understanding of the LEU system capabilities, and to develop new applications and missions for LEU ADS facilities. In order to more broadly disseminate the information of the experiences of working with the LEU systems, IAEA initiated the CRP T33002 in December 2015 on “Accelerator-Driven Subcritical Systems (ADS): Applications and use of Low-Enriched Uranium”, intended to follow upon the successful completion of CRP 32 006, and an International Research Collaboration, with 17 participants, devoted to gaining a better understanding of the physics of ADS with special emphasis on the investigation of various technical options for carrying out ADS research using LEU fuel.

The specific objectives of the CRP T33002 are to focus on the development of innovative applications for ADS facilities as well as on further development and validation of ADS technologies. Additionally, the expected work involves testing and validation of measurement and analytical techniques associated with the development of ADS technology and the use of LEU fuel.

Among the applications to be explored will be the use of ADS facilities to transmute nuclear waste, specifically, long-lived fission products and minor actinides (MA), and the development of robust techniques for online monitoring of ADS facilities. Attention will also be given to investigating the potential for use of ADS facilities as neutron sources for basic research, applied research and for the production of needed radioisotopes for medical and industrial applications.

In addition to gaining an understanding of both the static and dynamic properties of ADS systems, close collaborations between various research groups involved in previous research programmes have resulted in understanding and resolving a number of discrepancies initially observed for some of the results obtained during the course of the investigations. This has led to improved comprehension of the differences between various experimental methods used, between different employed nuclear data libraries, between the various nuclear system analysis codes used, and between the procedures employed in applying these resources in the context of ADS applications. Much of this analytical work will continue in this new CRP, and participating MSs have expressed their intentions to continue with exploring new applications and developing related analyses. Experiments are planned collaboratively at several facilities, such as the Kyoto University Critical Assembly (KUCA) and at Ukraine’s Kharkov Institute of Physics and Technology. Japan also plans to build a new Transmutation Experimental Facility (TEF) that will enable scale-up experiments to be performed.

The first meeting of the CRP T33002 was held in July 2016, with 24 participants from 14 MS. The outcome of the meeting was a matrix work plan, indicating which MS will contribute to the analytical and experimental work being performed at the ADS facilities. Additionally, each MS agreed to contribute to the work on one of the four main applications: Spent Fuel Transmutation; Thorium Fuel Cycle; Medical Isotope Production; and Material Irradiation.

The results of the CRP will be especially useful for MSs with new ADS facilities, and for those still using ADS with HEU fuel, helping them to convert their facilities to LEU to reduce nuclear proliferation risks. An IAEA publication will be prepared at the conclusion of the project.

Co-ordinated research projects on research and technology development of advanced fuels

Nuclear fuel is a highly complex material that has been subject to continuous development over the past 40 years and has reached a stage of development where it can be safely and reliably irradiated up to 65 GWD/ tU or more in commercial nuclear reactors. During this time there have been many improvements to the original designs and materials used. However, the basic design of uranium-
oxide fuel pellets clad with zirconium alloy tubing has remained the fuel of choice for the vast majority of commercial nuclear power plants.

Severe accidents, such as those at Three Mile Island and Fukushima Daiichi, have shown that under such extreme conditions, nuclear fuel will fail and the high temperature reactions between zirconium alloys and water will lead to the generation of hydrogen with the potential for explosions to occur, damaging the plant further.

Recognising that the current fuel designs are vulnerable to severe accident conditions, there is renewed interest in alternative fuel designs that would be more resistant to fuel failure and hydrogen production. Such new fuel designs would need to be compatible with existing fuel and reactor systems if they are to be utilised in the current reactor fleet and in current new build designs, but there is also the possibility of new designs for new reactor systems.

Globally, there is a great deal of experience with the performance of reactor fuel in off-normal conditions. Theoretical studies and experiments have been performed and there have been excursions from normal operating conditions in a few power reactors. During such an excursion, the difference between an incident of limited or no consequence and a severe accident, such as the one at Fukushima, depends on the conditions in the reactor and the performance of the fuel under those conditions. The CRP T12030 “Analysis of Options and Experimental Examination of Fuels for Water-Cooled Reactors with Increased Accident Tolerance” (ACTOF) with 14 participants from 11 MSs explores the potential to design and operate advanced fuel types that are intended to be more tolerant of severe accident conditions while retaining the capability of current fuel designs for safe operation under normal operation and anticipated transient conditions. The main objective of this CRP is the acquisition of data through experiments on new fuel types and cladding materials and to support modelling of new fuel designs with advanced cladding or fuel. The first RCM was held in November 2015.

The main expected outcomes of this CRP are the provision of information to MSs to support decision making on the available choices to improve the safety of nuclear power plants under severe accident conditions and the provision of data, analyses and advanced techniques to understand and predict the behaviour of the components and the integral performance of accident tolerant fuel designs under normal and transient conditions. Well checked experimental data on the behaviour of candidate materials for accident tolerant fuel designs, new models of materials behaviour in fuel modelling codes and results of computer modelling of advanced fuel types under normal and accident conditions can be expected as main results.

**IAEA databases related to partitioning and transmutation (P&T) activities**

The IAEA also develops and establishes databases as the “Integrated Nuclear Fuel Cycle Information System (iNFCIS)” [11] that was designed by the IAEA’s Nuclear Fuel Cycle and Materials Section (NFCMS) as a comprehensive resource for technical and statistical information about nuclear fuel cycle activities worldwide, as reported to the IAEA. The system includes among others:

- Nuclear Fuel Cycle Information System (NF CIS) that covers civilian nuclear fuel cycle facilities around the world. It contains information on operational and non-operational, planned, and cancelled facilities. All stages of nuclear fuel cycle activities are covered, starting from uranium ore production to spent fuel storage facilities.
• Post-Irradiation Examination Facilities Database (PIE) which is derived from a catalogue of such facilities worldwide. It includes a complete survey of the main characteristics of hot cells and their PIE capabilities.

• Minor Actinide Property Database (MADB) which is a bibliographic database on physico-chemical properties of selected minor actinide compounds and alloys. The materials and properties are selected based on their importance in the advanced nuclear fuel cycle options.

Advanced reactor information system (ARIS) [12] is an online database designed and maintained by the IAEA’s Nuclear Power Technology Development Section (NPTDS) since 2009. ARIS is a user-friendly, easy-to-navigate database that offers technical design descriptions for advanced reactors that are under design, in construction or in operation. It includes reactors of all sizes and types, from evolutionary nuclear power plant designs for near-term deployment to the latest innovative reactor concepts that are under development. ARIS enables users to easily get an overview of the current reactor technologies being developed and deployed by giving people access to the designers’ design descriptions. The latest upgrade of the ARIS database was released in the middle of 2016, which provides IAEA Member States with comprehensive and balanced information about all advanced reactor designs and concepts, including transmutation systems.

References


[5] Please see: https://www.iaea.org/INPRO/inpro_methodology/index.html, for links to all current manuals of the INPRO methodology


US Department of Energy, fuel cycle technologies R&D outlook

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Abstract

In order to maintain the US domestic nuclear capability and its scientific technical leadership, the Department of Energy, Office of Nuclear Energy (DOE-NE) invests in various R&D programmes to identify and resolve technical challenges related to the sustainability of the nuclear fuel cycle. Sustainable fuel cycles are those that improve uranium resource utilisation, maximise energy generation, minimise waste generation, improve safety and limit proliferation risk. DOE-NE chartered a study on the evaluation and screening (E&S) of nuclear fuel cycle options, to provide information about the potential benefits and challenges of nuclear fuel cycle options and to identify a relatively small number of promising fuel cycle options with the potential for achieving substantial improvements compared to the current nuclear fuel cycle in the United States. The identification of these promising fuel cycles helps in focusing and strengthening the US. R&D investment needed to support the set of promising fuel cycle system options and nuclear material management approaches. Under the auspices of DOE-NE, a number of partitioning technologies (aqueous recycling and electrochemical recycling) and advanced glass, ceramic-glass and ceramic waste forms are investigated for possible future implementation. Along with the finding of the E&S study will be presented a description of the major US research efforts on cost-effective used nuclear fuel recycling and waste management technologies. Discussion on transmutation will be illustrated by examining different fast reactors that possess different technical choices, such as advanced transmutation fuel, transmutation mode.
An update on nuclear energy and P&T programmes within the United Kingdom

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Abstract

Nuclear Energy is a mature, reliable low-carbon technology with a secure and abundant fuel source and is an essential contributor to the energy mix. It has provided around 20% of the UK energy mix for decades though initially a Magnox and latterly an AGR fleet of NPPs. From the 1960s to present day, a partially closed fuel cycle have been operated in the UK, where the used fuel from the Magnox and AGR reactors were reprocessed at facilities at Sellafield to recover valuable uranium and plutonium fuel. With much of the existing reactor fleet coming off line in the next decade, a decision has been made to refresh current nuclear energy plants to at least replace the existing nuclear fleet (16 GWe per annum). This new fleet consisting of EPR, ABWR and AP1000 reactors will not follow the closed fuel cycle of the past, but instead will follow a once-through cycle, where used fuel will be stored for a period of time once out of the reactor prior to final disposal in a geological disposal facility.

However, current future energy scenarios also recognise that providing 16 GWe of nuclear energy through a nuclear programme may not be enough, and hence expansion scenarios ranging from 16 to 75 GWe are being considered. For any such expansion programme, the type of fuel cycle required to support this will depend on a number of factors and is not limited to energy capacity, repository capacity, fuel requirements or public opinion. The implications for any future fuel cycle will become clearer over time but until then, there is a need to keep future fuel cycle options open.

Materials recycling technology has been pivotal to UK nuclear programmes since the early days of nuclear power through the development of new flow sheets for reprocessing plant, the separation and purification of specific radio nuclides and the enhancement of existing reprocessing plant flow sheets for enhanced economic and environmental benefits. With the move to a once through fuel cycle and the need to keep future fuel cycle options open, programmes involving partitioning R&D become even more important to ensure skills maintenance for future decisions. These programmes will need to be leveraged through other international partners to ensure maximum financial leverage, the sharing of best practice and enabling knowledge transfer and development of expertise.

The current status of nuclear energy in the UK, the challenges faced and the resulting implications for P&T will be explored together with examples of current P&T programmes.
Development of safe and responsible concepts for advanced nuclear fuel cycles in the European Union

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Abstract

The Horizon 2020 Research Framework Programme aims at reinforcing outcomes under the three main priorities: excellent science, industrial leadership and societal challenges. The EURATOM part puts a major focus on nuclear research and training activities to improve nuclear safety, security and radiation protection, notably to contribute to the long-term decarbonisation of the energy system in a safe, efficient and secure way.

In the area of nuclear safety, both co-funded research programmes (indirect actions) and direct research activities address key science and technology challenges to maintain and improve the safety level and competitiveness of fission technologies, covering existing (Generation II), evolutionary (Generation III) and advanced/sustainable (Generation IV) reactor concepts and related fuel cycles. This effort includes implementing long-term waste management solutions and closing the nuclear fuel cycle. Attention is particularly devoted to safety aspects of emerging trends and concepts with improved safety, reduced wastes and improved resistance to proliferation, including also innovation perspectives for energy and other applications.

The research is addressing the long-term safe operation of both Western and Russian type of nuclear power plants. To this end, advanced and refined safety assessment methodologies and corresponding analytical tools have to be extended and validated and include activities with regard to the safety of nuclear fuel concentrate on prevention and mitigation of the consequences of hypothetical accidents.

The H2020 EURATOM programme includes a responsible treatment of all kinds of radioactive wastes. Also here particular attention is being paid to the long-term safety of storage, the minimising of volume, chemical complexity and toxicity, the possibilities of recycling and material valorisation and the facilitating of handling and management. This action should be undertaken in close co-operation with the operational stakeholders concerned to facilitate the early uptake of the results in the development of disposal solutions. Recently, a Joint Programming on Radioactive Waste Disposal (JOPRAD) was launched to improve the co-ordination and to develop synergies of R&D programmes between Waste Management Organisations (WMOs), Technical Support Organisations (TSOs) and nationally funded Research Entities.
Review of research activities on partitionning and transmutation in Japan

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Abstract

The Government of Japan periodically formulates its basic energy plan in accordance with an article in the Basic Act on Energy Policy that entered into force in 2002. The latest version, called the Strategic Energy Plan, was issued in April, 2014. The largest change from the previous versions must be the accident at the Fukushima Daiichi Nuclear Power Plant. In spite of the accident, the plan defines nuclear power as an important baseload power source and as a low-carbon and quasi-domestic energy source, contributing to the stability of the energy supply-demand structure, on the major premise of ensuring its safety. The plan puts emphasis on the importance of activities to resolve the challenge of managing and disposing of spent fuel, as well as the Fukushima Daiichi restoration, safety operation of nuclear plants, and public acceptance. As for P&T technology, the government will promote technology development on volume reduction and mitigation of degree of harmfulness of radioactive waste. Specifically, development of technologies for decreasing the radiation dose remaining in radioactive waste over a long period of time and enhancing the safety of processing and disposal of radioactive waste, including nuclear transmutation technology using fast reactors and accelerators, will be promoted utilising global networks for co-operation.

Based on this new Strategic Energy Plan, research and development (R&D) on P&T are being accelerated in Japan. The Japan Atomic Energy Agency (JAEA) has been continuously implementing R&D on P&T technology to reduce the burden of the back-end of the nuclear fuel cycle. The R&D on P&T in JAEA are basing on two kinds of concepts: one is the homogeneous recycling of minor actinide (MA) in fast reactors and the other is the dedicated MA transmutation, so-called “double-strata” strategy, using an accelerator-driven system (ADS). In this work, recent R&D activities based on these policies are briefly shown.
Fuel cycle activities in Belgium: MYRRHA project status and new implementation plan

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Abstract
With the development of the MYRRHA accelerator-driven system (ADS), SCK•CEN has come to the forefront of heavy liquid metal (HLM) nuclear technology worldwide. MYRRHA is a flexible fast spectrum pool-type research irradiation facility cooled by lead-bismuth eutectic (LBE), and was identified by SNTP (www.snetp.eu) as the European Technology Pilot Plant for the Lead-cooled Fast Reactor. Since the FP5 EURATOM framework, MYRRHA is also serving as the backbone of the P&T strategy of the European Commission based on the “4 building Blocks at Engineering level” and fostering the R&D activities in EU related to the ADS and the associated HLM technology developments.

Since 1998 SCK•CEN has been developing the MYRRHA project as an accelerator-driven system (ADS) based on lead-bismuth eutectic as a coolant of the reactor and a material for its spallation target. The nominal design power of the MYRRHA reactor is 100 MWth. It is driven in subcritical mode ($k_{eff} = 0.95$) by a high power proton accelerator based on LINAC technology delivering a proton beam in CW mode of 600 MeV proton energy and 4 mA intensity. The choice of LINAC technology is dictated by the unprecedented reliability level required by ADS application. In the MYRRHA requirements the proton beam delivery should be guaranteed with a maximum number of beam trips lasting more than 3 seconds limited to 10 for a period of 3 months corresponding to the operating cycle of the MYRRHA facility. Since 2005, SCK•CEN and Belgium opened the MYRRHA project for the EU Member States for participation as well as to the major nuclear power countries for participation in the development of MYRRHA and further on during the construction and operation periods.

In this paper we present a brief review on the ongoing activities in Belgium related to the fuel cycle and will give the present status of the MYRRHA project in terms of the design of its subcritical reactor and high-level power accelerator, licensing, and perspective of implementation. The new implementation strategy of the MYRRHA project as endorsed by Belgian authorities will be developed from the end of 2015 on.
Development activities for recycling spent nuclear fuel in Korea

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Abstract
Korea’s extensive and growing fleet of conventional light water reactors faces a looming concern. At present, roughly 700 metric tonnes a year of spent nuclear fuel is being accumulated at the sites of the 24 operating nuclear units. This has already amounted to 14 000 tonnes of material being stored, which has filled about 75% of the present storage capacity. It is estimated that four of the operating sites will reach their capacity to store spent fuel in the time frame of 2024 through 2028. Clearly, Korea needs to solve the problems of the buildup. Korea is considering, as one of technical answers, burning the long-lived and radioactive materials in a sodium-cooled fast reactor (SFR). Recycling in SFR combined with pyroprocess technologies can reduce the size and the management period of the final repository by about 1/40 and several hundreds, respectively. To realise the recycling of spent nuclear fuel, Korea is developing prototype of Generation IV sodium-cooled fast reactor (PGSFR). The licensing process for the PGSFR will start at the end of 2017 and the construction will take place between 2023 and 2028. The main goal of the PGSFR is to prove the effectiveness of the recycling scheme on the spent nuclear fuel management. The recycling scheme in Korea is planned to be based on the combination of SFR and pyroprocess technology.
Plenary Session II: Fuel Cycle Strategies and Transition Scenarios

Chair: E. Touron and D. Matthers
Fuel cycle analysis of a long-term nuclear power scenario for the United Kingdom

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Abstract

The UK has operated a partially closed fuel cycle for decades but a fully open fuel cycle will be implemented beyond 2020 with the introduction of a 16GWe new build programme. However, further nuclear expansion scenarios are being considered of around 40 GWe to help curb greenhouse gas emissions, and to tackle energy security [1].

This study evaluates a viable 40 GWe UK nuclear scenario and considers the extent to which key drivers can impact a future UK nuclear energy strategy, including the ability to sustain a relatively high nuclear capacity, accumulation of spent fuel at reactor sites and the capacity of future geological disposal facilities (GDF) host sites.

This work follows from previous studies [2], [3] which examined UK fuel cycles involving the transition to a closed fuel cycle and deployment of a fast reactor fleet by the end of this century. The revised scenario now assumes that fast reactors (FRs) are required later in the scenario to replace the LWR fleet as reactor units come to the end of their operation and additionally to manage spent fuel waste volumes and to reduce repository footprint.

The ORION computer code [4] has been used to model the UK scenario and a new modelling tool called BADGER to assess the physical size of the repository required to dispose of all intermediate and high-level waste (HLW) while ensuring temperature limits imposed by the engineered barriers and surrounding geology are adhered to. Results were obtained to show:

- a viable energy scenario transition to a fast reactor fleet;
- spent fuel volumes;
- geological disposal facility footprint.

Introduction

Within the UK a closed fuel cycle has been in operation for decades but with the introduction of a new build programme to create 16 GWe capacity using light water reactor (LWR) technology and with the closure of the UKs reprocessing industry, an open fuel cycle will be implemented beyond 2020 [5].

However, it is expected that a 16 GWe nuclear capacity will not be enough for the UK to curb greenhouse gas emissions through increased demand for low-carbon electricity, and tackle energy security. As such, further nuclear expansion scenarios are being considered of around 40 GWe by 2050.
This study evaluates a viable 40 GWe UK nuclear scenario, evaluating the extent to which key drivers can impact a future UK nuclear energy strategy, including the ability to sustain a relatively high nuclear capacity, the accumulation of spent fuel at reactor sites and the capacity of future GDF host sites.

This work follows from previous studies presented in 2013 [6] and 2015 [7] which examined UK fuel cycles involving the transition to a closed fuel cycle and deployment of a fast reactor fleet by the end of this century.

The revised scenario (Figure 1) now assumes that fast reactors (FRs) are required later in the scenario, to replace the LWR fleet as reactor units come to the end of their operation and to additionally manage spent fuel waste volumes and to reduce repository footprint.

**Figure 1**: Scenario of 40GWe installed nuclear capacity

The ORION computer code [8] has been used to model the UK scenario. This new study has also used physics methods originally developed by SKB (Swedish Nuclear Fuel and Waste Management Company) to develop a new modelling tool called BADGER to assess the physical size of the repository required to dispose of all intermediate and HLW while ensuring temperature limits imposed by the engineered barriers and surrounding geology are adhered to.

Results were obtained to show the following:

- A viable energy scenario transition to a fast reactor fleet.
- Spent fuel volumes.
- Geological disposal facility footprint.
In addition to showing that it is possible to transition over to a fast reactor fuel cycle, through recycling Pu and U into the LWR fleet as mixed-oxide (MOX) fuel, fast reactor spent fuel cooling times and LWR-MOX utilisation rates were varied in order to show what would be required from back-end technologies to ensure that a 40 GW(e) nuclear capacity is maintained. Figure 2 illustrates the contribution of LWR and sodium-cooled fast reactor (SFR) technology to generating capacity and compares the impact of LWR-MOX utilisation rates.

Figure 2: Impact of spent fuel cooling times and LWR-MOX utilisation rates on electricity generation

An interim spent fuel storage capacity of 80 000 tHM is calculated for a 40GWe open fuel cycle with direct disposal (no recycle). The accumulation of spent fuel can be managed by disposing material in a repository (in this case after 100 years of spent fuel storage and cooling) or alternatively, by reprocessing and recycling the fissile material. Realistically, recycle can only be performed once through a LWR fleet due to the significant reduction in plutonium quality. In the ‘once through’ recycle case, spent fuel accumulation rates can be reduced by a factor of 10, compared to an open fuel cycle. Around 15 000 tHM of spent fuel is accumulated, mostly MOX, albeit at around 10 times lower rate. To further reduce the accumulation of spent fuel volumes, fast reactors are required to ‘close the fuel cycle’ enabling indefinite plutonium and minor actinide recycle. Figure 3 compares the impact of open versus closed fuel cycles on spent fuel accumulations. The various scenarios are outlined in Table 1.
To calculate a repository footprint, waste packages need to be arranged such that the surrounding bentonite backfill remains below 100°C and the gap between neighbouring waste canisters must be sufficient to allow heat to dissipate into the host rock. Calculating the evolution of
temperature in a repository is complex and requires detailed computational models. The BADGER code was developed to estimate the peak temperature reached in the bentonite backfill.

ORION estimates the total decay heat produced within the repository over time and the number of waste canisters which is then used by BADGER. BADGER then generates decay heat profiles for the canisters and the background temperature over time. Temperature distributions over time across canisters, gaps, bentonite backfill and the host rock are then used to predict the inner bentonite temperature for the central waste canister location. Decay heat calculations are illustrated in Figure 4 for various scenarios and repository footprints are predicted in Table 1.

Table 1: Fuel cycle scenario calculations of repository footprints

<table>
<thead>
<tr>
<th>Fuel cycle scenarios</th>
<th>VHLW Waste Containers</th>
<th>Spent Fuel Waste Containers</th>
<th>Repository footprint (relative to 1st Scenario)</th>
</tr>
</thead>
<tbody>
<tr>
<td>5b Open fuel cycles</td>
<td>1 x 10 GW(e) LWR fleet operating an open fuel cycle and recycling legacy Pu as MOX</td>
<td>4000</td>
<td>12000</td>
</tr>
<tr>
<td>6b Open 40 GW(e) UK fuel cycle, with recycle of Pu from legacy Pu stockpiles in LWR fuel, operating for only 2 successive future generations</td>
<td>4000</td>
<td>60000</td>
<td>480%</td>
</tr>
<tr>
<td>1b Open 40 GW(e) UK fuel cycle, with recycle of Pu from LWR-UO2 spent fuel in LWR fleet, operating for only 3 successive future generations</td>
<td>51000</td>
<td>7000</td>
<td>560%</td>
</tr>
<tr>
<td>2b Closed 40 GW(e) UK fuel cycle, 3 years min SFR spent fuel cooling time, Rn, Am recycle through 1st and 2nd SFR fleets</td>
<td>99000</td>
<td>0</td>
<td>80%</td>
</tr>
<tr>
<td>3b Culminating in a 40 GW(e) SFR-MOX fleet</td>
<td>Rn, Am recycle through 2nd SFR fleet only</td>
<td>91000</td>
<td>0</td>
</tr>
<tr>
<td>4b A total of 3 successive future generations are modelled</td>
<td>No Rn, Am recycle</td>
<td>81000</td>
<td>0</td>
</tr>
<tr>
<td>5b 5 years min SFR spent fuel cooling time, Rn, Am recycle through 1st and 2nd SFR fleets</td>
<td>77000</td>
<td>0</td>
<td>80%</td>
</tr>
<tr>
<td>6b Rn, Am recycle through 2nd SFR fleet only</td>
<td>71000</td>
<td>0</td>
<td>150%</td>
</tr>
<tr>
<td>7b Rn, Am recycle through 2nd SFR fleet only</td>
<td>65000</td>
<td>0</td>
<td>250%</td>
</tr>
</tbody>
</table>

The results from the repository model show that VHLW (vitrified high-level waste) and SF waste disposal zone footprints can be vastly reduced through transition to a fast reactor fleet if Am is recycled, this zone is approximately 6 times lower than an equivalent open fuel cycle. If Am is not recycled and instead vitrified and sent to a repository, the reduction in repository footprint for HLW is only halved. Noting that in a Pu and U-only recycle scenario the vast majority of decay heat originates from Am in the waste.
Conclusions

The scenarios described in this report have been performed to help understand the potential benefits of a fast reactor closed fuel cycle based on the most mature technologies currently available (i.e. MOX fuel in an SFR).

Results show how, for a UK 40GWe fuel cycle scenario:

- A transition to the fast reactor fleet is possible over the timescales considered
- Spent fuel volumes can be managed
- Geological disposal facility optimisation can be achieved

The number of fast reactors that could be phased in would be limited by:

- The quantity and quality of plutonium available, and
- The time it takes for spent SFR MOX fuel to be cooled, reprocessed, refabricated and reloaded back into the core.

In the scenario modelled, a transition to fast reactors is assumed to occur following extensive utilisation of LWR Pu in LWR-MOX fuel. A complete transition to a closed fuel cycle is only possible if spent SFR fuel is cooled for a maximum of 3 years (+ 2 years for reprocessing and refabrication). If fuel is cooled for 5 years (+ 2 years for reprocessing and refabrication), then LWR-MOX fuel fabrication needs to be limited to ensure the average quality (and quantity) of Pu available for the fast reactor fleet is high enough to maintain the same nuclear generation capacity.

Recycling LWR-UO$_2$ Pu as MOX through an LWR fleet reduces spent fuel accumulation rates by a factor of 10. Since plutonium can only be recycled once through an LWR, spent fuel volumes will ultimately increase. By recycling through an LWR fleet, it is possible to concentrate the plutonium inventory from LWR-UO$_2$ fuel into a smaller spent fuel volume. Therefore, reprocessing LWR-UO$_2$ and recycling the Pu as LWR-MOX is a convenient method of minimising spent fuel volumes in anticipation of a MOX fuelled fast reactor fleet commencing sometime in the future.

Denaturing of the plutonium in LWR-MOX fuel could lead to further problems regarding FR fuel fabrication, in-reactor fuel performance and subsequent spent fuel handling. Ultimately, spent fuel accumulation can only be stabilised once a transition to a fast reactor fleet has been made.

A new approach was developed to assess the GDF HLW/SF disposal zone footprint for the scenarios considered. The results show that the HLW and SF waste disposal zone footprint can be vastly reduced if the UK eventually transitions to a fast reactor fleet.

If americium is recycled through the fast reactor fleet, the disposal zone footprint is approximately six times lower than an equivalent open fuel cycle. If americium is not recycled and instead vitrified and sent to repository, the reduction in repository footprint for HLW is only halved.

Further considerations

The highest Am content will be in SFR fuel fabricated using Pu(+Am) from reprocessing LWR-MOX fuel. Once the Pu(+Am) has been recycled several times, the Pu quality will gradually increase and the Am reduce as a result of successive transmutation. Considering the high radiation and decay heat associated with Pu from LWR-MOX fuel, SFR fuel fabrication and fresh fuel handling will be challenging even without Am recycle. Likewise, the enhanced production of helium in SFRs will
potentially lead to fuel performance issues and the elevated levels of minor actinides (e.g. Cm) produced will potentially lead to back-end issues (storage, transport and reprocessing).

Further work would be required to assess the practicalities of recycling of LWR-MOX Pu(+Am). The inventories from this assessment could be used to assess the technologies needed to; operate reactors, handle spent fuel, reprocess spent fuel, immobilise waste streams from reprocessing, fabricate fuel, and dispose of the waste in a repository.

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References


Analysis of fast reactor deployment scenarios in the French nuclear power fleet

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Abstract

Fast reactors have a number of advantages when it comes to radioactive material management while being complementary with the current French thermal reactor fleet, which includes pressurised water reactors (PWR). Fast reactors can use the plutonium produced by water reactors (or by themselves) indefinitely, they increase the energy that can be extracted from a given mass of natural uranium by a factor of almost 100. Fast reactors are able to operate without natural uranium and provide a practically inexhaustible source of energy independence for a fast reactor fleet. Fast neutron spectrum also opens the door to minor actinide transmutation and thus to the possibility of reducing the inventory of these radionuclides in waste, while reducing the size of the deep geological repository needed for the disposal of ultimate waste.

Fast reactors therefore appear as a key link in the closed-cycle strategy by making it possible to fully exploit the materials contained in spent fuel.

The report submitted to the French government in December 2012, which assessed various options for sustainably managing radioactive materials and waste, highlighted the relevance of incorporating fast reactors into the French fleet. This report recommended “focusing on a progressive approach with scenarios that set out to first deploy a limited number of fast reactors in synergy with the water reactors comprising the French fleet”. It also stated that “studies of industrial scenarios would be led with AREVA and EDF to refine the approach”. CEA, AREVA and EDF have therefore started studying different options for introducing fast reactors into the French fleet based on a realistic approach which takes into account industrial assumptions and constraints. This has shed light on the subsequent possible benefits and conditions required for success.

Rather than imagining the large-scale replacement of PWRs by fast reactors in a short time span (the assumption of some past studies), it was decided to study a progressive deployment scenario taking into account existing materials and facilities. This solution appears to be better suited to the dynamics of technical progress in the field, while providing greater flexibility to adapt to societal changes. The path for this scenario is marked by successive milestones (stages), with each one corresponding to an increased deployment of fast reactors with their own objective of increasing ambition.
These various stages have been characterised and benchmarked against numerous criteria, particularly material and waste inventories and natural uranium consumption. This paper presents the main outcomes of this study.
Analysis of fuel cycle strategies and United States transition scenarios

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Abstract

The nuclear fuel cycle evaluation and screening (E&S) study that was completed in October 2014 enabled the identification of four fuel cycle groups that are considered most promising based on a set of nine evaluation criteria: (a) six benefit criteria of Nuclear Waste Management, Proliferation Risk, Nuclear Material Security Risk, Safety, Environmental Impact, Resource Utilisation, and (b) three challenge criteria of Development and Deployment Risk, Institutional Issues, Financial Risk and Economics. The E&S study was conducted at a level of analysis that is “technology neutral,” that is, without consideration of specific technologies, but using the fundamental physics characteristics of each part of the fuel cycle. The study focused on the fuel cycle performance benefits at the fuel cycle equilibrium state, with only limited consideration of transition and deployment impacts. Common characteristics of the four most promising fuel cycle options include continuous recycle of all U/Pu or U/TRU, the use of fast spectrum reactors, and no use of uranium enrichment once fuel cycle equilibrium has been established. The high-level wastes (HLWs) are mainly from processing of irradiated fuel, and there would be no disposal of any spent fuel.

Building on the findings of the E&S study, additional studies have been conducted in the last two years following the last information exchange meeting, the 13\textsuperscript{th} IEMPT, held in Seoul, Korea in 2014. Insights are presented from the recent studies on the benefits and challenges of recycling minor actinides, and transition considerations to some of the most promising fuel cycle options.

Introduction

The US Department of Energy, Office of Nuclear Energy (DOE-NE), chartered a study of nuclear fuel cycle options, called the nuclear fuel cycle evaluation and screening (E&S) study, which was completed in October, 2014 [1]. DOE-NE specified nine fuel cycle evaluation criteria representing broadly defined economic, environmental, safety, non-proliferation, security and sustainability goals to allow identification of promising fuel cycle options that have the potential for achieving substantial improvements as compared to the current nuclear fuel cycle in the United States. Six criteria addressed the potential for benefits with respect to fuel cycle performance, while the other three criteria addressed potential challenges for fuel cycle development and deployment.

DOE-NE also specified that the set of fuel cycle options to be evaluated in the E&S study was to be as comprehensive as possible with respect to potential fuel cycle performance. An approach based on the fundamental physics-based characteristics of nuclear fuel cycles rather than on fuel cycle examples using specific implementing technologies was developed to enable the creation of such a comprehensive set, which represented the performance of all once through and recycle fuel
cycles, and included thermal and fast reactors, critical and subcritical systems, and uranium and/or thorium for fuel along with other distinguishing fuel cycle features. The entire set of possible fuel cycle options was reduced to a set of 40 evaluation groups (EGs) by collecting fuel cycles with similar characteristics and performance into each EG. The E&S study considered the complete nuclear energy system from mining to disposal, and provided information about the potential benefits and challenges of nuclear fuel cycles that could be used to strengthen the basis of, and provide guidance for, the R&D activities undertaken by DOE-NE. The performance of each EG was assessed by using metrics developed for each evaluation criterion.

The process used to identify those fuel cycle options with potential for improvement included varying the relative importance of the six benefit criteria when considering multiple criteria simultaneously, resulting in promising fuel cycles that were relatively insensitive to viewpoints on the importance of the benefit criteria. The E&S study identified four groups of fuel cycles as being “most promising” for R&D, listed in Table 1 by their evaluation group (EG) number, based on the potential for performance improvement with respect to the benefit criteria provided by DOE-NE for the E&S study. The descriptions in Table 1 are only indicative of the fuel cycles included in each group, and the term "new natural uranium fuel" indicates that enrichment is not needed for either new uranium fuel resources or for the recycled uranium.

Table 1: Most promising fuel cycle groups from the evaluation and screening study

<table>
<thead>
<tr>
<th>Evaluation group</th>
<th>Most Promising Fuel Cycle Groups</th>
</tr>
</thead>
<tbody>
<tr>
<td>EG23</td>
<td>Continuous recycle of U/Pu with new natural-U fuel in fast critical reactors</td>
</tr>
<tr>
<td>EG24</td>
<td>Continuous recycle of U/TRU with new natural-U fuel in fast critical reactors</td>
</tr>
<tr>
<td>EG29</td>
<td>Continuous recycle of U/Pu with new natural-U fuel in both fast and thermal critical reactors</td>
</tr>
<tr>
<td>EG30</td>
<td>Continuous recycle of U/TRU with new natural-U fuel in both fast and thermal critical reactors</td>
</tr>
</tbody>
</table>

Note: U = uranium; Pu = plutonium; TRU = transuranic elements, i.e. atomic number higher than uranium (neptunium, plutonium, americium, curium, etc.); the term "U/Pu" indicates that uranium and Pu are recycled together, similarly the term "U/TRU" indicates that uranium and TRU are recycled together.

These four most promising evaluation groups share the following characteristics:

- Continuous recycle of actinides (U/Pu or U/TRU)
- Fast neutron-spectrum critical reactors
- High internal conversion (of fertile to fissile)
- No uranium enrichment required once steady-state conditions are established

The use of fast spectrum irradiation is beneficial compared to the thermal spectrum irradiation currently used in the US fuel cycle because of the more favourable fission/absorption ratio for isotopes of many of the higher actinide elements. The fast spectrum also has more favourable internal conversion of fertile $^{238}$U to $^{239}$Pu, facilitating the use of uranium without enrichment to sustain the fuel cycle.

The results of the E&S study have been used to identify the subsequent studies conducted in the past two years. This paper summarises the results and insights from several of these studies.

Minor actinide recycle – Benefits and challenges

In considering the four groups of most promising fuel cycles, it was observed that both U/Pu and U/TRU recycle in fast reactors appeared to provide comparable potential benefits for the criteria and
metrics used in the study, as shown by some examples in Table 2, where the performance of the current once-through US fuel cycle using thermal reactors (EG01) is compared to the performance of two of the most promising fuel cycles. However, these most promising groups exhibited differences with respect to the three challenge criteria, with fuel cycles in EG23 and EG29 estimated to have lower development and deployment challenges than those in EG24 and EG30 due to the use of U/Pu recycle as compared to U/TRU recycle.

### Table 2: Fuel cycle performance for selected evaluation metrics

<table>
<thead>
<tr>
<th>Evaluation Metric</th>
<th>EG01</th>
<th>EG23 (U/Pu)</th>
<th>EG24 (U/TRU)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Mass of SNF+HLW disposed per energy generated, t/GWe-yr</td>
<td>12 to &lt; 36</td>
<td>&lt; 1.65</td>
<td>&lt; 1.65</td>
</tr>
<tr>
<td>Activity of SNF+HLW (@100 years) per energy generated, MCi/GWe-yr</td>
<td>1.05 to &lt; 1.60</td>
<td>0.67 to &lt; 1.05</td>
<td>0.67 to &lt; 1.05</td>
</tr>
<tr>
<td>Activity of SNF+HLW (@100,000 years) per energy generated, kCi/GWe-yr</td>
<td>1.0 x 10^{-3} to &lt; 2.3 x 10^{-3}</td>
<td>5.0 x 10^{-4} to &lt; 1.0 x 10^{-3}</td>
<td>5.0 x 10^{-4} to &lt; 1.0 x 10^{-3}</td>
</tr>
<tr>
<td>Mass of DU+RU disposed per energy generated, t/GWe-yr</td>
<td>120 to &lt; 200</td>
<td>&lt;1</td>
<td>&lt;1</td>
</tr>
<tr>
<td>Volume of LLW per energy generated, m^3/GWe-yr</td>
<td>252 to &lt; 634</td>
<td>252 to &lt; 634</td>
<td>252 to &lt; 634</td>
</tr>
<tr>
<td>Natural Uranium required per energy generated</td>
<td>≥ 145.0</td>
<td>&lt; 3.8</td>
<td>&lt; 3.8</td>
</tr>
</tbody>
</table>

Note: HLW= High-level waste; DU = Depleted uranium; RU = Reactor-recovered uranium; SNF = Spent nuclear fuel; LLW = Low-level waste.

A study was conducted that compared the performance of U/Pu and U/TRU recycle in greater detail, going beyond the evaluation criteria and metrics used in the E&S study, to determine if there were any additional potential benefits associated with U/TRU recycle, and to further explore the nature of the development and deployment differences, that would help to justify the greater development and deployment challenges [2]. The basic difference between U/Pu recycle and U/TRU recycle is that the minor actinides (MA) are recycled along with the Pu in U/TRU recycle, while they are disposed with U/Pu recycle. In common with the E&S study, this study also compared the performance at an assumed "steady-state", where a fuel cycle is completely deployed and continuously operated.

Overall, the results of the study showed that both U/Pu and U/TRU recycle fuel cycles could each provide benefits with respect to the other depending on where the minor actinides are in the fuel cycle, as summarised in Table 3.
### Table 3: Relative benefits and challenges of U/Pu and U/TRU recycle

<table>
<thead>
<tr>
<th>Fuel Cycle Activity</th>
<th>EG23 (U/Pu)</th>
<th>EG24 (U/TRU)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Recycle fuel fabrication and</td>
<td>Potentially lower shielding requirements due to lower activity</td>
<td>Requires remote (hot cell) activities</td>
</tr>
<tr>
<td>handling</td>
<td></td>
<td></td>
</tr>
<tr>
<td>In-reactor fuel behaviour</td>
<td>Substantial experience with U/Pu fuel</td>
<td>Little experience with U/TRU fuel, especially for heterogeneous recycle, and development challenges have been identified</td>
</tr>
<tr>
<td>Used fuel handling</td>
<td>Determined by fission products in the used fuel</td>
<td>Determined by fission products in the used fuel; heterogeneous recycle may provide greater challenges</td>
</tr>
<tr>
<td>Used fuel reprocessing</td>
<td>Only uranium and plutonium are recovered</td>
<td>Uranium and the TRU are recovered, likely to be more complex and requiring greater development</td>
</tr>
<tr>
<td>HLW fabrication and handling</td>
<td>HLW will have minor actinides, increasing radioactivity and decay heat, potentially complicating HLW form development</td>
<td>HLW has lower activity and decay heat</td>
</tr>
<tr>
<td>HLW disposal (decay heat)</td>
<td>Higher initial decay heat due to presence of $^{241}$Am</td>
<td>Recycle of all TRU greatly lowers minor actinide content in the HLW</td>
</tr>
<tr>
<td>HLW disposal (radiotoxicity and</td>
<td>Long-term radiotoxicity is mainly determined by plutonium and uranium isotopes, which are in the HLW at process loss amounts in both cases. As a result, minor actinide content of HLW may not be relevant for long-term geologic repository performance. Mobile releases are fission product elements for many geologies.</td>
<td></td>
</tr>
<tr>
<td>long-term repository performance)</td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

For U/Pu recycle fuel cycles, whether only fast reactors are used (EG23) or if fast reactors provide the fissile materials to operate thermal reactors (EG29), the minor actinides are intentionally part of the high-level waste (HLW) and would only potentially be present in the rest of the fuel cycle as a contaminant from reprocessing activities. Implementing a fuel cycle that recycles U/Pu allows the use of fuels with a significant experience base, and these fuels could be fabricated and handled with less shielding than is needed when the minor actinides are in the fuel as is the case with U/TRU recycle. The irradiated U/Pu fuel also has lower decay heat than when U/TRU is recycled, allowing handling and shipping sooner with fewer shipments to processing facilities. Reprocessing technologies for recovering U/Pu are available that are similar to those that have been deployed industrially, potentially reducing R&D needs. However, at the same time, even though the presence of the minor actinides in the HLW is accommodated by existing waste form technologies, the minor actinides could complicate advanced waste form development and affect the durability of an advanced waste form due to the higher $\alpha$ radiation, potentially increasing R&D needs in this area. In addition, the HLW minor actinide content, especially $^{241}$Am, adds decay heat that needs to be managed during handling and storage, and which may either increase the size of a geologic repository due to the potential for requiring an increased area for disposal of HLW with higher decay heat, or pose greater engineering and operational challenges to manage the decay heat.

In contrast, recycle of the minor actinides in U/TRU recycle fuel cycles, whether only fast reactors are used (EG24) or if fast reactors provide the fissile materials to operate thermal reactors (EG30), keeps the minor actinides out of the HLW except for processing losses. This would allow the use of existing waste form technologies and has the potential to simplify advanced waste form development. The lower HLW decay heat may enable more compact disposal, reducing the size requirements for a geologic repository. However, these benefits also come with the development of advanced waste form development and the potential to simplify advanced waste form development. The lower HLW decay heat may enable more compact disposal, reducing the size requirements for a geologic repository. However, these benefits also come with the development of advanced waste form development and the potential to simplify advanced waste form development.
challenges associated with keeping the minor actinides within the fuel cycle. As mentioned above, U/TRU recycle fuels will require more shielding for handling and fuel fabrication, there is little to no experience with such U/TRU recycle fuels in a reactor, and the handling and storage of the irradiated fuel must deal with the higher decay heat associated with the minor actinides. Reprocessing technology also needs to be developed, and R&D has been ongoing to achieve U/TRU recovery.

It is also possible to consider recycle of just one or more of the minor actinides rather than all of them together. Prior studies and the considerations show that such approaches can modify the relative benefits and challenges between fuel cycle options but do not change the conclusions of the study [3]. For example, if americium is recycled along with the U/Pu, the decay heat from the americium is kept out of the HLW, potentially benefitting the space required for waste disposal, but there may still be complications for advanced waste form development due to α damage, mainly from curium. At the same time, the presence of americium in the recycle fuel increases the decay heat, affecting storage and handling, and there is little to no experience with such fuels although challenges related to the recycle of americium have been identified in other studies. This example illustrates that such partial recycle of the minor actinides may move specific benefits and challenges between the different parts of the fuel cycle but they will always be present.

Overall, this examination of the relative benefits and challenges of U/Pu and U/TRU recycle fuel cycles confirmed that either approach has both benefits and challenges. It appeared that the benefits are associated with the parts of the fuel cycle that do not contain significant amounts of minor actinides, and the R&D challenges we associated with the parts of the fuel cycle where the minor actinides are present. Given that both U/Pu and U/TRU recycle fuel cycles have both benefits and challenges, any preference for either U/Pu or U/TRU recycle would depend on the preferences for R&D either on waste forms or on recycle fuel fabrication, operation, and reprocessing, respectively. At this time, no further assessment was made of the relative difficulty of the R&D choices, and a strategy that pursued both U/Pu and U/TRU recycle options would appear to be the logical approach until such time that the results of R&D begin to differentiate the development and deployment challenge, if that should occur, with all four most promising groups from the E&S study as potential candidates for development.

Transition studies

While the recycle of fuel would have benefits identified by the E&S study, particularly in the steady-state implementation of such a fuel cycle, changing to such a fuel cycle would introduce practical issues that need to be addressed in the transition. Transition scenario studies have been conducted for the most promising fuel cycle options to develop an understanding of the requirements for a successful transition from the currently operating fuel cycle in the US to an alternative fuel cycle, especially identifying any issues or constraints that would inhibit or prevent such a transition. The scenario studies explored a range of effective transition strategies, with the goal of developing an understanding of transition issues, times, costs, and constraints in order to enable development of effective transition strategies; to identify robust transition pathways that consider economic conditions, energy demand, etc., and; to identify the decisions that need to be made, the time frame for such decisions, and the effects of delaying decisions. This ultimately requires consideration of a broad range of possible implementing technologies and future conditions to inform decision makers.
Transition to U/Pu recycle in fast reactors (EG23)

A base case was defined assuming US nuclear energy production growing at an annual rate of 1%, and where all LWR used nuclear fuel (UNF) produced beginning in 2015 is recycled and used to support deployment of fast reactors [4]. The transition simulations start in 2015 and extend to 2050. This growth rate is thought to be representative of nuclear energy roughly maintaining its current market share of electricity based on a projection of current electricity demand growth rates in the US. The base case assumes that existing inventories of UNF generated prior to 2015 will be disposed as spent nuclear fuel (SNF). The base case also included defining when each reactor in the current fleet is expected to shut down. For these analyses, the assumed lifetime of the existing fleet is that in terms of energy generation, half would operate for 60 years and half would have their lifetime extended to 80 years. It was additionally assumed that the implementing technologies for EG23 are sodium-cooled fast reactors (SFRs), off-site aqueous reprocessing of UNF, and glove-box fabrication of the recycled metallic fuel. The SFRs are assumed to be available for commercial deployment beginning in 2050. Prior to that, all demand for new or replacement generation to maintain the 1% growth rate is met by constructing new light water reactors (LWRs) with lifetimes of 80 years.

A major consideration during transition is having sufficient fissile material for starting the fast reactors before the fast reactor fleet is self-sustaining by recycling its own fuel. As Figure 1 shows, the FR fleet will go through a period of rapid growth as the fast reactor fleet expands from just a few reactors to 100 or more. During this phase, a separate fissile source will be required to supplement the fissile available from recycled fast reactor fuel, either from recycled LWR fuel or LEU, or new LWRs would be built until sufficient fissile is available to support the start-up of the fast reactors. As the fast reactor fleet expands, the fraction of fissile that needs to be added from outside the fuel cycle decreases and by the end of transition, the fleet has become self-sufficient, i.e. all new fissile material required will be produced by the fast reactor fleet.

Figure 1: Transition to fast reactor recycle of U/Pu and the reduction in the need for natural uranium as compared to the current US fuel cycle

Three pathways were explored in the study. The first pathway is the “base case” described above which uses only plutonium and uranium recovered from light water reactor (LWR) used nuclear fuel (UNF) of the current fleet, which is referred to as “LWR RU/Pu”, to start up the first fast reactors.
As Figure 1 shows, new LWRs need to be deployed after 2050, thereby extending transition. The second pathway involves using low-enriched uranium (LEU) fuel during initial fast reactor deployment to supplement the U/Pu available from recycled LWR fuel. This pathway is designated as the “LEU Support” pathway. The third is the “LEU only” pathway which uses only LEU fuel as the start-up fissile source for the fast reactors, and no LWR UNF is recycled, and represents a bounding case intended to inform on the tradeoffs of recycling the LWR UNF since fissile material availability was observed to be a key constraint on the ability to deploy fast reactors during transition. In the analyses, LEU and the necessary natural uranium (NU) and enrichment (SWU) capacity are assumed to be unconstrained, and removes the fissile material availability constraint to starting up fast reactors during transition [5].

The analyses indicated that for all scenarios envisioned, fast reactors need to be deployed as soon as possible in order to maximise the benefits associated with transition to a new fuel cycle. This may be achieved by fuelling the fast reactors with LEU when there is insufficient Pu available. In the base case scenario, LWRs may also be deployed during the transition to meet energy demand if fast reactors cannot be started due to insufficient fissile material to fuel them. In addition, existing LWRs shut down before the introduction of FRs must be replaced by new LWRs that are assumed to operate for another 80 years, thereby increasing the cumulative amount of energy generated with LEU fuel and delaying the end of transition to the new fuel cycle.

The study also found that life extension that keeps existing light water reactors operational until after the fast reactors are available will lead to better transition performance since fewer new LWRs will need to be built. The use of LEU to fuel the fast reactors reduces or perhaps even eliminates the need for LWR UNF recycle technologies. By decoupling the LWR UNF recycle technologies from the deployment of the fast reactors, significant investments in these technologies can be delayed, reduced, or eliminated.

In general, the total amount of fissile material required to sustain the operation of a reactor is the total amount of material that is in the reactor as well as all material that is in storage or held up in processes outside of the reactor including decay storage, separations, fabrication, and transportation. This total time from reactor discharge to recharge as fresh fuel is referred to in the study as the recycling time. The use of technologies or deployment of those technologies in a way that reduces recycling time will significantly reduce the fissile material required to start up and sustain the operation of a fast reactor. Shorter recycling time for the fast reactor fuel makes the fissile material produced in the fast reactors available sooner and reduces the material held up in processes and storage outside of the reactor. This allows for a much more rapid expansion of the fast reactor fleet. For transitions that are constrained by the availability of U/Pu from LWR UNF, there are significant performance benefits that result from a shorter recycling time of the fast reactor UNF. However, there appeared to be no significant benefits from reducing the recycling time of the LWR UNF, since there is already a large inventory of LWR UNF that exists when fast reactor deployment begins.

The study also found that for transitions potentially constrained by the availability of U/Pu from LWR UNF, using LEU to start up the fast reactors results in lower overall natural uranium and enrichment requirements by the end of transition, although there would be a significant increase during the early stages. An LEU-fuelled fast reactor requires significantly more natural uranium and a higher enrichment than an LEU-fuelled LWR to generate the same energy. However, the fast reactor only requires LEU until sufficient fast reactor recycled fuel is available (on the order of ~10 years depending on the scenario and recycle time) while LWRs require LEU for the entire 80-year lifetime of the plant, resulting in larger lifetime demand. In addition, the use of LEU to start-up fast reactors
may have large economic benefits during the early part of transition when availability of UNF and demand for recycled material is relatively low compared to the anticipated processing capacity of recycle facilities, potentially resulting in low capacity factors for reprocessing plants for many years. Unfortunately, the scenario that uses LEU to start-up fast reactors and eliminates the need for recycle of LWR UNF means that all of the LWR UNF will be disposed, resulting in larger quantities of material requiring geologic disposal, and the properties of these materials will be more challenging (higher activity, heat, and toxicity).

All three FR start-up pathways proved to be viable transition approaches, although the characteristics of each pathway affect the resulting performance of the fuel cycle during transition and the challenges of transition, which depend on the scale of the future nuclear energy system and the recycle time of the fast reactor UNF. There are other technology-dependent considerations that will have impacts on transition, but for most of these considerations, what is beneficial for the technology performance is also beneficial for transition performance. For example, the greater thermal efficiency and higher average discharge burnup for the fast reactor will both improve the economics of the fast reactor and the performance of the fuel cycle. However, while higher burnup is also better for LWR performance, less U/Pu would be available in the LWR UNF inventory to support start-up of the fast reactors, potentially delaying transition.

Transition to U/TRU recycle in fast reactors (EG24)

The analysis of transition to fuel cycles in EG24 (U/TRU recycle in fast reactors) was built on the knowledge developed in the analysis of EG23. As with EG23, there are four key technologies involved in recycle: 1) fast reactor; 2) fuel fabrication; 3) qualified fuel; and 4) material recovery (separation). As described above, other than the fast reactor, the recycle technologies for EG24 are less developed and there is less experience, potentially requiring more time and investment to advance them to commercial deployment. Additionally, there are challenges related to the recycling of higher heat and radiation emission materials that result from recycling of the MA, e.g. recycle times within the system due to the minimum cooling times for shipment and/or separation.

Three pathways were considered, equivalent to those for transition to U/Pu recycle as described in the preceding section, except now it is all TRU elements being recycled. These equivalent pathways are U/TRU from LWR UNF, U/TRU from LWR UNF supplemented by LEU, and LEU only without any LWR UNF recycled. For EG24 relative to EG23, it was found that the inclusion of the MA has little impact on the transition behaviour for the aspects driven by fissile material requirements. However, it was noted that achieving a particular recycle time becomes more challenging because of the higher decay heat and radiation emission levels that make shipping and separation more difficult. If a longer recycle time is required, this can significantly delay transition. The evaluation indicated that while homogeneous recycle (recycle of all TRU elements as a group) or heterogeneous recycle (recycle of the TRU elements in two or more groups) are not expected to have significant performance differences at steady state, there are significant differences that could result during transition.

Transition to U/TRU recycle in fast and thermal reactors fuel cycle (EG29)

Recent studies have also included an assessment of the key considerations that affect performance and the challenges of transition from the current US nuclear energy system to a future nuclear energy system in which U/Pu is continuously recycled in critical reactors using both fast and thermal reactors, but enriched uranium is no longer needed (EG29) since all of the fissile required to operate
the reactors is created in the fast reactors while the thermal reactors (e.g. advanced light water reactors (LWRs)) are net burners of fissile. The steady-state fleet after transition is a mix of U/Pu-fuelled fast reactors and U/Pu-fuelled thermal reactors. The proportion of fast reactors and thermal reactors can vary, with up to about 50% thermal reactors being possible for the maximum breeding ratio in the fast reactors in the case where there is no growth in nuclear power energy generation. Other factors may affect the proportion, such as the relative economics of the fast and thermal reactors, but in all cases, the overall fuel cycle benefits identified in the E&S study are attained. For this study, the assumption was that the objective would be to maximise the share of energy produced by the thermal reactors, which for the analyses performed in the study was about 30% LWRs for the assumed 1% growth rate for nuclear power.

As the results of the E&S study showed, the potential steady-state benefits of EG23 and EG29 are essentially the same since for both systems, there is no disposal of either uranium or spent nuclear fuel, and only minor actinides, fission products, and U/Pu losses are sent to HLW. As Figure 2 illustrates, the transition times are quite similar between EG23 and EG29 for the examples shown here (although the transition time for EG23 can be shortened by further optimising the fast reactor performance). The major differences between the two transitions are in the transition flexibility and the potential benefits of that flexibility since the ratio of thermal and fast reactors can be allowed to vary significantly during transition without necessarily delaying the end of transition. Almost all of the conclusions from the analysis of transition to EG23 can be applied to the transition to EG29. The major difference is the inclusion of the thermal reactors in EG29 as an end-state technology and not a technology to be phased out during transition.

**Figure 2:** Transition to fast and thermal reactor recycle of U/Pu

R&D needs and opportunities

Transition analyses typically involve significant complexity and consideration of a range of assumptions. The development of reactors and recycle systems designed for transition objectives is a key consideration, and the qualities and performance of reactors that facilitate transition need to be better understood. The design of recycle facilities that can efficiently handle different throughputs and material flows is needed to inform on the tradeoffs, such as those associated with operating at shorter cooling times, higher recovery efficiencies, and different scales of facility sizes.

The economics of transition adds complexity to the challenge of understanding and informing on the economics of future nuclear energy systems. For example, the relationship between fast reactor breeding ratio and the average discharge burnup can affect overall fuel cycle economic performance.
In the fast reactor, lower average discharge burnup for the driver fuel increases the reprocessing capacity required in terms of MTIHM/yr, but more fissile remains in the driver fuel and there is lower fission product content. If blanket assemblies are used, lower discharge burnup lowers both fission product content and fissile concentration. These and other relationships result in the economics of transition to a system using both fast and thermal reactors having even more complex cost behaviour.

Ongoing activities are designed to improve the understanding of the issues and factors that affect the potential transition to a new fuel cycle using recycle in fast reactors, including identifying the technology characteristics that lead to more favourable transition performance, such as overall time to complete transition, waste generation rates, and annual costs.

Conclusions

The Evaluation and Screening (E&S) study identified the most promising fuel cycle options for R&D within the US DOE Office of Nuclear Energy fuel cycle technologies programme for the specified evaluation criteria. Subsequent studies building on the results of the E&S study have provided insights on more focused issues associated with the development and deployment of technologies for an advanced fuel cycle system. Results of some of the completed and ongoing studies were discussed in this paper, and included the assessment of the impact of recycling minor actinides in the fuel cycle and the characterisation of transition and associated challenges.

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References


Economics of fuel cycle deployment

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Abstract

The transition from an existing to an alternative fuel cycle presents unique economic challenges not present in the final state of the alternative system, once fully deployed and operating as intended. This study identifies these unique economics issues that must be overcome for the deployment of an alternative fuel cycle option. Additionally, the study proposes approaches and methodologies to evaluate these issues for different deployment pathways and fuel cycle options of interest.

An underlying assumption is that deploying an alternative fuel cycle option is not the result of that alternative being financially preferred under the current economic conditions (or it would be naturally favourable for deployment). The significant upfront investment required to develop and deploy the alternative fuel cycle as well as the significant likelihood that it will have a higher average cost make it financially unattractive today. This requires that the economic analysis must inform on the other sources of value that have non-market valuation. This is needed to allow decision makers to evaluate if government intervention, by incentives, is warranted. It requires that the economic analysis include the material and labour flows (items that will be costed), cash flows (cost and revenue streams), and the benefit flows (value, positive or negative, not captured in the cash flow).

The challenge of appropriate discount rates is even more complex. There are issues relating to variation in the risk premium for demonstration facilities (high risks of new technologies) through Nth of a kind (well-established technologies with similar risks to the rest of the energy sector). Issues related to discounting monetary and non-monetary benefits, and issues related to intergeneration discounting because of the very long time horizons, must also be considered.

There are significant transition effects that need to be captured in the economic analysis. Some examples include learning, mismatch of supply and demand as systems ramp up and down and/or variation of facility scale, and variation in the properties and performance of the fuel cycle system.

System-level economics is considered. System-level economics is the aggregate performance of the entire system. The facility-level economics requires a much greater level of detail of modelling and assumptions than is currently in the transition analysis models in order to evaluate the revenue streams of each individual facility in the system. Extension to the facility-level is considered since it is the required extension of this work. The system level provides a lot of insight into the key issues and challenges, cost drivers and uncertainties.
Introduction

The recently completed Nuclear Fuel Cycle Evaluation and Screening study (E&S) [1] considered performance at steady state when evaluating the potential benefits of an alternative fuel cycle, and identified a set of challenges (Development and Deployment Risk, Institutional Issues, and Financial Risk and Economics) to achieving that eventual end state. Separate studies [2, 3] have looked at many of these issues for the transition of the current fuel cycle to alternative fuel cycle options. The deployment of an alternative fuel cycle system presents unique economic challenges that are not present in the final state of the alternative system, once fully deployed and operating as intended. This study identifies these unique economics issues that must be overcome for the deployment of an alternative fuel cycle system.

For consideration of systems at steady state, traditional levelised cost calculations provide good insight into the cost drivers and economic issues of one particular fuel cycle option relative to another. This approach was used by the E&S. There were some other economics related issues identified in the report as market metrics that pertain to specific facilities, and other economic challenges that are not captured in a levelised cost calculation.

The E&S focused on a broad spectrum of potential benefits. These benefits are for society as a whole. The economics approach will address system economics which are related to the costs and benefits to society as a whole. This area is useful in trying to inform on questions related to whether the benefits to society are worth the cost to society. This is a difficult question even for a simple steady-state system where all costs and benefits are assumed to occur concurrently. The economics of deployment involve costs and benefits that may be separated by large periods of time. Also, these costs and benefits may show a very significant time dependency during the transition period before achieving some steady-state performance that is sustained indefinitely.

While it is generally possible to develop equations based on well-known standard economic evaluations, finding the correct data to use with those equations is often very difficult. It is the reason that two different decision makers can and often do come to completely different conclusions from the same set of objective information. The study proposes approaches and methodologies to evaluate these issues for different deployment pathways and fuel cycle options of interest. The methods were tested on what is referred to as the example scenario for a specific scenario evaluated [2].

This study is has not focused on any specific deployment scenario. It is intended to identify the economic issues which need to be informed on, to understand the economics of transition sufficiently to make meaningful comparisons between different alternatives, identify issues that will need to be addressed as part of the challenges of deploying an alternative fuel cycle option, and the key drivers of the economics. From that information, approaches and methods to evaluate this information are developed. Initially, this is intended to help inform on better approaches for transition and the R&D that can address these issues and/or improve the economics of different options. Eventually, this study can be used for detailed quantitative comparison of specific options under consideration by decision makers.

Key considerations

*No financial driver for deployment*

The underlying assumption is that deploying an alternative fuel cycle options is not the result of that alternative being the financially preferred alternative under the current economic conditions. If it
were the financially preferred alternative, there would probably be no need for the government’s involvement, even in the initial research and development to enable that alternative.

A primary reason why an alternative fuel cycle might not be the financial preferred alternative is that the levelised cost of the future steady-state system is greater than or there is at least too high of a probability that it will greater than that of the current fuel cycle option. The high potential cost and large uncertain signifies an unattractive commercial investment under the current conditions which includes the significant costs associated with development and demonstration of the technologies. Figure 1 (Figure D-2.22.2. from E&S Report [1]) shows that nearly all fuel cycles are expected to be more expensive (including all future spent nuclear fuel (SNF) disposal costs) than the currently deployed one. However, there is generally large uncertainty in the final costs.

The E&S identified four fuel cycle groups as “Most Promising” and these are evaluation groups EG23, EG24, EG29, and EG30 of the study. The uncertainty for all of these groups is largely driven by the uncertainty in the eventual cost of the fast reactor, but there is also significant uncertainty related to the cost of recycling the fuel. The E&S broke down the cost of technology development, demonstration, and deployment into development and first of a kind (FOAK) deployment and represents data using broad cost bins. For the development stage, EG23 and EG29 were put in the USD 2B-USD 10B bin and EG24 and EG30 in the USD 10B to USD 25B. For the FOAK deployment, EG23 was in the USD 10B-USD 25B bin, and the other three evaluation groups were in the USD 25B-USD 50B bin. There are likely significant additional costs associated with the other transition effects (e.g. matching supply and demand during periods of rapid expansion) that are one the focus of this work.

Since there is not currently a financial driver, the quantification of the other sources of value that result from the alternative system must be considered. Obviously, a poorly designed system can be far too costly to justify the additional benefits. The question is whether or not there are well-designed systems where the additional benefits with non-market valuation (discussed below) are sufficient to justify the added cost and/or uncertainty. This drives the economic analysis to consider not just the financial (cash flow) issues, but also to consider the other benefits that result from deployment of the alternative fuel cycle option that enables decision makers to compare the costs and benefits in a meaningful way and to decide if government intervention is warranted to overcome the current conditions in order to make the alternative fuel cycle the financially preferred alternative.

This is in essence a cost/benefit analysis in line with what is typical for federal regulatory analysis. This work is not intended to provide the approach and methods for performing that cost/benefit analysis, but only to provide the approach and methods for developing many of the key inputs that would be needed.
System inputs and outputs

Whether studying a specific piece of the system or the entire system, understanding the inputs and outputs is pertinent to understanding the economics. These inputs and outputs are dynamic time-dependent values. A simple schematic in Figure 2 shows the three main types of flows that need to be understood for evaluating the entire system or a piece of that system.

An element is the typical inputs and outputs that are the materials and labour that go into the construction, operations, and decommissioning of all the physical facilities within the system of interest. Some of these inputs and outputs will be identified in the cost analysis while others will be identified in the fuel cycle analysis.

The cash flow related to the typical inputs is obviously a key part of understanding the economics. When looking at the cost, this directly ties to the cost of the inputs and outputs from that system. Most revenue can be tied directly to the inputs and outputs, but not shown is any supplemental revenue that does not tie directly to the typical inputs and outputs. This could include subsidies or other sources that result from value not directly captured by the inputs and outputs.

The benefits which have value that are not directly captured also need to be understood in order to have the complete economic picture. These are generally subjective in nature. They do have direct economic impact on which system is preferred and even in a purely commercial endeavour may be sufficient to justify a more financially costly alternative.
**Revenue and other sources of value**

While the financial benefit is most often the focus of economic analysis, there are other sources of value that play significant role in the economics. This is particularly true when it comes to matters of public policy. A critical part of economic evaluation is the non-market valuation of many of these other potential sources of value. A number of these were identified in the E&S study.

Suppose that based on an economic evaluation of alternatives where non-market values are included, a fuel cycle is found that maximises social net benefits. This alternative fuel cycle would be in society’s best interest. However, the per unit price of electricity of this alternative exceeds the per unit price today. This does not mean that the alternative, although socially beneficial, costs too much and therefore cannot be economically justified. It means that the monetised benefits become an important part of implementation. Monetising benefits means that the diverse set of social preferences are converted into a unit of measurement that is comparable across groups of people. An alternative with positive net benefits means that on the whole the diverse society is better off with the alternative in place.

The economic basis for implementing an alternative that is more costly than today’s technology is grounded in positive net benefits. The task, then, is for the decision maker to determine how best to incentivise the implementation of the preferred alternative. By internalising social value, through higher electric rates, subsidised rates, or incentives directly to firms undertaking such investments, decision makers facilitate the deployment of fuel cycle alternatives that maximise social well-being. Monetising the social value lets the decision maker do such facilitating on an economic basis.

**Discount rate**

Discounting is the central feature of present value analysis, which is the analytical technique to translate values in the future to values that are meaningful in terms of the present day. Discounting is based on the logic in the time value of money. It captures an element in decision making that economists refer to as opportunity cost. That is, the opportunity cost associated with investing in a particular alternative is the value foregone by not investing in the next best alternative. Moreover, discounting is a method of risk accounting because investing in the chosen alternative may return something less than the next best alternative.
While the logic of discounting is relatively straightforward, complexity presents itself to the analyst with respect to what value to choose for the discount rate \( r \). This is particularly complex in the context of transition to an alternative fuel cycle option.

Financial theory in economics introduces the notion of a “risk premium” (RP) to inform on what value to add to the “risk free” (RF) rate of return and therefore \( r \). The RF rate is typically taken to be the rate paid on short-term US Government securities [4]. The project’s return on investment is the sum of the RF (the investor reward for delaying consumption) and RP (the investor reward for taking on risk). In transition analysis, this is an important concept because the RP for investments in FOAK technology is not likely equal the RP for investments in NOAK technology. Therefore, the correct choice for \( r \) will be different for these two investment types. Intuitively, the RP for FOAK investments will exceed the RP for NOAK investments—possibly to the point where the private sector is not willing to take on such risk.

The issues of technology development and FOAK investment imply that the government will most likely be an important investor through at least FOAK technologies. For investments made by the US Government, Circular A-94 [5] outlines the discount rate policy. This document directs analyst to use a real (inflation held constant) discount rate of 7% when analysing cash flows of costs and benefits. The recommendation is based on the average, historic pre-tax market rate of return that is approximately 7%. The intuition in this recommendation is that if the government diverts funds from the private sector for public investments 7% approximates the opportunity cost associated with those funds. The document further directs the analyst to conduct sensitivity analysis on the outcomes of interest. A separate policy document, Circular A-4 directs regulatory impact analysis where discounting policy is addressed [6], and directs the analyst to report results using real discount rates of 3% and 7%.

A-94 has a clause for cost-effectiveness analysis (CEA) that points the analyst to a set of discount rates in what A-94 calls Appendix C. CEA is described in two ways. Projects where the flow of monetary benefits across alternatives is identical should use CEA. The flow of costs varies across alternatives but not benefits. The other situation where CEA should be applied is where benefits are not measured in monetary units, for example lives saved. For projects where CEA is the analysis to conduct, Appendix C provides a schedule of discount rates in terms of nominal and real rates. The schedule is based on maturities of US Treasury notes. If the analyst conducts CEA on a project where the time horizon is 30 years or greater then Appendix C directs the analyst to use a real discount rate of 1.4%.

Time is an important element to consider when choosing the discount rate for use in analysis. This is important to consider for analysis of transition and for issues in nuclear analysis in general. In the case where benefits are not generated for a very long time but costs result right away, the discount rate may in fact “discount away” the value of future benefits and make some alternatives look unrealistic. Take, for example, investment in disposal and storage facilities. Costs are incurred in the near term but the benefits are distributed over very long time horizons. The standard application of discounting reduces values far into the future to essentially zero in present day terms. This issue is not unique to analysis of investments in nuclear.

The Environmental Protection Agency (EPA) convened a meeting of 12 leading economists who have theorised and written much about discounting to address intergenerational discounting, a term coined by economists to refer to discounting over very long time horizons. A summary of the EPA meeting is found in Arrow [7]. The panel suggested using a discount rate that declines over time or DDR (for declining discount rate). There was consensus among the economists at the EPA meeting.
that the DDR is how intergenerational discounting should be handled although there was disagreement about how the DDR should be parameterised.

The key outcome is that the method needs to account for discount and be able to use discount rates that may vary with time. A range of discount rates will need to be applied in any analysis.

**Transition effects**

The deployment of an alternative fuel cycle will involve significant time-dependent behaviour of the nuclear energy system. The difference between the time-dependent behaviour and the time-independent steady-state behaviour are generically referred to as the transition effects. This by necessity requires understanding what the steady-state nuclear energy system performance is for the end-state alternative fuel cycle under the assumed conditions.

For a transition analysis, identifying the steady-state performance for the specified conditions (e.g. energy growth rate, discharge cooling time, etc.) is a significant part of that effort. The same is true for the economic analysis. Identifying the steady-state economics are needed to understand what the transitional effects are. Given that these are likely all negative relative to steady state, the difference will identify how efficiently the system is transitioning and where there is the potential for improvement. The assumption is that the economic analysis was performed on the steady-state nuclear energy system and it was determined that the alternative fuel cycle was in fact desirable under the end-state conditions. This evaluation of the transition economics is to help inform on whether the deployment of the alternative fuel cycle option with the transition effects included (e.g. higher costs) makes it desirable to deploy this system under the assumed conditions. Indirectly, this will help inform on how to best design the deployment strategy so that it is as economically efficient as practical given the other goals and constraints that apply.

These transition effects are the result of a wide variety of dynamic effects that are occurring as the existing system transitions to the final steady-state system on the 100-year time scale in most cases. These dynamic effects include things such as the time-dependent behaviour of identical facilities (learning), mismatch between supply and demand, scale effects (deployment at different scale than at steady state), material properties (non-equilibrium material sources and compositions), etc. The many things that evolve until the system achieves its economic steady-state conditions contribute to the transition effects and the economically significant effects must be identified and evaluated.

**Approach and methods**

The economic analysis of transition requires the detailed time-dependent behaviour of all relevant information. This consists of the financial (monetary) related information as well as the non-monetary information (e.g. waste production). The calculation of the non-monetary information is generally generated by most standard transition analysis codes and is assumed available. What was developed is the approach and methods to evaluate the financial or cash flow information required in the economic analysis.

**Expenditure profile**

System-level economics is considered and is the aggregate performance of the entire system. The facility-level economics requires a much greater level of detail of modelling and assumptions than is currently in the transition analysis models in order to evaluate the revenue streams of each individual facility in the system. Extension to the facility-level is considered since it is the required
extension of this work. The aggregate system must be economical, but it must also be piecewise economical, which is likely more challenging for initial facilities.

The system level provides a lot of insight into the key issues and challenges, cost drivers and uncertainties. The approach and methods developed produce a detailed year by year estimate of the cost for a particular transition scenario, referred to as the expenditure profile. Producing this data is a key input for most if not all subsequent economic analyses. This combined with the results from the transition analysis (modelling of facilities and material flows of a given system over time) provides a wealth of information that can be used to inform on most high-level economic related issues for deployment of future nuclear energy systems. The expenditure profile and systems analysis may eventually become an integrated analysis in order to evaluate the economic tradeoffs of the system, but at this point the assumption is that a transition analysis has been completed and the expenditure profile for that specific analysis is being developed.

The expenditure profile will need to extend from when the first dollar was spent until the last dollar is spent. Most transition analyses will start well after the system is operational. For example, the initial conditions will assume the current fleet of facilities exist and model from that point forward. The economics of an existing facility are very different than the economics of a future facility. This makes performing economic analysis on a system that is a mix of existing facilities and future facilities more complex. Therefore the entire expenditure profile from first dollar to last is necessary. Also, all transition analyses will only run for a finite period of time, but are assumed to continue indefinitely. The costs and benefits that occur beyond the end of simulation cannot be summarily dismissed as negligible. They may in fact be, but this information is needed to show that they are negligible or important to include their impact in the results. If they are not negligible, the results will be skewed because the front-loaded impacts (e.g. capital investment) of the facilities in operation at the end of simulation will be included, but the back-loaded impacts (e.g. energy generation) will be neglected. At the extreme, the last reactor completed at the end of the simulation will have 100% of the capital cost included, but the model will treat it as if no energy was ever generated which skews the economic picture. The question is whether this effect is negligible and that cannot be determined without an approach to quantify the impact.

The end result of this will be approach and methods that allow for a representative expenditure profile that can be used in a broad range of economic analysis. A common approach anticipated is looking at the delta between alternatives. When that delta is with the same alternative without transition it is the cost/benefit of transitioning under those conditions. When it is between transition alternatives to the same end state, it provides insight into the relative efficiency of alternatives.

The goal of developing the expenditure profile is to produce a method which allows for estimating the time-dependent expenditure profile by components. The component is really the smallest practical level of cost. For this study, the smallest level of cost is the elements included in the current transition analysis models. The expenditure profile for each component is estimated in one of two ways. The first are those for which the individual facility is modelled. The second is for those where the material flow is modelled. The approach and methods will allow for the easy expansion to include more facilities to be explicitly modelled and less based on material flows.

The results of the transition analysis are used to determine the mass flows, facility deployment, etc. Then for each cost contributor either the market price modelling or facility modelling is applied to generate the detailed annual expenditure profile for the system. Within this is the breakdown for all facilities modelled, at a minimum fleet average demand and other factors that will provide the timing and sources of cost differences between systems that are being compared.
The market price model is based on the price paid by the consumer. From the perspective of the consumer, they do not need to know the underlying costs that drive the price, but only what they will pay and when. The facility modelling is based on the investments and expenses of the facility and is from the perspective of the investor. The system-level economics is from the perspective of the “owner” of the entire system with assumptions on where the boundaries lie.

For certain items the facility that produces them is not explicitly modelled (e.g. uranium mines). The annual cost in the expenditure profile is simply the annual demand for that item times a unit price. The underlying assumption is that there is a competitive market at equilibrium where cost and price are equivalent. This simplifies modelling of costs associated with contributors whose detail cost makeup is not of interest and as long as the unit cost is equivalent to the results for explicitly modelling the facility that produced the item, the results are equivalent. For any items which represent a significant component of the cost, it may be desirable to explore this assumption by including a facility model approach. The method allows for time variance in both demand and cost of these items, but will generally be assumed to have a constant cost in real (inflation adjusted terms). Figure 3 shows a simple example of what this will generally look like when modelled. It will provide little insight in general.

Everything in the system is produced by some facility whether it is uranium from a mine or electricity from the reactors. Each of these facilities will have an explicit cost profile from the first dollar invested until the last dollar expended after the facility is decommissioned. The revenue they received from the sale of their products and any subsidies must be expected to be of equal or greater value to the investor than the costs. Obviously, some facilities will be judged as bad investments in hindsight, but the assumption is that on average all facilities will be viewed as a positive investment in hindsight. The assumption is that the present value of the revenues and costs are equal.

![Figure 3: Example cash flow for market price modelling](image)

The equivalent unit price for a given facility can be easily calculated using standard levelised cost calculations. The difficulty is that early in transition, some of the cost of certain facilities will almost certainly be recovered from sources other than the price of the goods produced. The system will pay these costs in some form, but it may be in the form of a subsidy or expectation of future profits. These must be equivalent to the costs, but are not easily modelled since they have to be assumed.
Facility models need to at a minimum include the capital investment profile, fixed operation and maintenance costs, and variable operation and maintenance costs. If these costs are likely to vary with scale or number of units deployed, these need to be taken into account in the modelling.

Figure 4 shows example cash flow for a system that involves the deployment of a facility starting in year 10 and a new facility every 10 years after. Each facility takes 5 years to construct and has a 60 year lifetime. This shows the periodic expenditure or capital and the growth in O&M which reaches equilibrium. For this example, the system in terms of this facility is at steady state when the 6th facility is deployed because that is when the first facility shuts down and is replaced and there is no more growth. Even at steady state, there is still periodicity in the capital investment that results from replacement of a facility every 10 years. This detail is lost in the market price modelling method. There was no learning or change in scale in this example.

Variable operating costs represent a challenge when not constant across facilities providing the same function. At the system level, the approach models aggregate material flows. Assumptions about facility specific material flows are required when variable component of O&M is different among alternative facilities that can perform the same function.

There may be significant early costs associated with developing, demonstrating, and initial deployment of technologies that are not captured in the market price modelling or the facility modelling. These costs need to be included because they are part of the overall system costs. They will likely be a small part of the overall system cost, but they are likely to represent a disproportionate challenge because they do not represent direct cost recovery. They are an investment in future benefits and those are generally benefits spread across society and not directly to the investor. A significant fraction, possibly as high as 100%, will likely come from government funds.

**Figure 4:** Example cash flow for facility modelling
Example results

The following are not final results, but are solely intended to show the type of information and level of detail that would result from the application of the approach and methods. These examples were taken from work performed in the transition analysis of an example scenario that we defined largely to develop our methods and understanding of transition for the type of scenarios we anticipated evaluating in the future. This requires developing the input and necessary assumptions in order to model the particular transition scenario. The key assumptions the example scenario are a 1% annual growth rate in demand for nuclear generated electricity, fast reactors would be available for commercial deployment in 2050, an interim period where both fast reactors and LWRs were deployed, and then only FR reactors beyond that point operating on a self-sustaining U/Pu recycle with LWR UNF recycled to provide U/Pu to start-up and sustain the FR fleet until it was self-sufficient. This is referred to as the base case. A case where LWRs operated indefinitely meeting the same demand for nuclear energy was also evaluated and referred to as the Basis of Comparison.

Figure 5 shows the annual electricity by source. Fast reactors with two different breeding ratios were modelled. One to support the steady-state requirements and the other to balance material demands during transition such that all used nuclear fuel was utilised. The Basis of Comparison is essentially identical with all the energy generated by fast reactors generated by the deployment of new LWRs.

Figure 6 shows the benefit of deployment of the alternative fuel cycle in terms of natural uranium required. There are likely significant societal benefits that result from avoiding the need to mine natural uranium. These include environmental benefits as well as security of the future supply of uranium as can be seen in the figure. If there is no transition to the alternative fuel cycle for this scenario, the demand just in the US will far exceed the current world identified resources. While sufficient resources may be identified at an acceptable price, this is not a certainty. The value of this benefit that results from deployment of the alternative fuel cycle is not easily captured in the economic analysis as discussed previously. The magnitude of the benefit can be quantified while the value is subjective. This and all other differences are the benefits of the alternatives being considered that must be weighed against the differences in costs.
Figure 7 and Figure 8 show examples of the type of information that will be generated by the system cash flow analysis and the resulting expenditure profile. It provides details on the source and timing of all costs incurred by the system. By comparing the two figures, the differences in costs, sources of costs, capital requirements, etc., can be evaluated. This information is shown by cost category. To generate this information, requires estimating the capital and O&M costs for every facility modelled within the system. There is an even greater level of detail available than represented in these figures. There is much that can be done with this information. Knowing this information will help to improve the design of the system and identify where R&D would be most valuable to understand and or reduce costs or improve performance.

These figures are for a 1% annual growth rate in demand for nuclear energy, which is why the general trend increases over time. Existing LWRs are assumed to be shut down and replaced in the two waves, one in the 2030s and one in the 2050s, which is why there are the two construction peaks. Fast reactors are deployed starting in 2050, which is why the related costs begin to appear in the 2040s. This was based on preliminary calculations and does not include all costs and is just an example. The shift in costs and their timing is evident from comparing these figures as is the source of the differences. While this is just an example, it shows that there will be a large transition in the dominant sources of cost when an alternative fuel cycle is deployed. Early on the existing systems will be the dominant cost contributors and given that these are undiscounted costs, when discounting is applied these earlier costs will become even more important.
Conclusions

The transition to an alternative fuel cycle system introduces unique economic challenges that are not present in the final end state of the system once fully deployed and operating as intended. Economic analyses of steady-state systems and individual facilities are challenging. This challenge is greatly increased for a dynamic system, particularly the one envisioned that transitions from a large existing system that takes on the order of a century to fully transition and then remains operational indefinitely.
By addressing and understanding the issues and challenges identified, and then applying appropriate assumptions (ranges of assumptions), a lot of valuable information about the performance of the system and relative performance of alternative transition scenarios can be obtained. Such assessment was limited in this study to the system level (aggregate) economic analysis, but was developed with the expectation that it would be expanded to allow facility-level economic analysis.

As in all economic analysis of future advanced systems, there is large uncertainty in terms of appropriate costs of these facilities with the expected ranges often quite large and greater than the differences between the alternatives. Even so, the method developed should show where the largest cost contributors are and why. The underutilisation of facilities (low capacity factor) or the construction of smaller facilities (loss of economy of scale) is likely one economic tradeoff that will need to be considered. This will identify which facilities that have greater economic challenges during transition and some insight into the magnitude of the potential improvement. It will identify when capital investment is required.

Over time, if desirable, these approaches can evolve to perform the type of standard cost benefit analysis that is used to inform decision makers on the tradeoffs in monetary and non-monetary benefits across alternatives. This can inform on differences between alternative transition scenarios envisioned and the business as usual alternative of no transition.

For now, the method allows for understanding the key economic drivers, key parameters (e.g. capital, fixed O&M, and variable O&M costs of a reprocessing facility of a certain scale and reprocessing certain materials), areas where the economic challenges are likely greatest (e.g. low capacity factors, large spikes in annual outlays), and just a wide variety of information that can be used to build the understanding of the economics of transition necessary to inform decision makers on the alternatives envisioned and the most pressing issues and challenges of moving forward along a proposed path.

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References


The effects of the uncertainty of input parameters on nuclear fuel cycle scenario studies

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Abstract

Nuclear systems, composed of reactors and fuel cycle facilities, are complex and in constant evolution. Fuel cycle scenario studies aim at evaluating the consequences of decisions taken in the near term on the eventual outcomes of a chosen fuel cycle.

Scenario studies require a large amount of data and hypotheses, such as the energy demand and its evolution or the performances and characteristics of existing and future technologies. Each hypothesis, represented by a parameter, is a potential source of uncertainty that can impact the scenario outcomes. The Nuclear Energy Agency’s Expert Group on Advanced Fuel Cycle Scenarios (NEA/EGAFCS) recently completed an activity that used sensitivity studies to evaluate the impact of uncertainty of the primary scenario input parameters on scenario results.

At the beginning of the study, a base case scenario was specified and evaluated. It considers a PWR UOX reactor fleet using low-enriched uranium-oxide fuel and its gradual replacement by a sodium-cooled fast reactor (SFR) fleet using uranium/plutonium mixed-oxide fuel. Each member of the group evaluated this scenario with its own scenario code (COSI, COSAC, FAMILY, SITON, TR_EVOL or VISION). It is noteworthy that the different codes show a good consistency in the results and that the differences have been investigated and documented in order to ensure a strong basis for comparison in the sensitivity study.

Then, 17 key input parameters and their range of variation as well as 22 scenario outputs were identified and sensitivity analyses were conducted. The key parameters were chosen among the general assumptions on the scenario (energy demand, introduction date of fast reactor...), reactors and facilities characteristics (burnups, losses at reprocessing...) and some minor actinides recycling parameters. As far as possible, each parameter was varied independently from the others. This sensitivity study shows the relative impact of each parameter on each output metric.

In order to compare the large amount of results, two methods of summarisation were adopted, as described in the paper. These comparisons have shown that some parameters, such as the energetic production or the introduction date of fast reactors, have a strong impact on the results whereas others, like the UOX fabrication time, have almost no impact on the studied scenario.
Introduction

Under the auspices of the NEA Nuclear Science Committee (NSC), the Working Party on the Scientific Issues of the Fuel Cycle (WPFC) has been established to co-ordinate scientific activities regarding various existing and advanced nuclear fuel cycles, including advanced reactor systems, associated chemistry and flowsheets, development and performance of fuels and materials, accelerators and spallation targets. Various expert groups were established to cover the abovementioned topics.

The Expert Group on Advanced Fuel Cycle Scenarios (EGAFC) was created in 2010, replacing the Expert Group on Fuel Cycle Transition Scenarios Studies, to study R&D needs associated with the transition from current or future advanced nuclear fuel cycles. The objectives of the Expert Group are to assemble, organise and understand the scientific issues of advanced fuel cycles, and to provide a framework for assessing specific national needs related to the implementation of advanced fuel cycles.

After conducting a benchmark study to compare existing codes in terms of capabilities, modelling and results [1], the expert group performed a parametric study to identify and communicate the importance of input parameters’ uncertainties in fuel cycle analyses using systems codes [2]. The benchmark was conducted in different phases: definition of a base case scenario, system codes normalisation, parameters identification, parametric studies, and uncertainties representation.

Description of the study

Fuel cycle scenario analysis is a common method for identifying and communicating potential nuclear energy futures, especially when assessing the impact of new technologies. System codes are used to assess the behaviour of different nuclear energy systems and to examine scenarios involving transition of infrastructure (reactors, fuel cycle facilities, etc.) to accommodate growth and adopt new fuel cycle management approaches (e.g. recycling).

By their nature, scenario analyses involve assumptions about the future, and the systems codes used in scenario analyses are imperfect representations of the real world. Uncertainties inherent in scenario specifications and system codes impact the accuracy of analyses. There are several sources of uncertainty in fuel cycle analyses using system codes, including scenario assumptions, facility operational values, reactor core physics calculations, etc., and their impacts vary based on the fuel cycle and the performance metrics of interest. The purpose of this study is to systematically identify these sources of uncertainty and use sensitivity studies to assess their impacts on system-level results.

Scenario codes

The study involved the use of multiple system codes, some of them used by more than one organisation. The different codes used to obtain those results are the following:

- **COSI**, used by CEA (France), ENEA (Italy) and KIT (Germany) [3];
- **COSAC**, used by AREVA (France) [4];
- **FAMILY 21**, used by JAEA (Japan);
- **SITON**, used by EK (Hungary) [5];
- **TR_EVOL**, used by CIEMAT (Spain) [6];
- **VISION**, used by CNL (Canada) and INL (US) [7].
All of the participants modelled the base case scenario and adjusted their models to produce similar behaviour. The analyses of the base case scenario by multiple organisations uncovered any remaining ambiguities in the scenario specification and helped clarifying the scenario documentation in order to ensure consistent interpretation. The remaining discrepancies revealed the impact of differences in code architectures. When these differences occurred, their drivers were identified and discussed. Finally, the base case results for each code provided a basis for identification of the effects of uncertainty of key parameters.

Description of the base case scenario

This study was conducted in several phases. First, a base case scenario was identified and carefully defined. The Expert Group has learnt from past efforts that a clear and concise scenario definition is necessary to reduce the potential for multiple interpretations of the specifications. The resulting base case scenario is 200 years in duration, begins with a fleet of thermal reactors (PWRs) in equilibrium using low-enriched uranium-oxide fuel (UOX) and transitions to a fleet of sodium-cooled fast reactors (SFRs) using a uranium/plutonium mixed-oxide fuel near mid-scenario. The SFR fleet reaches its equilibrium before the end of the scenario. The electricity production is maintained constant at 430 TWhe/y throughout (see Figure 1).

![Figure 1: Base case scenario – installed capacity](image)

In the base case scenario, the reactors lifetime is supposed infinite. The enrichment tail of $^{235}\text{U}$ is 0.25%. Fuel fabrication starts two years before the first load in each reactor type and SFR fuel is made with a mix of depleted uranium and plutonium (Pu). The minimum spent fuel cooling time before reprocessing is of 5 years for PWR fuels and 2 years for SFR fuels. Time for reprocessing is assumed to be null.

PWR UOX spent fuel reprocessing starts in year 35 and is fixed at 850tHM/y. SFR spent fuel is reprocessed when available and according to the need in Pu for fresh fuel fabrication.

The Expert Group selected 22 of the most important scenario outputs as indicators for the study. Standard outputs include reactor numbers and types, mass flows of fuel and waste materials, and inventories of materials in storage and disposal.

Identification of the key parameters

The most important component of this study was the identification of the key parameters to be evaluated. The Expert Group drew from its collective expertise a set of 17 parameters that most often are sources of uncertainty during the construction of scenario specifications. The second step in this process was to identify a variation range for each key primary parameter to be used as a basis
for the sensitivity analysis. Again, experience was used, with each parameter varied over ranges typically used in past analyses (see Table 1).

**Table 1: Identified key parameters and their range of variation**

<table>
<thead>
<tr>
<th>Parameters</th>
<th>Units</th>
<th>PWR UOX</th>
<th>SFR</th>
</tr>
</thead>
<tbody>
<tr>
<td>Fissile burnup (BU)</td>
<td>GWd/tHM</td>
<td>40, 50, 60</td>
<td>100, 136</td>
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<tr>
<td>Fresh fuel $^{235}$U enrichment</td>
<td>%</td>
<td>4.95 (adjusted with BU)</td>
<td>-</td>
</tr>
<tr>
<td>Equivalent Pu content</td>
<td>%</td>
<td>-</td>
<td>13.8 (adjusted with BU)</td>
</tr>
<tr>
<td>Cycle length</td>
<td>EFPD</td>
<td>410 (adjusted with BU)</td>
<td>340 (adjusted with BU)</td>
</tr>
<tr>
<td>Breeding gain</td>
<td>-</td>
<td>-</td>
<td>0.75, 0.9, 1, 1.1, 1.25</td>
</tr>
<tr>
<td>Reactor lifetime</td>
<td>y</td>
<td>Infinite, 60, 40</td>
<td>Infinity, 60, 40</td>
</tr>
<tr>
<td>Total nuclear energy demand</td>
<td>TWh/y</td>
<td>430</td>
<td>430 (steady), Increasing/Decreasing</td>
</tr>
<tr>
<td>Minimum cooling time</td>
<td>y</td>
<td>2, 5, 8</td>
<td>2, 5, 8</td>
</tr>
<tr>
<td>Fabrication time</td>
<td>y</td>
<td>1, 2, 3</td>
<td>1, 2, 3</td>
</tr>
<tr>
<td>Introduction date of FR</td>
<td>y</td>
<td>-</td>
<td>Year 70, 80, 90, 130</td>
</tr>
<tr>
<td>Rate of introduction</td>
<td>y</td>
<td>-</td>
<td>Over 20, 30, 40 years</td>
</tr>
<tr>
<td>First year of reprocessing</td>
<td>y</td>
<td>25, 35, 45</td>
<td>85, 95, 105</td>
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<tr>
<td>Annual reprocessing capacity</td>
<td>tHM</td>
<td>700, 850, 1000</td>
<td>400, 600, 800</td>
</tr>
<tr>
<td>Losses (U and Pu)</td>
<td>%</td>
<td>0.05, 0.1, 0.2</td>
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<tr>
<td>Reprocessing priority</td>
<td>-</td>
<td>FIFO to FILO</td>
<td></td>
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<tr>
<td>Enrichment tail</td>
<td>%</td>
<td>0.15, 0.25, 0.35</td>
<td></td>
</tr>
</tbody>
</table>

*In blue – Reference value of the parameter

**Output from the base case scenario**

The outputs from the base case scenario obtained by all participants were compared in order to identify the discrepancies between the different codes. It is noteworthy that the different codes show a good consistency in the results. As 22 outputs were studied, those for which a very good agreement between the different codes was observed are not detailed in this article. The remaining differences are detailed hereafter.

For the front-end cycle, a really good agreement between the different codes was observed for the evaluation of the natural uranium consumption, the enrichment need, the UOX fabrication need and the Pu flow for FR fuel fabrication.

Slight discrepancies were observed on the estimation of the FR fuel fabrication need (see Figure 2). They are due to inconsistencies in the initial data as this estimation was made from the energy production for some codes (e.g. COSI6) or from the installed nuclear power for others (e.g. FAMILY).
For the back-end cycle and the inventories, more discrepancies were observed on the estimation of outputs. One source of discrepancies is the modelisation of the reprocessing. Indeed, in the scenario specifications, the UOX spent fuel reprocessing starts in year 35 and is fixed at 850tHM/y in order to produce enough plutonium for SFR deployment. However, with the code COSAC, available spent fuel is reprocessed only when Pu is required and in function of the need in Pu, without the possibility to anticipate it. That creates discrepancies on the need in PWR UOX spent fuel reprocessing (Figure 3), and thus on the PWR UOX spent fuel and the interim reprocessed uranium storages (Figure 4). This issue is not encountered for the SFR spent fuel reprocessing, as it follows the need in Pu for all codes.

Another source of discrepancies is a slight difference in the breeding gain of the fast reactors considered by each organisation. As SFR spent fuel is reprocessed according to the need in Pu, the amount and fissile content of Pu in spent SFR fuel directly impact their flow at reprocessing. Thus,
differences on the breeding gain impact the need in SFR spent fuel reprocessing (Figure 5), and so the SFR spent fuel storage (Figure 6) and the Pu interim storage (Figure 6).

**Figure 5**: Base case scenario – SFR spent fuel flow at reprocessing

The discrepancies observed on the total inventory of Pu in cycle and on the minor actinides inventory in waste are the consequences of the differences described previously (Figure 7).

**Figure 6**: Base case scenario – SFR spent fuel (left) and interim Pu (right) storages

The discrepancies observed on the total inventory of Pu in cycle and on the minor actinides inventory in waste are the consequences of the differences described previously (Figure 7).

**Figure 7**: Base case scenario – Pu inventory in cycle (left) and MA inventory in waste (right)

Overall, the study of the base case scenario by all the benchmark participants lead to consistent results between the different codes and provided a strong basis for the sensitivity study.

**Sensitivity analysis**

The primary activity in this study was to conduct sensitivity analyses on the key parameters in order to identify the effects and to quantify the potential impacts of sensitivities of the outputs to the
inputs. To the extent possible, each parameter was varied independently, without change in any other assumption or parameter of the specification. During the sensitivity analyses, some scenarios “broke”, i.e. had insufficient fuel material for the SFR fleet. This was noted in the analysis and then an effort was made to “fix” the scenario. One option was to modify additional parameters (e.g. annual reprocessing capacity) to compensate for the imbalance in the scenario. The other option was to add an external source of fuel material and to note the amount of extra material required to complete the scenario.

Using the results from different scenario codes, the sensitivity of the 22 outputs to the 17 identified parameters has been evaluated. With each fuel cycle metric being evaluated for each year of the scenario, different ways of comparing and displaying the results had to be considered. Two methods were used and are detailed in this paper: the tornado diagrams and the sensitivity table.

To quantify the impact of the variation of an input parameter on the calculation, output indicators had to be defined. Three types of output indicators were retained:

- All the output parameters related to an annual flow (e.g. natural uranium consumption, enriched uranium needs, fuel fabrication and spent fuel reprocessing, ...) were associated to output indicators defined as the cumulated value of the concerned output parameter;

- All the output parameters related to an inventory in an interim storage (e.g. spent fuel storage, reprocessed uranium storage, depleted uranium storage, ...) were associated to output indicators defined as the maximum value of the concerned output parameter;

- All the output parameters related to a global inventory inside the cycle or the waste (e.g. plutonium inventory in the cycle, minor actinide inventory in the waste, ...) were associated to output indicators defined as the final value of the concerned output parameter.

**Tornado diagrams**

The tornado diagram shows the correlation of a single output parameter to changes in the input parameters, one bar per input, where the direction of the bars on the diagram shows whether the correlation is positive or negative. The length of the bars shows the magnitude of the coupling between the parameters. There are two values of \( q \) for each input parameter: one for the case in which the given parameter is at its low value, and one for the case in which the parameter is at its high value.

The sensitivity value \( q \) (see equation [1]) is used to quantify the change in an output indicator as a function of the change in the input parameter value, thus giving an estimation of the relative impact of each parameter on each output indicator. Note that the sensitivity cases that require changing more than one input parameter are excluded from this analysis.

\[
q = \frac{p_{\text{ref}} (R_{\text{ref}} - R_s)}{R_{\text{ref}} (p_{\text{ref}} - p_s)}
\]  

(1)

\( p_{\text{ref}} \) input parameter’s reference value, \( p_s \) input parameter’s value in the sensitivity case

\( R_{\text{ref}} / R_s \) output indicator’s corresponding to \( p_{\text{ref}} / p_s \)

The results of the sensitivity study, represented with Tornado diagrams, are detailed hereafter.
**Front-end cycle**

Overall, the beginning date of the SFR introduction has a strong impact on the front-end cycle results (see Figure 8). Indeed, it is directly correlated to the period of operation of PWR and SFR. A later introduction of SFR results in a longer operation time of PWR and so on higher natural uranium (NU) consumption and separative work unit (SWU) requirements. On the other hand, it reduces the SFR fleet operation time, resulting in reduced SFR fuel fabrication need and Pu flow to fabrication. For the same reasons but to a lesser extent, the duration of SFR introduction (their commissioning rate) impacts the results in the same way as their introduction date.

![Figure 8: Sensitivity of front-end cycle results to each parameter](image)

Obviously, the UOX enrichment tail impacts the SWU requirements. Due to the additional effort (SWUs) required extracting additional fissile material from lower assay tails, the relationships for tail enrichment are non-linear.

Due to the fabrication of final cores (resp. start-up), the PWR (resp. SFR) lifetime is negatively correlated to the PWR (resp. SFR) fuel fabrication need. This impact is slightly non-linear, as it is driven by the fraction of start-up cores over reactor life.

SFR fuel fabrication and Pu flow to fabrication are primarily impacted by the energy growth rate, which determines the total number of SFR required.

Finally, the SFR breeding ratio has a large, non-linear impact on the Pu flow through fuel fabrication. This is primarily due to SFRs with a negative breeding gain requiring an additional external Pu feed and impacting the fissile content and the quantity of the Pu.
**Back-end cycle**

The Tornado diagrams displaying the sensitivity of back-end cycle results to each parameter are represented on Figure 9.

**Figure 9:** Sensitivity of back-end cycle results to each parameter

- Spent fuel storages are sensitive to the reprocessing parameter as a later start of reprocessing results in an accumulation of spent fuel, but a higher reprocessing rate leads to a faster depletion of the storage. PWR UOX spent fuel reprocessing parameters also impact the amount of interim Pu stored. It is not the case for SFR spent fuel as it is reprocessed according to the need in Pu.

- The energy growth rate has a large impact on SFR spent fuel reprocessing as it determines the number of commissioned SFRs. This property is non-linear as with a higher growth rate there is relatively more SFR fuel in cooling inventory that is not yet available for reprocessing.

- As the SFR spent fuel flow at reprocessing is adapted according to the Pu need for fresh fuel fabrication, it depends on the amount of Pu available in the spent fuel and so is impacted by the breeding ratio. The breeding ratio also directly impacts the separated Pu inventory as a higher breeding ratio increases the available Pu without significantly changing the Pu needed.

- As the SFR introduction date is directly linked to the operational time of reactors and to the amount of available PWR and SFR spent fuel, it highly impacts the PWR and SFR spent fuel flow at
reprocessing. Separated Pu being used for SFR fuel fabrication, a later SFR introduction leads to an accumulation of separated Pu.

Finally, the reactors lifetimes are negatively correlated to the spent fuel reprocessing as a decreased lifetime increases the number of final (PWR) or start-up (SFR) cores, leading to an increased amount of available spent fuel.

**Inventories**

Total Pu in cycle is highly impacted by the SFR breeding ratio and the growth rate. A larger growth rate increases the total fleet size and total material inventory of the fleet, including the Pu in cycle (given that the breeding ratio is larger than 1). As the energy growth is exponential, the impact is non-linear and is bigger on the higher side of the sensitivity analysis. The SFR reprocessing parameters also impact the Pu in cycle in a non-linear way as, when a decreased SFR reprocessing results in broken scenarios and in the use of an external source of Pu, it increases the total Pu in the fuel cycle.

**Figure 10**: Sensitivity of front-end cycle results to each parameter

The final two tornado diagrams show the impact of input parameters on the total Pu and MA in waste. In both cases, the breeding ratio is the most significant input parameter. As explained previously, the breeding ratio is negatively correlated to the SFR spent fuel flux at reprocessing, which directly impacts the flux of materials being sent to the waste.

**Sensitivity table**

The sensitivity table provides a way to display the sensitivity analyses results, with all inputs and outputs together on a single table. The correlation of each input (table row) to each output (table column) is shown through colour shading of each cell in the table.

A sensitivity indicator \( S \), see equation (2)\) aims at quantifying the variation of an output indicator to the variation of an input parameter, provided the linearity of the output indicator as a function of the input parameter.

\[
S = \frac{P_{\text{ref}}}{R_{\text{ref}}} \cdot \frac{\partial R}{\partial p}
\]  

(2)
input parameter’s reference value, \( \frac{p_{\text{ref}}}{\partial p} \) its variation

output indicator’s corresponding to \( \frac{R_{\text{ref}}}{\partial R} \) \( \frac{p_{\text{ref}}}{p_{\text{ref}} + \partial p} \)

The linearity is checked by calculating the coefficient of determination \( r^2 \). The sensitivity indicator is retained as a valid parameter when \( r^2 \) is greater than 0.9. It must be noticed that each sensitivity indicator is calculated from a small amount of points (three points for most sensitivity indicators), so that the results should be considered as tendencies rather than absolute results.

Compared with the Tornado diagram that displays two values of sensitivity (qmin and qmax), the sensitivity indicator is more global as it is based on a least squares linear regression upon the three available points (reference, upper sensitive case and lower sensitive case).

On the sensitivity table (Figure 11), red sensitivity coefficient corresponds to a positive correlation and a blue coefficient to a negative one. A darker colour (red or blue) indicates a higher sensitivity of the output to the parameter. In cases where the relationship is non-linear or otherwise hard to quantify into a single value, the shading is replaced with a “?”.

The sensitivity table provides a big picture view of the analysis results. The most important input parameters from a sensitivity perspective can easily be identified by noting the rows with the strongest shading. In accordance with the Tornado diagrams, these are the overall growth rate (energetic production) and the introduction date of the FRs, followed by the rate of FR introduction and the reactor lifetime. Explanations for these impacts have been given in the description of Tornado diagrams.

The change in reactor lifetime impacts material flows without changing the rest of the system.

Additional input parameters with strong impacts on the scenario results are in the areas of breeding gain and reprocessing, where rows have some strongly shaded cells, but also have a number of “?” cells, indicating possible non-linear responses.
The sensitivity table also reveals more localised high impacts where one or two cells are strongly shaded relative to the rest of a row. For example, the enrichment tails only impacts Unat consumption and SWU requirements, but does not impact the rest of the fuel cycle.

Shifting to the columns, only a few outputs show significant sensitivity to a majority of the inputs, and none are sensitive to all inputs. It can be noted that outputs that are the most impacted are the storages and inventories.

Note that some of these conclusions are linked to the scenario hypothesis and cannot be extended to scenario studies in general. For example, the PWR spent fuel cooling time does not impact the results because PWR are being stored several years between the beginning of the scenario and the first year of reprocessing.

**Concluding remarks**

This study aimed at assessing and quantifying the importance of input parameters in scenario analyses. The expert group identified 17 input parameters and evaluated the sensitivity of 22 of the most important scenario outputs to those parameters.
Different scenario codes were used for this study. Where prior benchmarking by this group and others has mainly focused on the common code capabilities, this study used some capabilities only found in some codes. This allowed to include a broader range of cases and to provide fuller information on the sensitivity of the scenario outputs to its inputs.

Two methods, the Tornado diagrams and the sensitivity table, were used to display the results. For the considered scenario, both methods have put forward a strong impact of the overall growth rate and of the introduction date of the SFRs on the results. The rate of SFR introduction and the reactor lifetime are also important parameters from a sensitivity perspective. It is noteworthy that only a few outputs showed a significant sensitivity to a majority of inputs, and that none were sensitive to all inputs.

While the content of this study is most directly applicable to fuel cycle systems analysts, it should also be of value to researchers and developers working on the different parts of the nuclear energy system by providing a systems perspective for their work.

References


Plenary Session III: Transmutation Systems and Infrastructures

Chair: T. Taiwo and N. Stauff
Programme overview on development of innovative TRU burning fast reactor cycle with uranium-free metal fuel

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Abstract

Fast reactors have the capability to effectively burn plutonium and minor actinides (i.e. TRU: transuranic) due to its higher fission-to-neutron-capture ratio compared to LWR. When TRU in LWR spent fuel are recycled in fast reactor cycle, the radiotoxicity of the residual nuclear wastes from LWR spent fuel could be reduced to natural uranium level after several hundred years while more than hundreds of thousand years for no recycling case. This could significantly contribute to improve public perception regarding nuclear waste disposal and might reduce the burden of the disposal. However, almost the same capacity of fast reactors, even with low conversion ratio of 0.75, is needed to continuously burn TRU from LWRs, if U-TRU-fuel fast reactors are adopted.

Considering the above, minimisation of the required capacity of fast reactor system to burn TRU from LWRs is desirable. The most effective way is to use uranium-free TRU fuel since it does not produce any additional TRU. The benefits of such system are to reduce not only the required TRU burner units to about 1/5 but also the capacity of associated fuel cycle facilities to about 1/8. In order to explore feasibility of such fast reactor system, we have initiated the four years’ programme of the development on TRU burning fast reactor cycle using uranium-free TRU metal fuel since October, 2014 under the contract with Ministry of Education, Culture, Sports, Science and Technology (MEXT) in Japan. The most distinguished feature of the system under investigation is to use TRU-Zr metal fuel alloy. The major research items in this programme are, 1) evaluation of fundamental properties on uranium-free TRU metal fuel and its irradiation performances, 2) countermeasure against fuel-cladding chemical interaction (FCCI) for high-content-TRU metal fuel, 3) feasibility of pyroprocesss for TRU-Zr fuel and its waste treatment, 4) simultaneous achievement of enhanced Doppler feedback, low sodium void reactivity and reduced burnup reactivity swing, and 5) reduction of Doppler feedback uncertainty for alternative metal fuel alloys such as niobium and molybdenum.

In this paper, the overview of this programme and its current status are presented.

Introduction

Fast reactors burn plutonium and minor actinides (i.e. TRU: transuranic elements) effectively due to its higher fission-to-neutron-capture ratio compared to light water reactor (LWR). If TRU in LWR spent fuel are recycled into fast reactor cycle, the radiotoxicity of the residual nuclear wastes of LWR spent fuel is significantly reduced. It could also contribute to improvement of public perception on nuclear waste disposal and reduce the burden of the disposal.
However, U-TRU-fuel fast reactors also produce TRU while burning TRU because they use uranium together with TRU. For example, fast reactors, even with low conversion ratio of 0.75, burn only 0.18 tTRU/GWe/y, while LWRs produce about 0.22 tTRU/GWe/y. It means almost the same capacity of fast reactors as that of LWRs is needed to continuously burn TRUs from LWRs.

Considering that LWR would be the dominant nuclear power plants in the next few decades at least, it seems to be difficult to deploy many fast reactors in the near term. Therefore, in order to burn TRU from LWRs using fast reactors under such circumstance, TRU burning capability of a fast reactor needs to be improved. The most effective way is to use uranium-free TRU fuel since it does not produce any additional TRU. Such system can reduce the capacity of the TRU burner units and the associated fuel cycle facilities to about 1/5 and 1/8 respectively.

There have been many studies on uranium-free or fertile-free fuel system in the past. For example, variety of inert matrix fuel concepts such as rock-like oxide fuel, MgO-based oxide fuel, Tc-based oxide fuel and W-based oxide fuel for fast reactors were studied [1-3]. The French study showed potential solutions using a neutron moderator against the uranium-free core issues such as the reduced Doppler feedback and the increased sodium void reactivity [3]. However, such types of the inert matrix fuel seem to need new reprocessing technology. Also, many processing steps seem to be required upon fabrication, and thus may cause economic penalty. On the contrary, uranium-free TRU metal could be reprocessed and fabricated based on pyroprocess and injection casting technologies without large modifications.

As for uranium-free metal fuel concept, there were many studies in the US. One of them was the US accelerator-driven transmutation of waste (ATW) programme which investigated an accelerator-driven transmutation system coupled with a subcritical fast reactor using uranium-free metal fuel [4-7]. Although the ATW system may relax the issue of the reduced Doppler feedback on account of its subcritical system, accelerator facility causes economic penalty too due to its large electricity consumption for the beam production. The US had also studied sodium-cooled Advanced Burner Reactor cores with various conversion ratios from 1.0 to 0.0 to burn TRU based on a critical fast reactor. It had shown that the Doppler feedback was significantly reduced and the sodium void reactivity was increased to greater than USD 10 for the uranium-free core [8]. However, the more recent study has shown that introduction of neutron moderator in-core region would enhance the Doppler feedback with low sodium void reactivity for uranium-free metal fuel core [9].

Considering the above past studies, metal fuel fast reactor cycle is considered to be one of the most promising ways for a uranium-free TRU burning system. Therefore, we have initiated the four years’ programme of the technology development on TRU burning fast reactor cycle using uranium-free TRU metal fuel since October 2014 under the contract with Ministry of Education, Culture, Sports, Science and Technology (MEXT) in Japan.

This paper describes an overview of this research programme and some preliminary results at the present.

**Program overview**

**Objective**

The target system in this programme and its benefit are shown in Figure 1. It aims to burn TRU from LWRs continuously with the minimum capacities of the TRU burning reactors and its associated fuel cycle facilities, having another role as a commercial power station as well. For example, the 5 GWe system with this uranium-free TRU metal fuel burns 4 tTRU/GWe/y approximately and the consumed
TRU is resupplied from 20 GWe-LWRs continuously, while conventional U-TRU fuel fast reactors with the conversion ratio of 0.75 needs around 25 GWe system to burn the same amount of TRU. Thus, the required capacities of the reactors and fuel cycle facilities are reduced to 1/5 and 1/8 respectively.

The objective of this four years’ development programme is to clarify the feasibility of the uranium-free TRU metal fuel fast reactor cycle.

**Research plan**

There are technological issues about this system. Most of them come from its unique fuel composition and reduced Doppler feedback. According to the previous study, the fuel composition would be 60-70wt.%TRU and 40-30wt.%Zr in case of using Zr metal alloy fuel [9]. Taking them into account, the important technological issues and our approach in this program have been identified as shown in Table 1. The planned schedule and the framework of the participating organisations are shown in Table 2 and Figure 2, respectively [10].
Table 1: Technical Issues and approach

<table>
<thead>
<tr>
<th>Area</th>
<th>Technical issues</th>
<th>Approach</th>
</tr>
</thead>
<tbody>
<tr>
<td>Fuel</td>
<td>Fundamental properties of uranium-free TRU metal fuel and irradiation behaviour</td>
<td>Thermodynamic calculation and its validation by test using simulator</td>
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<td>Mechanistic analysis of irradiation behaviour using CRIEPI's ALFUS code</td>
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<td>Increased fuel-cladding chemical interaction (FCCI) due to high-TRU-content fuel</td>
<td>FCCI tests for cladding barriers using Pu and minor actinides at INL in the US</td>
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<td>Increased impurities of lanthanide (Ln) FPs due to high-TRU-content fuel</td>
<td>Clarify the allowable inclusion of Ln to keep its uniformity in fuel slug by tests</td>
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<td>Reprocessing</td>
<td>Need to Improve decontamination factor for Ln FPs</td>
<td>Modification of process flowsheet, liquid cathode material etc</td>
</tr>
<tr>
<td></td>
<td>Electro refining and its waste treatment for high-Zr-alloy fuel</td>
<td>Electro-refining test using high-content-Zr alloy uranium metal fuel, waste treatment tests specific to high-content of Zr &amp; TRU fuel pyroprocess</td>
</tr>
<tr>
<td>Core</td>
<td>Simultaneous achievement of enhanced Doppler feedback, reduced burnup reactivity and sodium void reactivity</td>
<td>Develop the optimised core design by sensitivity analysis for core characteristic parameters</td>
</tr>
<tr>
<td></td>
<td>Severe accident behaviour</td>
<td>Accident analysis for the developed core</td>
</tr>
<tr>
<td></td>
<td>Doppler feedback uncertainty of alternative materials for metal fuel alloy</td>
<td>Measurement of Doppler absorption effect for Zr, Nb, Mo by LINAC at Kyoto Univ.</td>
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</table>

Table 2: Schedule

<table>
<thead>
<tr>
<th>Items</th>
<th>JFY’14</th>
<th>JFY’15</th>
<th>JFY’16</th>
<th>JFY’17</th>
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<tbody>
<tr>
<td>1-1 Fundamental property evaluation</td>
<td>Thermodynamically calculation</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>1-2 Irradiation behavior</td>
<td>Past data evaluation, mechanistic analysis</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>1-3 FCCI countermeasure test</td>
<td>Test 1</td>
<td>Test 2</td>
<td></td>
<td></td>
</tr>
<tr>
<td>1-4 Allowable inclusion of Ln in injection casting</td>
<td>Solution tests</td>
<td>Evaluation</td>
<td></td>
<td></td>
</tr>
<tr>
<td>2-1 High-DF pyroprocess for Ln</td>
<td>Process improvement</td>
<td>Overall evaluation</td>
<td></td>
<td></td>
</tr>
<tr>
<td>2-2 Electro-refining test of high-Zr alloy metal fuel</td>
<td>Preparation</td>
<td>Tests</td>
<td></td>
<td></td>
</tr>
<tr>
<td>2-3 Waste treatment of Ln waste in pyroprocess</td>
<td>Preparation</td>
<td>Tests</td>
<td>Process improvement</td>
<td>Ln wastes tests</td>
</tr>
<tr>
<td>3-1 Core optimization method</td>
<td>Design method development</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>3-2 Establish uranium-free TRU burning core</td>
<td>Core design</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>3-3 Measurement of Doppler absorption effect for Zr, Nb, Mo</td>
<td>Preliminary measurement</td>
<td>Doppler measurement</td>
<td></td>
<td></td>
</tr>
<tr>
<td>4 TRU burning scenario evaluation</td>
<td>Scenario, plant concept, demonstration plan</td>
<td></td>
<td></td>
<td></td>
</tr>
</tbody>
</table>
**Fuel research**

In order to clarify the fundamental properties and irradiation behaviour of uranium-free TRU metal fuel, fundamental properties such as melting temperature, specific heat and thermal conductivity were planned to be evaluated by thermodynamic calculation and some validation tests using simulator. The irradiation behaviour is also evaluated by mechanistic analysis code, ALFUS [11] using the evaluated properties.

The TRU content of uranium-free fuel is higher than that of uranium-containing fuel. It is anticipated that fuel-cladding chemical interaction (FCCI) will increase, mainly due to its higher content of plutonium. According to previous research, some barrier materials for cladding inner wall such as chromium and vanadium proved effective to reduce FCCI [12]. However, the effectiveness has not been confirmed for uranium-free fuel. Therefore, it will be examined by tests using Pu and MA.

Another concern is the increase of lanthanide (Ln) fission products (FPs) in the fuel recycled by pyroprocess due to the absence of uranium and low decontamination factor (DF) of Ln. It could cause non-uniformity together with MA in fuel slug that may results in unfavourable irradiation performance. However, the allowable limit of Ln content in fuel slug is still unclear. Then, the dissolution test for Ln uniformity in metal fuel is planned. The result is used as the target performance of DF in electro refining for uranium-free metal fuel.
Reprocessing research

Improvement of DF for Ln FPs may be needed in this system and various countermeasures such as alternative materials to liquid cadmium and, improvement in treatment process design for FP removal from electro-refining spent salt are investigated.

Another potential issue is the feasibility of electro refining for high-content-Zr metal fuel. Electro-refining tests using U-20wt%Zr alloy and U-40wt%Zr alloy will be conducted. Uranium is used as a substitute of TRU since it is easier to use and uranium is considered to give a conservative test condition for the evaluation of Zr separation because of the smaller difference of Redox potential between U and Zr rather than between TRU and Zr.

Also, waste treatment tests specific to high-content Zr & TRU metal fuel pyroprocess are planned.

Core research

Although some countermeasures to increase Doppler feedback such as the introduction of neutron moderator has been found in the previous study [9], simultaneous achievement of enhanced Doppler feedback, reduced burnup reactivity swing and low sodium void reactivity needs to be further investigated for core design optimisation. An optimisation method for uranium-free fuel core design will then be explored using sensitivity analysis on core design parameters such as core height, neutron moderator, burnable poison, heterogeneous core geometry, alternative materials of metal fuel alloy, etc. An optimised uranium-free fuel core will be developed using the method.

With regards to safety, severe accident behaviour could be somehow different from conventional fast reactor core because the large excess reactivity would be suppressed by neutron moderator during normal operation, reduced core height or something else. Therefore, severe accident behaviour for the developed core will be investigated.

Regarding enhancement of Doppler feedback, the use of alternative fuel alloy such as niobium (Nb) and molybdenum (Mo) instead of Zr might be one of potential countermeasures [9]. However, there could be large uncertainty in analysis of Doppler feedback due to the lack of precise neutron resonance cross section measurement. To reduce the uncertainty, measurements of Doppler absorption effect for Zr, Nb and Mo by neutron irradiation are planned using the LINAC facility in Kyoto University Research Reactor Institute.

Scenario evaluation

In order to examine how to deploy this TRU burning system, the deployment scenario, the commercial plant concept and the engineering demonstration plan are studied in combination with the above technology developments.

Current status and preliminary results

Current status and preliminary results on some items in this research are presented in the following sections.

Fundamental properties of fuel

The melting temperatures for Pu-MA-Zr alloys were calculated as a function of MA concentration using the Thermo-calc software [13]. The thermodynamic database used for the calculation was U-Pu-Zr-Np-Am-Cm-Fe alloy system which was developed by adding Cm data to the previously established database [14-19]. As the results, the solidus temperature and the liquidus temperature of the Pu-MA-Zr alloy will increase with Am and Cm content, and slightly decrease with Np content [19].
Melting temperatures for the assumed compositions, 2.2Np-54.6Pu-7.0Am-1.2Cm-35wt%Zr and 58.8Pu-8.2Am-35wt%Zr, were also calculated to evaluate the effect of Np and Cm. In the latter composition Np and Cm are replaced by Pu and Am respectively. The composition is equivalent to the multi-recycle composition of the uranium-free TRU metal fuel fast reactor. The result is shown in Figure 3. The solidus temperatures and the liquidus temperatures are almost the same for the two compositions. Thus, it is found that the melting temperature for Np-Pu-Am-Cm-Zr fuel can be estimated by the simplified composition, Pu-Am-Zr [19].

The thermal conductivity of Pu-40Zr was estimated based on Vegard’s law taking into account the lowering effect of Zr. The result is shown in Figure 4 in comparison with conventional metal fuel, U-20Pu-10Zr [20]. It indicates that the thermal conductivity of Pu-40Zr or TRU-40Zr could be approximately 60% of conventional one [21]. However, some differences were found between this result and past INL paper [22], this needs to be investigated further.

Figure 3: Calculated liquid phase ratio and melting temperatures for Pu-MA-Zr metal fuel

Figure 4: Comparison of the evaluated thermal conductivities on Pu-40Zr and U-20Pu-10Zr
Fuel-cladding chemical interaction tests

The diffusion couple tests to assess fuel-cladding barrier materials against FCCI on uranium-free TRU metal fuel alloys are being conducted in Idaho National Laboratory (INL). The first test result for Pu-40Zr alloy will be presented at the 2016 ANS Winter Meeting on November, 2016 [21].

Allowable inclusion of lanthanide fission products during injection casting

Lanthanide (Ln) dissolvability tests using U-8Zr metal fuel were done in order to evaluate the allowable limit of Ln FPs during injection casting. The used Ln elements for the test are Cerium (Ce) and Neodymium (Nd) which are typical elements among FP Ln. The test temperature was 1 200°C. Results are shown in Figure 5. It indicates that the dissolved amounts for each element are more than that of solubility in uranium metal. Therefore, allowable inclusion of Ln could be determined based on solubility based on major fuel material, i.e. Pu in TRU-Zr fuel. According to this result, allowable inclusion of total Ln FPs has been conservatively set to 2 wt.% in TRU-Zr metal fuel.

Electro-refining tests

Electro-refining tests of high-Zr alloy metal fuel using U-Zr are in progress. The preliminary results are shown below. Chronopotentiometry measurements have been conducted to evaluate electro-refining behaviour of high-content-Zr metal fuel. The test conditions were:

- Test materials: U-10Zr, U-20Zr, U-40Zr, U, Zr
- Temperature: 500°C
- Salt: LiCl-KCl-UCl₃
- Electrode material (anode, cathode): stainless steel
- Electric current density: constant (parameters: 10 ~ 1 000 mA/cm²)

The test results are shown in Figure 6 – 8. The electric current densities at the anode to dissolve U only, while most of Zr is kept remained at anode, are considered to be about several hundred mA/cm², 100 mA/cm² and 20 mA/cm² for U-10Zr, U-20Zr and U-40Zr respectively. Thus, it is found that 40wt% Zr alloy metal fuel can be reprocessed by conventional electro-refining method. However, the process speed would be lowered at about 1/10 of 10wt% Zr fuel due to the low electric current density. Countermeasures to improve the process speed are being investigated at this moment. Also, alternative methods such as chemical dissolution are under consideration.
Table 3 shows the comparison of U-TRU-Zr fuel core and TRU-Zr fuel core based on a 300MWe conventional core without blanket. The Doppler coefficient of the uranium-free core is about 1/5 of the uranium-containing core. The sodium void reactivity is reduced in a uranium-free core. However,
the sodium void reactivity increases if countermeasure is applied to enhance the Doppler coefficient. The burnup reactivity swing increases in uranium-free fuel since fissile conversion ratio is almost zero.

Table 4 shows the investigated countermeasures against the technical issues of a uranium-free core design. A number of calculations have been done to obtain the effect of the countermeasures. The engaged core design including the trade-off parameters is shown in Table 5. The assumed reference core layout and fuel subassembly geometry are shown in Figure 9 [10].

**Table 3**: Comparison of U-TRU-Zr fuel core and TRU-Zr fuel core based on a conventional core

<table>
<thead>
<tr>
<th>Core characteristics</th>
<th>U-TRU-Zr</th>
<th>TRU-Zr</th>
</tr>
</thead>
<tbody>
<tr>
<td>Doppler coefficient of fuel nuclide and alloy material (Tdk/dT)</td>
<td>-0.0032</td>
<td>-0.0005</td>
</tr>
<tr>
<td>Sodium void reactivity of core and upper gas plenum (%dk/kk')</td>
<td>2.2</td>
<td>-1.0</td>
</tr>
<tr>
<td>Burnup reactivity (%dk/kk' for cycle length of 148 days)</td>
<td>1.7</td>
<td>6.3</td>
</tr>
<tr>
<td>Delayed neutron fraction, Jeff</td>
<td>0.0035</td>
<td>0.0025</td>
</tr>
<tr>
<td>Prompt neutron life time (μs)</td>
<td>0.34</td>
<td>0.65</td>
</tr>
</tbody>
</table>

Note: 300 MWe fast reactor without blanket

**Table 4**: Technical issues and countermeasures for uranium-free fast reactor core

<table>
<thead>
<tr>
<th>Technical issues</th>
<th>Countermeasures</th>
</tr>
</thead>
<tbody>
<tr>
<td>Decrease of Doppler coefficient</td>
<td>Alternative fuel alloy material with large resonance absorption</td>
</tr>
<tr>
<td></td>
<td>Spectral softening by neutron moderator</td>
</tr>
<tr>
<td>Increase of sodium void reactivity due to countermeasure for decrease of Doppler coefficient</td>
<td>MgAl&lt;sub&gt;2&lt;/sub&gt;O&lt;sub&gt;4&lt;/sub&gt;, BeO</td>
</tr>
<tr>
<td>Increase of burnup reactivity swing</td>
<td>Burnable poison (Absorber mixed with neutron moderator)</td>
</tr>
<tr>
<td></td>
<td>Decrease of core height</td>
</tr>
</tbody>
</table>

**Table 5**: Core design characteristics for parametric calculation

<table>
<thead>
<tr>
<th>Items</th>
<th>Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>Reactor thermal power</td>
<td>714 MW</td>
</tr>
<tr>
<td>Operation cycle length</td>
<td>148 days</td>
</tr>
<tr>
<td>Batch number of refueling</td>
<td>5</td>
</tr>
<tr>
<td>Metal fuel alloy material</td>
<td>Zr (reference) / Mo / Nb</td>
</tr>
<tr>
<td>Fuel smear density</td>
<td>75%TD</td>
</tr>
<tr>
<td>Core height</td>
<td>93 (reference) / 65 / 40 cm</td>
</tr>
<tr>
<td>Core volume</td>
<td>2335 L (constant)</td>
</tr>
<tr>
<td>Core diameter (see note below)</td>
<td>179 cm (reference) - 273 cm</td>
</tr>
<tr>
<td>Fuel pin diameter</td>
<td>cm</td>
</tr>
<tr>
<td>Total number of pins per S/A</td>
<td>331</td>
</tr>
<tr>
<td>Fuel pins / Empty or neutron moderator pins</td>
<td>169 / 162</td>
</tr>
<tr>
<td>Neutron moderator</td>
<td>None / MgAl&lt;sub&gt;2&lt;/sub&gt;O / BeO</td>
</tr>
<tr>
<td>TRU composition</td>
<td>LWR discharged 10 years cooled</td>
</tr>
</tbody>
</table>

Note: core diameter was varied according to core height change keeping core volume constant.
Figure 9: Reference core layout and fuel subassembly geometry

Figure 10 shows the calculated results on Doppler coefficient and sodium void reactivity for various countermeasures. It is found that those reactivity parameters can be roughly correlated with the selected three metrics, i.e. core height, slowing-down power of the composition of the core region, and capture cross section of metal fuel alloy material. In order to more easily find the optimum design point for the target Doppler coefficient and sodium void reactivity simultaneously, a contour map for these reactivity parameters depending on the three metric have been developed as shown in Figure 11. If the target reactivity coefficients are given, the corresponding core design point(s) can be easily found through core height, slowing-down power of the composition of the core region, and capture cross section of metal fuel alloy material.

Figure 10: Calculated results on Doppler coefficient and sodium void reactivity
Regarding burnup reactivity swing, calculations were performed for the mixture of MgAl$_2$O$_4$ (as neutron moderator) and B$_4$C loaded in the empty pins shown in Figure 9. The volume ratio of B$_4$C to (B$_4$C+MgAl$_2$O$_4$) was varied from 0 to 10%. Natural boron was assumed for B$_4$C. As a result, burnup reactivity swing is reduced 27% and 37% for B$_4$C volume ratio of 5% and 10%, respectively. In addition, the calculated burnup reactivity swing for the various countermeasures on Doppler and sodium void reactivity indicated in the above is plotted in Figure 12. It has almost linear relationship to discharged burnup regardless of various countermeasures on Doppler and sodium void reactivity [10].
Figure 12: Relationship between burnup reactivity swing and discharged burnup

Thus, the core design database to develop uranium-free TRU metal fuel core has been produced. We are now working to clarify the target Doppler coefficient and sodium void reactivity to ensure core safety such as UTOP and ULOF events via transient analysis considering the reduction of thermal conductivity of uranium-free TRU metal fuel. Then, we will establish the optimised core using those results.

Doppler measurement of alternative fuel alloys

Measurements of Doppler absorption effect for Zr, Nb and Mo to reduce the uncertainty are now in progress [23]. The experiment uses pulsed neutrons produced by the LINAC facility of Kyoto University Research Reactor Institute. In the experiment, neutron capture rates of the sample at 300K and 600K are obtained by prompt gamma-ray measurement with the time of flight (TOF) method, and the Doppler effects are defined as the ratio of neutron capture rates at 600K and 300K. The measurement apparatus is shown in Figure 13 and the incident neutron spectrum is shown in Figure 14. The measurement method has been confirmed by the preliminary tests in JFY2015. The main measurements are planned in 2016 and 2017.

Figure 13: Doppler measurement apparatus
Figure 14: Incident neutron spectrum
Conclusions

The four years’ research programme on TRU burning fast reactor cycle using uranium-free TRU metal fuel and its current status were presented. This programme will continue until March 2018. At the end of this programme, the feasibility of the uranium-free TRU metal fuel fast reactor cycle will be clarified. According to the current preliminary results, the feasibility of the core design is being clarified, which is one of the most key technical challenges on a uranium-free TRU fuel fast reactor. We continue to move forward on this programme as planned, and the other fundamental issues are expected to be solved by the end of this programme.

Acknowledgements

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References


R&D on heavy liquid metal technology in support of MYRRHA: State of the art

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Abstract

SCK•CEN is at the forefront of heavy liquid metal nuclear technology worldwide with the development of the MYRRHA accelerator-driven system (ADS). MYRRHA is a flexible fast spectrum pool-type research reactor cooled by lead-bismuth eutectic (LBE). Because of its characteristics, MYRRHA is foreseen to be a groundwork facility for international partitioning and transmutation efforts.

In this paper we present the state of the art of the R&D at SCK•CEN on LBE coolant chemistry, thermal hydraulics, ultrasonic techniques and robotic remote handling system components – technologies that are essential for the engineering design and safety of MYRRHA.

An important part of the chemistry R&D in support of MYRRHA focuses on the control and measurement of dissolved oxygen in LBE, the determining species in various safety-related chemical processes. Accurate oxygen control in MYRRHA conditions has now been achieved in several medium to large-scale installations. In addition, extensive studies of the evaporation of safety-critical radioactive elements – such as polonium and iodine – from LBE were carried out.

Two unique thermal-hydraulic facilities have been built at SCK•CEN to qualify the design for licensing and to validate numerical tools. The E-SCAPE facility is a 1/6-scale model of the MYRRHA reactor pool with an electrical core simulator to study forced and natural circulation flow patterns in nominal and accidental conditions. The COMPLPLOT facility represents one core position or in-pile section at full height to test full-scale mock-ups of fuel assemblies, spallation target and control and safety rods.

Ultrasonic based instrumentation systems are proposed to monitor fuel manipulations in the reactor vessel, as visual feedback is not possible due to the opacity of the liquid metal. In the MYRRHA ultrasonic R&D program LBE-compatible and fail-safe fuel identification and lost fuel localisation systems have been developed.

Mechanical components submerged in LBE are used in MYRRHA equipment such as the fuel loading system, control and safety rods, fuel recovery and pumps. RHAPTER is designed to test the behaviour of components like bearings, spur and bevel gears or electrical cables.

Introduction

One of the principal goals of the MYRRHA accelerator-driven nuclear system under design at SCK•CEN is to be a groundwork facility for international partitioning and transmutation efforts, in which it contributes as ADS prototype and flexible irradiation facility. MYRRHA will provide a fast neutron environment for the qualification of materials and fuel for future industrial transmuters. It
will be capable of irradiating minor actinide (MA) fuel samples, but it is also foreseen to house fuel pins or even a small number of fuel assemblies heavily loaded with MAs [1].

In MYRRHA, lead-bismuth eutectic (LBE) functions as both primary coolant and spallation target. The use of LBE and the choice to implement several advanced safety features in the MYRRHA design presents several safety and technological challenges, which have been under investigation at SCK•CEN during the past years.

It is well known that control of the chemistry of the LBE coolant and in particular the concentration of dissolved oxygen is essential to reduce corrosion rates and prevent blockage formation by coolant or impurity oxidation products. In addition, many important chemical processes in the coolant are not well understood or lack sufficiently accurate data to allow predictive calculations [2]. With the conditioning and chemistry programme, SCK•CEN has addressed several technological challenges and scientific gaps, in particular oxygen control, impurity chemistry and release of safety-critical radionuclides from LBE.

An accurate description of the thermal-hydraulic phenomena in the coolant of MYRRHA is essential to assess the performance of several specific MYRRHA safety features and components. On a more fundamental level, there is a need to develop and validate suitable turbulent heat transfer models in heavy liquid metal [2]. Unique, heavily instrumented large facilities have been recently constructed at SCK•CEN that allow detailed thermal hydraulics studies in geometries representative for the primary system globally and core channel specifically.

In MYRRHA, in-vessel manipulations of the fuel and inspection require ultrasonic techniques because of the opacity of LBE towards electromagnetic radiation. A specific technological challenge with the use of ultrasonic transducers in LBE is the proper wetting of stainless-steel acoustic lenses by LBE. An approach to overcome this wetting issue has now been developed, as well as strategies and algorithms for the fail-safe identification of fuel assemblies and their localisation when lost.

Finally, robotic systems will be implemented in MYRRHA to manipulate fuel assemblies while submerged in LBE. A dedicated installation has been developed to test the performance and long-term reliability of robotic components in LBE, including mechanical parts such as bearings and electrical components such as signal transmission cables.

Because of their importance, coolant chemistry, thermal hydraulics, ultrasonics and component testing constitute a significant part of the R&D effort in support of MYRRHA. In the following sections we give an overview of the state of the art of SCK•CEN activities in these fields.

Coolant chemistry

The chemistry of lead-bismuth eutectic coolant involves the reactions of the main coolant constituents Pb and Bi, of dissolved oxygen and of a large variety of impurities, present either already in the non-irradiated LBE, released in the LBE by steel corrosion (Fe, Ni, Cr, Mn) or formed by nuclear reactions with the coolant. In a LBE-cooled ADS such as MYRRHA, critical nuclear reaction products are spallation products with masses close to that of the coolant atoms (such as Hg) and coolant activation products, most importantly Po. Chemical reactions of LBE and its impurities have a strong influence on several safety related processes (Figure 1). Dissolved oxygen has been shown to affect corrosion of steel used as fuel cladding and other components. Reactions of corrosion products may cause the formation of solid particles and deposits, frequently as oxides, and nuclear reaction products may evaporate from the LBE into the cover gas space.
Figure 1: Sketch of the MYRRHA reactor vessel illustrating safety issues caused by chemical reactions in the LBE coolant

Oxygen measurement and control

Within the multitude of possible chemical interactions in the coolant of MYRRHA, oxygen is a central element because of its high chemical affinity with most other elements. At SCK•CEN, the project on oxygen control is a continuous effort, with the aim to develop systems that are able to accurately measure and control the oxygen concentration in the LBE of MYRRHA to avoid coolant oxidation and reduce corrosion.

Oxygen sensors capable of measuring accurately down to 200 °C have been developed [3,4]. Oxygen control systems based on gas-LBE interaction, and controlled dissolution of lead oxide in a so-called PbO mass exchanger (PbO MX) have been evaluated experimentally and numerically [5-7]. A new approach towards oxygen control in LBE called electrochemical oxygen pumping (EOP) has been developed [8] and tested in various experimental conditions.

LBE chemistry in large systems has been studied at SCK•CEN in the MEXICO, HELIOS 3 and CRAFT installations. MEXICO is a pilot-scale non-isothermal loop with an LBE inventory of 7 tonnes, where cold and hot leg temperatures are representative for MYRRHA, i.e. ~200 °C and ~400 °C, respectively (Figure 2). At present MEXICO reached 10,000 h of operation, during which it has been used for oxygen control, lead oxide deposition, and filtering experiments (Figure 3). Accurate oxygen control in MEXICO has been achieved using the PbO MX technology (Figure 3, inset) [9] and electrochemical oxygen pumping (4-1 in Figure 3). Planned experiments include a study of lead oxide deposition and possible associated increase of the pressure drop in a fuel pin bundle. Such experiments are only feasible because of the very accurate control and measurement of dissolved oxygen in both hot and cold legs of the MEXICO loop.
In parallel, several small setups are used to test new designs of oxygen sensors and to perform well-controlled experiments for the measurement of key chemical properties of dissolved oxygen in LBE, such as the Sieverts constant which describes the equilibrium of dissolved oxygen with oxygen gas.

**Impurity management**

Corrosion products are expected to be released continuously in the LBE of MYRRHA in amounts that can potentially cause blockages of narrow flow paths inside the primary system. To assess the behaviour of corrosion products in LBE, the thermodynamics and kinetics of solids-forming reactions is studied and as well as the implementation of these chemical reactions in CFD models to predict solids formation, transport and deposition in the primary system. These models will be used for safety calculations and to engineer LBE filter systems for MYRRHA. Recently, for example, the reaction between dissolved iron and oxygen in liquid LBE has been characterised quantitatively and was found to have a strong impact on the local oxygen concentration and precipitate formation in the CRAFT corrosion loop [10].
**Figure 3**: Oxygen concentration history of the MEXICO loop. Each section represents a different experiment. Inset: enlarged section 3-1, in which accurate oxygen control by the PbO MX technology was demonstrated. Cold and hot legs were at 210°C and 430 °C, respectively [9]

**Fission and spallation product evaporation**

Volatile fission products (I, Cs, Te, ...) may be released into the LBE coolant after a breach in the fuel cladding. As input for safety calculations, the release of fission products from LBE needs to be known quantitatively under various MYRRHA relevant conditions and scenarios. Iodine evaporation from LBE has been characterised by the so-called transpiration method. A more advanced system to characterise fission product evaporation from LBE using stable isotopes has been constructed. This system consists of a cell containing an LBE sample doped with fission products. The gas atmosphere in this cell can be sampled and characterised by a mass spectrometer coupled to the evaporation cell by a high temperature transfer line.

Spallation products are unique to ADS systems in which coolant and spallation target form a single entity. One important spallation product is mercury, which is both volatile and produced with high yield. In the frame of MYRRHA, mercury evaporation from LBE has now been characterised using a dedicated setup [11].

**Polonium chemistry in LBE**

The presence of 55% bismuth in the LBE leads to the production of relatively large quantities of polonium-210 during operation of MYRRHA. The chemistry of polonium and especially its release has been one of the focus points of the conditioning and chemistry programme at SCK•CEN. Using the transpiration method, the release of polonium from dilute solution in LBE has been quantified under various experimental conditions [12-15]. Recent important results obtained in collaboration with the Paul Scherrer Institute in Switzerland include the characterisation of adsorption (sticking) of polonium-bearing vapour molecules on surfaces, and the derivation of the corresponding thermochemical interaction properties. The latter results are used in safety studies to assess transport of polonium vapours, after their release from LBE, in various components of MYRRHA such as the proton beam tube.
Thermal hydraulics

The use of passive systems to achieve safety functions is one of the guiding principles of the MYRRHA design, embedded in the MYRRHA safety approach. Natural circulation in the primary system is the chosen option for the design in terms of decay heat removal. The demonstration that this is viable is indispensable to support this approach. Next to passive decay heat removal, the understanding of the thermal-hydraulic phenomena occurring in the upper and lower plena of the reactor pool is a critical issue in the design of the MYRRHA system. A good knowledge of convection patterns, flow mixing and stratification in operational and accidental conditions (e.g. partial or complete loss of flow, loss of heat sink) etc. is absolutely necessary. Model experiments are necessary for understanding the physics, for validating numerical tools and to qualify the design for the licensing.

The E-SCAPE (European SCAled Pool Experiment) facility is a thermal hydraulic 1/6-scale model of the primary system of the MYRRHA reactor, with an electrical core simulator, cooled by LBE. It provides experimental feedback to the designers on the forced and natural circulation flow patterns. Moreover, it enables to benchmark and validate system thermal hydraulic (STH) and computational fluid dynamics (CFD) codes for their use with LBE. The facility is currently in commissioning.

Replicas of all main components of MYRRHA are placed in the main vessel in order to maintain a geometric similarity. The main vessel outer diameter of E-SCAPE is 1400 mm. The total LBE inventory is 2.5 m³. Total LBE mass-flow rate can amount to 120 kg/s; temperature range is 200°C-340°C. E-SCAPE is heavily instrumented to allow detailed characterisation of the thermal-hydraulic phenomena in view of the validation of numerical codes:

- [13] 300 thermocouples determine temperature distributions and identify possible stratification
- [14] pressure sensors measure absolute and/or differential pressures; also levels and levels differences can be deduced through these
- [15] a traversable Pitot tube characterises the velocity profile at the pump tube outlet
- [16] UDV (Ultrasonic Doppler Velocimetry) sensors in 45 positions characterise the velocity

The design of the facility involved an in-depth scaling study based on non-dimensional analysis, confirmed by STH and CFD modeling of the complete system [16]. The computational models were also used to predict the transient behaviour of the system and to determine the optimal positioning of instrumentation. Figure 4 presents an isometric view and the as-built condition of the E-SCAPE LBE facility [17].

E-SCAPE will work mainly in two conditions: in forced convection (with the pumps on) or in natural convection (by means of buoyancy forces related to temperature differences in the loop). Transient test scenarios involve pumps running, shut down or in coast down (loss of flow); heat exchangers being active or excluded (loss of heat sink); powers being reduced or increased.
Next to pool thermal hydraulics, knowledge of the hydraulic and hydrodynamic behaviour of individual reactor core components is also of primary importance in the design and licensing process.

The COMPlot LBE facility is a closed-loop facility, designed to characterise the hydraulic behaviour of various full-scale MYRRHA components in LBE [18]. The system is capable of circulating a maximum flow rate of 36 m$^3$/h of LBE. The loop is isothermal, meaning it operates at a constant LBE temperature for a given test, although the temperature can be varied up to a maximum of 400°C to investigate temperature effects. Figure 5 illustrates a schematic of the COMPlot loop with the constituent components and the as-built condition of the COMPlot LBE facility.

The core components of interest and the relevant hydraulic phenomena are listed as follows:

- Fuel assembly – Pressure drop and flow induced vibration
- Spallation target – Pressure drop and LBE velocity profile at the beam window
- Control rod – proof of operating principle and rod insertion time
- Safety rod – proof of operating principle and rod insertion time
Each of these components are full-scale mock-ups of the MYRRHA design and are mounted into a specially designed test section, representative of a single MYRRHA reactor (core) channel and suitably instrumented for the intended experiment.

Innovative double-walled bayonet-tube heat exchangers with leak monitoring, submerged in the reactor pool, remove the power generated in to core towards a secondary steam/water system. Thermal and mechanical behaviour, reliability and safety of such a system have to be experimentally confirmed.

For this purpose, the COMPLOT facility will be upgraded in 2017 to host a new test section containing a single bayonet tube of the double-wall heat exchanger at full length. A 100 kWth LBE-heater will be added to COMPLOT and a secondary water/steam cooling circuit at 16 bar will be joined to the facility in order to provide the corresponding cooling capacity at conditions representative for the MYRRHA reactor.

**Ultrasonic techniques**

The opacity of the LBE coolant in MYRRHA necessitates the development of ultrasonic techniques to provide feedback during in-vessel fuel manipulations as regular camera based inspection systems are unusable in liquid metal cooled reactors. In this respect, ultrasonic measurement techniques have been proposed and even developed in the past for operation in sodium-cooled reactors. No such systems have ever been deployed in HLM cooled reactors and SCK•CEN is the first to have a research programme in this direction.

An extensive survey on the acoustic properties of reactor coolants shows that LBE is in many ways superior to the properties of liquid sodium and water, resulting in simpler transducer designs and longer detection ranges while being less disturbed by temperature gradients inside the liquid [19]. This allows us to leverage on the results of half a century of ultrasonic research for sodium-cooled reactors. Moreover, as the speed of sound in water, LBE and sodium are comparable, new and innovative ultrasonic techniques can first be tested in water before their final validation in hot liquid LBE. However, the high-surface tension of LBE combined with the naturally occurring oxide layer on the stainless-steel acoustic lenses of the ultrasonic sensors prevents proper wetting with liquid metal, which drastically decreases the performance of the overall ultrasonic system. This wetting issue has been solved by the development of a sacrificial coating that prevents oxide formation on the lens prior to submersion in liquid metal [19].

**Ultrasonic visualisation in LBE**

In order to prove the feasibility of ultrasonic instrumentation in LBE we have done an experiment where a properly wetted ultrasonic transducer (5 MHz, 8 mm diameter, bismuth titanate piezo) is scanned over a submersed stainless-steel object to render the first ever ultrasonic image in LBE [19]. Figure 6 shows the results of this experiment. An area of 130x120 mm was scanned in a grid-like pattern with 1 mm step-size at a distance of approximately 11 cm from the top of the submerged object. The visualised object is a stainless-steel mockup of the inflow nozzle of a MYRRHA fuel assembly (Figure 6, left). A unique combination of "fuel identification" marks have been machined at the top edge of the nozzle. Additionally, two stainless-steel bars with 100 µm standoff distance have been added to simulate a cracked surface. The centre image visualises the amplitude of the strongest ultrasonic reflection. The right image shows the depth of the strongest reflection where the white areas indicate the locations where the object is closest to the ultrasonic sensor. The experiment was done in an oxygen saturated pool of LBE at 200°C.
For certain tasks in MYRRHA full visualisation is not strictly required and will only result in excessively long scanning times without having an added value. In MYRRHA we foresee to have two dedicated non-visualising ultrasonic systems: the "Ultrasonic fuel identification system" and the "Ultrasonic lost fuel localisation system" as described in following sections.

**Figure 6**: Illustration an ultrasonic visualisation experiment in LBE at 200°C [19]

The ultrasonic fuel identification system

Each MYRRHA fuel assembly will carry a unique pattern of 12 notches of different depths that are machined at the inflow nozzle of the fuel assembly. A full ultrasonic visualisation of the pattern of the notches to identify a fuel assembly is clearly overkill and only hinders the real-time operation that is required during the fuel reloading process. Therefore it is proposed to use 12 dedicated ultrasonic transducers mounted on the fuel manipulation device in such a way that each of them is positioned right above an edge of the identification notches. Figure 7 depicts a test of this system in water. A single ultrasonic pulse on each of the transducers allows readout of the depths of the notches and nearly instant identification of the fuel assembly. Proper implementation of the code with integrated error correction techniques makes this system robust against reading errors or failing transducers. More details on the fuel identification system can be found in a previously published paper [20]. Since this paper, further research has resulted in an updated version of the code, with improved error correcting capabilities. The improved code is able to correctly identify a fuel assembly, even when five out of the 12 fuel identification transducers are faulty. The details of the new code are subject of a dedicated paper that is currently in its editing phase.
**The ultrasonic lost fuel localisation system**

Localisation of objects that are lost in the reactor vessel do not require a detailed visualisation as we can make use of the *a priori* known size of the lost object and optimise the search algorithm. Rough object localisation can be done by using a grid-like scanning path that is fine enough to not miss the object, but coarse enough to speed up the scanning process. Once located, a detailed inspection renders more information on the orientation and state of the object. A similar strategy will be used in the unlikely event that a wrong fuel manipulation action has resulted in a fuel assembly being lost in the lower plenum of the reactor vessel. Due to the high density of LBE, the assembly will be found floating just underneath the diaphragm that separates the hot upper plenum from the cold lower plenum. In order to ease the fuel recovery process, the diaphragm is designed as flat as possible without any protrusions. This forces the fuel assembly to float horizontally under the diaphragm, thus simplifying the recovery and detection process. For the localisation of the lost assembly, we propose to use a scanning system that moves a single ultrasonic transducer in a plane parallel to the diaphragm. The transducer measures the distance to the diaphragm for each scanning position. By proper alignment of the transducer, it is possible to receive the high intensity specular echo of the diaphragm. The reflected pulse can then be compared with what is expected from the known location of the diaphragm. Anomalies in reflected pulse arrival time or pulse amplitude indicate the presence of an object which is blocking the sound path between transducer and diaphragm. This proposed algorithm has been validated in a dedicated water setup and the results have been presented at the ANIMMA2015 conference in Lisbon [21].

**Testing of robotic remote handling system components**

The MYRRHA design requires the fuel assemblies to be loaded in the liquid LBE from below. A combination of robotic systems transports the fuel from the top of the reactor to the correct position in the core. Due to the particular nature of the reactor, there is little test data available for robotics component properties and failure. Thus one of the central goals of the remote handling research is to test all potential components for the fuel manipulation system (i.e. bearings, springs, electrical cables) to gain an overview of the possibilities of working in LBE under the specific circumstances present in MYRRHA.
Testing of remote handling components is performed in RHAPTER, the Remote HAndling Parts TEst Rig at SCK•CEN (Figure 8). The test rig consists of a vessel containing liquid LBE in which different test modules can be submerged, with external drive and load motors, a vacuum and cover gas system and diagnostic instrumentation such as accelerometers and torque sensors. Test modules have been developed for ball bearings, electrical cables and journal bearings.

Figure 8: The RHAPTER test rig

The facility has been in use since 2011, with the main focus on ball bearings (Figure 9) and springs for remote handling operations. This has resulted in bearings which operate for a number of motions equivalent to over 50 years of service in MYRRHA. For springs, fatigue and relaxation tests resulted in a design envelope for reliable operation in all operational conditions.

The current research expands these results to long-term reliability tests for ball bearings and to other components like electrical cables and sliding bearings. Electrical cables that connect instrumentation for fuel identification and lost fuel localisation (cf. previous section) need to comply with all the movements of the fuel manipulation robot. Mechanical systems that allow this kind of motions have been developed and are currently in the final validation phase. Sliding bearings such as journal and sleeve bearings are used in a number of places in the fuel manipulation system and are under development.

Figure 9: Different ball bearings after testing in LBE
In a next phase, test modules for axially loaded bearings (thrust bearings), combined axial-radial loads and for gear transmission system will be developed. A second RHAPTER facility will be constructed for high-speed pump bearing tests. In addition to the component tests, integrated tests of the fuel manipulation system will be built to demonstrate the operation of the system under MYRRHA conditions, using scaled models of the fuel manipulation robots.

Conclusions

SCK•CEN’s extensive R&D programme on LBE technology has resulted in the construction of several large testing facilities, many of which are one-of-kind. Installations such as the chemistry loop MEXICO, and the component test rig RHAPTER have already operated for a significant time and produced a large number of results that contributed to solving technological and safety issues during the development of the MYRRHA ADS. Specific technologies such as coolant oxygen control and ultrasonic techniques have now reached a high degree of maturity. Future work will include the start of the experimental programme in the E-SCAPE facility focusing on pool thermal hydraulics, an upgrade of the COMPLIT facility to host a section to test double-walled heat exchangers, and the development of new component test facilities. In the field of coolant chemistry, an experimental campaign in which the blockage of a fuel assembly mockup by oxides will be studied, is one of the priorities.

Acknowledgements

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References


MATADOR: An innovative sub-assembly design for heterogeneous minor actinides transmutation

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Abstract
In the frame of next generation fast reactor design, the minimisation of minor actinide production is one of the key objectives for current R&D. At CEA, studies focused on sodium-cooled fast reactors (SFR) are being conducted with the intent to identify the advantages and immediate challenges for near-term industrial deployment. One option is to consider minor actinide multirecycling in a homogeneous way (in the fuel assembly). Unfortunately, the strong impact of minor actinides on sodium void coefficient allows only a small fraction of the total heavy nuclei mass to be loaded with minor actinides in the core (~2-3%vol).

An alternative consists of inserting a larger amount of minor actinides in some specific locations such as inner core targets, radial, and axial blankets. These locations are to be optimised in order to lower the impact on the core standard management. In this framework, multirecycling management based on minor actinides-bearing blankets (MABB) has been widely studied as a promising solution concerning the minimisation of feedback coefficients degradation. This concept enables the total decoupling of the management of the fuel and actinides by putting minor actinides in radial blankets in a depleted UO$_2$ matrix. It enables high resident time for minor actinides which allows significant transmutation rates. However, the maximum initial minor actinide content in the MABB is driven by the maximum decay heat level allowed by back end fuel cycle technologies (irradiated S/A washing and transport limits). Another limiting factor comes from the constraint on the maximum DPA rate seen by pins near the core/MABB interface that put restrictions on the irradiation time.

In order to satisfy these limitations, a new design (MATADOR) is presented here. It is based on an axially-segmented structure that consists of superimposed fuel discs. Each elementary disc has an external structure (steel or ceramic) that contains granular fuel (coated particles, sphere-pac, ...). These discs have dedicated channels which allow coolant flow and power extraction with the capability to rotate in situ to smooth the flux gradient during irradiation. Using such modular design, the decay heat of the elementary object to be handled is reduced by a factor ten compared to a traditional monolithic pin-bundle subassembly design. This major breakthrough could be used to increase the initial minor actinides content and thus to lower the associated fuel cycle inventory in advanced transmutation strategies.

Introduction
The nuclear reactors of next generation have to demonstrate their capacity to reduce the rate of their used nuclear fuel (UNF) production as well as those of existing reactors (PWR). The harder neutron spectrum of GenIV reactors can be utilised to facilitate the transmutation of minor actinides
(MA), which are fissile in the fast neutron spectrum. Among the many possible strategies, two main approaches of MA multirecycling are being studied at CEA:

- The homogeneous mode, in which the actinides are diluted in fuel
- The heterogeneous mode, in which the actinides are introduced in dedicated assemblies in core or periphery as minor actinides-bearing blankets (MABB) allowing decoupling their management from the standard fuel.

Within this framework, it is necessary to evaluate the transmutation capacity of these two approaches for current sodium-cooled fast reactors (SFR) being studied at CEA in the frame of the French fleet scenario [1]. The corresponding MA transmutation rate and consumption (or mass balance) are of major importance regarding industrial issues such as heavy nuclei inventories, fabrication, material reprocessing, and the evaluation of the space required in a nuclear waste repository.

From a core safety point of view, the heterogeneous MA transmutation approach offers large flexibility in SFR core design since it does not significantly impact important core feedback coefficients such as sodium thermal expansion or the Doppler constant. This transmutation mode has been widely studied in the frame of the French Nuclear Waste act and optimisations are being performed in order to obtain a subassembly design that can comply with MABB specifications. The basic subassembly design relies on a “traditional” pin bundle located inside a hexagonal wrapper. Inside those pins, unitary uranium and americium oxide ceramic pellets are axially stacked to form a metre long active zone.

Minor actinide transmutation performance has several limitations:

- Manufacturing: based on current fuel manufacturing processes, the maximum achievable amount of minor actinide content without imposing major modifications of shielding level in uranium/minor actinides fuel mixtures is considered to be 20% by volume.
- Pin behaviour under irradiation: mechanical stress on irradiated cladding material and adherence to an acceptable margin of fuel melting are of major importance. Helium production coming from americium and curium alpha decay is the main contributor to pin pressurisation and consequently the americium content and the irradiation time need to be tuned to meet failure-safe mechanical criteria.
- Back-end fuel cycle: once discharged from the SFR core, MABB subassemblies have to be cooled in dedicated pools until they are able to be sent for reprocessing. The decay heat thresholds corresponding to in-core subassembly handling, washing, storage, and transportation must be adhered to with actual or expected industrial capabilities [2] as well as with the timescale of MA transmutation scenarios. Once again both americium content and irradiation time need to be tuned to meet these requirements.

Among all of these limitations, the decay heat thresholds are seen as the most stringent so the associated initial content of minor actinides is lowered from 20% to 10% for americium MABB in the current “industrial” version of such subassemblies which leads to large minor actinide inventories in the associated transmutation scenario.

A way to enhance transmutation capability could be found in the subassembly design: as decay heat threshold focuses on an “elementary object” to be handled, we need to go beyond a “monolithic” subassembly design to a “removable” subassembly design in which unitary parts could exhibit lower decay heat levels when compared to their monolithic counterparts. In one way, pins
themselves could be seen as candidates of such unitary objects. However, the typical size of fuel pins (2 metres high with ~1 cm diameter) makes them not so convenient to unpack from a MABB subassembly and to handle in a safe way from mechanical point of view. This sort of “radial” splitting of the subassembly could be done by gathering inner subassemblies in a hexagonal wrapper but it leads to a somewhat prohibitive reduction of the fuel content of the subassembly, which does not benefit MA transmutation performance.

Another way to achieve this relies on an axial “splitting” of the subassembly design. This is the purpose of the minor actinide transmutation advanced device for optimised reprocessing (MATADOR) concept. It is based on axially-segmented structure that consists of fuel discs superposition. Each elementary disc has an external structure (steel or ceramic) that contains fuel under granular form. Those discs have dedicated channels passing through for the coolant flow and power extraction and are able to rotate in situ to smooth the DPA gradient during irradiation.

The MATADOR concept

Conceptual design

The MATADOR concept is based on the following characteristics:

- Analogously to the pin, the first containment barrier is made of a sealed hollow cylinder crossed by numerous coolant channels. The height of this cylinder is small compared to its diameter so that it could be compared as a thick “disc”. The disc has a central channel.

- The fuel is granular in order to fill the inner part of the previous “disc”. The fuel nature is generic; it could be oxide, carbide or metallic fuel alloy. The granular form could be either sphere, powder, grains or coated particles. The remaining part of the inner disc is filled with helium just as in pin design.

- Discs can be added axially to be put into hexagonal wrapper just like pin bundle.

- The disc stack is maintained by central rod inserted in the central channel. This central rod does ensure an in situ rotation of the disc stack in order to smooth the flux as well as damage dose in the radial direction during irradiation. This helps reduce the maximal damage rate of the subassembly.

A radial and axial cut of the disc design is presented in the Figure 1. If the disc concept is close to prismatic design often used in High Temperature Reactor design, it differs by the fact that the fuel is not diluted in an inert matrix. Here, the disc is only a container that can be filled by fuel or a mixture of fuel and other dedicated material such as moderator or absorber. As the inner shape of the disc is quite complex, the suitable fuel form is grain-like in order to be able to fill the inner cavity in the most efficient way. This offers great flexibility in terms of volume fraction since granular fuel could be either spherical [3], composed of particles like in HTR designs [4], granular similar to the VIPAC concept [5] or powder. Using such fuel form leads to a large free volume that is needed to accommodate the fission gas release during irradiation. As each disc remains closed, initial heavy nuclei content can be tuned to a desired value according to irradiation configuration (flux level, irradiation time…) and foreseen transmutation performances. The number, size and locations of the coolant channels have to be settled in order to keep an acceptable margin on fuel maximum temperature all along irradiation time.
Instead of pin bundle, the inner part of the wrapper of the MATADOR subassembly gathers an axial accumulation of elementary discs described below. To do so a central steel rod is used to maintain the disc stack and extra external rods help keep the coolant channels aligned between the different discs. The disc stack and central rod rely on a lower rotating grid support. On the upper part of the subassembly another rotating grid is used as support for neutron shielding device.

When MABB are located on core periphery, damage dose accumulates near the fuel/MABB interface due to the inherent flux gradient. A solution would be to rotate the subassembly by 60° or 180° at specific time scale (for instance, 180° at mid life). For traditional pin-bundle design, it means handling and moving the subassembly to another position at each end of cycle, which increases the core shut-down delay. With the MATADOR concept, the disc stack design ensures the possibility of in situ rotation (without handling stage). It only requires a dedicated and adapted rotating handling device. Several solutions have been proposed in order to prevent disc rotation during irradiation and their designs are in progress at CEA.
At the end of irradiation and after some cooling time, the subassembly can be unpacked into elementary discs with low unitary decay heat level to be transported to reprocessing stage. This drop in the decay heat level helps minimise the time gap between the end of irradiation and the reprocessing compared to a monolithic subassembly design. Furthermore, the compactness of the disc design makes them easier to transport in a dedicated device rather than large subassemblies.

- **MATADOR concept vs MABB pin bundle design**

  Compared to the pin-bundle MABB design, the MATADOR concept brings promising solutions to minor actinide transmutation:

  - It gives a new flexibility on the axial composition of the fuel (several content zones and/or fuel volume fractions...)
  - The in situ rotation capability helps evenly distribute the natural damage rate across the subassembly as well as the peaking fuel temperature with minimal subassembly handling.
  - The maximum decay heat level of the unitary disc is far below the one of the “monolithic” subassembly which allows it to be compatible with the different thresholds associated to the washing and transportation stages. The gain in cooling delay helps minimise the minor actinide inventory over the fuel cycle.
  - Once unpacked, the compactness of the unitary disc makes it easy to handle.

  This concept is faces numerous technological challenges, among them the manufacturing process of the disc and fuel, the cooling capability of the subassembly, the unpacking process, etc. Beyond the technological issues, simulation capabilities are also questionable since this new design is far from the traditional assembly geometry for which calculation codes where primarily developed.

  At this preliminary stage, the MATADOR still needs a full cross-discipline design in order to quantify realistic transmutation performances. Such a process is currently in progress at CEA and needs to be completed. However, some basic inputs can give some perspective on the typically expected characteristics:

  - In order to get acceptable transmutation performances, the volume fuel fraction across the subassembly has to be close to what is expected for the corresponding traditional pin-bundle design. At least 30%vol is expected to make it viable.
  - The inner volume of the disc has to be large enough to meet the previous requirement. As it is filled by granular fuel, the typical size of the grain-like particle can be tuned to achieve between 66% to 85% of the corresponding volume.
  - As MABBs do experience quite low power levels compared to fissile subassemblies (roughly 1 MW at the end of irradiation), the volume fraction of coolant can be as low as 20% without significant impact on fuel maximum temperature. The channel number and typical size still need to be obtained accurately to have an acceptable margin with regard to fuel melting.
  - As helium production of such minor actinide bearing fuel is quite large during irradiation, the structural material of the disc has to face high internal pressures. Advanced steel or ceramic-like design need to be developed.

  Starting from a widely studied SFR V2B core, an illustration of a preliminary image of a MATADOR design is given in the following section.
MATADOR preliminary design

- Application to SFR V2B core

Starting from an existing pin-bundle design of a minor actinide-bearing blanket (MABB) [6], a preliminary image of a MATADOR subassembly has been derived. The reference core is based on SFR 3600 MWth design (named SFR V2B) that has been widely described in the past [7].

The design of the fuel assembly is based on a concept with large pins (10.73 mm diameter) and a spacer wire of small diameter (1 mm). The performances of the core reach a burnup of 100 GWd/t with a residence time of fuel of approximately 2 000 effective full-power day (EFPD) and a near zero internal breeding gain (IBG), this last point being an objective of design. Since the tightening of the pin bundle in the fuel assembly requires a cladding material which does not swell significantly while reaching the expected burnup, the material considered is an advanced ferritic steel (ODS).

<table>
<thead>
<tr>
<th>SFR V2B – main characteristics</th>
<th></th>
</tr>
</thead>
<tbody>
<tr>
<td>Power (MWth/MWe)</td>
<td>3600/1450</td>
</tr>
<tr>
<td>Power density (W/cm³)</td>
<td>207</td>
</tr>
<tr>
<td>Number of fuel elements (inner/external)</td>
<td>453 (267/186)</td>
</tr>
<tr>
<td>Number of MABB</td>
<td>84</td>
</tr>
<tr>
<td>Life time</td>
<td>5 × 410 = 2050 EFPD</td>
</tr>
<tr>
<td>Fuel</td>
<td>(U,Pu)O2</td>
</tr>
<tr>
<td>PuO2/(PuO2+UO2)</td>
<td>14.7</td>
</tr>
</tbody>
</table>

The reference core design is presented in Table 1 and Figure 3. It consists of:

- 453 fuel assemblies divided into two different Pu content zones (yellow and red S/A)
- 18 control rods and 9 shut-down rods (dark and gray S/A)
- A provision of 84 MABB (magenta S/A) positioned in the first external row beyond the fissile core. In the reference configuration these assemblies are replaced by steel assemblies.
- 2 additional rows of radial reflectors (light green S/A)
- 2 rows of radial neutron shielding zones (absorbers, green S/A).
Figure 3: Radial cut of the SFR V2B core

At the core periphery, the average flux level is four times lower than the fissile core. Thus the irradiation time of the MABB has been set to defined as $4 \times 10^3$ EFPD. The current design of the MABB for this application leads to a reduced pin size with an adapted gas plenum height which differs from the reference fissile pin. This change enables the requirements on pin pressurisation for the corresponding irradiation time to be met. The pin characteristics are summarised in the Table 2.

Table 2: Fuel and MABB pin description

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Fissile pin</th>
<th>MABB pin</th>
</tr>
</thead>
<tbody>
<tr>
<td>Pellet diameter (mm)</td>
<td>9.43</td>
<td>7.34</td>
</tr>
<tr>
<td>Pin diameter (mm)</td>
<td>10.73</td>
<td>8.64</td>
</tr>
<tr>
<td>Clad thickness (mm)</td>
<td>0.50</td>
<td>0.50</td>
</tr>
<tr>
<td>Spacing wire diameter (mm)</td>
<td>1.00</td>
<td>1.23</td>
</tr>
<tr>
<td>Number of pins</td>
<td>271</td>
<td>331</td>
</tr>
<tr>
<td>Active zone height (cm)</td>
<td>100</td>
<td>100</td>
</tr>
<tr>
<td>Gas plenum height (cm)</td>
<td>95</td>
<td>155</td>
</tr>
<tr>
<td>Fuel volume fraction (%)*</td>
<td>42.7</td>
<td>38.4</td>
</tr>
<tr>
<td>Fuel</td>
<td>UPuO$_2$</td>
<td>UAmO$_2$</td>
</tr>
</tbody>
</table>

*Theoretical density

Three preliminary designs have been proposed based on very basic evaluations which adhere to the objectives of the MATADOR concept. The main assumptions were the following:

- Tuned granular fuel form to enable high smearing factor (at least 2/3 of the inner volume)
- Identical structural thickness across the disc
- Large number of coolant channels across the disc
- Reduced disc height in order to lower the internal pressure during irradiation

The associated disc parameters are summarised in Table 3. Among all possibilities, a quite large disc thickness has been chosen considering predicted stress levels at the end of irradiation. The corresponding value is ranging from 1.0 to 2.0 mm compared to 0.5 mm for ODS cladding in the
current MABB pin design. Obviously, accurate mechanical studies are needed to determine a realistic value.

As there is a need of a very low decay heat level, the unitary disc size has to be small enough to exhibit a compromise between initial fuel and structural mass. The disc height has been set to 2.5 cm leading to 40 unitary objects for each subassembly in the frame of the SFR V2B core implementation. The preliminary designs presented here are an attempt to obtain boundary values for fuel volume fraction across the active zone of the MATADOR subassembly (see Table 3):

- The “Optimistic” option relies on a “reduced” steel (or ceramic) thickness and a maximum fuel smearing factor leading to roughly 41% fuel fraction.
- The “Conservative” option considers a large value for the steel thickness and a low value for the smearing factor. The fuel fraction drops to 30%.
- The “Medium” option stands in between with 36% fuel fraction close to the current value of the MABB pin bundle design.

All designs show significant coolant fraction that seem quite sufficient for power extraction.

**Table 3: Main characteristics of preliminary MATADOR discs**

<table>
<thead>
<tr>
<th></th>
<th>Medium</th>
<th>Conservative</th>
<th>Optimistic</th>
</tr>
</thead>
<tbody>
<tr>
<td>Inner disc fuel smearing factor (%)</td>
<td>75</td>
<td>66</td>
<td>80</td>
</tr>
<tr>
<td>Disc thickness (mm)</td>
<td>1.5</td>
<td>2.0</td>
<td>1.0</td>
</tr>
<tr>
<td>Disc height (mm)</td>
<td>2.5</td>
<td>2.5</td>
<td>2.5</td>
</tr>
<tr>
<td>Coolant channel</td>
<td>20</td>
<td>20</td>
<td>20</td>
</tr>
<tr>
<td>Coolant fraction (%)</td>
<td>28.46</td>
<td>26.97</td>
<td>30.04</td>
</tr>
<tr>
<td>Disc structure fraction (%)</td>
<td>13.55</td>
<td>17.81</td>
<td>9.17</td>
</tr>
<tr>
<td>Wrapper fraction (%)</td>
<td>8.17</td>
<td>8.17</td>
<td>8.17</td>
</tr>
<tr>
<td>Fuel fraction (%)</td>
<td>36.24</td>
<td>30.12</td>
<td>40.83</td>
</tr>
<tr>
<td>Void fraction (%)</td>
<td>12.45</td>
<td>16.00</td>
<td>10.52</td>
</tr>
<tr>
<td>Disc « void » volume (cm³)</td>
<td>112</td>
<td>154</td>
<td>101</td>
</tr>
</tbody>
</table>

**Main characteristics of the “Medium” design**

The transmutation performances of the “Medium” design have been compared to those of the MABB reference design. The calculations have been carried out with the CEA reference codes:

- ERANOS [8] for neutronics, based on JEFF3.1 cross section library [9]. The core depletion used a RZ modification and averaged depletion option.
- DARWIN [10] for comprehension isotopic depletion and decay heat characterisation

Both 10% and 15% americium (75% Am²⁴¹, 25% Am²⁴³) content were used for MABB and MATADOR design loading. Irradiation time was twice that of the fuel, i.e. 4 100 EFPD. The mass balance of the average subassembly is summarised in Table 4. As one can see, the americium transmutation performances are quite similar for both concepts.
### Table 4: Main characteristics of preliminary MATAODR discs

<table>
<thead>
<tr>
<th></th>
<th>MABB pin</th>
<th>MATADOR- Medium</th>
<th>MABB pin</th>
<th>MATADOR- Medium</th>
</tr>
</thead>
<tbody>
<tr>
<td><strong>Initial loading</strong></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Am content (%)</td>
<td>10</td>
<td>15</td>
<td>10</td>
<td>15</td>
</tr>
<tr>
<td>Heavy nuclei mass (kg)</td>
<td>141.19</td>
<td>134.60</td>
<td>141.19</td>
<td>134.60</td>
</tr>
<tr>
<td>Am initial mass (kg)</td>
<td>14.13</td>
<td>13.46</td>
<td>21.18</td>
<td>20.20</td>
</tr>
<tr>
<td><strong>End of Irradiation (4 100 EFPD)</strong></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Am transmutation rate (%)</td>
<td>44.5</td>
<td>44.5</td>
<td>41.8</td>
<td>42.1</td>
</tr>
<tr>
<td>Am mass balance (kg)</td>
<td>-6.28</td>
<td>-5.98</td>
<td>-8.87</td>
<td>-8.50</td>
</tr>
</tbody>
</table>

The time dependence of the decay heat level at the end of irradiation is also very close with only a 1% maximum difference after ten days of cooling. The decay heat level of the 10% and 15% Am content MATADOR “Medium” design are displayed on Figure 4. Plain curves refer to whole subassembly values whereas hashed curves refer to maximum values of the unitary discs. Disc and subassembly curves show the same trends, the only differences are in the absolute decay heat levels which are 38 times lower for the unitary discs.

**Figure 4: Radial cut of the SFR V2B core**

If we assume a 7.5 kW threshold on S/A decay heat for the subassembly washing stage, a MATADOR-like subassembly could be handled out of the sodium pool storage between 500 and 1,000 days depending on the initial americium content. If we assume a 3 kW threshold for transportation purposes, a MATADOR-like subassembly would have to wait for 4,200 days for 10% Am content and up to 11,000 for 15% content before it is compliant. Now, if there is a possibility to unpack the subassembly into elementary disc, the limiting decay heat level drops below 300 W which gives a comfortable margin for transportation issues.

Beyond the technological R&D needs of manufacturing the in situ rotation and the unpacking process of the MATADOR design, the major concern of this concept relies on the end of irradiation disc pressurisation level. Since the available “free” volume to accommodate the released helium and fission gas during irradiation is quite reduced compared to a pin design, the disc has to “survive” to large axial and/or radial mechanical stresses due to an internal pressure that may be as high as...
300 bars. Further optimisation based on best-estimate calculations will be needed to obtain an adapted disc design and assess the feasibility of the concept.

Conclusions and perspectives

The heterogeneous minor actinides transmutation in fast reactors, in which the actinides are introduced in dedicated assemblies in the core or periphery (minor actinides-bearing blankets) permits large flexibility in terms of fuel management while having a small impact on core behaviour (almost no impact on feedback coefficients). However, such subassemblies exhibit high decay heat levels mainly driven by alpha decay of curium isotopes with half-lives longer than a typical fuel cycle length. In order to obtain acceptable transmutation scenarios, the initial americium content is limited to 10%vol in the current MABB pin design.

A new innovative subassembly concept, the minor actinide transmutation advanced device for optimised reprocessing (MATADOR) concept, has been presented to satisfy this limitation. It is based on an axially-segmented structure that consists of superimposed fuel discs. Each elementary disc has an external structure (steel or ceramic) that contains granular fuel. These discs have dedicated channels which allow coolant flow and power extraction with the capability to rotate in situ to smooth the flux gradient during irradiation.

The transmutation performances of a preliminary design of such a disc and subassembly have been compared to a current MABB design. Both americium transmutation rate and mass balance were found to be quite similar. Concerning decay heat, the possibility to unpack the subassembly into a unitary disc does shorten the required delay before transportation stage by several years, which could lead to a significant reduction on minor actinide inventory for transmutation scenarios.

This promising concept is facing numerous technological challenges, such as the manufacturing process of the disc and fuel, the cooling capability of the subassembly, the unpacking process, etc. The most penalising effect relies on the pressurisation level which could be four times than those experienced in traditional MABB pin designs. Beyond these technological issues, simulation capabilities are also questionable since this new design is far from the traditional geometry for which calculation codes where primarily developed. An optimisation of the design process based on a multi-disciplinary approach is in progress at CEA to establish a realistic image of such a device for heterogeneous americium transmutation.

References


Transmutation scoping studies for a chloride molten salt reactor

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Argonne National Laboratory, United States

Abstract

Over the past few years, there has been strong renewed interest from private industry, mostly from start-up enterprises, in molten salt reactor (MSR) technologies because of the unique properties of this class of reactors. These are reactors in which the fuel is homogeneously mixed with the coolant in the form of liquid salts and is circulated continuously into and out of the active core region with online fuel management, salt treatment, and salt processing. In response to such widespread interest, Argonne National Laboratory is expanding its well-established reactor modelling and simulation expertise and infrastructure to enable detailed analysis and design of MSRs. The tools being developed are able to simulate the continuous fuel flow, the complex online fuel management and elemental removal processes (e.g. fission product removal) using depletion steps representative of a real MSR system.

Leveraging these capabilities, a parametric study on the transmutation performance of a simplified actinide-burning MSR concept that uses a chloride-based salt was performed. This type of salt has attracted attention over the more commonly discussed fluoride-based salts since no tritium is produced as a result of irradiation and it is compatible with a fast neutron spectrum. The studies discussed in this paper examine the performance of a burner MSR design with a fixed core size and power density over a range of possible fuel salt molar ratios with NaCl-MgCl\(_2\) as the carrier salt. The intent is to quantify the impact on the required transuranics content of the make-up fuel, the actinide transmutation rates, and other performance characteristics for typical burner MSR designs.

Introduction

Work on molten salt reactors (MSR) started in the 50s with the US Aircraft Reactor Experiment, and despite an initial strong interest in this type of reactor, limited work and progress have been made over the last four decades. Recently, a number of new and established companies have shown a renewed interest in MSRs because of their unique properties such as a negative salt expansion reactivity coefficient, small burnup reactivity swing, and good heat transfer characteristics [1]. In these reactors, the fuel is in the form of molten salt and is mixed with a carrier salt to insure chemical stability. The salt is flowing through the critical core region and upon exiting the core is circulated through a heat exchanger and a number of other components, including the salt processing stages for MSR concepts in which salt needs to be processed.

There exist many variations of the MSR concept since thorium based fuel could be used in lieu of uranium based fuel, several different fuel salt and carrier salts can be envisioned, and salt processing can be performed in many different ways. However, it is possible to differentiate between the MSRs offering a thermal spectrum and those offering a fast spectrum. Advantages and disadvantages of
these two types of spectrum are independent from the reactor technology considered. Of interest for the study discussed here, the ability to transmute actinides in a fast spectrum system is essential. Use of chloride-based salts is often considered in order to achieve a fast spectrum [2, 3], so they are used in this study.

A major unknown for simulating MSRs is the lack of reliable data about the various salts considered. Some data exist for a large number of salts [4, 5], but often times different values are reported in other publications [6]. Discrepancies could possibly be due to the temperature dependency, which strongly impacts the salt characteristics, and temperature-dependent data only exist for a few salts. Furthermore the molar composition of the salt, specifically the molar ratio between the fuel salt and the carrier salt, is not well established and different concepts propose using different molar compositions. Although a specific molar ratio is known to yield the lowest salt melting temperature [5], this molar ratio is not constant with burnup since the fuel composition changes through depletion.

The objective of this work is to use the MSR analysis tools recently developed at Argonne to provide a simple assessment of the impact of the molar composition of a chloride-based salt on the transmutation characteristics of a MSR. A brief overview of the capabilities of the MSR analysis tools developed at Argonne is provided, followed by a discussion of the assumptions and core model used for the MSR system simulated. Results of the simulations are then presented and analysed, and conclusions of the study are discussed. It should be noted that this is a synthetic study providing some insight into the expected trends. Specific MSR designs, fuel management approaches, and fuel processes will impact the results presented, and the analysis would need to be repeated for other MSR concepts if transmutation is part of their mission objectives.

**Modeling tools**

In 2016, Argonne has started to leverage its well-established system analysis capabilities to extend it to studying MSRs. These new capabilities enable simulating a full MSR core, allowing depletion calculations to be performed while taking into consideration the very short residence time of the fuel in the core (less than a minute) as well as fission product removal or any other isotope removal. Removal of those elements could be either total or partial, which typically represents a challenge for MSR modeling. Furthermore, equilibrium characteristics of an MSR core, when such a state exists, can be determined in a time efficient manner with minimum effort without having to resort to unit cell modeling [7].

Different modes of operation can be simulated by the MSR analysis tools recently developed, but in this study only a single mode is considered. The fuel and coolant salts circulate through the core and heat exchanger, carrying out the heat generated in the core. As it exits the core, a small fraction of the salt is continuously diverted towards the fuel processing section of the reactor, where it is treated as it flows through different stages. This diverted salt is then continuously re-injected with the bulk of the salt flowing back into the reactor.

Another unique feature of the MSR analysis tools developed is that they can work with a number of neutronics and depletion codes. In this study, MCNP6 and ORIGEN2.2 were selected. The cross section data used are based on the ENDF/B.VII libraries, and 1-group cross sections for the 90 most important actinides and fission product isotopes are tallied in MCNP6 and passed to ORIGEN2.2. Numerous iterations between these two codes and processing were performed to determine the equilibrium core characteristics and fuel composition.
Assumptions and system description

The MSR core model used is schematically shown in Figure 1. The reflector region is assumed to be made of Hastelloy-N [8] and to be 50 cm thick on the sides and above and below the active core regions. Heat will need to be removed from the reflector, but this has not been considered as part of this study. The active core region is modelled as a cylinder having a radius of 1.90 metres and a total height of 3.25 metres. This does not include the pipes coming in and out of the core. There is no solid component in the active core region which is entirely filled with molten salt. Reactivity control is assumed to be achieved through controlling the heavy metal content of the salt and through external control mechanisms, e.g. rotating drums in the reflector region. The core geometry used is very basic and is not representative of a real system in which the transition from the active core region to the inlet/outlet pipes should be more progressive.

Figure 1: Core layout

For this study, the fuel salt is taken to be UCl$_3$-TRUCI$_3$, and the carrier salt is NaCl-MgCl$_2$. The reason for using a carrier salt made of both sodium and manganese is that it allows reducing the melting temperature of the salt to enable a lower inlet salt temperature. In order for the NaCl-MgCl$_2$ system to be at the eutectic point, there needs to be 51.5 moles of NaCl for 48.5 moles of MgCl$_2$ [5], in which case the melting temperature of the carrier salt is about 450°C instead of 800°C for NaCl alone.

Given that there is very limited data for the solubility of the different actinides and fission product elements in chloride-based salt, even considering only a single element at a time, no solubility limit was used in this study. Instead, by providing parametric results showing the concentration of plutonium and fission products as a function of the molar concentration of the fuel salt in the carrier salt, it is expected that one could easily check future solubility limits against those results.

The salt density used are interpolated using existing data for the UCl$_3$-NaCl system for various molar ratios [2, 3], for the single NaCl and MgCl$_2$ systems [4], as well as for the PuCl$_3$-NaCl system [2]. Furthermore, existing data have been extrapolated to the average temperature assumed for the core, which is about 700°C. Five different salt compositions have been studied, with the fuel molar ratio varied to 10, 15, 20, 30, and 40 molar percent. Table 1 summarises the different densities assumed for the various salt compositions used at 700°C. In this table “An” denotes the actinides, including both uranium and transuranics. It is important to note that those values are simply extrapolations and are not measurements so they should be used with care.
Table 1: Salt densities assumed

<table>
<thead>
<tr>
<th>Salt Composition</th>
<th>Density, g/cm³</th>
</tr>
</thead>
<tbody>
<tr>
<td>40AnCl₃-35.1NaCl-24.9MgCl₂</td>
<td>3.87</td>
</tr>
<tr>
<td>30AnCl₃-41.0NaCl-29.1MgCl₂</td>
<td>3.42</td>
</tr>
<tr>
<td>20AnCl₃-46.8NaCl-33.2MgCl₂</td>
<td>2.96</td>
</tr>
<tr>
<td>15AnCl₃-49.7NaCl-35.3MgCl₂</td>
<td>2.65</td>
</tr>
<tr>
<td>10AnCl₃-52.7NaCl-37.4MgCl₂</td>
<td>2.37</td>
</tr>
</tbody>
</table>

This study focuses only on the equilibrium characteristics of the MSR core described in this section. The equilibrium state is dependent on the salt processing considered and on the make-up material used. The initial fuel loading composition does not have any effect on the equilibrium core characteristics. The fuel depletion is performed with a specific power of 30 W/gram of heavy metal. It is assumed that 0.001% of the salt coming out of the core is continuously diverted and flowing through the salt processing stages and that during this process 50% of all fission products are removed from the salt. This assumption is not realistic since different fission products elements are known to have different removal rates, such as the gaseous fission products which would be entirely removed. However, this approximation is sufficient for the purpose of this study. With an assumed fuel residence time in the active core region of about 22 seconds, the fission product concentration in the fuel at equilibrium is predicted to be about 0.15 wt%.

To make up for the fission products removed from the fuel, TRU recovered from light water reactor (LWR) used fuel and cooled for 10 years is loaded along with depleted uranium. For each salt composition considered, the TRU concentration in the make-up material was determined to be the amount needed to make the core critical at equilibrium. The TRU feed vector used is shown in Table 2.

Table 2: Transuranics feed vector

<table>
<thead>
<tr>
<th>Isotope</th>
<th>Atom %</th>
</tr>
</thead>
<tbody>
<tr>
<td>²³⁷Np</td>
<td>4.72%</td>
</tr>
<tr>
<td>²³⁸Pu</td>
<td>2.18%</td>
</tr>
<tr>
<td>²³⁹Pu</td>
<td>47.34%</td>
</tr>
<tr>
<td>²⁴⁰Pu</td>
<td>22.82%</td>
</tr>
<tr>
<td>²⁴¹Pu</td>
<td>8.42%</td>
</tr>
<tr>
<td>²⁴²Pu</td>
<td>6.84%</td>
</tr>
<tr>
<td>²⁴¹Am</td>
<td>5.61%</td>
</tr>
<tr>
<td>²⁴²mAm</td>
<td>&lt;0.01%</td>
</tr>
<tr>
<td>²⁴³Am</td>
<td>1.56%</td>
</tr>
<tr>
<td>²⁴³Cm</td>
<td>&lt;0.01%</td>
</tr>
<tr>
<td>²⁴⁴Cm</td>
<td>0.46%</td>
</tr>
<tr>
<td>²⁴⁵Cm</td>
<td>0.04%</td>
</tr>
</tbody>
</table>

Results

The MSR core characteristics at equilibrium, including the fuel composition, have been determined for each of the salt compositions shown in Table 1. Only equilibrium characteristics are discussed here, but it is acknowledged that reaching equilibrium may take several years depending on the initial fuel composition used and specific design characteristics of the MSR core considered. For all
calculations, the same specific power of 30 W/gram of heavy metal is used. Since all the salts considered contain a different molar fraction of AnCl₃ and the core volume and specific power are constant between all cases, the total reactor power is different for all cases. The fuel density in the salt and the corresponding core power are summarised in Table 3. The specific power was kept constant to ensure the same fuel burnup accumulation rate. The yearly burnup accumulation rate provided in Table 3 is only indicative since burnup for molten salt being continuously reprocessed is not meaningful. In practice, since the carrier salt is also carrying heat out of the core, the specific power relative to the mass of heavy metals (and fission products) is not relevant. Instead, the specific power per unit mass of salt, including both the fuel salt and the carrier salt, is what should be kept constant to maintain the same temperature rise across the core for a given flow rate (i.e. fuel residence time). This was not done in this study to avoid introducing a bias caused by a different burnup accumulation.

As a result of the simulation, the fraction of TRU that needs to be included in the make-up material has been determined. The values are provided in Table 3, and it is observed, as would be expected, that as the salt contains a lower fraction of fuel, a larger amount of TRU needs to be included in the make-up material. For 40AnCl₃-35.1NaCl-24.9MgCl₂, the make-up material only needs to contain 12% TRU while for 10AnCl₃-52.7NaCl-37.4MgCl₂, it needs to contain 95.3%. For a salt having an AnCl₃ molar fraction of 5%, it is not possible to make the MSR core critical, even when using pure TRU. Although not studied here, using chloride enriched in ³⁷Cl instead of the natural chloride composition could enable making the core critical due to its lower capture cross section. Issues arising from extracting pure or nearly pure TRU from the LWR used fuel have not been considered in this study, but this is acknowledged as being a potential challenge. The high concentration of TRU in the MSR fuel itself is a lesser concern as it is in a virtually closed system and would not need to be shipped to any off-site facility for the lifetime of the reactor.

The equilibrium concentration of TRU contained in the fuel flowing through the core is different from that of the TRU used in the make-up fuel. This is due to the fact that when uranium is present in the fuel, plutonium is produced, which increases the TRU concentration in the fuel. As shown in Figure 2, the neutron spectrum does not vary significantly between the cases with the highest and lowest AnCl₃ molar fraction, indicating that the plutonium production from uranium is proportional to the uranium concentration and neutron flux. Furthermore, only a small fraction of the fuel is continuously being processed, and a very small amount of make-up fuel is added to the core, meaning that the TRU introduced in the make-up fuel only slightly affects the overall concentration of TRU in the core.
Table 3: Core performance characteristics

<table>
<thead>
<tr>
<th>AnCl₃ mol%</th>
<th>40</th>
<th>30</th>
<th>20</th>
<th>15</th>
<th>10</th>
</tr>
</thead>
<tbody>
<tr>
<td>Specific power, W/gHM</td>
<td>30</td>
<td>30</td>
<td>30</td>
<td>30</td>
<td>30</td>
</tr>
<tr>
<td>Total reactor power, MWth</td>
<td>1 528</td>
<td>1 190</td>
<td>832</td>
<td>624</td>
<td>422</td>
</tr>
<tr>
<td>Heavy metal mass in core, bim</td>
<td>50.9</td>
<td>39.7</td>
<td>27.7</td>
<td>20.8</td>
<td>14.1</td>
</tr>
<tr>
<td>Burnup accumulation rate, GWd/bim-yr</td>
<td>10.95</td>
<td>10.95</td>
<td>10.95</td>
<td>10.95</td>
<td>10.95</td>
</tr>
<tr>
<td>Equilibrium k eff value (within 200 pcm)</td>
<td>1.0</td>
<td>1.0</td>
<td>1.0</td>
<td>1.0</td>
<td>1.0</td>
</tr>
<tr>
<td>Salt density, g/cm³</td>
<td>3.87</td>
<td>3.42</td>
<td>2.96</td>
<td>2.65</td>
<td>2.37</td>
</tr>
<tr>
<td>Heavy Metal + FP density, g/cm³</td>
<td>2.02</td>
<td>1.57</td>
<td>1.10</td>
<td>0.83</td>
<td>0.56</td>
</tr>
<tr>
<td>TRU density, g/cm³</td>
<td>0.40</td>
<td>0.42</td>
<td>0.45</td>
<td>0.46</td>
<td>0.48</td>
</tr>
<tr>
<td>Fuel fraction in salt, wt%</td>
<td>52.2</td>
<td>46.0</td>
<td>37.2</td>
<td>31.2</td>
<td>23.6</td>
</tr>
<tr>
<td>TRU fraction in equilibrium fuel, wt%</td>
<td>20.0</td>
<td>26.9</td>
<td>40.7</td>
<td>55.4</td>
<td>85.7</td>
</tr>
<tr>
<td>Atomic fraction of TRU in make-up fuel, at%</td>
<td>12.0</td>
<td>29.0</td>
<td>53.5</td>
<td>71.3</td>
<td>95.3</td>
</tr>
<tr>
<td>Normalised make-up fuel, kg/yr-GW th</td>
<td>370.9</td>
<td>370.9</td>
<td>370.9</td>
<td>370.9</td>
<td>370.9</td>
</tr>
<tr>
<td>Normalised make-up TRU, kg/yr-GW th</td>
<td>44.5</td>
<td>107.5</td>
<td>198.4</td>
<td>264.4</td>
<td>353.4</td>
</tr>
</tbody>
</table>

Figure 2: Normalised neutron spectra

For this study a constant fuel core residence time of 22 seconds has been assumed for simplicity. In practice, this value should be adjusted for all cases in order to maintain a given core temperature rise, which would depend on the heat capacity of the salt and its melting temperature. This would result in lower flow rates for the cases having a lower reactor power. Flow rates in the core and through the processing stages are summarised in Table 4. Depending on the salt composition, the 0.001% of salt diverted towards the processing stages corresponds to a salt flow rate of 27 to 44 kg per second, which is equivalent to a heavy metal flow rate through the processing stages of 6.4 to 23.2 kgHM per second.

When normalising those numbers to one GW th, the heavy metal flow through the processing stage is constant for all cases and equal to 15.2 tHM/s-GW th. These results in the make-up fuel, normalised per GW th, to represent about 371 kg per year. Given the different TRU fractions contained in the different cases studied, the normalised TRU loading ranges from 44.5 to 353.4 kilograms of TRU per year and per GW th. The TRU consumption is equal to the TRU loading. Although the same specific power is used, the TRU consumption rate is lower when the salt contains a larger molar fraction of AnCl₃. This is due to more plutonium being produced from the larger fraction of uranium contained in the fuel.
Table 4: Flow rates

<table>
<thead>
<tr>
<th>AnCl₃ mol%</th>
<th>40</th>
<th>30</th>
<th>20</th>
<th>15</th>
<th>10</th>
</tr>
</thead>
<tbody>
<tr>
<td>Salt through core, t/s</td>
<td>4.43</td>
<td>3.92</td>
<td>3.39</td>
<td>3.03</td>
<td>2.71</td>
</tr>
<tr>
<td>Heavy metal + fission products through core, t/s</td>
<td>2.32</td>
<td>1.80</td>
<td>1.26</td>
<td>0.95</td>
<td>0.64</td>
</tr>
<tr>
<td>Salt through processing, kg/s</td>
<td>44.3</td>
<td>39.2</td>
<td>33.9</td>
<td>30.3</td>
<td>27.1</td>
</tr>
<tr>
<td>Heavy metal + FP through processing, kg/s</td>
<td>23.2</td>
<td>18.0</td>
<td>12.6</td>
<td>9.5</td>
<td>6.4</td>
</tr>
</tbody>
</table>

A summary of the fuel composition for each of the cases studied is provided in Table 5 along with the isotopic composition for the transuranics. It is observed that the quality of the TRU contained in the fuel is significantly lower (i.e. fewer fissile isotopes) when the salt contains a lower molar fraction of AnCl₃. This is consistent with the fact that in this case, almost no uranium is contained in the fuel. In fact, for all cases studied, the equilibrium TRU composition in the fuel has a lower quality than that of the TRU used for make-up.

In Table 5, it is shown that as the AnCl₃ molar fraction is reduced, the atomic fraction of minor actinides in the fuel is increased, and is more than 10 times larger with 10% AnCl₃ than with 40% AnCl₃. However, since the fuel density in the salt is reduced as the AnCl₃ fraction is increased, the minor actinide content for the case with 10% AnCl₃ is only about three times larger than for the case with 40% AnCl₃. In context, in the 10% AnCl₃ case, eight times more TRU was flowed through the core as well.

Table 5: Fuel composition at equilibrium

<table>
<thead>
<tr>
<th>AnCl₃ mol%</th>
<th>40</th>
<th>30</th>
<th>20</th>
<th>15</th>
<th>10</th>
</tr>
</thead>
<tbody>
<tr>
<td>Heavy metals at%</td>
<td>99.8</td>
<td>99.8</td>
<td>99.8</td>
<td>99.8</td>
<td>99.8</td>
</tr>
<tr>
<td>TRU at%</td>
<td>19.9</td>
<td>26.7</td>
<td>40.5</td>
<td>55.1</td>
<td>85.6</td>
</tr>
<tr>
<td>Minor actinides at%</td>
<td>1.4</td>
<td>2.7</td>
<td>5.5</td>
<td>8.7</td>
<td>15.5</td>
</tr>
<tr>
<td>Fission products at%</td>
<td>0.15</td>
<td>0.15</td>
<td>0.15</td>
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<td>0.15</td>
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</tbody>
</table>

<table>
<thead>
<tr>
<th>Transuranics isotopic composition, at%</th>
<th>237Np</th>
<th>239Np</th>
<th>238Pu</th>
<th>239Pu</th>
<th>240Pu</th>
</tr>
</thead>
<tbody>
<tr>
<td>239Np</td>
<td>0.88</td>
<td>1.29</td>
<td>1.75</td>
<td>2.07</td>
<td>2.56</td>
</tr>
<tr>
<td>238Pu</td>
<td>0.04</td>
<td>0.02</td>
<td>0.01</td>
<td>0.00</td>
<td>0.00</td>
</tr>
<tr>
<td>239Pu</td>
<td>2.34</td>
<td>3.51</td>
<td>4.90</td>
<td>5.78</td>
<td>6.79</td>
</tr>
<tr>
<td>240Pu</td>
<td>51.86</td>
<td>42.01</td>
<td>31.79</td>
<td>26.22</td>
<td>20.27</td>
</tr>
<tr>
<td>241Pu</td>
<td>30.32</td>
<td>32.61</td>
<td>34.92</td>
<td>36.17</td>
<td>37.53</td>
</tr>
<tr>
<td>242Pu</td>
<td>3.84</td>
<td>4.37</td>
<td>4.70</td>
<td>4.67</td>
<td>4.34</td>
</tr>
<tr>
<td>243Pu</td>
<td>4.86</td>
<td>7.43</td>
<td>10.08</td>
<td>11.52</td>
<td>13.02</td>
</tr>
</tbody>
</table>

| 244Pu                                  | 2.70  | 3.84  | 5.29  | 6.32  | 7.75  |
| 242Am                                  | 0.26  | 0.37  | 0.52  | 0.62  | 0.76  |
| 243Am                                  | 1.45  | 2.31  | 3.21  | 3.69  | 4.20  |
| 244Am                                  | 0.08  | 0.10  | 0.11  | 0.11  | 0.10  |
| 242Cm                                  | 0.01  | 0.01  | 0.01  | 0.01  | 0.01  |
| 243Cm                                  | 1.02  | 1.57  | 1.98  | 2.05  | 1.93  |
| 244Cm                                  | 0.21  | 0.34  | 0.44  | 0.47  | 0.45  |
| 245Cm                                  | 0.14  | 0.22  | 0.29  | 0.30  | 0.29  |
Conclusions

The MSR analysis tools recently developed at Argonne have been used to determine the impact of the fuel molar fraction on the transmutation capabilities of an MSR core using chloride-based salts. Under the stated assumptions, it was found that having a smaller molar fraction of fuel salt allows having a lower amount of uranium in the fuel which results in a significantly higher transuranics consumption rate. However, this also results in a significantly higher concentration of minor actinides in the fuel, which may prove challenging as the fuel is continuously circulating out of the core. This may result in challenges for adequate shielding and could potentially affect the chemical processes used to continuously treat the salt.

Had the core layout not been fixed, it could have been possible to use the salt having a high AnCl$_3$ molar fraction and reduce the core size to increase neutron leakage probability and therefore allow loading more TRU in the fuel salt. By changing some of the other assumptions used in this study, it is expected that the results presented here would also be affected. In particular, with a different removal rate of fission products, the concentration of fission product in the fuel could end up being higher. This would not affect the reactor spectrum much, but would result in a lower $k_{eff}$ value which means that a higher TRU fraction would be needed in the make-up fuel. If that were to become a limitation, it would be possible to consider using chloride containing mostly $^{37}$Cl since it has a lower capture cross section than $^{35}$Cl and would result in a significant increase of the core multiplication factor. However, there are currently a number of uncertainties surrounding the accuracy of the chloride isotope cross sections. Overall, a chloride-based MSR could be configured as a burner and by adequately selecting the salt composition it is possible to achieve a very low conversion ratio.

Acknowledgements

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References


Investigation of beam window structure for accelerator-driven system with subcriticality adjustment rod

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Japan Atomic Energy Agency

Abstract

To reduce the burden of the geological disposal of high-level waste (HLW), the Japan Atomic Energy Agency (JAEA) has investigated an accelerator-driven system (ADS) to transmute minor actinide (MA) included in HLW. Since the ADS is a hybrid system of an accelerator and a subcritical core, there are various inherent issues in the research and development of the ADS. As one of the critical issues, the design of a beam window which is a boundary of the accelerator and the subcritical core, has been pointed out. In the previous study, JAEA showed a feasible beam window concept, however, more durable concept with a simple shape has been required from the viewpoint of its fabrication and reliability.

In this study, new beam window concept is investigated by changing the design condition. The most important factor for the beam window design is the proton beam current, and the design condition will be mitigated if the required proton beam current to keep the same core power will be reduced. To reduce the proton beam current, a subcriticality adjustment rod (SAR) which was a $\text{B}_4\text{C}$ control rod was employed.

Based on the mitigated calculation condition by the installation of SAR, the design of the beam window was revised by the particle transport analysis by PHITS code, the thermal hydraulics analysis by STAR-CCM+ code and the structural analysis by ANSYS code. Through these coupled analyses, more feasible beam window concept which was the hemispherical shape, the outer diameter = 470mm, the thickness at the top = 4.0mm, was presented.

Introduction

To reduce the burden of the geological disposal of high-level waste (HLW), the Japan Atomic Energy Agency (JAEA) has investigated an accelerator-driven system (ADS) to transmute minor actinide (MA) included in HLW. Since the ADS is a hybrid system of an accelerator and a subcritical core, there are various inherent issues in the research and development of the ADS. As one of the critical issues, the design of a beam window which is a boundary of the accelerator and the subcritical core, has been pointed out. In the previous study, JAEA showed a feasible beam window concept which was the ellipse shape, the outer diameter = 470mm, the thickness at the top of the beam window = 2.0mm and factor of safety = 4 [1]. However, more durable concept with a simple shape has been required from the viewpoint of its fabrication and reliability.

In this study, new beam window concept is investigated by changing the design condition. The most important factor for the beam window design is the proton beam current, and the design condition will be mitigated if the required proton beam current to keep the same core power will be reduced. To reduce the proton beam current, a subcriticality adjustment rod (SAR) which was a $\text{B}_4\text{C}$
control rod was employed. The neutronics calculation of ADS concept with SARs was performed and the mitigated calculation condition for the beam window was presented.

Based on the mitigated calculation condition, the design of the beam window was revised by the particle transport analysis, the thermal hydraulics analysis and the structural analysis. Through these coupled analyses, more feasible beam window concept was represented.

**Calculation conditions and flow**

The ADS investigated in the ADS feasibility study [2] was employed in this study. It was the LBE-cooled tank-type ADS with 800MW thermal power. Main parameters of this reference ADS are summarised in Table 1. For the accelerator, a superconducting linac with 1.5GeV proton beam energy was supposed. Based on the previous neutronics design study [3], the maximum proton current was 20mA in the reference ADS design.

In this study, burnup, particle transport, thermal hydraulics and structural analyses were performed. The burnup analysis was performed by ADS3D code [4], which can calculate the transport of neutrons, the burnup calculation and the fuel exchange in three-dimensional calculation models. This analysis aimed to get a feasible ADS core concept with SARs.

The particle transport analysis was carried out for protons and neutrons by using the proton beam current as the output of the burnup analysis. The flux distributions of protons and neutrons above 20 MeV were calculated by PHITS code [5]. PHITS code is a general purpose particle and heavy ion transport Monte Carlo code. As the result, heat generation distributions in the spallation target region were calculated and used in the thermal hydraulics analysis as the input.

The thermal hydraulics analysis was performed by STAR-CCM+ code [6], which is a computational fluid dynamics (CFD) code. From this analysis, temperature distributions at the spallation target region and the beam window were obtained. The temperature distributions at the beam window were reflected to the input data for the structural analysis.

The structural analysis was performed by ANSYS code [7], which is a finite element method (FEM) calculation code. This analysis outputs stress and strain distributions and buckling pressure of the beam window. Based on the analysis results, the feasibility of the beam window was evaluated.

<table>
<thead>
<tr>
<th>Table 1: Main parameters of reference ADS</th>
</tr>
</thead>
<tbody>
<tr>
<td>Thermal power</td>
</tr>
<tr>
<td>Coolant and spallation target material</td>
</tr>
<tr>
<td>Coolant inlet temperature</td>
</tr>
<tr>
<td>Maximum $k_{\text{eff}}$</td>
</tr>
<tr>
<td>Operation period</td>
</tr>
<tr>
<td>Transmutation amount</td>
</tr>
<tr>
<td>Proton beam energy</td>
</tr>
<tr>
<td>Maximum proton beam current</td>
</tr>
</tbody>
</table>
Neutronics calculation with subcriticality adjustment rods

Calculation condition

Figure 1 shows the ADS calculation model for the burnup analysis as the reference case. Seven fuel assemblies (FAs) at the centre were corresponds to the spallation target region. Then, there were 84 FAs which contained MA fuel, with four layers as shown in Figure 1. A duct-less assembly was employed and 391 fuel pins were bundled in one FA. Nitride MA fuel, (MA+Pu)N+ZrN, was considered in this model. The ratio of ZrN was adjusted to set the $k_{\text{eff}}$ value as 0.97 at BOC. When the SAR was installed to the core, the number of fuel assembly (FA) was maintained as the reference one, 84.

As the SAR, $\text{B}_4\text{C}$ type control rod which was the conventional one, was employed in this study. The natural abundance of boron was assumed for the $\text{B}_4\text{C}$ pellet. For the operation of the SAR, the following operation was assumed in the neutronics calculation. At the BOC, all SARs were inserted fully in the core. The maximum reactivity worth for all CRs was limited 1.5%dk to prevent a critical accident by the SAR drawing out. During the operation, all SARs were drawn gradually to keep the $k_{\text{eff}}$ value to 0.97. In the neutronics calculation, due to the size of calculation mesh, burn-up period and computational time, the SARs were drawn 20cm by each 100days during the operation. In reality, it could be drawn by mm/sec unit like control rods used in conventional nuclear power plants.

Calculation results

Figure 2 presents the calculation model of the ADS concept with SARs. In this case, three SARs were installed as shown in Figure 2. The burnup calculation for 2 burn-up cycles was performed. Figures 3 and 4 illustrate the changes of the criticality and the proton beam current, respectively.

It was shown that the $k_{\text{eff}}$ value would be maintained 0.97 during the cycle. Due to the calculation limitation as mentioned above; the size of calculation mesh, the burn-up period, the $k_{\text{eff}}$ value was slightly larger than 0.97 and the proton beam current was slightly smaller than 10mA. However, in reality, the SARs could be drawn by mm/sec unit. So, it was supposed that this concept could maintain the proton beam current near 10 mA. As the result, the proton beam current became half by the installation of SARs and 10 mA was employed as the new design condition for the beam window.
**Figure 2: ADS calculation model with SARs**

![ADS calculation model with SARs](image)

**Figure 3: Criticality changes for the ADS concept with SARs**

![Criticality changes](image)

**Figure 4: Proton beam current changes for ADS concept with SARs**

![Proton beam current changes](image)
Particle transport analysis

Calculation condition

In the particle transport analysis by PHITS code, the hemispherical window in the calculation model shown in Figure 5 was employed. The thickness of the beam window was 2.0 mm. Table 2 presents main parameters for the particle transport analysis in the reference case investigated in Ref. [1] and this study. All parameters except the beam current and the shape of the beam window were the same. The proton beam current in this study was 10 mA based on the burnup calculation result. For the shape of the beam window, a simple hemispherical model was employed from the viewpoint of fabrication.

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Reference [1]</th>
<th>This study</th>
</tr>
</thead>
<tbody>
<tr>
<td>Proton beam energy [GeV]</td>
<td>1.5</td>
<td>←</td>
</tr>
<tr>
<td>Proton beam current [mA]</td>
<td>20</td>
<td>10</td>
</tr>
<tr>
<td>Beam duct radius [mm]</td>
<td>235</td>
<td>←</td>
</tr>
<tr>
<td>Shape of beam window</td>
<td>Ellipse</td>
<td>Hemispherical</td>
</tr>
<tr>
<td>Thickness of beam window at top [mm]</td>
<td>2.0</td>
<td>←</td>
</tr>
<tr>
<td>Thickness of beam duct [mm]</td>
<td>10.0</td>
<td>←</td>
</tr>
<tr>
<td>1σ of Gaussian profile for proton beam [mm]</td>
<td>111.6</td>
<td>←</td>
</tr>
</tbody>
</table>

Figure 5: Calculation model for PHITS

Calculation results

Figure 6 presents the heat density distribution in the spallation target region. The co-ordinate of radial and axial directions corresponds to the one shown in Figure 5. It was observed that the maximum heat density was about 40 W/cc/mA in the spallation target region. Figure 7 shows the heat density distribution in the beam window. It was confirmed that the heat density at the top of
the beam window was 27 W/cc/mA. The values shown in Figures 6 and 7 were used for the input of the thermal hydraulics analysis.

**Figure 6**: Heat density distribution in the spallation target region

![Heat density distribution in the spallation target region](image)

**Figure 7**: Heat density distribution in the beam window

![Heat density distribution in the beam window](image)

**Thermal hydraulics analysis**

**Calculation condition**

The STAR-CCM+ code was employed for the thermal hydraulics analysis in the spallation target region. Figure 8 illustrates the calculation model for the analysis. This calculation model is two-dimensional axial symmetry one. T91 steel was assumed as the material of the beam window and other structures, so the property of T91 steel was applied to the solid region.

The velocity and temperature of LBE at the inlet (bottom in Figure 8) was 2.0 m/s and 300 °C, respectively. The standard k-ε model was employed as the turbulence model and the high Reynolds number model was adopted at the wall boundary condition.

For the heat generation, the calculation results in the previous section were used. In this calculation, 15% beam loss and 20% safety margin which is based on the PHITS calculation accuracy
were taken into account. Namely, the heat density of 10 W/cc/mA in the 10mA beam current condition gave 10 W/cc/mA x 1.15 x 1.20 x 10 mA = 138 W/cc.

**Figure 8:** Calculation model for thermal hydraulics analysis

![Calculation model](image)

**Calculation results**

Temperature distributions in the spallation target region and beam window are shown in Figures 9 and 10, respectively. The maximum temperature and difference of the temperature in the beam window were 409 °C and 27 °C, respectively. In the previous design with 20mA beam current, these values were 516 °C and 55 °C, so it was confirmed that the design condition of the beam window was mitigated significantly. These temperature distributions in the beam window were used for the input of the structural analysis.

**Figure 9:** Temperature distribution in the spallation target region

![Temperature distribution](image)
Structural analysis

Calculation condition

The structural analysis was performed by ANSYS code with two-dimensional calculation models shown in Figure 11. The temperature distribution derived by the thermal hydraulics analysis was given to the calculation model.

Parametric survey by changing the thickness of the beam window was performed. Since the temperature distribution varies by the change of the thickness, the approximate value derived from the following equation was employed,

\[
T = T_0 + Q(r) \left(\frac{r_2^2}{2} - \frac{r_1^2}{2}\right) \left(1 - \frac{r_1^2}{r_0^2}\right) / \lambda
\]  

(1)

where \( T \) °C is temperature at the position \( t \) cm (\( t=0 \) corresponds to the inner surface), \( T_0 \) °C is the temperature at the outer surface, \( Q(r) \) W/cc is the heat density and \( \lambda \) W/cm/°C is the thermal conductivity of T91 steel.
Calculation results

Figures 12-14 present the relationship between the beam window thickness and the maximum temperature, Von Mises stress and buckling pressure, respectively. For the maximum temperature, even if the thickness increased to 4 mm, the maximum value was less than 500 °C which was the upper limitation to use T91 steel in LBE. From the viewpoint of the equivalent stress, all cases satisfied the criteria $3S_m$ ($S_m$ is the intensity of design stress) as shown in Figure 13. These results indicate that the beam window with 4 mm thickness is acceptable.

For the buckling pressure, the value increased as the thickness increased. The buckling pressure with 4 mm thickness was 3.6 times larger than the value with 2 mm thickness. The mitigated design condition by the installation of SARs indicates the hemispherical beam window with 4mm thickness and its buckling pressure is at least 3.6 times larger than the previous ones.

**Figure 12**: Relationship between beam window thickness and maximum temperature

![Figure 12](image1)

**Figure 13**: Relationship between beam window thickness and Von Mises stress

![Figure 13](image2)
Conclusions

In this study, a new beam window concept was investigated by changing the design condition. To mitigate the design condition for the beam window, SAR which was a B₄C control rod was employed. The results of the neutronics calculation by ADS3D code presented that the proton beam current was reduced from 20mA to 10mA by the installation of SARs.

Based on the mitigated calculation condition, the design of the beam window was revised by the particle transport analysis by PHITS code, the thermal hydraulics analysis by STAR-CCM+ code and the structural analysis by ANSYS code. Through these coupled analyses, more feasible beam window concept which was the hemispherical shape, the outer diameter = 470mm, the thickness at the top = 4.0mm was presented. The buckling pressure of this new concept was at least 3.6 times larger than the previous ones.

Acknowledgements

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References


Am-bearing blanket transmutation strategies in sodium-cooled fast reactors

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²Idaho National Laboratory, Idaho, United States

Abstract

Reactor physics and fuel cycle analyses were conducted to assess different minor actinide (MA) transmutation strategies based on the two Sodium-cooled Fast Reactors: 600 MWe ASTRID with oxide fuel and the 1 000 MWt ABR with both metallic and oxide fuels. In the ASTRID core, 24 americium-bearing blankets (AmBB) assemblies loaded with 15% Am content allow self-sustainable recycling of the Am, while 66 AmBB assemblies (one row surrounding the driver fuel) allows continuous recycling and transmutation from about 1.8 GWe of PWR. The impact of AmBB assemblies on ASTRID performance and safety were confirmed to be small, but helium production and decay heat are especially large in these assemblies. The AmBB transmutation performance characteristics with metallic and oxide fuels were assessed in the ABR.

The ABR concept displays similar core performance characteristics between continuous U/Pu and U/TRU homogeneous recycling, and U/Pu+AmBB heterogeneous recycling strategies. However, in order to achieve self-sustainable recycling of the Am, the metallic-fuelled core configuration requires fewer AmBB blanket assemblies compared to the oxide-fuelled core due to the harder neutron spectrum and smaller MA production rate. Finally, detailed analyses were performed to determine irradiation conditions of AmBB fuels loaded in metallic ABR-1000 and ASTRID cores to support the development of irradiation experiments of AmBB fuel concepts in the advanced test reactor (ATR).

Introduction

The Evaluation and Screening study performed by the Fuel Cycle Technology Office of the US Department of Energy [1] identified continuous recycling of uranium together with plutonium and/or transuranic elements in fast reactors, either alone or with supporting thermal reactors, as the most promising options for improving resource utilisation and reducing high-level waste (HLW) generation.

A recent follow up analysis on minor actinide (MA) recycling [2] assessed the impacts of keeping MA’s together with Pu in reactor and showed that benefits of MA recycle are mostly associated with HLW handling and repository; however, they present challenges in fuel fabrication, core performance, and reprocessing.

One potential path investigated in France [3] considers Am-only recycle in americium-bearing blankets (AmBB) and direct disposal of Np and Cm. This allows recycling Am in a few specific radialblanket assemblies of a fast reactor, minimising the repository impacts by transmuting Am, minimising fuel handling impacts from Cm, and minimising impacts on reactor performance and safety. In this context, reactor physics and fuel cycle analyses are conducted to assess different minor
actinide (MA) transmutation strategies and to identify the irradiation experiments needed. These strategies are investigated in this paper based on the ASTRID with oxide fuel [4] and the ABR-1000 [5] with both metallic and oxide fuels.

**SFR cores description**

**The ASTRID core**

The Advanced Sodium Technological Reactor for Industrial Demonstration (ASTRID) was studied as part of a US/France bilateral agreement. Irradiation experiments of transmutation in the advanced test reactor (ATR) are being prepared, which requires analysing the fuel form and flux spectrum needed to represent French interests. The ASTRID reactor targets a power level of 1500 MW thermal or 600 MW electric and uses mixed-oxide fuel with a conversion ratio close to unity. The current analysis of the ASTRID core is based on the ASTRID-V0 design [4] and its radial layout is displayed in Figure 1. The ASTRID core uses an axially heterogeneous core to reduce the sodium void reactivity feedback, with an internal layer of blanket in the inner core in order to reduce the flux in this lowleakage region, and on a sodium plenum at the top of the core in order to favour neutron leakage during coolant voiding events. It is noted that the reference ASTRID design [4] does not have radial blanket, but for transmutation of Am in AmBB assemblies, one row of radial blanket with 66 assemblies were added to the ASTRID core shown in Figure 1. The radial blanket was designed to be similar to the outer core assembly, which has an axial height of 120 cm of packed (U,Am)O2 fuel pellets for Am-bearing blanket (AmBB) or UO2 fuel pellets for regular blanket. In this study, there was no attempt to optimise the blanket design. The irradiation lifetime of AmBB assemblies is limited with the same peak discharged fast (>0.1MeV) fluence as evaluated in the driver fuel. Radial blanket assemblies will remain 8 cycles in the core (~8 years), which is twice longer than driver fuel assemblies. Future work should confirm the AmBB fuel pin performance parameters associated with helium generation remains acceptable at the end of life. For ASTRID, a cooling and fabrication times of 5 and 2 years, respectively, are assumed.

**Figure 1**: Radial layout of the ASTRID core with radial blankets

![Figure 1: Radial layout of the ASTRID core with radial blankets](image)

**The ABR-1000 concepts**

The ABR-1000 core concept was developed for the study of future fast reactor design options in the United States under the Global Nuclear Energy Partnership (GNEP) programme. Compact core concepts with a transuranics (TRU) conversion ratio of ~0.7 were developed for a one-year cycle length generating 1000 MW thermal (400 MW electrical) [5]. Both metallic and oxide fuels options were considered. Self-sustaining U/Pu or U/TRU recycle concepts were developed by replacing
30 driver fuels assemblies from the outer row by radial blanket assemblies. Figure 2 shows the radial core layout of the self-sustaining 1 000 MW(th) ABR metallic core [5]. For ABR-1000, a cooling time of 1 year and a fabrication time of 0.5 year are assumed. Driver fuel and blanket assemblies remain in the core during 4 and 6 cycles, respectively.

**Figure 2**: Radial layout of the breakeven version of the ABR-1000 core

Calculation methodology

The ASTRID and ABR-1000 cores were modelled using consistent methodologies with the ANL fast reactor analysis suite. The MC2-3 code [6] is used to calculate the 33-group cross sections for the REBUS-3 code. MC2-3 employs the ENDF/B-VII.0 nuclear data library [7]. The whole-core flux calculation is performed with the DIF3D code [8] using the variational nodal transport solver VARIANT [9]. The order for the angular flux and scattering approximations, and the polynomial order for the flux within a node, on the boundary, and for the source within a node were selected appropriately for the different cores considered. Core calculations were done with “all rods out” in a three-dimensional core model where fuel assemblies are radially homogenised. The REBUS-3 code [10] was used for fuel cycle analysis at equilibrium state, which includes fuel fabrication, irradiation, discharge cooling, reprocessing, and reloading. The code was also used for the enrichment search at the equilibrium state. The cross sections used for the equilibrium analysis were calculated with MC2-3 for the BOC configuration through an iterative process. The neutronic feedback coefficients such as the sodium void worth and the Doppler coefficients are calculated with the PERSENT perturbation theory code [11], which is based on neutron transport equation in a Hex-Z core. The Monte Carlo code SERPENT [12] is also employed to provide detailed flux and power information for several pins of a specific AmBB assembly. It is a continuous-energy Monte Carlo reactor physics burnup calculation code developed at the VTT Technical Research Centre of Finland.

The ORIGEN-2 and ORIGEN-S codes [13, 14] were used to estimate the detailed charged and discharged fuel compositions, activity, and decay heat in the driver fuel and blanket assemblies. Similar approach is being used with both versions of the ORIGEN code where the one-group cross sections from the REBUS-3 code are being used for irradiation simulations with ORIGEN. This procedure allows generating detailed actinides and fission products compositions for fuel cycle analysis with good agreement with the physics calculations. Decay heat values are calculated for the charged fuel assemblies, and for the discharged fuel assemblies after 5 years of cooling. These information help evaluate the challenges for handling and fabricating a fresh fuel assembly, and the challenges for handling the irradiated assembly prior to transportation and reprocessing. The System Analysis Module (SAM) is an advanced system analysis tool for SFRs being developed at Argonne.
under the US Department of Energy (DOE) Nuclear Energy Advanced Modeling and Simulation Programme (NEAMS) programme [15]. It is used in this work to assess the fuel and cladding temperature distributions in an AmBB assembly.

**AmBB blanket transmutation study**

**Fuel cycle scenarios investigated**

Two sets of transmutation scenarios with Am-bearing blankets are investigated: Am self-sustaining mode and Am-burning mode scenarios, as described in Figures 3 and 4, respectively. For both scenarios, Pu is continuously recycled and both non-recovered MAs (Np, Cm and higher actinides) and fission products (FP) are sent to repository. In the Am self-sustaining mode, the recovered Am was recycled without external Am feed. In order to achieve Am self-sustaining scenario, the number of AmBB assemblies was adjusted with a fraction of Am of about 15 – 20w% of heavy nuclei. In the Am-burning mode, the maximum Am-burning rate was searched by loading Am-bearing fuels into all radial blankets, and Am recovered from the PWR used nuclear fuels were used as make-up Am feed. In this case, the isotopic content of external Am feed is representative of discharged PWR fuel with burnup of 50 MWd/kg and 5 years of cooling before separation.

**Figure 3**: Overview of AmBB concept for Am self-sustaining mode.

**Figure 4**: Overview of AmBB concept for Am-burning mode.

**Transmutation performance of AmBB in ASTRID**

Both Am self-sustaining and burning transmutation modes with Am-bearing blankets were investigated based on the ASTRID core to be representative of potential French fuel cycle strategies, and results are summarised in Table 1. The Ref. ASTRID case indicates that the recovered Pu is recycled in the ASTRID core and MA is sent to a repository. The initial Am content in the AmBB blanket assemblies is set to 15% (the target Am content in the French AmBB programme is 10 – 20%). In the Am self-sustaining mode, 24 radial blankets are required at equilibrium to burn all the Am generated in the driver fuel, which indicates that each AmBB assembly supports about 12 driver fuel assemblies. In the Am-burning mode, 66 AmBB assemblies surrounding the core burn about 26 kg of
americium per cycle, which is about 45 kg/GWe-yr, while a PWR generates about 15 kg/GWe-yr of Am (when accounting for 5 years of cooling of the spent fuel). This represents a support ratio of about three, meaning that 0.6 GWe of ASTRID can transmute Am from 1.8 GWe of PWR while also minimising the Am generation from ASTRID.

The results in Table 1 show that the amount of MA sent to the repository from SFR fuel cycle decreases from ~21 kg/cycle (all minor actinides and loss from ASTRID fuel cycle) without transmutation down to ~5 kg/cycle or ~14 kg/cycle for self-sustaining mode or Am-burning mode, respectively. For Am-burning case, the amount of MA sent to repository from SFR fuel cycle is larger than in the self-sustaining mode since there are more AmBB assemblies where Curium buildup is favoured by the high concentration of americium. However, the burning mode leads to the reduction of 26 kg/cycle of americium from PWR fuel cycle.

In terms of reactor physics, it was confirmed that negligible effects on the sodium void worth and Doppler coefficients are expected from heterogeneous transmutation strategies. The Am content in AmBB is mainly limited by fuel cycle performance parameters such as the maximum decay heat for the most constraining type of assembly, which is displayed in Table 1. For the Ref. scenario, the most constraining assembly is the inner driver fuel that contains higher fissile content. Significant impact is observed in the radial blanket assemblies of both Am self-sustaining and burning modes in terms of decay heat for the charged assembly and discharged assembly after 5 years of cooling: those are due to the larger Am content in the AmBB blanket.

Finally, the mass of helium (in kg per initial heavy metal metric tonne, IHMMT) generated in the AmBB during irradiation was estimated. Helium is generated from decay of Cm-242 (half-time is 0.45 yr), which is continuously produced through beta-decay of Am-242. The AmBB self-sustaining case generates 0.43 kg/IHMMT of helium, while 0.33 kg/IHMMT of helium is generated by the AmBB burning case. These results can be compared with the mass of 0.01 kg/IHMMT of helium generated by the reference case. High helium generation leads to higher pressure in the pin of the blanket, which should be properly taken into account for designing the assembly in order to avoid failure of the cladding. Helium production is mostly affected by the presence of Am-241 in the AmBB. The differences in He generation observed between transmutation modes are explained by the different isotopic contents of Am charged in the AmBB, since PWRs generate higher content of Am-243 than SFRs.
Table 1: AmBB transmutation characteristics in ASTRID

<table>
<thead>
<tr>
<th>Fuel Cycle Strategy</th>
<th>Ref. ASTRID No recycle of Am</th>
<th>Am self-sustaining mode</th>
<th>Am burning mode</th>
</tr>
</thead>
<tbody>
<tr>
<td>Pu content in driver fuel</td>
<td>~24%</td>
<td>23.9%</td>
<td>23.6%</td>
</tr>
<tr>
<td>Am fraction in AmBB fuel</td>
<td>N/A</td>
<td>15%</td>
<td>15%</td>
</tr>
<tr>
<td>No. AmBB blanket assemblies</td>
<td>0</td>
<td>24</td>
<td>66</td>
</tr>
<tr>
<td>Supporting ratio (driver/AmBB)</td>
<td>N/A</td>
<td>12.1 (= 291/24)</td>
<td>N/A</td>
</tr>
<tr>
<td>Avg burnup AmBB blanket, GWd/t</td>
<td>-</td>
<td>25</td>
<td>19</td>
</tr>
<tr>
<td>Sodium void worth, pcm</td>
<td>14</td>
<td>28</td>
<td>-1</td>
</tr>
<tr>
<td>MA sent to disposal, kg/cycle</td>
<td>20.7</td>
<td>4.6 (Np, Cm+)</td>
<td>14.1 (Np, Cm+)</td>
</tr>
<tr>
<td>External Am burning, kg/cycle</td>
<td>-</td>
<td>-</td>
<td>25.9</td>
</tr>
<tr>
<td>Decay heat, W/assembly</td>
<td>a) 116</td>
<td>b) 2,000</td>
<td>b) 1,720</td>
</tr>
<tr>
<td>- Charge</td>
<td>a) 461</td>
<td>b) 4,660</td>
<td>b) 4,790</td>
</tr>
<tr>
<td>- Discharge + 5 years</td>
<td>a) 0.01</td>
<td>b) 0.43</td>
<td>b) 0.33</td>
</tr>
<tr>
<td>He production, kg/iHMMT</td>
<td>a) Driver, b) Blanket</td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

**Transmutation performance of AmBB in ABR-1000**

In the GNEP project, homogeneous recycling of Pu or TRU together with U in the driver fuel assemblies was studied. In Table 2, the core performance characteristics of heterogeneous recycling with AmBB target assemblies in the metallic-fuelled ABR-1000 are compared with the homogeneous Pu and TRU recycling. In Table 3, the difference of the heterogeneous recycling core characteristics of metallic and oxide-fuelled ABR-1000 are compared. Only Am self-sustaining case is considered with ABR-1000.

The AmBB scenario applied to the metallic core ABR-1000 provides high supporting ratio: at equilibrium state, a single AmBB assembly with 16.9% Am can burn the Am generated from 50 driver assemblies. This high supporting ratio is due to the harder neutron spectrum associated to metallic fuel when compared with an oxide fuel. In fact, the supporting ratio of the oxide-fuelled ABR-1000 core with AmBB strategy shown in Table 3 is down to ~17 driver fuel assemblies supported per AmBB target assembly. The difference still observed with ASTRID also comes from shorter cooling and fabrication times targeted with ABR-1000 cores, those reduce the fraction of Pu-241 that decays into Am-241.

An important challenge associated with homogeneous TRU recycle observed here comes from the higher charged and discharge decay heat, which affects every driver fuel assembly. As shown in Table 2, only the three target AmBB assemblies are affected by increase in decay heat due to Am loaded. The helium production is not significantly increased in the TRU recycling case, it is much larger in the AmBB assemblies with 0.35 and 0.39 kg/iHMMT of helium generated in the oxide and metallic cases, respectively.
Table 2: AmBB transmutation characteristics in metallic ABR-1000 concept

<table>
<thead>
<tr>
<th></th>
<th>Homogeneous Pu recycle</th>
<th>Heterogeneous Driver</th>
<th>Heterogeneous AmBB</th>
<th>Homogeneous TRU recycle</th>
</tr>
</thead>
<tbody>
<tr>
<td>Pu content in driver fuel, %</td>
<td>14.6</td>
<td>14.5</td>
<td>-</td>
<td>14.4</td>
</tr>
<tr>
<td>Am fraction in AmBB fuel, %</td>
<td>-</td>
<td>-</td>
<td>16.9</td>
<td>-</td>
</tr>
<tr>
<td>No. assemblies</td>
<td>a) 150 / 30</td>
<td>a) 150 / 27</td>
<td>3</td>
<td>a) 150 / 30</td>
</tr>
<tr>
<td>Supporting ratio</td>
<td>N/A</td>
<td>50.0</td>
<td>N/A</td>
<td>8.2</td>
</tr>
<tr>
<td>Average burnup, %</td>
<td>8.4</td>
<td>8.4</td>
<td>3.9</td>
<td>8.2</td>
</tr>
<tr>
<td>MA sent to disposal, kg/year</td>
<td>5.0</td>
<td>1.8 (Np,Cm)</td>
<td>0.8 (Np,Cm)</td>
<td>~0</td>
</tr>
<tr>
<td>Decay heat, W/Assembly</td>
<td>b) 80</td>
<td>b) 81</td>
<td>1,923</td>
<td>b) 438</td>
</tr>
<tr>
<td>- Charge</td>
<td>b) 410</td>
<td>b) 410</td>
<td>6,195</td>
<td>b) 652</td>
</tr>
<tr>
<td>- Discharge + 5 years</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>He production, kg/MHMNT</td>
<td>b) 0.002</td>
<td>b) 0.002</td>
<td>0.39</td>
<td>b) 0.012</td>
</tr>
</tbody>
</table>

a) Driver / Radial blanket, b) Driver

Table 3: Comparison of AmBB Transmutation Characteristics in ABR-1000 Metal and oxide cores

<table>
<thead>
<tr>
<th></th>
<th>Metal</th>
<th>Oxide</th>
</tr>
</thead>
<tbody>
<tr>
<td>Pu content in driver fuel, %</td>
<td>Driver</td>
<td>AmBB</td>
</tr>
<tr>
<td>Am fraction in AmBB fuel, %</td>
<td>-</td>
<td>16.9</td>
</tr>
<tr>
<td>No. assemblies</td>
<td>150 / 27</td>
<td>3</td>
</tr>
<tr>
<td>Supporting ratio</td>
<td>50.0</td>
<td></td>
</tr>
<tr>
<td>Average burnup, %</td>
<td>8.4</td>
<td>3.9</td>
</tr>
<tr>
<td>MA sent to disposal, kg/year</td>
<td>1.8 (Np,Cm)</td>
<td>0.8 (Np,Cm)</td>
</tr>
<tr>
<td>Decay heat, W/Assembly,</td>
<td></td>
<td></td>
</tr>
<tr>
<td>- Charge</td>
<td>81</td>
<td></td>
</tr>
<tr>
<td>- Discharge + 5 years</td>
<td>410</td>
<td>6,195</td>
</tr>
<tr>
<td>He production, kg/HHMMT</td>
<td>0.002</td>
<td>0.39</td>
</tr>
</tbody>
</table>

AmBB transmutation characteristics

The typical irradiation environment of AmBB assemblies located in ASTRID and metallic configuration of ABR-1000 was researched to support the development of representative irradiation experiments. Calculations were performed using the stochastic code SERPENT [12] for generating radial pin-by-pin power and flux profiles during depletion. The axial power profile was obtained with the DIF3D code [9]. This calculation is aimed at providing the trends in the flux and temperature of different pins of one selected AmBB blanket assembly for ASTRID and ABR-1000. In order to track the evolution of the isotopic compositions depending on their position in the selected assembly, the different pins #1 to #8 displayed in Figure 5 are tallied separately with SERPENT. Thermal-hydraulic calculations were performed with the SAM code [15] in the flow channels defined surrounding the fuel pin located at each assembly corner. This calculation takes into account variations in the power of the different fuel pins of the AmBB assembly, the lower velocity at the border of the assemblies, and the flow mixing within the channels. The total velocity was calculated so that the average outlet coolant temperature of the AmBB assemblies are 510°C and 550°C near the end of their lifetime in the ABR-1000 and ASTRID cores, respectively.
The peak fuel temperature, the radial power peaking, and the change in spectrum hardness are displayed in Table 4. In this table, the radial power peaking factor was defined by the pin power to the average fuel pin power in the assembly. The results shown here display especially large variation in the power peaking for both the ABR-1000 metallic core and the ASTRID core. Higher power peaking is especially found in the pin closer to the driver fuel. This pin is also associated with higher flux level as observed in Figure 6 and harder neutron spectrum, as also shown in Table 4. Consequently, the americium content in these specific pins is burnt faster, as shown in Figure 7. The peak fuel temperature in Table 4 displays large variations from one pin to another, especially in the case of the ASTRID core due to the use of oxide fuel pins. In the AmBB fuel assembly of the ASTRID core, there is about 60°C of variation in the coolant and cladding temperatures, and about 300°C of variation in the peak fuel temperatures at the end of life. The variation in peak fuel temperature is about 150°C in the metallic-fuelled ABR-1000 AmBB assemblies. The temperatures also increase significantly during irradiation due to the increased fission power generated in the assembly and the reduced thermal conductivity, more specifically for the oxide fuel, with increased burnup.

**Table 4:** Comparison of AmBB power peaking and fuel temperature at end of life, and spectrum shift in AmBB assembly for ASTRID and ABR-1000

<table>
<thead>
<tr>
<th>Pin #</th>
<th>ASTRID</th>
<th>ABR-1000</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Peak fuel temp</td>
<td>Power peaking</td>
</tr>
<tr>
<td>#3</td>
<td>656</td>
<td>0.8</td>
</tr>
<tr>
<td>#4</td>
<td>666</td>
<td>0.8</td>
</tr>
<tr>
<td>#5</td>
<td>843</td>
<td>1.3</td>
</tr>
<tr>
<td>#6</td>
<td>988</td>
<td>1.7</td>
</tr>
<tr>
<td>#8</td>
<td>-</td>
<td>1</td>
</tr>
</tbody>
</table>

The results summarised here show a significantly different behaviour within the pins of a typical AmBB assembly situated in the radial blanket region of the SFR core, as also observed in [16]. Such
detailed analysis is important for properly designing AmBB assemblies to meet peak cladding and fuel temperature limitations. The current suite of neutronic deterministic codes traditionally used is poorly suited for performing such analysis, especially for pin-wise depletion calculations. Monte Carlo codes have the potential for providing invaluable information as shown in this section but at the price of large computational cost in order to reduce the statistical uncertainty in a fine-mesh pin-wise model. Advanced code systems such as PROTEUS [17] developed in the framework of the NEAMS programme display high promises since they allow detailed modeling while taking advantage of the deterministic approach and of the capability of multi-physics coupling. This analysis also shows that those target assemblies could benefit in being turned inside out during their irradiation to homogenise radially the burnup and fluence seen in the different blanket pins.

**Figure 6**: Comparison of neutron flux spectrum in different fuel pin of AmBB assembly for ASTRID and ABR-1000

**Figure 7**: Evolution of Am fraction in different fuel pin of AmBB assembly for ASTRID and ABR-1000

**Conclusions**

Several potential minor actinide transmutation paths involving SFRs that can be used in a US and in a French strategy were investigated with the objective to prepare irradiation experiments in the
Advanced test reactor (ATR). Minor Actinides can be transmuted in an SFR with a homogeneous strategy where they are mixed in the driver fuel, or with a heterogeneous strategy where they are mixed with depleted uranium in a blanket region such as the americium-bearing blanket (AmBB) concept. Both types of transmutation modes were analysed based on oxide and metallic-fuelled SFRs, and are found to be efficient with specific benefits and challenges with a focus on the evaluation of core and fuel cycle performance change for using AmBB in ASTRID and ABR-1000. The heterogeneous strategy concentrates the fuel cycle challenges on a smaller number of target assemblies. Those target assemblies display large decay heat with larger challenges associated with their handling during operations such as loading, discharge and cleaning. Helium generation in target assemblies is linked to the content of americium transmuted and will require adapted pin design. However, these target assemblies can benefit from a modular management to optimise the irradiation and reduce the fuel cycle challenge.

In the ASTRID core, 24 AmBB assemblies loaded with 15% Am content allow continuous recycling of the Am, while 66 AmBB assemblies (one row surrounding the driver fuel) allows continuous recycling and transmutation of the Am from about 1.8 GWe of PWR. The impact of AmBB assemblies on ASTRID performance and safety were confirmed to be small, but helium production and decay heat are especially large in these assemblies. The ABR-1000 concept displays similar core performance characteristics between continuous U/Pu and U/TRU homogeneous recycling, and U/Pu+AmBB heterogeneous recycling strategies. However, the metallic-fuelled core configuration requires fewer AmBB blanket assemblies compared to the oxide-fuelled core due to the harder neutron spectrum and smaller MA production rate.

Finally, detailed analyses were performed to determine irradiation conditions of AmBB fuels loaded in metallic ABR-1000 and ASTRID cores to support the development of irradiation experiments of AmBB fuel concepts in the ATR. Significant differences in linear power, neutron flux and irradiation performance were found within an AmBB assembly. Those need to be taken into account for proper modeling and also for design optimisation.

Acknowledgements

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References


Plenary Session IV: Transmutation Fuels and Targets

Chair: N. Chauvin and L. Ramond
Advances in metallic fuels for high burnup and actinide transmutation

S. L. Hayes, J. M. Harp, H. J. M. Chichester, R. S. Fielding, R. D. Mariani, W. J. Carmack
Idaho National Laboratory

Abstract

Research and development activities on metallic fuels in the US are focused on their potential use for actinide transmutation in future sodium fast reactors. As part of this application, there is a desire to demonstrate a multifold increase in burnup potential.

A number of metallic fuel design innovations are under investigation with a view towards significantly increasing the burnup potential of metallic fuels, since higher discharge burnups equate to lower potential actinide losses during recycle. Promising innovations under investigation include: 1) lowering the fuel smeared density in order to accommodate the additional swelling expected as burnups increase, 2) utilising an annular fuel geometry for better geometrical stability at low smeared densities, as well as the potential to eliminate the need for a sodium bond, and 3) minor alloy additions to immobilise lanthanide fission products inside the metallic fuel matrix and prevent their transport to the cladding resulting in fuel-cladding chemical interaction.

This paper presents results from these efforts to advance metallic fuel technology in support of high burnup and actinide transmutation objectives. Highlights include examples of fabrication of low smeared density annular metallic fuels, experiments to identify alloy additions effective in immobilising lanthanide fission products, and early post-irradiation examinations of annular metallic fuels having low smeared densities and palladium additions for fission product immobilisation.

Introduction

Metallic fuels have a history of use that spans the entire nuclear age, especially in association with the development of liquid metal fast breeder reactors. Initially, metallic fuels were employed due to their ease of fabrication and high heavy metal density (which led to the most favourable breeding efficiencies in fast reactors). During subsequent decades of use, metallic fuel technology has matured significantly, leading to the realisation of major benefits in the areas of fuel reliability and burnup, proliferation-resistant recycling, remote fabrication, and passive reactor safety [1]. Such characteristics continue to make metallic fuels a highly attractive technology, especially for use with future sodium-cooled fast reactors (SFR).

Current research and development activities in the US on metallic fuels are focused on their potential use for actinide transmutation in advanced SFRs. In such a mission, metallic fuels would be employed as part of a closed fuel cycle into which plutonium and minor actinides (Am, Np) recovered from LWR spent fuel would be incorporated. All actinides in this metallic fuel would be fissioned or transmuted in a fast reactor to eliminate the extremely long-lived, heat-producing actinides in order to reduce the volume, heat load, and radiotoxicity burden on a future geological repository [2]. Thus,
the need exists to demonstrate reliable performance of metallic fuels with Pu and minor actinide constituents to very high burnup.

In order to address increasingly high burnup demands on metallic fuel, a number of design innovations are under investigation. Promising innovations include: 1) lowering the fuel smeared density in order to accommodate the additional swelling expected as burnups increase, 2) utilising an annular fuel geometry for better geometrical stability at low smeared densities, as well as the potential to eliminate the need for a sodium bond, and 3) minor alloy additions to immobilise lanthanide fission products inside the metallic fuel matrix and prevent their transport to the cladding resulting in fuel-cladding chemical interaction.

This paper presents results from these efforts to advance metallic fuel technology in support of high burnup and actinide transmutation objectives. Examples are included from fabrication experiments of low smeared density annular metallic fuels, experiments to identify alloy additions to immobilise lanthanide fission products, and early (i.e. low burnup) post-irradiation examinations of annular metallic fuels having low smeared densities and palladium additions for fission product immobilisation.

Low smeared density, annular metallic fuels

Within the first 2% burnup, metallic fuels swell approximately 30% due to fission gas generation, nucleation into bubbles, and bubble growth. At 30% swelling, a significant fraction of the fission gas bubbles begin to interconnect to each other as well as to fuel slug free surfaces, such that a majority of the fission gases are released from the fuel matrix into the gas plenum from that point forward. Fuel swelling continues beyond 2% burnup due to the accumulation of solid fission products, but at the much slower rate of about 0.5% swelling per percent burnup [3].

Metallic fuel fabricated at the historical value of 75% smeared density does not begin to experience fuel-cladding mechanical interaction (FCMI) due to fuel swelling of any significance until between 10% and 20% burnup. However, the desire to demonstrate metallic fuel performance to ultra-high burnups (i.e. 30-40%) will result in unacceptably high FCMI caused by fuel swelling, almost certainly resulting in fuel pin failure, for fuel pins at 75% smeared density. For this reason, current experiments are investigating the fabrication and irradiation performance of metallic fuels with lower smeared densities (as low as 55%).

Fuel pins that include solid, cylindrical metallic fuel slugs at 55% smeared density have a small fuel slug diameter relative to the cladding, resulting in a very large fuel-cladding gap. Since metallic fuel pins are traditionally sodium-bonded, this presents no problem from a thermal performance point of view. However, there could be some concern regarding the geometric stability (i.e. slumping) of a very long, small diameter metallic fuel slug operating at high temperature inside a relatively large diameter cladding tube. Although a previous low smeared density metallic fuel experiment irradiated in the Experimental Breeder Reactor II (EBR-II) showed no evidence of fuel slumping, some anomalous structural features and development of a non-uniform fuel geometry were observed [4].

As an alternative design option for low smeared density fuels, use of an annular metallic fuel slug is being investigated. Annular metallic fuels offer two possible benefits over solid, cylindrical fuel slugs for low smeared density designs: 1) the annular fuel slug can be in mechanical contact with the cladding at fabrication, which may provide an added geometrical stability to the fuel during irradiation, thereby mitigating slumping concerns, and 2) with essentially no fuel-cladding gap at the time of fabrication, the sodium bond generally needed for efficient heat transfer may be eliminated.
The performance issue associated with this design feature, however, is fuel swelling. Will the large amount of swelling associated with metallic fuel be adequately accommodated by the void volume located in the fuel slug annulus without excessive cladding strain?

Annular metallic fuel slugs have been fabricated for testing in order to evaluate their performance during irradiation. Although not expected to be a fabrication method amenable to commercial scale operations, a small number of annular fuel slugs were prepared for irradiation testing by casting as solid, cylindrical slugs, sectioned to the final length, and machined to final inner and outer diameters. An example of a 55% smeared density U-10Mo fuel slug fabricated for irradiation testing in the advanced test reactor (ATR) is shown in Figure 1.

Figure 1: Annular fuel slug fabricated at 55% smeared density

For the irradiation experiments in ATR, the annular metallic fuel slugs were slip fit into cladding tubes with a nominal fuel-cladding gap of 40 µm at fabrication; thermal analyses indicated that this small gap would be closed by differential thermal expansion of the fuel and cladding when the fuel pin was at normal operating conditions in ATR.

Alloy additions for fission product immobilisation

The principal failure mechanism of concern for metallic fuels is creep rupture of the stainless-steel cladding driven by plenum pressurisation from released fission gases. Cladding creep is accelerated with increasing temperature and fluence, FCMI (which is largely precluded by the low smeared density fuel pin design), and fuel-cladding chemical interaction (FCCI). FCCI attack on the cladding results in the growth a brittle interaction layer on the cladding inner surface; this interaction layer has essentially no strength, so its growth into the cladding thins the unreacted, load-bearing wall thickness, increasing cladding stress and accelerating cladding creep. The rate of cladding attack by FCCI increases with temperature and burnup. Lanthanide fission products (principally La, Ce, Pr, Nd, and Sm) are the species that are responsible for cladding attack. Furthermore, the interaction product formed by these lanthanide elements reacting with the iron-based cladding generally exhibits liquefaction at a relatively low temperature (~700°C) and can reduce the thermal margin of the fuel pin, of concern during certain transient scenarios [5].

Coatings/liners applied to the inside surface of the cladding to act as a diffusion barrier against fission product attack are under investigation. However, the reliable application of these coatings/liners to the inside surface of a very long, small diameter cladding tube presents a manufacturing challenge. As an alternative to coatings/liners for FCCI mitigation, minor alloy
additions to metallic fuel that would chemically bind in situ with the lanthanide fission products and immobilise them in the fuel matrix are being sought.

Preliminary out-of-pile experimentation with palladium as an alloying element is showing promise. Casting experiments on metallic fuels spiked with lanthanide elements, followed by microstructural characterisation, indicate that metallic fuels that incorporate a small amount of palladium into the fuel alloy result in microstructures in which the palladium and lanthanide elements precipitate together as a distinct, stable, intermetallic phase [6], as seen in Figure 2.

**Figure 2:** Backscattered electron image of U-15Zr-3.86Pd-4.3Ln (Ln = 53Nd-25Ce-16Pr-6La, wt.%) with distinct Pd-Ln precipitates

In addition, isothermal inter-diffusion studies between iron and rare earth elements showed remarkably different behaviour when palladium was introduced into the system. As illustrated in Figure 3, a diffusion couple of Fe vs. Nd held at 700°C for 100 hours experienced extensive inter-diffusion, including liquefaction at the Fe-Nd interface, whereas under identical conditions no inter-diffusion is when the Nd side of the couple is changed to a 1-to-1 ratio of Pd:Nd [7].

Assuming this out-of-pile behaviour could be realised during irradiation, the quantity of palladium needed to immobilise the lanthanide fission products produced during irradiation would depend on the anticipated discharge burnup of the fuel. At 8% burnup, metallic fuel contains approximately 2 wt.% lanthanides and would require 1.6 wt.% palladium to bind lanthanides at a 1:1 stoichiometric ratio [6]. This means 2 wt.% palladium in the metallic fuel alloy would be needed to control lanthanides up to 10% burnup, or 4 wt.% palladium for fuel going to 20% burnup.

**Figure 3:** Diffusion couple results at 700°C for 100 hours. (a) Fe vs. Nd resulting in inter-diffusion and liquefaction, and (b) Fe vs. Pd:Nd shows no inter-diffusion
Palladium additions could also prove to be beneficial for fuels fabricated with transuranic feedstock coming from electrochemical recycling of metallic fuels. Some lanthanide fission product carryover is expected as part of this recycling process, which could accelerate FCCI attack relative to the rate observed in metallic fuels fabricated using pure actinide feedstocks. An additional measure of palladium to stabilise lanthanides from carryover could mitigate this concern.

Irradiation testing and post-irradiation examinations

To investigate the performance of the innovative design features outlined above (namely, low smeared density, annular metallic fuels with the addition of palladium to the fuel alloy), a series of irradiation experiments has been initiated in the ATR. These experiments irradiate miniature fuel pins (designated “rodlets”) of prototypic radial, but much reduced axial, dimensions inside of a cadmium-shrouded test train. Although not enhancing the fast neutron flux, the cadmium-shrouded experiment virtually eliminates the thermal flux and results in a fairly prototypic radial temperature profile within the test rodlets [8].

The first irradiation test in this series (AFC-3A) included 4 test fuel rodlets irradiated to approximately 3% burnup at linear powers between 330 and 360 W/cm. The test matrix for this experiment is shown in Table 1. The metallic fuels included in this experiment did not contain plutonium or minor actinides (greatly simplifying their fabrication), since the design features that were the focus of the experiment were not expected to be strongly dependent on transuranic content of the fuel.

<table>
<thead>
<tr>
<th>Rodlet</th>
<th>Fuel Alloy</th>
<th>Slug Form</th>
<th>Smeared Density</th>
<th>Burnup (% HM)</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>U-10Mo</td>
<td>Solid</td>
<td>75</td>
<td>2.3</td>
</tr>
<tr>
<td>2</td>
<td>U-10Mo</td>
<td>Annular</td>
<td>55</td>
<td>3.3</td>
</tr>
<tr>
<td>4</td>
<td>U-10Zr</td>
<td>Annular</td>
<td>55</td>
<td>3.2</td>
</tr>
<tr>
<td>5</td>
<td>U-1Pd-10Zr</td>
<td>Solid</td>
<td>75</td>
<td>2.5</td>
</tr>
</tbody>
</table>

Post-irradiation examination of the test rodlets from AFC-3A is underway, but not yet fully complete. Nonetheless, several observations can be made. Figure 4 shows transverse metallography performed on Rodlet 1 containing 75% smeared density U-10Mo fuel. The metallic alloy system based on U-Mo has been proposed as an alternative to the traditional U-Zr system, since it should not be subject to fuel constituent redistribution as is U-10Zr [9]. Although there is no indication of constituent redistribution, considerable fuel-cladding interaction has occurred even at this low burnup level. The upper left portion of the cladding has been thinned to 340 µm from its original 440 µm (a 23% reduction). The attack on the HT9 cladding is from the U-10Mo fuel itself, not lanthanide fission products. This fundamental fuel-cladding incompatibility appears to be so significant as to rule out the U-Mo alloy system as a viable alternative to U-Zr as a fast reactor fuel, unless an acceptable coating/liner were available for use.
Figure 4: Cross section of U-10Mo solid fuel (Rodlet 1)

Figure 5 (a) shows transverse metallography performed on Rodlet 4 containing annular, 55% smeared density U-10Zr fuel. There is some localised FCCI apparent at reduced number of contact points between fuel and cladding on the right side of the image. It would appear that the slip fit of the annular fuel slug into this cladding tube resulted in poor thermal contact in this area of the fuel-cladding interface. The limited number of contact points in this area would have increased the local heat flux at these locations, resulting in higher cladding temperatures than expected. This probably explains the FCCI associated with these locations. Subsequent investigation suggests that this effect was probably the result of poor contact between fuel slug and cladding at fabrication, which did not correct itself on heat up in reactor, but that it can be eliminated with more precise machining. As for fuel swelling, it is clear that swelling into the central annulus has occurred, and no cladding strain is measurable at this low burnup level.

Finally, Figure 5 (b) shows transverse metallography performed on Rodlet 5 containing 75% smeared density U-1Pd-10Zr fuel. At this low level of palladium, the U-1Pd-10Zr fuel alloy appears to perform analogously to U-10Zr, including the apparent onset of constituent redistribution, but with the notable exception that there is no indication of any FCCI observable. Since some FCCI occurred in the U-10Zr fuel of Rodlet 4, operating under essentially identical conditions, this suggests that the palladium addition to the fuel alloy may be having the desired effect of immobilising lanthanide fission products.
**Figure 5**: Cross section of U-10Zr annular fuel (left, Rodlet 4) and U-1Pd-10Zr solid fuel (right, Rodlet 5)

(a) (b)

A more detailed SEM examination of the U-1Pd-10Zr fuel microstructure in Rodlet 5 is scheduled for later this year, which should shed additional light on the role played by palladium relative to lanthanide fission mobility.

**Conclusion**

With a view to applying the US’s historic metallic fuel technology to an advanced fuel cycle that includes recycle, and may include actinide partitioning and transmutation, a number of innovative design features have been under investigation for the past several years. These include low smeared density, annular fuels having the potential to achieve ultra-high burnup without limitations from fuel-cladding mechanical interaction. Also included is the addition of palladium as a minor addition to the metallic fuel alloy, with the expectation that it will react with lanthanide fission products in situ during irradiation, form stable and immobile intermetallic phases with them in the fuel matrix, and thereby mitigate or completely eliminate fuel-cladding chemical interaction. Finally, there has been some interest in the possible advantages that could come from the U-Mo system over the U-Zr system, especially relative to fuel constituent redistribution.

Having initially investigated these possible design changes out-of-pile, positive results from these investigations have motivated an irradiation test series to assess their performance in an integral fuel experiment. While this test series is still in its infancy, results from the first low burnup test (AFC-3A) are becoming available. Conclusions at this early time include: 1) although constituent redistribution is not observed in U-10Mo under irradiation, a fundamental incompatibility between U-10Mo fuel and HT9 cladding probably precludes its use as a fast reactor fuel; 2) annular, low smeared density U-10Zr fuel appears to be geometrically stable, accommodates the large amount of swelling associated with metallic fuel in the central annulus, and does not induce cladding strain; and 3) a very preliminary result at low burnup seems to confirm that palladium additions to metallic fuel do act to mitigate FCCI by immobilising lanthanide fission products in the fuel matrix.
It appears that modest design enhancements to the historic metallic fuel design can make it even more attractive for application in advanced fast reactors operating with a closed fuel cycle, including the option of actinide partitioning and transmutation.

Acknowledgements

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References


State-of-the-art report on innovative fuels

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Abstract

Spent fuel management has become one of the major concerns of the last few decades and an option, in some countries, would be to reprocess the fuel by implementing new innovative technologies in various nuclear fuel cycle schemes like minor actinides transmutation. Several member states have initiated R&D programmes to study the safety aspects of new types of fuel containing minor actinides for burning under fast neutrons. To review those programmes and the most recent advancements in the field, the Expert Group on Innovative Fuels (working under the guidance of the Working Party on Scientific Issues of the Fuel Cycle [WPFC]) prepared a state-of-the-art report on innovative fuels in 2014.

This report covers the study of any fuel containing minor actinides (MA) to be introduced in advanced nuclear systems and reviews the available data on such MA actinides bearing fuels. Different fuel types were reviewed (metal, oxide, nitrides, dispersions fuels and special mechanical fuel forms) mainly for fast neutron reactors examining different technical issues associated with their fabrication, characterisation, irradiation performance, design and safety criteria. The technical maturity of each fuel was also assessed.

Among these fuels, metal and oxide fuels containing minor actinides have been widely investigated and irradiations tests and PIE have been performed. Additional results, however, are needed to gain further insights into He-gas release and FCCI (fuel cladding chemical interactions). Nitrides fuels have numerous advantages for use in ADS compared to other types of fuels due to their higher density. However, these fuels are still at an early stage of development and more fundamental research is needed to determine their safety performance.

Dispersion fuels are a relatively new concept. The first irradiation tests on inert matrix fuel containing MA started in the mid-90s as part of the irradiation programme EFTTRA and were followed by other irradiation programmes. Three types of dispersion fuel have now been subjected to safety tests. Finally, minor actinides can be integrated into special mechanical fuel forms (particles) which are considered as a new concept, namely VIPAC and SPHEREPAC. Some irradiation experiments have marked important milestones in the R&D of transmutation fuels. However, the examination of the behaviour and safety issues of minor actinide bearing fuels are limited to laboratory scale (grams quantities of TRU) and irradiation tests have been limited to small samples or rodlets. More experimental data coupled to modelling are needed to gain better understanding of fuel behaviour under operating conditions.
Microstructural characterisation of metallic transmutation fuels

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Abstract

Metallic fuels are considered for application in advanced fast reactors because of their high burnup, high fissile and fertile density capability, and high thermal conductivity with significant safety benefits [1]. Uranium-plutonium-zirconium (U-Pu-Zr) alloys are considered to be one of the most promising metallic fuels. The addition of Zr in U-Pu matrix was sought to increase the melting temperature of U-Pu alloys and to enhance compatibility between the fuel and stainless-steel cladding by suppressing the inter-diffusion of fuel and cladding constituents during steady-state reactor operations.

Extensive studies have been carried out to understand fuel swelling, fuel performance during transients, failure mechanisms, behaviour following benign failure and complete failure of fuel pins, and mechanical and chemical fuel-cladding interactions [2-4]. Constituent migration between radial zones of a cylindrical fuel element and their redistribution can lead to inhomogeneity of the initially uniformly mixed alloys. Inhomogeneity of metallic fuels can lead to phase transformations, solidus temperature change, and local changes in fissile atom density, which can alter mechanical and physical properties of the fuel and affect its behaviour and performance [4]. Understanding fuel constituent redistribution that alters alloy composition and affects fuel performance is imperative for safe operation of the reactor. However, complete understanding of the irradiation behaviour of U-Pu-Zr transmutation fuels cannot be achieved without accurate knowledge of the phases and microstructure of un-irradiated fuels. In addition, to comprehend irradiation-induced changes in U-Pu-Zr alloys and consequent fuel-cladding chemical interaction (FCCI), the phases and microstructure of un-irradiated fuels should be characterised first. A detailed structural and chemical compositional analysis of phases formed in these fuel alloys and phases formed within FCCI layer is yet to be conducted.

This contribution reports the results from ongoing work on microstructural characterisation of un-irradiated U-Pu-Zr and Pu-Zr fuels. Phase identification in these alloys has been performed using energy and wavelength dispersive spectroscopy (EDS/WDS), scanning and transmission electron microscopy (SEM/TEM), selective area electron diffraction (SAED), and atom probe tomography (APT). In addition to characterisation of fresh fuels, this work investigates FCCI between U-Pu-Zr fuel and Fe cladding at elevated temperatures, understanding of which is critical for evaluation of the fuel performance.

This work was supported by the Fuel Cycle Research and Development (FCRD) programme of the US Department of Energy.
References

Overview of post-irradiation examination results on minor actinide bearing fuels within the FUTURIX-FTA international collaboration

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Abstract

Transmutation of minor actinides and long-lived fission products in fast neutron spectrum systems is a promising possibility to reduce the long-term radiotoxicity of spent nuclear fuel. In order to investigate the safety aspects of irradiation behaviour of a wide range of conceptual Am-bearing uranium-free fuels for accelerator-driven systems (ADS) as well as low-fertile fuels for Fast Spectrum Fission Reactors, the FUTURIX/FTA irradiation experiment has been implemented as an international co-operation between CEA, JRC-ITU, US/DOE and JAEA. The programme started in 2004 and irradiation was performed from 2007 to 2009 in the Phénix fast spectrum reactor in Marcoule, France. Eight Am-bearing fuel compositions (two metallic, two nitride, two Mo-CERMET and two MgO-CERCER fuel types with Am contents ranging from 0.3 to 2.6 g.cm\textsuperscript{-3}) have been investigated.

The pins were irradiated for up to 235 EFPD to burnups from 2 to 18 at% at Linear Heat Rates (LHR) ranging from 80 to 450 W.cm\textsuperscript{-1} according to the fuel composition, the location in the core and the irradiation duration. After irradiation the pins were transported to hot cells of JRC-ITU (CERMET fuels), CEA Cadarache (CERCER fuels) and INL (metallic and nitride fuels) for execution of post-irradiation examinations (PIE).

After a review of the fuel preparation conditions, the pins features and the irradiation conditions, the paper gives an overview of the performed post-irradiation examinations and the results gained thus far.
Preliminary post-irradiation examination comparison between AFC-1 and FUTURIX-FTA

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Abstract
The destruction of long-lived minor actinide isotopes generated in irradiated nuclear fuel by transmutation in fast reactors is a goal of the US DOE Fuel Cycle Research & Development programme. Transmutation would dramatically decrease the volume of material requiring disposal and reduce the longer-term radiotoxicity and heat load of high-level waste (HLW) sent to a geologic repository. In an effort to better understand the fuel performance implications of adding minor actinides to a fuel system, the Advanced Fuel Cycle (AFC) experiments examine the performance of metallic, nitride, and ceramic fuels in approximated fast reactor neutron spectra at the Idaho National Laboratory Advanced Test Reactor. Fast neutron reactor neutron spectra are simulated through the use of a cadmium filter placed in an ATR irradiation position. In addition to ATR testing, the FUTURIX-FTA irradiations were performed at the Phénix reactor in France to confirm that behaviour observed in ATR testing was representative of a true fast neutron reactor spectrum. This irradiation experiment subjected a subset of fuel compositions from AFC testing in ATR to a real fast neutron spectrum. Irradiation tests designated AFC-1B, AFC-1D, AFC-1F, AFC-1G, AFC-1H, FUTURIX-FTA DOE1 and FUTURIX-FTA DOE2 contain pins with low-fertile and non-fertile actinide bearing metallic alloy fuel compositions. Irradiation tests designated AFC-1Æ, AFC-1G, FUTURIX-FTA DOE3 and FUTURIX-FTA DOE4 contain pins with both low-fertile and non-fertile actinide bearing nitride fuel compositions. The AFC-1 series tests were irradiated to a range of burnups from 3.0 to 30.2 atom % heavy metal (2.7x10^{20} – 3.96x10^{21} fissions/cm^3) while FUTURIX-FTA were irradiated a range of burnups from 1.57 to 15.5 at% heavy metal. Preliminary results such as optical microscopy, fission product distribution, and final actinide composition will be compared between the FUTURIX-FTA pins and the AFC-1 experiments to assess the applicability of using a modified thermal spectrum reactor (e.g. ATR with cadmium filters) for testing fast reactor fuels. Performance data from all these irradiations can be used to inform the feasibility of minor actinide transmutation in future reactor systems.
Post-irradiation examination of fast reactor metal fuel for minor actinide transmutation

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2European Commission, Joint Research Centre, Karlsruhe, Germany

Abstract

METAPHIX is a collaborative programme between the Central Research Institute of Electric Power Industry (CRIEPI) and the Joint Research Centre, Karlsruhe (JRC Karlsruhe) for understanding the irradiation behaviour of fast reactor metal fuels containing minor actinides (MAs: Np, Am, Cm) and demonstrating the MA transmutation performance in fast reactors. Four U-Pu-Zr-based alloys, U-19Pu-10Zr, U-19Pu-10Zr-2MA-2RE (REs=rare-earth elements: Y, Ce, Nd, Gd), U-19Pu-10Zr-5MA-5RE and U-19Pu-10Zr-5MA (wt%), were fabricated at JRC-ITU (currently JRC Karlsruhe) in this programme. REs were added to simulate the contamination of products recovered by the pyrometallurgical reprocessing of LWR spent fuel. These fuel alloys were irradiated in the Phénix fast reactor with the support of CEA, achieving peak burnups of ~2.5 at.%, ~7.0 at.% and ~10.0 at.%. Post-irradiation examinations (PIEs) were performed at JRC Karlsruhe. Non-destructive tests (NDTs) such as cladding profilometry and axial gamma-ray spectrometry have been completed on all the irradiated METAPHIX fuel pins, which showed no appreciable change in the irradiation behaviour of the metal fuels caused by the addition of up to 5 wt% MAs and REs. Destructive examinations including plenum gas analysis, optical microscopy (OM), scanning electron microscopy (SEM) and electron probe microanalysis (EPMA) are currently in progress for the ~2.5 at.% and ~7.0 at.% burnup fuels. Chemical analysis using inductively coupled plasma-mass spectrometry (ICP-MS) was also performed. The irradiation behaviour of MA-bearing metal fuels was analysed using the ALFUS code. The results of the ALFUS calculation were consistent with the NDT results, regardless of the addition of MAs and REs. The change in the actinide compositions was calculated using a multigroup three-dimensional diffusion and burnup model code. The MA transmutation ratio during the METAPHIX irradiation experiment was reasonably well predicted by the calculation. The results of the PIEs obtained so far indicate that the presence of MAs in the metal fuel did not cause significant changes in the irradiation behaviour and that the MAs can be transmuted in fast reactor metal fuels as expected.

Introduction

The deployment of fast reactors (FRs) and related nuclear fuel cycles is an important option for achieving a sustainable nuclear energy supply to meet worldwide future demand. In addition, the long-term radiotoxicity of high-level waste (HLW) can be reduced by recovering long-lived minor actinides (MAs: Np, Am, Cm) as well as U and Pu from spent nuclear fuel and burning them in FRs [1-3]. In particular, metal fuel FRs facilitate the effective burning of MAs owing to the high heavy metal density and the high-energy neutron spectrum [4]. The METAPHIX programme is a collaborative
programme between the Central Research Institute of Electric Power Industry (CRIEPI, Japan) and the Joint Research Centre – Karlsruhe (JRC Karlsruhe, European Commission) to understand the irradiation behaviour of MA-bearing metal fuels and demonstrate the MA transmutation performance in FRs.

In this programme, metal fuel pins that include U-Pu-Zr-MA alloys were fabricated at JRC – Institute for Transuranium Elements (JRC-ITU, currently JRC Karlsruhe) and irradiated in the Phénix fast reactor up to three different burnups, 2.5at.% (METAPHIX-1), ~7at.% (METAPHIX-2) and ~10at.% (METAPHIX-3), with the support of the Commissariat a l’Energie Atomique et aux Energies Alternatives (CEA, France) [5]. This paper summarises the results of the METAPHIX programme obtained so far.

**Fuel composition**

Since the MAs recovered pyrometallurgically from light water reactor (LWR) spent fuels can be accompanied by a comparable amount of rare earth (RE) fission products owing to their chemical affinities, the effect of the contamination of MAs with REs must be taken into account. The characterisation of U-Pu-Zr alloys containing MAs and REs indicated that practically homogeneous fuel alloys can be prepared whose basic properties do not change, provided that the content of individual MAs and REs is limited to 5wt% or less [5]. Thus, three MA-bearing fuel alloys, U-19Pu-10Zr-2MA-2RE, U-19Pu-10Zr-5MA-5RE and U-19Pu-10Zr-5MA (wt%), were selected along with a standard ternary fuel alloy, U-19Pu-10Zr, for an irradiation experiment [5, 6]. Table 1 summarises the analysis results for the elemental compositions of the four fuel alloys prepared by the arc melting process [7]. These results indicate that the prepared fuel alloys have large uncertainties in the composition, which depends on the sample position, presumably due to the heterogeneity of the molten alloys. Table 2 shows the isotopic compositions of U, Pu, Am and Cm. The isotope composition of each actinide was expected to be uniform regardless of the homogeneity of the cast fuel alloy [7]

<table>
<thead>
<tr>
<th>Table 1: Measurement results for initial elemental compositions of fuel rods (wt%) [7]</th>
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<tbody>
<tr>
<td>U-19Pu-10Zr</td>
</tr>
<tr>
<td>-----------------------------------------------</td>
</tr>
<tr>
<td>Target</td>
</tr>
<tr>
<td>-----------------------------------------------</td>
</tr>
<tr>
<td>U</td>
</tr>
<tr>
<td>Pu</td>
</tr>
<tr>
<td>Np</td>
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<td>Am</td>
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<tr>
<td>Ce</td>
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<tr>
<td>Nd</td>
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<tr>
<td>Gd</td>
</tr>
</tbody>
</table>

*: Average of 14 sample analyses. #upperbound, #lowerbound, Impurities < 0.4 wt%.
Table 2: Measurement results for initial isotopic compositions of U, Pu, Am and Cm (wt%) [7]

<table>
<thead>
<tr>
<th>U</th>
<th>Composition</th>
<th>Pu</th>
<th>Composition</th>
<th>Am</th>
<th>Composition</th>
<th>Cm</th>
<th>Composition</th>
</tr>
</thead>
<tbody>
<tr>
<td>234U</td>
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<td>238Pu</td>
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<td>241Am</td>
<td>40.92</td>
<td>244Cm</td>
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<td>0.29</td>
<td>245Cm</td>
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<td>236U</td>
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<td>240Pu</td>
<td>4.620</td>
<td>243Am</td>
<td>58.79</td>
<td>246Cm</td>
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<tr>
<td>238U</td>
<td>99.597</td>
<td>241Pu</td>
<td>0.084</td>
<td></td>
<td></td>
<td>247Cm</td>
<td>0.29</td>
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<td></td>
<td></td>
<td>242Pu</td>
<td>0.007</td>
<td></td>
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</table>

Irradiation experiment

Three metal fuel pins, two of which include MA containing alloy segments of 100mm length in the U-19Pu-10Zr fuel stacks, were fabricated. The total length of the active fuel was 485mm. The cladding material was 15-15Ti austenitic steel [8-10] and the fuel-cladding gap was filled with sodium for thermal bonding. A schematic view of metal fuel pins No 1, No 2 and No 3, compared with that of the oxide driver fuel pin, is shown in Figure 1. These metal fuel pins were arranged in an irradiation capsule along with 16 oxide fuel driver pins for irradiation in the Phénix fast reactor as shown in Figure 2.

Three irradiation capsules with identical configurations were prepared to achieve different target burnups. The irradiation conditions were evaluated to predict the temperature conditions at the beginning and end of irradiation using the ALFUS code [11, 12].

Figure 1: Schematic view of metal fuel pins and oxide fuel driver pin used in irradiation experiment in Phénix [7]

Figure 2: Schematic view of irradiation capsule with Phénix geometry [7]
Non-destructive tests

**Cladding profilometry**

The cladding diameter of the irradiated fuel pins was measured using a laser shadowgraph from four directions: 0°, 45°, 90° and 135°. Figure 3 shows the mean values obtained from the measurement for the METAPHIX-1, -2 and -3 pins compared with the ALFUS predictions [7, 13].

The localised peaks on the spectra correspond to measurement fluctuations, whereas the two large peaks observed near and below the bottom end of the fuel stacks correspond to the positions of the orbital welds sealing the plugs.

METAPHIX-1 exhibits a slight deformation of <0.1% in part of the fuel stack. For the METAPHIX-2 and -3 pins, the maximum deformation of the cladding reached 0.3% and 0.9%, respectively [13]. No significant differences were found among the fuel pins with or without added MAs or REs for the same burnup. These results are within the range of the deformation predicted by the ALFUS code [13]. The irradiation properties of the cladding material, 15-15Ti steel, were taken from the literature [8-10], and other characteristics were assumed to be those of a typical Type 316 or D9 austenitic steel [14, 15].

**Gamma-ray spectrometry**

The measurement results for the axial distribution of the relative gamma-ray intensity emitted from the $^{137}$Cs and $^{106}$Ru in metal fuel pin No 3 are shown in Figure 4 for METAPHIX-1, -2 and -3. In these fuel pins, the U-19Pu-10Zr-5MA and U-19Pu-10Zr-5MA-5RE alloys were loaded in zone B as shown in Figure 1. The gamma-ray intensity from $^{106}$Ru, which has low mobility in the fuel alloy, shows local depressions at intervals of approximately 20 to 50mm. These depressions indicate the boundaries between fuel rodlets. From the boundary positions, fuel zones A, B and C after irradiation could be identified. Table 3 summarises the axial elongations of fuel zones A, B and C as well as those of the total fuel stack of pins No 1, No 2 and No 3 [7, 16]. In this table, ALFUS calculation results are also given for comparison. The axial elongation of the fuel alloy is larger in the upper zone than in the lower zone; it reaches 13.0-16.0% in zone A of the METAPHIX-3 high-burnup fuel pins, but is 1.0-2.4% in zone C. This axial distribution of the fuel elongation can be qualitatively predicted by ALFUS [7].

The differences in the axial elongations in zone B among pin No 1 (U-19Pu-10Zr), pin No 2 (U-19Pu-10Zr-2MA-2RE) and pin No 3 (U-19Pu-10Zr-5MA/U-19Pu-10Zr-5MA-5RE) were 1.0% in METAPHIX-1 and -2 and 3.0% in METAPHIX-3, which were not significant compared with the fluctuation of 2.0-3.0% in the elongation of zone A, in which the U-19Pu-10Zr alloy was loaded, in all pins. These results suggest that the irradiation swelling behaviour of metal fuels is hardly affected by the addition of MAs and REs of 5wt% or less.

Cs is an alkali metal with relatively high mobility that is easily dissolved into Na; therefore, $^{137}$Cs acts as a tracer of bonding Na relocation, particularly its migration into the plenum in the case that the irradiated metal fuel swells and the fuel-cladding gap closes. The ALFUS calculation predicts that up to ~80% of the total bonding Na has already migrated into the plenum at 2-3at.% burnup; the remaining bonding Na infiltrates into the open pores of the irradiated fuel [17].
**Figure 3:** Measurement results for cladding deformation of METAPHIX-1, -2 and -3 pins [7]

(a) METAPHIX-1

(b) METAPHIX-2

(c) METAPHIX-3

**Figure 4:** Axial distributions of gamma-ray intensity from pin No 3 (U-19Pu-10Zr-5MA/-5MA-SRE) [7]

(a) METAPHIX-1

(b) METAPHIX-2

(c) METAPHIX-3

**Destructive post-irradiation examinations**

**Plenum gas analysis**

Plenum gas analysis revealed that 45.7-51.3% and 63.1-68.2% of the fission gases generated in the METAPHIX-1 and -2 fuels were respectively released to the plenum from the fuel alloys; the total amount of fission gas generated by each fuel was calculated using the ORIGEN-2 code. At the same burnup, no significant difference was found in the fission gas release behaviour, regardless of MA.
and RE addition. Furthermore, these results are consistent with reported data for U-Pu-Zr test fuels irradiated in EBR-II \cite{18} as shown in Figure 5.

\textbf{Figure 5:} Burnup dependence of fission gas release fraction \cite{5}

![Figure 5: Burnup dependence of fission gas release fraction](image)

\textbf{Metallography}

Cross-sectional metallography of the METAPHIX-1 and -2 fuels showed several distinctive deposits in the MA- and RE-containing alloys. These were inferred to be MA/RE-rich precipitates. In particular, in the high-temperature region, the size of the MA/RE-rich precipitates increased to > 100μm diameter during irradiation as shown in Figure 6.

\textbf{Figure 6:} Optical metallographic images of U-19Pu-10Zr-5MA-5RE alloy \cite{7}

![Figure 6: Optical metallographic images of U-19Pu-10Zr-5MA-5RE alloy](image)

In addition, the concentric phase structure of the fuel matrix was found to depend on the irradiation temperature as shown in Figure 7. From the metallographic image of each fuel sample, the irradiation temperature at the fuel centre was estimated and is summarised in Table 4 along with the ALFUS predictions. The fuel temperature predicted by ALFUS is consistent with the matrix phase structure of the irradiated metal fuels with or without the addition of MAs and REs.
Figure 7: Cross-sectional overview and fuel morphology distribution from centre to periphery for METAPHIX-1 fuel sample [5, 7].

Electron probe microanalysis

Electron probe microanalysis (EPMA) was performed on a METAPHIX-2 sample of U-19Pu-10Zr alloy irradiated at ≲ 650°C [19-23]. X-ray element distribution maps for the fuel constituents (U, Pu, Zr) and selected fission products (e.g. Nd, Mo, La, Ru, Xe, Cs) were obtained. Figure 8 shows the qualitative redistribution of U, Pu and Zr in the irradiated fuel and includes a table listing the main phases and features identified. The irradiation temperature deduced from the constituent redistribution profiles corresponds well with the ALFUS prediction.

From the mid-radius to the periphery of the fuel, the microstructure is very porous. U and Pu exhibit symmetric radial profiles and the Pu concentration decreases close to the periphery. The Zr content in the fuel matrix is close to its nominal as-fabricated value. The "missing" Zr from the central region is located in segregated phases, which are present throughout the fuel.

Figure 9 shows X-ray maps of the fuel components, the cladding constituents and representative fission products of noble metals and REs in the fuel-cladding interface region. The cladding material employed in this irradiation experiment, 15-15Ti steel, contains 14-16wt% Ni and Cr. The maps in Figure 9 reveal several features of interest in this region. A schematic representation of the different defined zones is shown in Figure 10.

In addition to the main components and fission products, the fuel phase contains Fe and Ni originating from the cladding. The diffusion of other cladding elements, such as Cr and Ti, was not
observed. Discrete precipitate particles (Zr-Ni phase) enriched with Ni (6.7 wt%) and containing Fe are visible on the fuel side.

Pu depletion (the "Pu-free" area in Figure 10) was measured near the outer surface of the fuel. This fuel region is not homogeneous and it includes discrete precipitates of Zr-Ni and Zr-rich phases.

The outermost fuel region is characterised by the presence of a Zr-rich phase forming a layer of 5 μm thickness. This phase mainly consists of Zr (33-47 wt%), Fe (18 wt%), Cr (6 wt%) and U and Pu (15 wt% and 3 wt%, respectively). Small amounts of REs (less than 4 wt%) are also included.

The Pu-rich phase at the inner surface of the cladding consists of a layer of μm-order thickness. Its small thickness made the layer difficult to analyse; thus, a limited number of analysis points were available. The main constituents are Pu, measured to be up to 57 wt%, and Fe (29 wt%). In the cladding, the presence of Pu (0.7 wt%) and Rh (0.1 wt%) was observed up to a depth ~20 μm from the initial interface of the fuel cladding. The presence of Pu is probably due to the depletion of Pu, which was detected in the outermost radial region of the fuel.

A Cr-rich (23 wt%), Ni-depleted layer of 5-8 μm width also formed on the cladding side. In addition to Cr, REs (Ce, La, Nd) were also present.

Figure 8: Redistribution of constituents in METAPHIX-2 fuel obtained by EPMA, and estimated fuel phases [22, 23]
Chemical analysis

The irradiated MA-bearing fuel samples were cut into discs with ~4 mm thickness from the METAPHIX-1 and -2 fuel pins, and individually dissolved with the cladding in nitric-hydrofluoric acid. The fuel materials including Zr and all the FPs were completely dissolved over several hours. Then, the isotopic compositions of these dissolved samples were analysed by inductively coupled plasma-mass spectrometry (ICP-MS).

MA transmutation performance

On the basis of the results of chemical analysis, the changes in the actinide composition after fuel fabrication were evaluated and compared with the results of multigroup three-dimensional diffusion and burnup calculations. Table 5 summarises the obtained MA transmutation ratio of the U-19Pu-
10Zr-5MA alloy irradiated up to ~5.7at.% burnup in the METAPHIX-2 experiment. The MA transmutation ratio was defined as the relative depletion of the elements or isotopes during the irradiation and evaluated using the following equation:

$$\text{Transmutation ratio} = \frac{(\text{Quantity before irradiation}) - (\text{Quantity after irradiation})}{(\text{Quantity before irradiation})} \times 100\%.$$  

A negative transmutation ratio means that neutron irradiation results in a net increase in the element or nuclide due to the additional production by the neutron capture of parent nuclides. There was a decrease of about 20% in the total quantity of MA elements in the U-Pu-Zr-5MA alloy during the METAPHIX-2 irradiation experiment. The chemical analysis results for the $^{237}$Np and $^{241}$Am transmutation performance are in reasonable agreement with the previously reported results for the SUPERFACT experiment [24].

As shown in Table 5, the transmutation ratios of Np, Am and Cm elements were predicted by core burnup calculations with C/E ratios of 0.94-1.00. However, these C/E ratios contain uncertainties of ~21%-24%, which are due to the variations in the initial fuel composition as well as the uncertainty in the chemical analysis. By improving the homogeneity of the as-fabricated fuel alloys and by reducing the uncertainty in the chemical analysis of the irradiated fuel samples, the MA transmutation ratio is expected to be predicted more precisely.

Summary

METAPHIX, a collaborative study between CRIEPI and JRC Karlsruhe, has been carried out to determine the irradiation performance of MA-bearing metal fuels and demonstrate MA transmutation in fast reactors. In this study, metal fuel pins that included U-Pu-Zr-MA alloys were irradiated up to peak burnups of ~2.5 at.%, ~7 at.% and ~10 at.% in the Phénix fast reactor.

NDTs were performed on all the irradiated METAPHIX fuel pins and no significant differences were found in the results obtained with or without MA addition. Then, destructive examinations were performed only on the low- and medium-burnup fuel pins.

Table 5: MA transmutation ratio of U-19Pu-10Zr-5MA alloy irradiated up to ~5.7at.% burnup [25]

<table>
<thead>
<tr>
<th>Element</th>
<th>Isotope</th>
<th>Transmutation ratio [%]</th>
<th>Chemical analysis (E)</th>
<th>Calculation (C)</th>
<th>C/E</th>
</tr>
</thead>
<tbody>
<tr>
<td>Np</td>
<td>$^{237}$Np</td>
<td>23.3 ± 4.8</td>
<td>23.3 ± 0.2</td>
<td>23.3 ± 0.2</td>
<td>1.00 ± 0.21</td>
</tr>
<tr>
<td>Am</td>
<td>$^{241}$Am</td>
<td>23.5 ± 5.4</td>
<td>22.1 ± 0.8</td>
<td>24.8 ± 0.9</td>
<td>0.94 ± 0.22</td>
</tr>
<tr>
<td></td>
<td>$^{242m}$Am</td>
<td>-475.5 ± 40.4</td>
<td>-47.5 ± 40.4</td>
<td>-328.9 ± 11.8</td>
<td>0.94 ± 0.22</td>
</tr>
<tr>
<td></td>
<td>$^{243}$Am</td>
<td>23.7 ± 5.4</td>
<td>21.9 ± 0.8</td>
<td>21.9 ± 0.8</td>
<td>0.94 ± 0.22</td>
</tr>
<tr>
<td>Cm</td>
<td>$^{244}$Cm</td>
<td>-47.2 ± 11.4</td>
<td>-45.1 ± 2.8</td>
<td>-50.8 ± 3.2</td>
<td>0.96 ± 0.24</td>
</tr>
<tr>
<td></td>
<td>$^{245}$Cm</td>
<td>-52.3 ± 11.6</td>
<td>-50.8 ± 3.2</td>
<td>-50.8 ± 3.2</td>
<td>0.96 ± 0.24</td>
</tr>
<tr>
<td></td>
<td>$^{246}$Cm</td>
<td>-47.5 ± 13.0</td>
<td>-36.9 ± 2.3</td>
<td>-36.9 ± 2.3</td>
<td>0.96 ± 0.24</td>
</tr>
<tr>
<td></td>
<td>$^{247}$Cm</td>
<td>-13.5 ± 7.6</td>
<td>1.0 ± 0.1</td>
<td>1.0 ± 0.1</td>
<td>0.96 ± 0.24</td>
</tr>
<tr>
<td></td>
<td>$^{248}$Cm</td>
<td>-82.3 ± 16.1</td>
<td>-59.8 ± 3.7</td>
<td>-59.8 ± 3.7</td>
<td>0.96 ± 0.24</td>
</tr>
<tr>
<td>Total MAs</td>
<td></td>
<td>19.8 ± 5.1</td>
<td>19.4 ± 0.2</td>
<td>19.4 ± 0.2</td>
<td>0.98 ± 0.25</td>
</tr>
</tbody>
</table>

Plenum gas analysis revealed that the fission gas release behaviour was essentially the same as that of conventional U-Pu-Zr fuel pins up to ~7at.% peak burnup regardless of MA addition.
The morphology, composition and distribution of the fuel matrix and secondary phases were characterised by optical and scanning electron microscopy and by EPMA. The distribution of the matrix phases in the irradiated fuel containing MAs and REs is essentially similar to that observed in the basic ternary alloy fuel. In the high-temperature phase, some large precipitates considered to be inclusions of MAs and REs were observed. Second phases were also present in the low-temperature region at the radial periphery of the fuel. Interactions between the fuel and cladding occurred to a limited extent and were characterised. The first EPMA analysis was performed on a sample of the U-19Pu-10Zr fuel with ~7 at.% burnup, providing further insight into the complex configuration of the irradiated ternary alloy fuel. The redistribution behaviour of the fuel constituents (U, Pu, Zr) was consistent with the literature, and many secondary phases were present with a variety of compositions.

Furthermore, the chemical analysis results for the changes in the actinide composition during the irradiation experiment for up to ~5.7 at.% burnup demonstrated that MAs are transmuted as expected in U-Pu-Zr-MA(−RE) alloys in a fast reactor.

The results obtained so far confirm that MA-bearing metal fuels are a promising option for the recycling of MAs. Ongoing and planned investigations are expected to extend the range of compositions and burnup levels for the practical and effective use of U-Pu-Zr fuel containing MAs.

Acknowledgements

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References


Fabrication of (U,Ce)O$_2$ and (U,Am)O$_2$ pellets with controlled porosity from oxides microspheres

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Abstract

$U_{1-x}Am_xO_{2+\delta}$ mixed oxides are considered as promising compounds for americium heterogeneous transmutation in Sodium Fast Neutron Reactor [1–3]. Comparison between dense (95% of the theoretical density (TD)) and porous (88% TD) oxide microstructure in terms of behaviour during irradiation is currently under study through experimental irradiation test [3,4]. Porous microstructure is envisaged in order to facilitate helium and fission gas release and to reduce pellet swelling during irradiation and under self-irradiation.

In this study, we focus on determining the experimental parameters necessary to the fabrication of porous ceramic pellets with controlled porosity (8-10% open porosity) from mixed-oxide microspheres obtained by the weak acid resin (WAR) process [5]. More precisely, uranium-cerium mixed-oxide microspheres were first synthesised by co-loading uranyl and cerium cations onto carboxylic resin beads, Ce being used as a surrogate of Am. Calcination under air of these metal loaded spheres led to the mineralisation of the organic matter and to the formation of (U,Ce)O$_2$ microspheres. The major part of this as-treated spherules was then submitted to a second thermal treatment under reductive atmosphere (Ar/4vol.%H$_2$) to form the (U,Ce)O$_2$ solid solution. The specific mix of (U,Ce)O$_2$ and (U,Ce)$_3$O$_8$ microspheres allowed to obtain, after pelletisation and reductive sintering, an optimised porous microstructure, as previously reported by E. Rémy et al. for simple uranium dioxide [6]. Indeed, the reduction of (U,Ce)$_3$O$_8$ into (U,Ce)O$_2$ during the sintering treatment is accompanied by the formation of open porosity, mainly due to a 3% decrease of the lattice volume and to an oxygen release in the form of H$_2$O. Porous pellets (D~88% TD), with an open porosity of about 8% were thus fabricated.

Parameters found for (U,Ce)O$_2$ were successfully applied to the fabrication of the highly active $(U_{0.90}Am_{0.10})O_2$ pellets. Finally, an $(U_{0.90}Am_{0.10})O_2$ pellet with controlled porosity of 89% TD was fabricated to demonstrate the scientific feasibility of this spherule metallurgy process.

Introduction

Even though neptunium, americium and curium, known as the minor actinides (MA) represent less than 0.1 wt.% of the spent nuclear fuel, they are the main responsible for long-term radioactivity and heat load of the ultimate waste after plutonium recycling [7,8]. A possible option to reduce the long-term radiotoxicity of spent fuel nuclear wastes is to deploy MA transmutation into lighter short-lived elements, for instance in fast neutron reactors (FNRs) developed in the framework of GEN-IV International Forum [1,9]. The programme specially focuses on the transmutation of
americium since this element is the most abundant and the most active among MA [2,10]. In France, among the different MA transmutation modes [2,11-13], heterogeneous transmutation is one of the most considered options for the future nuclear fuel cycle [1]. It consists in irradiating uranium-americium mixed-oxide compounds \((U_{1-x}Am_xO_{2±δ})\) located at the core periphery. Such irradiation targets are called americium-bearing blankets (AmBB) [3]. The content of americium \([Am/(Am+U)]\) in such materials can reach 15 at.%

In this context, several processes are being developed to fabricate such mixed-oxide pellets which are to meet the specifications required to perform analytical irradiations such as Am content, O/M ratio (oxygen to metal with M = U + Am), chemical and microstructural homogeneity, pellet dimensions and density, open porosity ratio, etc [2,10,14,15].

The development of a dustless innovative route, adapted from the weak acid resin process (WAR) is currently studied. It consists in impregnating resin microspheres with the desired element(s) in their cationic form and then in submitting the metal loaded resin to a calcination step to obtain the single or mixed-oxide microsphere precursors. Those microspheres are then shaped into pellets which are sintered to produce single or mixed-oxide ceramic. The overall process is the so-called Calcined Resin Microsphere Pelletising (CRMP) process [14].

The fabrication of dense pellets by CRMP process was demonstrated for compounds such as UO\(_2\) [16] and \((U,Am)O_2\) [17]. Tailored-porous samples from oxide spheres were fabricated only for UO\(_2\) [6]. As the main objective of MARIOS and DIAMINO irradiations was to assess the influence of the fuel microstructure (dense and porous), it could be important to also demonstrate the feasibility of \((U,Am)O_2\) pellets fabrication with controlled density from CRMP process for an eventual future irradiation. Indeed, the porous microstructure is considered to facilitate the release of helium produced during irradiation but also to limit pellet swelling under self-irradiation.

In this study, we focus on determining the experimental parameters necessary to the fabrication of porous ceramic pellets with controlled porosity (8-10% open porosity) from mixed-oxide microspheres obtained by the WAR process [5]. Firstly, \((U,Ce)O_2\) porous pellets (D~88% TD), with an open porosity of about 8% were fabricated. Secondly, experimental parameters found for \((U,Ce)O_2\) were applied to the fabrication of the highly active \((U_{0.90}Am_{0.10})O_2\) pellets. Finally, an \((U_{0.90}Am_{0.10})O_2\) pellet with controlled porosity (89%TD) and with an open porosity of 9% was fabricated.

**Fabrication of \((U,Ce)O_2\) pellets with controlled porosity**

**Resin preparation**

The ion exchange resin used for the fixation was an IMAC HP333 carboxylic resin supplied by Dow Chemicals (Chauny, France). The resin was manually sieved under deionised water and the 630-800 µm diameter size range was collected for the fixation. The resin was introduced in a column to be washed by successive percolations of 1 M nitric acid (HNO\(_3\) – Fisher Chemical, Certified ACS Plus), demineralised water, 1 M ammonia solution (Merck, Pro Analysis) and demineralised water again. Eventually, the washed resin was prepared in its protonated form by a final percolation of 1 M HNO\(_3\), followed by a demineralised water rinse cycle.

**Preparation of the loading solution (U/Ce)**

As the fixation of cerium on the resin is not congruent, a content of \([Ce/(U+Ce)] = 25\) at.% in the solution was selected in order to obtain 15 at.% Ce in the resin [16].

The loading solution was prepared by first dissolving 64 g of cerium (III) nitrate hexahydrate \((Ce(NO_3)_{3.6}H_2O – 99.99\%\) Sigma Aldrich) in a 0.347 M HNO\(_3\) solution (volume = 1.65L). Then uranium
tetroxide solid (UO₃) was added to this solution. The latter was an Acid Deficient Uranyle Nitrate solution (ADUN) [18]. After 12h stirring, the solution was filtered to remove UO₃ excess. The final concentration of uranium and cerium was 270 mmol and 89 mmol, respectively, which corresponds to a Ce/(Ce+U) ratio of 24.8%, and the pH value was 3.3.

**Impregnation**

Resin beads were introduced in a glass chromatography column (2.5 cm diameter and 30 cm height – Biorad). Fixation of both UO₂⁺ and Ce³⁺ cations was performed by percolating the ADUN solution by recirculation through the column during 6 h. The loaded resin was rinsed with demineralised water before being dried at 105°C overnight in an air oven. 50 g of dried loaded resin were prepared in one go.

In order to measure the amount of metal cations fixed by the resin, a sample of 150 mg of dried metal loaded microspheres was contacted with 5 mL of 1 M HNO₃ in a column for 1 h and then rinsed 4 times with same amount of acid. All fractions were collected in a 25 mL jauged flask and the obtained solution was finally analysed by inductively coupled plasma-atomic emission spectroscopy (ICP-AES). The cerium content versus total metal content [Ce/(Ce+U)] on the loaded resin microspheres was determined to be equal to 13.8 at.% which is closed to the targeted 15 at.%.

**Calcination – heat treatment**

Two thermal treatments were then applied to the loaded resin in an instrumented tubular furnace.

The resin was first calcined at 700°C for 4h under air (flow 600 L.h⁻¹) with a low heating rate of 1.5K.min⁻¹ preventing any microsphere degradation and guaranteeing a proper homogeneity of the calcination temperature. A part of the latter was then submitted to a second heat treatment under reductive atmosphere Ar/4vol.%H₂ (flow 600 L.h⁻¹) at 700°C for 5h with a heating rate of 10 K.min⁻¹.

In terms of mass loss, total weight loss of 56.2% and 4.2% was observed during the first and second thermal cycle, respectively. Those values are consistent with values generally encountered for uranium loaded resin bead [6,14].

**Figure 1:** Microspheres a) before calcination under air (total weight~50g) b) after calcination under air 700°C-4h (total weight~22g)
Oxide microsphere characterisation

Crystalline structure analyses on the obtained microspheres were performed by X-ray diffraction (XRD) using a Bruker D8 Advance diffractometer (Madison, WI) with a Cu X-ray source (Kα1/α2, λ = 1.54059/1.54439 Å) and equipped with a special sample holder for radioactive material measurements. The microspheres were first milled manually and then mixed in grease with Au powder (Sigma Aldrich, >99.9%). Gold is used as standard to control any potential instrumental deviation.

The XRD patterns obtained from the powdered oxide microsphere after calcination under air and after reduction in Ar/4vol.%H2 are presented in Figure 2 in red (bottom) and blue (top), respectively.

Figure 2: XRD patterns for microspheres after the first calcination (air, bottom) and after second heat treatment (Ar/4%H2, top) (*gold standard)

After the calcination under air, the XRD pattern is characteristic of a single hexagonal phase (P-62m) with the following refined parameters, a=6.811 (1) Å and c=4.161 (1) Å.

After the reduction step under Ar/4vol.%H2, only peaks derived of a single fluorite-type phase (Fm-3m) are visible. The refined lattice parameter was found equal to 5.459 (1) Å. No trace of the (U,Ce)3O8 hexagonal phase is revealed which proves the total reduction of (U,Ce)3O8 to (U,Ce)O2.

A nuclearised Zeiss Supra 55 VP field emission gun (FEG) scanning electron microscopy (SEM – Oberkochen, Germany) was used for the morphology and the microstructure observations of the oxide microspheres. SEM observation (Figure 3) revealed that the spheres have a porous microstructure after calcination under air as well as after the second heat treatment under Ar/4vol.%H2. Energy dispersive spectroscopy revealed homogeneity of U and Ce distributions between microspheres.

Fabrication and characterisation of U-Ce mixed-oxide pellets with controlled porosity

To fabricate porous pellets, reduced microspheres (U,Ce)O2 were mixed with oxidised microspheres (U,Ce)3O8 (Figure 4). Different contents of oxidised spheres (30, 35 and 40 wt.%) were used in order to obtain samples with a density in the 86-90% TD range, with a homogeneous open porosity network. After the pelletisation step in a three-part matrix (5.0 mm diameter) , the green pellets (about 500 mg each) were sintered under reductive atmosphere at 1 850°C-5min in a horizontal dilatometer or at 1 700°C-4h in a high temperature furnace.
XRD diffractograms obtained on the sintered pellets are presented in Figure 5. These patterns are characteristic of the fluorite-type phase, with a refined parameter of 5.462(1), 5.463(1) and 5.464(1) Å, for a \((U,Ce)_3O_8\) content of 30, 35 and 40wt.%, respectively. The sintering step under Ar/4vol.%H\(_2\) ensured the total reduction of \((U,Ce)_3O_8\) to \((U,Ce)O_2\), with a volume lattice decrease from an hexagonal to a fluorite phase.

**Figure 3**: FEG-SEM micrographs in secondary electron mode of oxide microspheres:
(a) \((U,Ce)_3O_8\) and (b) \((U,Ce)O_2\)

**Figure 4**: Porous pellets fabrication flowchart using oxide microsphere precursors
Sintered pellets were cut longitudinally then mirror-polished before being observed with an optical video microscope (Olympus BX 30). Figure 6 presents the microstructure of pellets sintered at 1850°C-5min, for 30, 35 and 40 wt.% of (U,Ce)₃O₈. The observations clearly exhibit the porous microstructure of these samples which have a density between 86 and 89% TD (the final density of each sintered sample was obtained via the Archimedes method using de-ionised water as the immersion medium). The percolating porosity network was created during the reductive sintering by volume shrinkage of (U,Ce)₃O₈ phase in (U,Ce)O₂. The results for 30 and 35 wt.% are quite similar; the open porosity is about 8%. A significant difference can be observed for the 40 wt.% sample where the open porosity reaches 10%. This high ratio of porosity induced a lower mechanical resistance and grains removal during polishing as observed. To insure good mechanical resistance while keeping a correct ratio of open porosity (≥ 8%), a content of 30-35 wt.% will be selected for the next fabrications, and in particular for the fabrication of porous (U,Am)O₂.

Figure 5: XRD patterns of sintered pellets (1850°C-5min) for different (U,Ce)₃O₈/(U,Ce)O₂ ratios (*gold standard)

Figure 6: Optical microscopic observations of the internal microstructure of sintered pellets

<table>
<thead>
<tr>
<th>Composition</th>
<th>Density (D&lt;sub&gt;rel&lt;/sub&gt;)</th>
</tr>
</thead>
<tbody>
<tr>
<td>30% (U,Ce)₂O₈</td>
<td>~88%</td>
</tr>
<tr>
<td>35% (U,Ce)₂O₈</td>
<td>~89%</td>
</tr>
<tr>
<td>40% (U,Ce)₂O₈</td>
<td>~86%</td>
</tr>
</tbody>
</table>
Fabrication of \((U,\text{Am})_2\text{O}_2\) pellet with controlled porosity

- **Uranium-amercurium mixed-oxide microsphere synthesis**

The microspheres used for the fabrication of the pellet were synthesised according to previously published procedure [14]. The resin preparation was the same than described in first section for U/Ce (sieving of resin bead in the 630-800 µm size range, extensive washing cycle, resin prepared in its protonated form).

The resin microspheres were impregnated by an acid deficient nitrate uranyl solution (ADUN) mixed to amercurium (III) nitrate (with a concentration of amercurium \([\text{Am}/(\text{Am}+\text{U})]= 10.1 \pm 0.2\) at.%) during 5h by recirculation through a plexiglass column. After drying, the microspheres were first calcined in air at 700°C for 4h. A part of these oxide microspheres was secondly reduced in Ar/4vol.%H\(_2\) at 700°C for 5h (same heat treatment parameters than \((U,\text{Ce})_2\text{O}_2\)).

**Figure 7**: Pictures of U-Am oxide microspheres synthesis in hot cell in ATALANTE facility. From left to right: loaded resin in plexiglass column, loaded resin in the quartz crucible before calcination in airflow and oxidised \((U,\text{Am})_2\text{O}_3\) microspheres

Characterisation of U-Am mixed-oxide microspheres

A content of 11.1 ±0.2 at.% Am measured by TIMS analysis by dissolving 20 mg of reduced oxide microspheres \((U,\text{Am})_2\text{O}_2\) in nitric acid (5 M), which can demonstrate the quasi-congruence of the Am fixation (~10 at.% in the initial solution).

The XRD patterns obtained from the powdered oxide microsphere after calcination under air and followed by a reduction step under Ar/4vol.%H\(_2\) are presented in Figure 8 in red (bottom) and blue (top), respectively.

After the calcination under air, the XRD pattern is characteristic of a single hexagonal phase (P-62m) with the following refined parameters \(a=6.838\) (1) Å and \(c=4.165\) (1) Å.

After the reduction under Ar/4vol.%H\(_2\), only peaks derived of a single fluorite-type phase (Fm-3m) are visible which proves the quantitative reduction of \((U,\text{Am})_2\text{O}_3\) to \((U,\text{Am})_2\text{O}_2\). The refined lattice parameter was found equal to 5.467 (1) Å. This parameter is higher than the one previously obtained by E. Remy et al. [17] (5.4511 Å) with the same CRMP process, which indicates a higher reduction for our sample.
Fabrication and characterisation of U-Am mixed-oxide pellet with controlled porosity

The ultimate goal of this study was to demonstrate the fabrication of a porous AmBB pellet (86-90% TD) with an open porosity higher than 8% (expected specifications for AmBB porous pellets). The same process presented in Figure 4 was used for the fabrication of porous (U,Am)O$_2$.

A proportion of 65 wt.% of reduced microspheres (U,Am)O$_2$ were mixed with 35 wt.% of oxidised (U,Am)$_3$O$_8$ microspheres. This ratio of 65/35 was chosen in order to get the specified density (about 88% TD) while keeping a sufficient mechanical resistance of the sintered pellet. After a pelletisation step at 700 MPa, the green pellet was sintered at 1700°C-4h under reductive atmosphere (Ar/4vol.%H$_2$). After sintering, the final pellet (m=560 mg, h=4.4mm, diameter =4.1mm) is rectilinear and neither deformations nor particular defects were observed.

The final density was obtained via the Archimedes method in deionised water. It was found a density of 89% TD with 9% of open porosity and 2% of closed porosity. These characteristics strongly fit with the required porous pellet specifications for previous irradiation programmes such as MARIOS [19,21].

Figure 9 presents a longitudinal observation of the sintered (U,Am)O$_2$ pellet by optical microscopy. The porosity is homogeneously distributed through the pellet and the open porosity is interconnected on a long path. Memory form of ex-(U,Am)$_3$O$_8$ microspheres is rather hemispherical and could be compared with the memory form of ex-U$_3$O$_8$-AmO$_2$ agglomerates observed in MARIOS samples which had no specific form.

XRD pattern obtained on the sintered pellet is characteristic of the fluorite-type phase, with a refined parameter of 5.470(1) Å. The latter is very close to usual lattice parameter found for U$_{0.99}$Am$_{0.10}$O$_{2.15}$ sintered pellets fabricated by metallurgical process [19,20]. The sintering step provided the total reduction of (U,Am)$_3$O$_8$ to (U,Am)O$_2$, with a volume lattice decrease for an hexagonal to a fluorite phase creating the visible open porosity network (Figure 9).
Conclusion

This work focused on the fabrication of mixed-oxide pellets with controlled porosity by implementing the CRMP process. The synthesis of mixed-oxide microspheres by impregnation and calcination of loaded resin beads really limits the production of fine particles. Moreover, the CRMP process is a simplified one, with only two steps after the oxide microsphere synthesis: pelletising and sintering. The feasibility of porous AmBB fabrication was demonstrated for a potential heterogeneous transmutation of americium in FNRs by the fabrication of an 89% DT porous pellet with 9% of open porosity. The developed process allows a homogeneous distribution of the open porosity through the entire pellet created by the complete reduction of hexagonal (U,Am)\(_3\)O\(_8\) into fluorite-type phase (U,Am)\(_2\).

This porosity network could be tuned by modifying the initial resin bead diameter or by doing a specific mix between several microsphere diameters.

Acknowledgements

The authors are thankful to P. Grangaud, J.M. Pomarède, Y. Sinot for uranium-ameriicum mixed-oxide microsphere synthesis, N. Bousquet, A. Achabouni for the calcination of (U,Ce)O\(_2\) spherules, E. Pascal for optical microscopy observation, J.R. Sevilla for SEM observation, R. Vauchy and A. Joly for XRD on sintered (U,Am)O\(_2\). The authors also acknowledge the CEA PACFA programme for financial support.

References


Plenary Session V: Waste Management for Partitioning and Transmutation

*Chair: T. Matsumura*
Partitioning and transmutation and the prevention of underground criticality

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Abstract
Partitioning and transmutation (P&T) broadens the appeal of geological disposal in terms of proliferation resistance, reduced heat emission, and radiological risk to the future population. In addition, it enables reduction in the total inventory of fissile nuclides in high-level waste (HLW) packages generated by partitioning, which can eliminate the potential for critical masses to accumulate in the near- and far-field of the geological disposal system over time. The direct geological disposal of spent nuclear fuel, on the other hand, contributes a large source term for radionuclide transport upon waste package failure, which may lead to underground criticality and thus additional dose in the biosphere. This paper compares the transport results between compromised canisters for used nuclear fuel (UNF) and HLW packages from the end-products of P&T. The used fuel inventory is based on a PWR at 45 GWd/MTU burnup and 4.5% initial enrichment and 1050 years of total interim cooling. The P&T process employing this UNF is based on an accelerator-driven system (ADS) employing a stream of transuranics from UREX-based reprocessing. A simple dual porosity model is employed for a fractured crystalline granitic rock matrix using transport parameters that conservatively enhance the mobility of the nuclides of interest. Nuclides are assumed to be released congruently from the HLW due to reduced inventories in it whereas actinides in the used fuel undergo a solubility-limited release after a common 1000-year failure period. Arbitrary locations in the far-field were chosen for the superimposed precipitation of nuclides from multiple waste packages comprising a 32 000 MTU source term from PWR operation. Total accumulation was compared to past criticality analyses involving similar magnitudes of heavy metal deposition in geological media. The models established in the present study and the numerical exploration by those models indicate that if mass reduction from P&T is substantial, the possibility of accumulating a critical mass can be completely excluded. This bears implications on the effectiveness of P&T in the context of improving the long-term criticality safety of geological disposal.

Introduction
A permanent geological repository for used nuclear fuel (UNF) is the final component of the open nuclear fuel cycle. Upon the inevitable and extensive failure of waste canisters underground, and the subsequent transport of radionuclides in the host rock, scenarios have been suggested involving the accumulation of a critical mass in the far-field. If such a critical mass is autocatalytic, sustained chain reactions can release a large amount of energy, causing catastrophic failure to the host rock and a dose of radiation to the biosphere. The total amount of UNF in interim storage numbers in the tens of thousands of metric tonnes in the United States,[1] which presents a large source term for criticality in a performance safety assessment. Partitioning and transmutation (P&T) is suggested as a
measure to address the large inventory of UNF with regard to such a scenario. Among the many benefits of utilising P&T in the nuclear fuel cycle is the reduction in the inventory of actinides. This is hypothesised to reduce the risk (or possibly exclude) the possibility of accumulating a critical mass by removing fissile isotopes and their precursors from the waste canisters. If a critical mass is plausible for scenarios involving direct disposal, it may be the case that end-products from P&T will not be susceptible to criticality under any circumstance. Given the public perceptions towards geological disposal in the United States, utilising this technology may enhance the public image of waste disposal and bring about a viable long-term solution to final high-level waste (HLW) disposition.

The objective of this paper is to demonstrate the reduction in risk of attaining a critical mass by comparing nuclide transport results for direct disposal (from pressurised water reactors) and the end-products of P&T. In meeting this end, nuclide transport calculations will be conducted for a virtualised granitic repository arranged to accommodate a set source term for either disposal option. The analysis will be based on a framework of assumptions meant to purposely overestimate the amount of precipitate that could form in the far-field. Results will be scrutinised based on the total accumulated mass in the far-field and the overall composition of fissile material, which determines the relevance to criticality in a fractured, saturated system.

Methodology

The model consists of a finite array of waste forms, each surrounded by a porous bentonite buffer, emplaced in a fully-saturated granitic host rock intersected by planar fractures (qualitatively diagrammed in Figure 1). Each waste form is separated by a finite centre-to-centre pitch distance, and groundwater flows through the repository in one direction from a hydraulic gradient. The buffer is assumed to be saturated, and the canisters are assumed to fail concurrently, upon which the radionuclides contained within immediately begin dissolution according to their solubility. If the nuclide has a solubility-limited release, it is expected to precipitate on the surface of the waste form until the concentration plume allows for dissolution. The nuclides are then transported diffusively through the bentonite clay and then subsequently into the alluvial entrances of the fractures. Transport directly into the rock from the waste form is considered to be negligible. The proximity of each waste package is assumed to play no role in influencing the concentration plume; that is, the concentration of each plume is assumed to the highest possible with no solubility limitations proceeding from upstream to downstream in the array (as would be expected in reality). The parallel fractures have a uniform aperture and spacing and serve as the main conduit of advective mass transport, while rock matrix infiltration in the fractures is a diffusive transport mechanism. The fracture spacing is assumed to be large enough (relative to the fracture length) to impart no concentration gradient in between fractures.
For the model, the density, porosity, and tortuosity of the buffer and host rock must be known, and measured nuclide transport properties (sorption distribution and/or diffusion) are needed for each. The fractures are assumed to only contain solution with material imparting porosity, and there are no colloids that would enhance the transport of otherwise immobile species through attachment. Solubilities are also needed for each nuclide relevant to the particular buffer/groundwater chemistry. To maximise total precipitation, it is assumed that all transport pathways lead to a common point of precipitation in the far-field, which in this study is a set distance extending from the edge of the array. This is justified in that the point of accumulation does not have to be in the same geology as that of the host rock, and that geochemical conditions more antagonistic to dissolution are prevalent in that location.

The analysis required for the model is based on the classical advection-dispersion equation applied to species in a decay chain, where the transport phenomena on the waste form, in the buffer, in the rock, and through the fractures are all coupled. These coupled equations are utilised in a code titled Transport-to-Biosphere (TTB),[2] which was originally used to model vitrified high-level waste (VHLW). The assumptions further required for using this code will be explained later. It is first necessary to characterise the two different waste forms to be considered.

**Used nuclear fuel**

For direct disposal, depending on the reactor and its operational characteristics, a sizeable portion of fissile nuclides remain in the UNF that were not utilised for thermal energy. In the United States and Japan, commercial power reactors come in the form of light water reactors (LWRs), and a prevalent type is the pressurised water reactor (PWR). The reference used fuel type in this paper is taken to be that of a 17x17 PWR, which has the advantage of utilising a consistent initial enrichment throughout the whole active fuel length and allows for convenient, representative depletion simulations. The inventory of used PWR fuel is based on the 45 GWD/MTU burnup of uranium-oxide fuel enriched to 4.5 wt% with a specific power of 38 MW/MTU. ORIGEN is used to calculate the nuclide inventory after 50 years of interim cooling and 1000 years of emplacement before expected canister failure. [3]
Figure 2: Nuclides of PWR spent fuel at 45 GWd/MTU burnup and 4.5 wt% initial enrichment plotted by half-life and mass (normalised to 1 MTU of fuel) after 1 050 yr of cooling.

Nuclides in green were selected for transport analysis.

The elements of interest are those whose isotopes are fissile or give rise to fissile isotopes through radioactive decay. In particular, focus is on the uranium isotopes U-233 and U-235 and the plutonium isotopes Pu-239 and Pu-241; therefore, the transport analysis must follow at least all uranium and all plutonium isotopes in the canister for consistency. Although Pu-241 is short-lived, it is kept in consideration in order to keep track of all plutonium species and because it is a parent of the much longer-lived U-233. Therefore, the original, non-truncated selection of isotopes corresponded to the following decay chains, where the nuclides with high thermal neutron cross sections for fission are in bold:

1. 4n: Cf-252 → Cm-248 → Pu-244 → Pu-240 → U-236 → Th-232 →
2. 4n+1: Cf-249 → Cm-245 → Pu-241 → Am-241 → Np-237 → Pa-233 → U-233 → Th-229 →
3. 4n+2: Cm-246 → Pu-242 → U-238 → Th-234 → Pa-234 → U-234 → Th-230 →
6. (4n) Cm-244 → Pu-240 → U-236 → Th-232 →
7. (4n) Np-236 → U-236 → Th-232 →

The chains 5, 6, and 7 are branches of 4n+2, 4n, and 4n, respectively. In order to impart the proper level of complexity to the problem, so as to reduce computation time and numerical errors in the computer code, Figure 2 is used to identify both long-lived radionuclides and those prominent in the spent fuel. That is, engineering-informed decisions were made to exclude isotopes with miniscule quantities and half-lives shorter than the expected canister failure time of 1 000 years. Auxiliary decay chains generating uranium isotopes were also removed, with those precursors lumped into the daughter participating in the main series of decay chains. The thorium isotopes (originally included by virtue of Th-232 being fertile) were further removed since they are not expected to play an immediate role in achieving criticality. Therefore, the amended selection of species is as follows:

1. 4n: Pu-240 → U-236 →
Partitioning and transmutation

The goal of the P&T system is to reduce the long-term hazard of criticality by reducing the amount of uranium along with Np, Pu, Am, and Cm as the most prominent TRU elements (and uranium precursors) in the UNF in terms of mass. To meet this end, a number of mass-flow pathways are involved before a final waste form is produced, as shown in Figure 3. The UNF is assumed to be cooled for five years after discharge before being sent to a reprocessing facility, where all cladding materials are removed and the oxide fuel is dissolved. Fission products and 0.1% of all actinides are separated as HLW, while the majority of uranium is assumed to be extracted through UREX for use in the LWR fuel cycle. Fuel for transmutation is fabricated from the stream of readily-available TRU nuclides and zirconium (as a fuel matrix material [4]) from the reprocessing facility, which further serves as the make-up stream (D) after the partitioning process.

Figure 3: Mass-flow diagram for P&T system, based on Ahn et al 2006. [5]
The flows of zirconium from fuel cladding are not included

Transmutation is a process where neutron absorption reactions deplete the concentration of long-lived fission products and actinides from reprocessed UNF. The stream of TRU from fuel fabrication \((f)\) is burnt in a “transmuter,” which is usually either a high-burnup reactor or ADS. The latter employs a subcritical “burner” reactor coupled to the neutron spallation source downstream from a proton accelerator, and this paper will adopt characteristics of this particular implementation. The effective role of this component is modelled by employing a coefficient of destruction \((d)\), which describes the exponential rate at which a nuclide is transmuted via a neutron flux (in conjunction with decay). While considered a constant, in reality, this quantity changes with time as neutron flux and cross sections vary with the nuclide composition in the fuel.

In this section, the flow of a TRU nuclide with initial concentration \(C_1^0\) is analysed along with its transmutation products \((C_P^0)\). For example, Np-237 would have the transmutation products of U-233 from decay, Np-238 from the \((n,\gamma)\) interaction, Np-236 from \((n,2n)\), and fission products from fast fission. Therefore, although UREX can remove the majority of uranium from the UNF, some will be present in the HLW as transmutation products. Furthermore, Np-237 is also the transmutation product of another TRU nuclide Am-241, which is an example of how the analysis can be more
complicated. This coupling necessitates another simplification of the decay chains mentioned in the previous section in order to use a simpler first-member mass-flow analysis.

Among the TRU, only the fissile nuclides Pu-239, Pu-241, Am-242m, and Cm-245 would be of importance for criticality, although Np-237 and Am-243 maintain importance in giving rise to fissile isotopes through decay. Table 1 shows the TRU nuclides appreciable for the mass-flow analysis, where the relations between nuclides appear interweaved. While certain TRU precursors can be lumped due to small quantities, it is clear that the 4n+1 and 4n+3 chains involve significant multimember contributions. Therefore, a major assumption will be made that the fission transmutations dominate over capture and decay and all waste will consist of fission products and leftover TRU. The half-lives (14 to 10^6 yr) relative to the time needed for P&T cycling (years) adds some reasonability to this simplification. Also, by these assumptions, all uranium in the HLW will be from the 0.1 wt% of actinides leftover in the reprocessing waste stream.

Table 1: TRU nuclides in UNF after 5 years of cooling along with transmutation products, with fissile nuclides in bold

<table>
<thead>
<tr>
<th>Chain</th>
<th>TRU</th>
<th>Mass [g/MTU]</th>
<th>t_{1/2} [yr]</th>
<th>Transmutation Products</th>
</tr>
</thead>
<tbody>
<tr>
<td>Other (4n)</td>
<td>Cm-244</td>
<td>4.86E+01</td>
<td>1.81E+01</td>
<td>Pu-240 SF</td>
</tr>
<tr>
<td>Other (4n)</td>
<td>Np-236</td>
<td>3.19E-04</td>
<td>1.55E+05</td>
<td>U-236 Pu-236 Pa-232 FP</td>
</tr>
<tr>
<td>4n</td>
<td>Pu-244</td>
<td>2.32E-02</td>
<td>8.00E-07</td>
<td>U-240 SF</td>
</tr>
<tr>
<td>4n+1</td>
<td>Pu-240</td>
<td>2.54E+03</td>
<td>6.56E-03</td>
<td>U-236 Pu-241 FF SF</td>
</tr>
<tr>
<td>4n+1</td>
<td>Pu-241</td>
<td>1.32E+03</td>
<td>1.43E-01</td>
<td>Am-241 Pu-242 FP</td>
</tr>
<tr>
<td>4n+1</td>
<td>Am-241</td>
<td>4.11E+02</td>
<td>4.33E+02</td>
<td>Np-237 Am-242 Am-242m FF SF</td>
</tr>
<tr>
<td>4n+2</td>
<td>Np-237</td>
<td>6.56E+02</td>
<td>2.14E+06</td>
<td>U-233 Np-236 Np-238 FF</td>
</tr>
<tr>
<td>4n+3</td>
<td>Cm-246</td>
<td>2.45E-01</td>
<td>4.76E+03</td>
<td>Pu-242 SF</td>
</tr>
<tr>
<td>4n+3</td>
<td>Pu-242</td>
<td>6.71E+02</td>
<td>3.74E+05</td>
<td>U-238 Pu-243 SF</td>
</tr>
<tr>
<td>4n+3</td>
<td>Pu-243</td>
<td>1.71E+02</td>
<td>7.39E+03</td>
<td>U-239 Pu-243 Am-243 SF</td>
</tr>
<tr>
<td>4n+3</td>
<td>Pu-239</td>
<td>6.24E+03</td>
<td>2.41E-04</td>
<td>U-235 Pu-240 FP</td>
</tr>
<tr>
<td>Other (4n+2)</td>
<td>Am-242m</td>
<td>1.14E+00</td>
<td>1.41E+02</td>
<td>Am-242m Cm-242 Pu-242 Am-243 FP</td>
</tr>
<tr>
<td>Other (4n+2)</td>
<td>Pu-238</td>
<td>2.55E+02</td>
<td>8.77E+01</td>
<td>U-234 Pu-239 FF</td>
</tr>
</tbody>
</table>

SF: spontaneous fission products, FP: thermal fission products, FF: fast fission products

After irradiation, a portion (f) of the fuel (C1 and Cp) is discharged to partitioning, which can be achieved through means such as TRUEX or pyrochemical processing. The discharge of TRU and products from transmutation as waste for final disposal occurs during this part of the system (D), where the transmutation products and a fraction (a) of the TRU is discarded as HLW. Using material from the makeup stream, the fraction of recovered TRU (1 – a) is recycled as new fuel for another cycle (i) in the transmuter. These interactions are shown in equations (2) and (3).

\[
C_0^{(i-1)} = C_0^{(i)} e^{-d} \quad \text{(1)}
\]

\[
C_0^{(0)} = (1 - f) C_0^i + f \quad \text{(2)}
\]

The assumed values for the P&T quantities are shown in Table 2. After non-dimensionalising the system based on the total concentration of all constituents, the initial concentration of TRU is set at 1 for normalisation. The initial concentration of products is set to zero, although this was previously shown to be unrealistic. The value of 1/3 for f is based on the accelerator-driven transmutation of waste (ATW) system.[6] For a, a value of 0.001 was chosen arbitrarily since PUREX and Pyroprocessing are known to recover more than 99% of TRU in UNF.[6][7] The most arbitrary choice of parameter was 0.05 for the destruction coefficient. Ahn et al 2006 used different values of d (0.05 and 1) for various cases.[5] Since the framework of conservatism would be to purposely overestimate...
the fraction of TRU per canister (and thus to use a lower $d$) the former value of 0.05 is borrowed to enhance the quantity of TRU exiting P&T to waste.

The one-member mass-flow analysis achieves steady state after about 6 cycles, and results are shown in Table 3. The use of a small destruction coefficient is evident in that 87% of TRU still exists in the P&T stream after successive irradiations. The waste reduction ratio, defined as the accumulate loss of TRU into waste up to a cycle $i$ relative to the total TRU fed into the system ($\Delta_i^f$) up to the same cycle, is defined at steady state in equation (3). Had $d=1$ been employed instead, 16% of TRU would remain in the stream after irradiation and $\rho_1^\infty$ would have been just 0.02%

Table 2: Parameters for mass-flow modeling in P&T

<table>
<thead>
<tr>
<th>Symbol</th>
<th>Quantity</th>
<th>Value</th>
<th>Ref</th>
</tr>
</thead>
<tbody>
<tr>
<td>$C_0^p$</td>
<td>Initial amount of transmutation product before first irradiation</td>
<td>0</td>
<td></td>
</tr>
<tr>
<td>$C_0^i$</td>
<td>Initial amount of TRU nuclide before first irradiation</td>
<td>1</td>
<td></td>
</tr>
<tr>
<td>$f$</td>
<td>Fraction of the fuel in the transmuter discharged for partitioning</td>
<td>1/3</td>
<td>[5]</td>
</tr>
<tr>
<td>$d$</td>
<td>Coefficient of destruction</td>
<td>1/20</td>
<td></td>
</tr>
<tr>
<td>$\alpha$</td>
<td>Fraction of TRU going to waste stream</td>
<td>1/1000</td>
<td></td>
</tr>
</tbody>
</table>

$$\rho_1^\infty = \lim_{i \to \infty} \rho_1^{(i)} = \lim_{i \to \infty} \sum_{j=1}^{\infty} \Delta_j^{(i)} = \frac{\alpha f \exp(-d)}{1 - (1 - \alpha f) \exp(-d)} \quad (3)$$

With the transmutation products and TRU lost to waste, along with the original actinides and HLW from reprocessing, 20 kg of high-level waste is generated per 1 MTU employed in the PWR, where about 1 kg is comprised of actinides (mostly uranium escaping the UREX process). Fission products in the P&T waste were assumed to proportionately contribute to the masses of pre-existing fission products from reprocessing, although the fast fission spectra from the transmuter knowingly discredit this procedure. Using all nuclides above 0.1g, ORIGEN was used to calculate the composition after 45 years of decay in interim storage followed by an additional 1,000 years until the same repository failure point as UNF. The cross sections specific to borosilicate glass were adhered to.

Table 3: Steady-state values for mass-flow model

<table>
<thead>
<tr>
<th>Symbol</th>
<th>Quantity as $i \to \infty$</th>
<th>Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>$C_1^{(i)}$</td>
<td>Fraction of TRU in transmuter core before irradiation cycle</td>
<td>0.911</td>
</tr>
<tr>
<td>$C_1^{(i-1)}$</td>
<td>Fraction of TRU in transmuter core after irradiation cycle</td>
<td>0.866</td>
</tr>
<tr>
<td>$C_1^{(i)}$</td>
<td>Fraction of transmutation product in transmuter core before irradiation</td>
<td>0.089</td>
</tr>
<tr>
<td>$C_1^{(i-1)}$</td>
<td>Fraction of transmutation product in transmuter core after irradiation</td>
<td>0.133</td>
</tr>
<tr>
<td>$\rho_1^{(i)}$</td>
<td>TRU reduction ratio</td>
<td>0.00646</td>
</tr>
</tbody>
</table>

Repository

In a performance safety assessment, the full extent of repository features is taken into account for developing the transport scenarios, including the rock properties, tunnel configurations, and waste form orientations. While transport parameters are at hand for the actinides of interest, there is no information on the repository layout. Therefore, certain assumptions are made to construct a representative array of waste forms, where quantities are detailed in Table 4. The source term is compared to the mass loadings of each waste type to construct a discrete square array, which is assumed in order to remove the arbitrariness of a rectangular or multi-tiered layout.

The exact observation point that would be chosen in a safety assessment is governed by the specific features of the site, such as changes to geological stratigraphy or nearby populations. Without a specific site at hand, a value has to be chosen to justifiably represent such a distance while keeping in mind that any area beyond the engineered barrier system can be considered the far-field.
In the present analysis, the distances of 10 and 100 metres away from the downstream edge of the repository array are chosen. That is, the array is placed on the xy plane and the observation points are located 0.1 and 1 km away from the repository edge on the y axis at a point y0. Here, the precipitation is calculated by a superposition of contribution by individual canisters in the repository. The transport length is determined by applying the distance formula \( \sqrt{(x_i)^2 + (y_i - y_0)^2} \) for each canister \( i \).

The spacing between the canisters in the array, or the “pitch”, is chosen primarily to allow for proper heat dissipation through the buffer and host rock. If heat is not transferred efficiently over time, alteration of the buffer material and thermal strain on the host rock can compromise the effectiveness of the repository as a natural barrier to solute transport. Using the known alteration temperature of the buffer and the extrapolated heat emission of the LWR used fuel, thermo-hydromechanical codes such as TOUGH2 can be used to ascertain whether a particular spacing is adequate over time (typically about few thousand years). While such an analysis was not performed in this study, a pitch value of 20 metres was held to be a representative figure.

For UNF, the PWR assembly configuration, along with the canister and buffer designs, were borrowed from the specifications set forth by the Japanese Atomic Energy Agency (JAEA) in their direct disposal studies.[8] Equation (4) was used to calculate the total heavy metal loading of each canister based on the original tonnage of uranium used in the reactor. For a 0.92 MTU/canister loading, a simple square array with 186 canisters to a side accommodates the assumed 32 000 MTU source term. To put this into perspective, this inventory could be generated by Japan’s 42 operational LWRs with 1 GWe capacity, 80% capacity factor, 45 GWd/MTU burnup and 33% thermal efficiency for a total of 45 years.

\[
\ell_{PWR} = \frac{2 \, \text{asm}}{\text{can}} \times 264 \, \frac{\text{rods}}{\text{asm}} \times 1.096 \times 10^4 \, \frac{\text{kg}}{\text{m}^3} \times 0.95 \times \pi \left( \frac{0.0082}{2} \right)^2 \times 3.6 \, \text{m} \times 10^{-3} \frac{\text{MT}}{\text{kg}} \times 238 \, \text{MTU} \times \frac{270 \, \text{MTU}}{270 \, \text{MTU}} = 0.92 \, \frac{\text{MTU}}{\text{can}} \tag{4}
\]

The HLW from the P&T system is assumed to be vitrified in borosilicate glass, wherein two glass logs are surrounded by a steel overpack in the repository. Nuclides are loaded into the waste glass in a manner that avoids in-canister criticality and maintains heat emission, plutonium concentration, waste volume fraction, and overall weight below regulatory limits while maximising waste loading. The loading constraints suggested by the Japanese Nuclear Cycle Development Institute are used to determine total waste loading into the waste glass,[9] some of which are shown in Table 4. No more than 0.25 wt% of the waste glass should be taken up by HLW, and with a 400 kg glass loading limit, this implies

\[
\ell_{new} = 0.1 \, \frac{\text{MT}}{\text{can}}.
\]

After reprocessing and P&T, there is 0.02 MT of HLW per 1 MTU used in the PWRs, implying that one VHLW canister can accommodate the waste from 4.87 MTU of original fuel. For the source term, this suggests 6 575 vitrified waste canisters. This can be roughly approximated by a square array with 81 canisters to a side.

**Nuclide transport**

The Transport-to-Biosphere (TTB) code is used to model the transport of radionuclides in decay chains through parallel planar fractures intersecting a rock matrix. Results were obtained using repeated applications of the code where inputs were prepared for each discrete waste form with their characteristic transport distances to the precipitation location. A script was made to read
precipitate data for each isotope in the output after each execution of the code, and when the last canister was calculated, all data was summed for each point in time and for each isotope by virtue of the common precipitation point. Non-atomic and non-positive molar amounts resulting from numerical error were removed from consideration.

It is important to note that TTB was programmed for the simulation of VHLW, whose dissolution behaviour is markedly different than UO2 fuel. For direct disposal, given the pre-eminence of uranium, only so much of the U species can enter the aqueous phase before falling out in a precipitate. The precipitate deposits on the surface of the fuel and then undergoes decay or reenters solution when the plume passes and the concentration gradient allows for it. However, the use of solubility-limited boundary conditions in the code results in unrealistic uranium isotopic abundances at the precipitate location. Therefore, congruent-release boundary conditions are employed in the calculation but with a leach period calculated from the solubility-limited calculation.

Table 4: Parameters for the canisters and repository layout

<table>
<thead>
<tr>
<th>Item</th>
<th>Value</th>
<th>Ref</th>
</tr>
</thead>
<tbody>
<tr>
<td>Total mass of heavy metals from fuel cycle [MTU]</td>
<td>32,000</td>
<td></td>
</tr>
<tr>
<td>Waste form pitch [m]</td>
<td>20</td>
<td></td>
</tr>
<tr>
<td>Interim cooling period before emplacement [yr]</td>
<td>50</td>
<td></td>
</tr>
<tr>
<td>In-situ waste form failure period [yr]</td>
<td>1000</td>
<td></td>
</tr>
<tr>
<td>PWR (17x17) Array</td>
<td>186×186 (34596)</td>
<td></td>
</tr>
<tr>
<td>Uranium oxide density [kg/m³]</td>
<td>10.960</td>
<td>[10]</td>
</tr>
<tr>
<td>UO₂ fuel fraction of theoretical density</td>
<td>0.95</td>
<td></td>
</tr>
<tr>
<td>Fuel pin radius [m]</td>
<td>0.0082</td>
<td></td>
</tr>
<tr>
<td>Fuel rod height [m]</td>
<td>3.6</td>
<td></td>
</tr>
<tr>
<td>Fuel rods per assembly</td>
<td>264</td>
<td></td>
</tr>
<tr>
<td>Assemblies per canister</td>
<td>2</td>
<td></td>
</tr>
<tr>
<td>Canister Height (Hc) [m]</td>
<td>4.780</td>
<td></td>
</tr>
<tr>
<td>Canister Radius (Rc) [m]</td>
<td>0.419(5)</td>
<td></td>
</tr>
<tr>
<td>Buffer Height [m]</td>
<td>6.18</td>
<td></td>
</tr>
<tr>
<td>Buffer Radius [m]</td>
<td>1.144(5)</td>
<td></td>
</tr>
<tr>
<td>Used fuel canister loading [MT/canister]</td>
<td>1.10</td>
<td></td>
</tr>
<tr>
<td>Uranium canister loading [MTU/canister]</td>
<td>0.922</td>
<td></td>
</tr>
<tr>
<td>HLW (P&amp;T) Array</td>
<td>81×81 (6561)</td>
<td></td>
</tr>
<tr>
<td>Borosilicate Glass Matrix Porosity</td>
<td>0.001</td>
<td></td>
</tr>
<tr>
<td>VHLW Canister Radius [m]</td>
<td>0.215</td>
<td></td>
</tr>
<tr>
<td>VHLW Canister Height [m]</td>
<td>1.340</td>
<td></td>
</tr>
<tr>
<td>Carbon Steel Overpack Thickness [m]</td>
<td>0.0245</td>
<td></td>
</tr>
<tr>
<td>Borosilicate Glass Volume [m³]</td>
<td>0.160</td>
<td></td>
</tr>
<tr>
<td>Overall Canister Mass [MT/canister]</td>
<td>0.5</td>
<td></td>
</tr>
<tr>
<td>Borosilicate Glass Mass [MT/canister]</td>
<td>0.4</td>
<td></td>
</tr>
<tr>
<td>Buffer Radius [m]</td>
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<td></td>
</tr>
<tr>
<td>Buffer Height [m]</td>
<td>4.2</td>
<td></td>
</tr>
</tbody>
</table>

For the vitrified waste, the proportion of actinides relative to glass is very low, so the species of interest are assumed to be released congruently with the glass. With this in mind, per given case, the leach period of the glass has a remarkable effect on the accumulation; a shorter dissolution period allows more of a soluble nuclide to accumulate per given time before the effects of decay are realised. Given the low surface area-to-volume ratio of 11 m⁻¹ for the spherical equivalent in this study, a pessimistic leach period of 109 yr can be employed with respect to the extremely low mass release rates of borosilicate glass in groundwater. However, Ahn et al 1994 used a minimum leach period of 4×104 years,[12] which would conservatively overestimate precipitation. Therefore, these two extremes can be compared, considering that the long period results in a mass release rate similar to the release of uranium from oxide waste.
In accordance with the analytical simplifications used by the code, a spherical equivalent is defined for the waste forms and their surrounding buffers that maintain the same surface area, since this is most important aspect governing dissolution at the surface and subsequent diffusive mass transport. For direct disposal, the actual surface area of the fuel pins is not important since the spacing in between them would be saturated. For the P&T case, in the same manner as the UNF canisters, the spherical equivalent is chosen based on the external surface area of the overpack, with the material properties of the steel overpack assumed to have no effect on mass transport. At the surface of the spherical buffers, nuclides immediately flow into the alluvial entrance.

Table 5: Parameters describing system for TTB input

<table>
<thead>
<tr>
<th>Item</th>
<th>Value</th>
<th>Ref</th>
</tr>
</thead>
<tbody>
<tr>
<td>Fracture half-aperture [m]</td>
<td>0.000315</td>
<td></td>
</tr>
<tr>
<td>Granite-matrix tortuosity correction factor</td>
<td>0.055</td>
<td></td>
</tr>
<tr>
<td>Bentonite porosity</td>
<td>0.30</td>
<td></td>
</tr>
<tr>
<td>Fracture half-spacing [m]</td>
<td>0.5</td>
<td>[13]</td>
</tr>
<tr>
<td>Granite-matrix porosity</td>
<td>0.01</td>
<td></td>
</tr>
<tr>
<td>Bentonite tortuosity-correction factor</td>
<td>1</td>
<td></td>
</tr>
<tr>
<td>Fracture-filling material porosity</td>
<td>1.0 (none)</td>
<td></td>
</tr>
<tr>
<td>Granite density [kg/m³]</td>
<td>2600</td>
<td></td>
</tr>
<tr>
<td>Bentonite density [kg/m³] (wet)</td>
<td>2100</td>
<td></td>
</tr>
<tr>
<td>Groundwater velocity in the fracture [m/yr]</td>
<td>1</td>
<td>[16]</td>
</tr>
<tr>
<td>Dispersion coefficient in the fracture [m²/yr]</td>
<td>1</td>
<td>[17]</td>
</tr>
<tr>
<td>PWR</td>
<td>Spherical equivalent fuel radius [m]</td>
<td>1.044</td>
</tr>
<tr>
<td></td>
<td>Spherical equivalent buffer radius [m]</td>
<td>1.817</td>
</tr>
<tr>
<td>HLW</td>
<td>Spherical equivalent fuel radius [m]</td>
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</tr>
<tr>
<td></td>
<td>Spherical equivalent buffer radius [m]</td>
<td>1.612</td>
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<tr>
<td></td>
<td>Leach Period [yr]</td>
<td>1: 1e9</td>
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<tr>
<td></td>
<td></td>
<td>2: 4e4</td>
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</tbody>
</table>

Table 6: Transport parameters for highly-mobile uranium scenario (relative to TRU)

<table>
<thead>
<tr>
<th>Solubility [mol/m³]</th>
<th>Buffer Kd [m³/kg]</th>
<th>Buffer Free Water Diffusion [m³/yr]</th>
<th>Buffer Retardation [#]</th>
<th>Rock Kd [m³/kg]</th>
<th>Rock Capacity Factor [#]</th>
<th>Fracture Retardation [#]</th>
</tr>
</thead>
<tbody>
<tr>
<td>U 1.00E-05</td>
<td>1.00E-01</td>
<td>2.00E-02</td>
<td>4.910E+02</td>
<td>1.00E-04</td>
<td>2.67E-01</td>
<td>1.00E+00</td>
</tr>
<tr>
<td>Np 1.00E-09</td>
<td>1.00E+02</td>
<td>2.00E-02</td>
<td>4.900E+05</td>
<td>1.00E+02</td>
<td>2.57E+05</td>
<td>1.00E+00</td>
</tr>
<tr>
<td>Pu 1.00E-07</td>
<td>1.00E+01</td>
<td>2.00E-02</td>
<td>4.900E+04</td>
<td>2.50E+01</td>
<td>6.44E+04</td>
<td>1.00E+00</td>
</tr>
<tr>
<td>Am 1.00E-06</td>
<td>1.00E+00</td>
<td>2.00E-02</td>
<td>4.900E+04</td>
<td>2.50E+01</td>
<td>6.44E+04</td>
<td>1.00E+00</td>
</tr>
<tr>
<td>Cm 1.00E-06</td>
<td>1.00E+00</td>
<td>2.00E-02</td>
<td>4.900E+04</td>
<td>2.50E+01</td>
<td>6.44E+04</td>
<td>1.00E+00</td>
</tr>
</tbody>
</table>

PWR uranium release rate: 8.05E-07 mol/yr; HLW mass release rate: (1) 2.07E-07 mol/yr, (2) 5.18E-03 mol/yr

Table 7: Transport parameters for highly-mobile TRU scenario (relative to uranium)

<table>
<thead>
<tr>
<th>Solubility [mol/m³]</th>
<th>Buffer Kd [m³/kg]</th>
<th>Buffer Free Water Diffusion [m³/yr]</th>
<th>Buffer Retardation [#]</th>
<th>Rock Kd [m³/kg]</th>
<th>Rock Capacity Factor [#]</th>
<th>Fracture Retardation [#]</th>
</tr>
</thead>
<tbody>
<tr>
<td>U 1.00E-07</td>
<td>1.00E-01</td>
<td>2.00E-02</td>
<td>4.910E+05</td>
<td>1.00E+02</td>
<td>2.57E+05</td>
<td>1.00E+00</td>
</tr>
<tr>
<td>Np 1.00E-05</td>
<td>1.00E-01</td>
<td>2.00E-02</td>
<td>4.910E+02</td>
<td>1.00E-03</td>
<td>2.58E+00</td>
<td>1.00E+00</td>
</tr>
<tr>
<td>Pu 1.00E-05</td>
<td>1.00E+00</td>
<td>2.00E-02</td>
<td>4.910E+03</td>
<td>2.50E-02</td>
<td>6.44E+01</td>
<td>1.00E+00</td>
</tr>
<tr>
<td>Am 1.00E-04</td>
<td>1.00E+00</td>
<td>2.00E-02</td>
<td>4.910E+02</td>
<td>1.00E-02</td>
<td>2.58E+01</td>
<td>1.00E+00</td>
</tr>
<tr>
<td>Cm 1.00E-04</td>
<td>1.00E+00</td>
<td>2.00E-02</td>
<td>4.910E+02</td>
<td>1.00E-02</td>
<td>2.58E+01</td>
<td>1.00E+00</td>
</tr>
</tbody>
</table>

PWR uranium release rate: 8.05E-09 mol/yr; HLW mass release rate: (1) 2.07E-07 mol/yr, (2) 5.18E-03 mol/yr

For the specific nuclide transport properties, considerable variation in the parameter space is observed that can only be addressed in full through a comprehensive uncertainty analysis. Rather,
cases are made instead choosing from within the scope of variation. While previous studies have based such cases on the arrival of nuclides to the surface of the buffer,[18] in this paper, two extreme cases have been identified that could be relevant to objective of maximising precipitate mass:

1. Uranium is highly-mobile relative to other species, with upper range solubility and lower range retardation (Table 6).
2. TRU is highly-mobile relative to uranium, with upper range solubility and lower range retardation for those nuclides (Table 7).

Results and discussion

Direct disposal scenario

Figure 4 shows the data for precipitates forming in the far-field at the different observation points for the highly-mobile uranium parameter space. No other element besides uranium has a precipitate greater than 1e-10 mol at the final location, and the fissile content is therefore described in full by the uranium enrichment. Table 8 shows the time points to accumulate 0.1, 1, and 10 metric tonnes of uranium along with the associated U-233/U-235 mass fractions. These total masses were chosen as indicative of the heavy metal depositions comprising critical configurations in repeated fractured geometries. Enrichment reads a maximum of 1.778 wt% and 1.776 wt% for the 10m and 100m observation points, respectively. It is plausible for criticality to occur with at least 1 wt%, although it should be kept in mind that the reactor zones of the Oklo natural analogue was at least 4 wt% enriched at the start of chain reactions. Nonetheless, the possibility of criticality cannot be excluded after the first two million years from package failure.

Figure 5 shows the data for the inverse case of highly-mobile TRU at different points in the far-field. Although there is greater diversity in the species being observed, the total mass of precipitate does not exceed the milligram range even in close proximity. Therefore, even though the fissile mass fraction reaches 7.217 wt% at 100 m, the overall mass is far too small to provide the multiplicity necessary for criticality.
**Partitioning and transmutation scenario**

The transport of HLW was modelled using two different leach periods at opposite extremes to demonstrate the effect of glass dissolution on far-field accumulation after overpack failure. Figure 6 shows the results for the highly-mobile uranium transport case for the two observation points in the situation where glass dissolution is very prolonged. The results have very minimal differences due to overall closeness to the repository array, apart from the expected time lag resulting from retardation. The effects of decay are most-obvious with the short-lived U-233 and U-236, while U-234 is in secular equilibrium with its U-238 parent. Enrichment reaches a maximum of 1.969 wt% and 1.966 wt% for the points at 10m and 100m, respectively, which both exceed the maxima observed for direct disposal in the same parameter space. However, it takes an order of magnitude more time to accumulate just 0.1 MT of precipitate, as shown in Table 8. Figure 7 shows the effect of decreasing the dissolution period to 40 000 years. At 10m away from the repository edge at 10 000 years, the amount of U-238 is four orders of magnitude higher than the previous case. This means that more uranium can enter solution at an earlier time with the shorter dissolution period, and the first 0.1 MT is achieved at a time two orders of magnitude lower than the prolonger leaching case. Compared to direct disposal, 1 MT of uranium can accumulate in 850 000 years with an enrichment of 1.948 wt%, as opposed to 6 million years with a lower enrichment. This is caused by a combination of the following:

1. A higher surface area-to-volume ratio of the HLW (~11 m-1 compared ~3 m-1) compared to the UNF canisters, which enhances the diffusive mass transport into the buffer from the surface of the dissolving waste
2. The oxide solubility not being a factor for TRU precursors entering solution
Table 8: Time required for precipitates to accumulate with magnitudes representative of critical masses, with fissile mass fraction

<table>
<thead>
<tr>
<th>Observation Point</th>
<th>0.1 MT</th>
<th>1 MT</th>
<th>10 MT</th>
</tr>
</thead>
<tbody>
<tr>
<td>Case 1 PWR</td>
<td>10 m</td>
<td>1.96e6 yr (1.775 wt%)</td>
<td>6.18e6 yr (1.769 wt%)</td>
</tr>
<tr>
<td></td>
<td>100 m</td>
<td>2.32e6 yr (1.774 wt%)</td>
<td>6.85e6 yr (1.768 wt%)</td>
</tr>
<tr>
<td>Case 1 HLW T₁=1e9</td>
<td>10 m</td>
<td>1.48e7 yr (1.959 wt%)</td>
<td>6.87e7 yr (1.874 wt%)</td>
</tr>
<tr>
<td></td>
<td>100 m</td>
<td>1.60e7 yr (1.947 wt%)</td>
<td>7.18e7 yr (1.870 wt%)</td>
</tr>
<tr>
<td>Case 1 HLW T₁=4e6</td>
<td>10 m</td>
<td>1.28e5 yr (1.778 wt%)</td>
<td>8.54e5 yr (1.948 wt%)</td>
</tr>
<tr>
<td></td>
<td>100 m</td>
<td>2.61e5 yr (1.851 wt%)</td>
<td>1.22e6 yr (1.950 wt%)</td>
</tr>
</tbody>
</table>

Figure 6: Data for HLW repository with leach period of 109 years for highly-mobile uranium transport case at (a) 10 and (b) 100 metres

Figure 7: Data for HLW repository with leach period of 4*10^4 years for highly-mobile uranium transport case at (a) 10 and (b) 100 metres

Figure 8 shows the inverse transport case of highly-mobile TRU for the pessimistic dissolution period. As was observed in the direct disposal case, there is a significant proportion of fissile nuclides but a miniscule amount of metal overall, numbering in the hundreds of milligrams. Increasing the observation distance from 10 to 100 metres greatly impacts the ability for TRU to arrive in appreciable amounts despite the increased mobility. Decay quickly diminishes the plutonium.
isotopes while the arrival or uranium is extremely retarded, save for the U-233 generated by the decay of Np-237.

Figure 9 shows the effect of decreasing the dissolution period to 40,000 years, where the actinides are once again allowed to enter solution at a much earlier time. For example, at 10 m, the mass of Np-237 at 100,000 years is five orders of magnitude higher than the previous dissolution case. Nonetheless, the upper-bound of precipitate mass does not exceed the tens of grams.

**Figure 8**: Data for HLW repository with leach period of $10^6$ years for highly-mobile TRU transport case at (a) 10 and (b) 100 metres

**Figure 9**: Data for HLW repository with leach period of $4 \times 10^4$ years for highly-mobile TRU transport case at (a) 10 and (b) 100 metres

**Conclusions**

Criticality cannot be excluded from consideration in the repository performance assessment based on the scenarios involving both the direct disposal of UNF or HLW from a P&T process coupled to reprocessing with UREX. Only transport scenarios involving highly mobilised uranium (both in the buffer and the fractured granite) can result in the tonnage necessary required for criticality in the far-field. The inverse transport case with highly-mobile TRU is not liable to resulting criticality, but will allow solute transport with a higher proportion of fissile content. Neutronics simulations are still
required to investigate through which exact geological parameters (e.g. porosity and water content) the observed proportions of uranium are liable to result in sustained chain reactions.

For P&T, results from TTB code indicate that the prevention of precipitates indicative of criticality is highly dependent on the characteristics of the borosilicate glass, i.e. surface area and dissolution rate, despite utilising a much more compact array in the repository. If the waste glass can be engineered to vastly inhibit the mass release rate from the surface, perhaps by increasing volume while limiting heat and obeying the other constraints, then performance will at least marginally exceed that of direct disposal. A less-pessimistic P&T process with a higher coefficient of destruction will certainly reduce the amount of TRU in the HLW, but this benefit may not be completely realised if the 0.1 wt% loss of actinides from reprocessing is still observed. As was demonstrated, uranium is the major species involved in both scenarios, so reducing its presence in the final waste stream may be key to preventing criticality in any case.

Acknowledgements

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References


An innovative way of thinking partitioning and transmutation: Neutron physics of a reactor directly operating on spent nuclear fuel

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\textsuperscript{2}National Nuclear Laboratory, Chadwick House, Warrington, United Kingdom
\textsuperscript{3}Helmholtz-Zentrum Dresden – Rossendorf (HZDR), Dresden, Germany

Abstract

The partitioning and transmutation (P&T) technology is the cutting edge of nuclear research. However, the current generation of nuclear reactors producing the nuclear waste are evolutionary in design and in many cases based on the technology originally designed to power submarines, and continue to be dominated by light water reactors (LWRs) for power generation. The future generations of reactors have to be designed and developed driven by sustainability, safety and reliability, economics, resistance to nuclear material proliferation and physical protection. In consequence their development has to follow the ultimate and universal vision for electric energy production, the “perpetuum mobile” – at least as close as is practical.

To achieve this vision, we have designed a nuclear reactor which:

- uses no resources for the operation
- produces no waste during operation
- continues to be safe, secure, reliable, and economic

Developing a reactor following this demand would incorporate P&T into the initial power production system. The results from first simulations demonstrate that, from a theoretical perspective, it is possible to fulfil the sustainability goals by the reuse of spent nuclear fuel from currently operating reactors as the fuel for a new reactor creating almost no additional waste mass.

This approach addresses two of the extended goals we have outlined, however, the safety, reliability and operational economics will remain a challenge and will need to be demonstrated, as basis for the long-term success of nuclear power production as a major carbon free, sustainable, and applied highly reliable energy source. This R&D challenge should be ideally brought forward by a broad international co-operation and the formation of a new, international consortium.

Introduction

Strategic development is the key to the any long-term success of industrial innovation. During the development of a new technology important decisions have to be made due to current external drivers like political, economic or technical boundary conditions. Unfortunately, these drivers often undergo significant changes during the long-term development of a technology. This leads to a change in the objectives and the final outcome differs very often significantly from the objectives given at begin of the development. In the 1960s, Everett Rogers described the development of
innovation through S-curves. He fixed these thoughts in the theory of diffusion of innovations [1]. Rogers’ argues that the application of a technology to a market and the penetration phases from low to high follow an S-curve which is characterised by innovators and early adopters in the first phases and laggards joining the development at the end. The economist Fredmund Malik extended this thinking and titled it a “Symphony of S-curves: Seeing the Future Clearly” in his book on strategy development [2] to motivate the people to leave the beaten tracks when it is requested by changed boundary conditions. He describes the development not with a single s-curve anymore. He invented the idea of changing from one s-cure to another more advanced when radical innovation is required by the market surrounding.

In the first part of this publication the development concept is applied to analyse the current development of nuclear reactor systems. The historic boundary conditions are reviewed against the current boundary conditions. In the second part the updated boundary conditions will be used to define a reactor system which is as close as possible to the requests of a wide spread future electric energy production by nuclear power. The requests of the P&T technology will provide here one of the major keys since it is in our view a major request to create the basis for the long-term success of nuclear reactors. Applying innovative technologies will be the basis for nuclear to act as a major contributor providing reliable carbon free, sustainable electric energy.

The development of nuclear technologies

Development of nuclear reactors started in 1942, with the “Chicago Pile 1” which was the world’s first nuclear reactor, built by Nobel Prize winner Enrico Fermi [3]. During the initial years of the development nuclear technologies, the purpose of a nuclear reactor was based around three objectives

1. Demonstration of a self-sustained chain reaction and its potential application
2. Production of material for military purposes and/or commercial or medical applications
3. Demonstrating energy generation using the chain reaction, either as electricity, or heat

Today’s main focus of nuclear reactors operation is the generation of reliable carbon-free energy in a sustainable way, literally an advancement of objective 3. The self-sustained and safe use of the chain reaction has been extensively proven as well as the energy production in the last centuries. Today, the production of medical isotopes is secured using more specialised technologies.

The currently operating LWRs are still essentially based on technology developed to power nuclear submarines, even if they have undergone a successive evolution to the Generation III/III+ reactors. The location on the current s-curve of the development of current reactors (black curve in Figure 1) is very much advanced after the evolution from the very first prototype reactors of the first generation via second generation to the Gen III/III+ reactors, see red cross in Figure 1. This status is characterised by a competitive market, almost converged solutions from different providers, and small evolutionary development steps to complete the optimisation. LWRs are to be seen as almost ideal solution under the currently given boundary conditions.

The Generation IV (GenIV) goals [4] (sustainability, safety and reliability, economics, proliferation resistance and physical protection) build the bridge between the current S-curve and a possible future one (red curve in Figure 1). The GenIV programme is the planned next stage of reactor design, the application of ‘new, innovative’ concepts for nuclear power generation. Such concepts could lead to the design of a reactor that can continually breed, and burn its own fuel, i.e. become a perpetual
reactor as it were. However, we should not forget when the development of these reactor concepts had been started. Most of the concepts go back to the time of the “atoms for peace” speech and the foundation of the IAEA [6]. “Seen from the historic point of view given above almost all systems are based on the very early developments of nuclear reactors when the development objective has been formed with completely different boundary conditions and requests as we have it today – producing nuclear materials and powering submarines versus wide spread sustainable nuclear power production.”[5]

![Figure 1: The nuclear reactor development in S-curves](image)

**Demand driven research**

Most of the important decisions during the development of nuclear power have been made on the historic demand cited above to develop the ideal system for these given demand. It is time to revisit these old decisions posing the question “Would these decisions be taken the same way under today’s boundary conditions? Would we take the branch-off once more into the same direction, even if we are travelling to another destination?” The community should question their decision makers permanently; “Are we on the right way to the destinations of the future?”

Some very interesting examples can be found for these historic decisions. One of the most prominent could be the decision between the molten salt breeder reactor and the sodium-cooled fast reactor [8]. The sodium-cooled fast reactor with solid fuel is advantageous for the production of high quality nuclear material. Thus supporting this system was the right decision to fulfil the demand of the 60ies but will this decision be advantageous for the wide spread sustainable nuclear power production, too? Another point is the decision for the Plutonium Uranium Redox EXtraction (PUREX) process. What are heading for in reprocessing? We try to take out what we need in the past the Pu like the name of the process defines. Thus the combination of the fast breeder reactor and the PUREX process was ideal for the demand at the time of the development – producing and separating high quality fissile material. For the today’s demand we have to achieve extremely high recovery factors to avoid plutonium being forwarded to the final disposal, but why do we not only take out what disturbs/prevents the reactor operation? Maybe we have to invent new processes for this purpose. We should ask “Can we fulfil the demand in easier and less complex processes?” Do we really need this extremely high purity or is it just a drawback of the way we have chosen? Do we need this extremely complex fuel cycle or could we design a reactor for energy production including...
breeding for sustainable fuel use and a significantly reduced process scheme around? Can we adopt our system to reduce the requests e. g. for reprocessing, mining, enrichment...?

**Figure 2**: The nuclear reactor development based on a roadmap example

Yes we can and we have to! We could compare the development with a road trip. Let’s assume we started our trip in Berlin 1960 with the destination Rome (representing the development destination of 1960 – high quality Pu production). We are now in the year 2016 and we are somewhere south of the Alps and we decide that we have to go to Paris instead of the Rome. Our destination has changed from high quality Pu production to sustainable long-term electricity production. Nobody sitting in a car would further head south when he has to go to Paris – everybody would turn right and head to the west, maybe on different routes but to the direction of Paris.

A comparable process has to be started in nuclear development. If we are willing to leave the evolutionary development to disrupt the chain defining the direction/way we have decided for in the 60ies and 70ies under from today’s demand wrong boundary conditions. Sodium-cooled fast reactors, ideally with metal fuel, to breed plutonium in a highly efficient way and PUREX to win the Pu to enlarge the nuclear armament of the cold war. This isn’t the world of tomorrow. Why should we build an advanced technology to solve a part of the problems of the historic and future nuclear power generation (spent fuel and long-term toxicity of actinides) following a development track defined by historic decisions taken under the historic boundary conditions? We should define a new way to solve the problems of the future taking into account the current demand or even better the envisaged future demand. P&T is a part of a future technology which is even more advanced than Generation IV. The technology has been proven using SFRs and processes developed from PUREX [7], but we are now obliged to find an optimal scheme for the whole fuel cycle if we want to be successful.

Applying the basic requests for a wide spread sustainable nuclear power production in the future, the upcoming next level S-curve should focus on the ultimate, universal vision for electric energy production which is independent of the observed ‘power plant’ system itself. This general vision is
much more advanced and broader than the development goals of the first reactors, than the GenIV goals. The ultimate, universal vision can be given with one simple, old fashioned historic phrase – ‘perpetuum mobile’. It is consensus, among engineer and physicists and, there are the laws of thermodynamics which prevents this vision from operating. However, it is a clear vision and provides a destination that will drive research and innovation to the right point, into developing a reactor with the requested features: breed and burn its own fuel, limiting the waste production as much as possible. This will be required to achieve the vision as closely as possible thus it will form the essential basis for a successful wide spread application of nuclear reactors as carbon-free energy source. The key words for such a translated vision are:

- As little resources as possible requested
- As few waste as possible produced
- Highly economic, reliable, and secure
- Safe [5]

Developing a reactor following the above principles will lead to a system providing the key for a long-term success of nuclear power reactors to become a reliable major carbon free, sustainable energy source for the future. Obviously, a reactor can’t be operated completely without resources but it would be a smart option to better use already existing resources which we currently consider as waste. Such a reactor wouldn’t require fresh resources, when operated on the basis of spent nuclear fuel (SNF) which exists in vast amounts. In addition, it would not produce additional waste since SNF is already considered as waste but it would be used to fuel the reactor to produce additional energy out of the given waste. The ideal design should also make misuse of the plutonium unattractive; it has to be reliable, safe, and economically competitive compared to other sources of electricity generation. The operation of a reactor based on SNF could even excel the objectives of the current P&T strategies. The solution for the problem would be assured by using the electricity production system itself. P&T does not have to be performed in special dedicated reactors by definition if we are able to fulfil the requests directly in the system dedicated to electric energy generation. The identical can be assured in the fuel cycle, if we fulfil the requests in the energy production fuel cycle using a smart design we are not obliged to operate a complicated fuel cycle for P&T. We should design a fuel cycle or reprocessing scheme which is shaped to fulfil the demand of the reactor – separate the materials which prevent the rector from long-term operation.

The sustainability defined above is a hard challenge which has to be solved within the core physics of a proposed reactor and the fuel cycle scheme has to be built around. The limitation of the complete nuclear system is to provide enough excess neutrons for a self-sustained long-term operation on the basis of SNF used. Given this the first requests is to design a fast system which can produce enough excess neutrons.

The provided feasibility study for a reactor operating on SNF is based on a molten salt fast reactor configuration. However, it is only a proposal which definitively needs further exploration and optimisation to realise the requested goals. The potential advantageous safety behaviour and the excellent operational flexibility of molten salt reactors have already been extensively discussed [9, 10, 11]. The challenges in the development of molten salt reactors have already been described in several projects [5, 12, 13]. A short list of the main R&D challenges is given in a final paragraph.
Materials and methods

Reference configuration

The calculations are based on the core dimensions and boundary conditions given in the EVOL benchmark definition (see Figure 3), and focus on a reference molten salt fast reactor (MSFR) of 3 000 MWth. The core is a single cylinder where the reactions occur, with flowing fuel salt within the cylinder [12]. The dimensions are shown in Figure 3. The inner core is surrounded by a blanket/reflector ring, and a protection ring, with both components having the function of protecting the pumps and heat exchanger from the high neutron flux arising within the inner core.

Figure 3: (Right): Simplified scheme of the MSFR system including the core, blanket and heat exchangers (IHX) – (Left): Benchmark definition [12]

The spent nuclear fuel composition for this analysis is based on the “OECD/NEA AND US NRC PWR-MOX/UO₂ CORE TRANSIENT BENCHMARK” [15]. The UOX fuel assembly, with 4.5% U-235, has been modelled using HELIOS 2.1 using an expected burnup of 50 GWd/THM to develop the material configuration for the SNF used as feed for the MSFR. To simulate storage time, the Pu-241 content has been halved and turned to Am-241, representing a postulated storage of 14 years. Besides this change no further adaption for storage has been introduced, which leads to a conservative assumption from a neutronic point of view. The fissile material for reactor start-up TRU as given in Table 1.

Table 1: Used TRU isotopic vector – EVOL benchmark data [12]

<table>
<thead>
<tr>
<th>Isotope</th>
<th>Percentage</th>
</tr>
</thead>
<tbody>
<tr>
<td>Np 237</td>
<td>6.30%</td>
</tr>
<tr>
<td>Pu 238</td>
<td>2.70%</td>
</tr>
<tr>
<td>Pu 239</td>
<td>45.90%</td>
</tr>
<tr>
<td>Pu 240</td>
<td>21.50%</td>
</tr>
<tr>
<td>Pu 241</td>
<td>10.70%</td>
</tr>
<tr>
<td>Pu 242</td>
<td>6.70%</td>
</tr>
<tr>
<td>Am 241</td>
<td>3.40%</td>
</tr>
<tr>
<td>Am 243</td>
<td>1.90%</td>
</tr>
<tr>
<td>Cm 244</td>
<td>0.80%</td>
</tr>
<tr>
<td>Cm 245</td>
<td>0.10%</td>
</tr>
</tbody>
</table>

1. The raw data of the calculations and the used PYTHON script are available under http://datacat.liverpool.ac.uk/id/eprint/194.
The basic salt configuration is also based on the EVOL benchmark, and consists of LiF (18 m$^3$) with mainly UF4 (applied as SNFF4 as approximation) in the core. However, in contrast to the EVOL benchmark, the share of SNF has been determined in a material search, to enable acceptable breeding of fissile material to keep the reactor critical for long-term operation without further feeding of fissile material. The power of the salt clean-up system has to be adapted, since provides an important degree of freedom for the optimisation the neutron economy and thus for the breeding. The blanket region is filled with pure LiF4 (7.7 m$^3$).

**Modelling and simulation tool**

For the study, the HELIOS 2.1 licensing grade code system is used with the internal 177 group library [16]. The HELIOS code is a 2D spectral code with wide unstructured mesh capabilities and a transport solver, based on the collision probability method [17]. The benchmark configuration is transferred to a volume corrected 2D HELIOS model (see Figure 4) with vacuum boundary conditions for a reproduction of the 3D structure. The leakage in the third dimension has been fixed by a comparison of 2D and 3D calculations within the EVOL benchmark exercises. Additionally, the model has been adapted to improve the representation of the 16 heat exchanger pipes instead of the smeared treatment in the benchmark configuration.

![Volume corrected 2D HELIOS model of the molten salt reactor](image)

The HELIOS code is an industrial standard software for the neutron transport calculation, the burn-up calculation, and if requested the cross section preparation in defined calculation areas. Originally, as HELIOS was written for the simulation of solid structured fuel assemblies as used for the determination of the SNF configuration above. Neither online refuelling nor online salt clean-up was foreseen. To deal with these special features of molten salt reactors a PYTHON script has been developed, based on the special features of the HELIOS package. All information, which is constant during the whole reactor operation, is stored in a so-called expert input, the changing material configuration is given in the user input, and is written new in every cycle using the PYTHON script. Within each cycle 5 burnup steps are calculated to the target burnup of 5 000 MWh/tHM. Both inputs are merged in the pre-processor code AURORA, which creates the updated input for the next HELIOS run. The results are evaluated for each cycle in the post-processor ZENITH, where it is decided which isotopes will be fed back into the next user input, created with the help of the PYTHON script (see Figure 5). Using this scheme it is possible to simulate a molten salt reactor...
precisely by using small time steps in the calculation loop. However, in a real MSR two different time scales for salt clean-up can be observed, which are given due to the different foreseen extraction methods for fission product removal. These are the helium bubbling for gaseous and volatile fission products with a comparably short half-life time, and the online salt clean-up for dissolved fission products with a significantly longer half-life. To simulate these different time scales a full removal of all gaseous and volatile fission products takes place after each 5 000 MWD/THM cycle, but only a partial removal of dissolved fission products is established at the end of cycle. The lower efficiency in the separation of lanthanides is considered by a 20% lower reduction than for the other dissolved fission products. The time foreseen for a full throughput of the 18 m$^3$ salt is an optimisation parameter, which needs to be determined in conjunction with the quality of the salt clean-up process chemistry. However, what is most likely is that throughput through the salt clean-up system will have to be increased when compared to the EVOL benchmark, due to the fission products which are inserted with the SNF.

**Figure 5**: Description of the calculation cycle for the simulation of a MSR

Due to the characteristics of HELIOS, some approximations have to be accepted, for example, there is no fuel salt movement, and thus an undesired burnup distribution arises during one calculation cycle. The materials are only re-distributed when a new user input is defined via the script. As HELIOS was designed for use with LWR reactors, a LWR spectrum is used for the within group weighting of the 177 group master library. However, comparisons to SERPENT on the isotope accumulation during the burnup in a fast reactor configuration have shown an acceptable agreement for major isotopes [18]. Additionally, different versions of the HELIOS 1 code have been used in the EVOL benchmark calculations for MSFRs [20] and in the benchmark calculations in the ESNII+ project for SFR [19]. In both benchmarks the results were comparable to the other codes, with no major systematic discrepancies observed. The approximations and use of the HELIOS code package seems to be adequate for the level required for this kind of long-term study. The major uncertainties are predominantly given by the current preliminary design, with a more detailed design required for the next phase. Such design uncertainties are expected to impact have significantly higher impact on the results than the approximations made within the modelling and applied code. In general the calculations are performed to demonstrate the feasibility of such operation of a critical reactor on the basis of spent LWR fuel with high burnup.

**Results**

**Initial core**

The initial core configuration for the simulation is based on 70 tonnes of SNF with 15.5 tonnes of TRU which accumulates to 82 tonnes of HM, with the blanket filled with pure LiF salt. Such a configuration
leads to a start-up core with an averaged $\Delta k_{\text{eff}}$ of 0 over the first burnup cycle of 5 GWd/THM. The values are in a comparable range to other kinds of nuclear reactors, e. g. ~86 tonnes of HM for a 1 000 MWe LWR core [21], or ~130 tonnes of HM for a 1 450 MWe LWR core, or 42 tonnes for a SFR core with 1 450 MWe [22]. In this simulation, an optimisation strategy has been used loading the initial core with an increased amount of SNF and TRU compared to the ideal configuration. In this configuration the initial fission product content of the SNF load acts as a kind of burnable poison which is slowly reduced by the salt clean-up system.

**Simulation over lifetime**

For the simulation over longer-term variable TRU feeding is used to keep the $\Delta k_{\text{eff}}$ in the range of ±500 pcm, Figure 6. In the initial phase until the first tip ~0.5% of the initial TRU loading is fed in every calculation cycle. After ~10 years the system has transformed far enough for the initial LWR configuration of the major part of the core (SNF loading) to a fast reactor configuration that the reactor is operating in self-sustained mode. In this early operational phase less than 2 tonnes of TRU is fed in addition to the initial loading. After this initial phase, the reactor operates on SNF only and the breeding processes support the long-term operation with sufficient fissile material to keep the reactor critical. The use of an optimised loading scheme has reduced the initial feeding period by ~50% compared to earlier simulations. [5]

**Isotopic contents**

Due to the optimised starting configuration the atomic number density of U-238 doesn't remain almost constant like in former calculations. The feed of SNF is now slightly smaller since a reduction of the U-238 content can be accepted to create enough breeding for the long-term sustainable operation, Figure 7.

The atomic number density of the most important fissile isotope Pu-239 increases during the the TRU feeding period, as shown in Figure 8. However, the Pu-240 content in the core increases to an asymptotic value almost twice as high as the initial value, whereas the Pu-241 content drops during TRU feeding below the initial amount produced within the LWR, even after this amount has already been reduced to half simulating the long-term storage of SNF. Both Pu-242, and Pu-238 content increases during TRU feeding, with the Pu-242 decreasing during operation, while the Pu-238 remains fairly constant.
Figure 7: Number density of U-238 particles in the fuel salt over the observed operational period

Isotopic analysis of the Pu compositions at the end of the model shows clear changes, when compared to the initial TRU feed, see Table 2, with the share of LWR fissile Pu isotopes, decreasing from the already low content of TRU, itself taken from high burnup LWR fuel. The primary cause is significant build-up of Pu-240, see Figure 8, with the observed decrease of Pu-241, which is itself typical for the operation of a nuclear reactor with a fast neutron spectrum. In these reactors the accumulation of higher isotopes is strongly reduced compared to LWR with thermal spectrum.
Table 2: The plutonium vector appearing in the reactor after long-term operation compared to the Pu vector of the TRU used for loading

<table>
<thead>
<tr>
<th>Pu</th>
<th>After 60 years</th>
<th>Load</th>
</tr>
</thead>
<tbody>
<tr>
<td>Pu-238</td>
<td>3%</td>
<td>3%</td>
</tr>
<tr>
<td>Pu-239</td>
<td>47%</td>
<td>52%</td>
</tr>
<tr>
<td>Pu-240</td>
<td>38%</td>
<td>25%</td>
</tr>
<tr>
<td>Pu-241</td>
<td>6%</td>
<td>12%</td>
</tr>
<tr>
<td>Pu-242</td>
<td>5%</td>
<td>8%</td>
</tr>
<tr>
<td>Pu\text{fiss}</td>
<td>53%</td>
<td>64%</td>
</tr>
</tbody>
</table>

Discussion

The results presented here demonstrate that a molten salt fast reactor such as that proposed in the EVOL project could operate using pure spent nuclear fuel (SNF) from LWRs. However, for the start-up and the initial transformation phase, some plutonium or TRU fissile material would have to be added, see Figure 6. The addition of fissile material such as separated LWR TRUs is required to keep the system critical during the transformation phase, where the fuel configuration is changed from the LWR to the fast reactor configuration. Once this transition has occurred, the required feeding can be performed using SNF solely. The ideal timeframe for the transformation is about 10 years and depends on the core size, the initial material configuration, the feeding strategy, the salt clean-up system, and the quality of the SNF used for feeding the system. In the study the salt clean-up system has been enhanced compared to the EVOL benchmark.

For the operation of the reactor ~17 tones of TRU are required and ~72 tonnes of SNF are fed, during an operation time of 60 years. A quick calculation value for burnt HM leads to 66 tonnes, with a burning rate of 42 kg/TWh which is characteristic for a fertile-free system where nothing leaves the reactor \([10]\). The comparison of both values in conjunction with the U-238 content of ~95% in the SNF confirms the quality of the very complicated and innovative simulation result.

The reactor operation based on SNF will lead to a significantly reduced fuel cycle (see Figure 9) compared to current nuclear reactor operation scheme or to the double-strata strategy of P&T. New, fresh resources are required which avoids mining which causes significant toxicity, “Clearly, mining is the only contributor with more than 99% of the potential impact both for the eco and the human toxicity” \([26]\). The fuel cycle requirement and complexity could be reduced, too and the separation of specific minor actinides as well as the possibly required production of transmutation targets is not required.
The arrangement of the molten salt reactor typically has co-location of reactor, salt clean-up (reprocessing), and fuel production without further transport, thus there appears no ‘plutonium economy’ like the current requirement for a closed fuel cycle which requires the separation of actinides. No risky handover of Pu at the different stages is required. No separated fresh fertile material appears in the reactor since all fuel components are immediately mixed in the liquid state as already mentioned. This is combined with the Pu staying within the reactor until it undergoes fission. This was already highlighted in 1978 by Engel et al, as one of the most attractive features of liquid fuelled reactors. Once the fissile material, is put into a molten salt reactor system it is not necessary to separate the material, in order to keep the fission process going [23].

A comparison to classical closed fuel cycle operation of fast reactors and the linked double strata is given in Figure 10. If a reactor would be operated on SNF directly, the gains of the closed fuel cycle operation of a fast reactor could be made accessible without a complicated fuel cycle system. There would be no separation of Pu and minor actinides required as well as no double strata to keep the fast reactor fuel cycle free of minor actinides. There would be only one reactor required for the sustainable operation making use of U-238 via breeding and the transmutation of the built TRUs would be solved in one go. From demand driven point of view the low-carbon request as well as the nuclear waste treatment request could be fulfilled in this single machine. Additionally, the waste mass would stay the same since no additional material is used. However, the activity of the used SNF would increase related to the mass as it is known from conventional reprocessing. This is a facto of the increased energy produced form the ~ 20 times more energy is produced, thus the activity of the fission products would be expected to be ~20 higher while the activity per produced MWe will stay almost the same.
Besides the energy production all requirements of waste management driving the development of partitioning and transmutation (P&T) would be fulfilled [10, 13] since only fission products would remain and all transuranic isotopes would stay in the reactor, and in turn be burnt. Hence, only the short-term activity of the nuclear waste will increase when compared to the original SNF. However, a strategy for handling and storage of the separated fission products from the clean-up system has to be developed and a final disposal with reduced observation time requests will be required. In addition to this, a strategy for the capturing and handling of volatile fission products, evaporating from the salt during reactor operation has to be developed. Both challenges are well known in standard molten salt reactor development, with significant work already completed [12, 24, 25].

However, the described system is a demand driven vision which has to fulfil additional objectives safe, secure, and reliable energy production on a competitive cost level compared to other energy technologies. This drives the demand for multi-disciplinary R&D to tackle the major challenges of the molten salt reactor technologies to create a real design which can be manufactured and operated.

Engineering:
- Optimisation of burning used LWR fuel, i.e. demonstration of principles with validation
- Optimisation of reactor design, i.e. better than EVOL
  - Fully understanding the fluid dynamics under both normal and accidental conditions
  - Componentry for molten salt system
- Development of materials capable of operating under such extreme conditions, i.e. temperature, induced radiation damage, corrosion

Chemistry and Thermodynamics:
- Optimisation of salt purification, i.e. removal of fission products from within the liquid phase
- Design, implementation and capture of volatile fission products, helping to keep the liquid phase pure
- Preparing the fuel – choosing the optimal method for converting used LWR fuel in to a form sufficient for use within the reactor
Appreciation of the chemical thermodynamics, and limitations in using molten salts with high levels of actinide loading

Safety:
- Develop the safety methodology and implementation protocols for such a novel liquid reactor design
- Assurance in safety of such a co-located site, i.e. both reactor and reprocessing

Other:
- Economic viability, ensuring the reactor is economically viable and competitive with current technology
- Ensure the public acceptance of such a new technology, without which the reactor is unviable [5]

Besides the above challenges gaining operational experience will be a significant request starting with a small low power experimental machine, which would lead to improved implementable designs. Such a machine has to be planned, built, and financed [10].

Conclusions

Invention and innovation in nuclear reactor development can be described with the concept of developments in s-curves. These s-curves are defining the vision for the future and the vision should be ideally demand driven. The identified key demands which should drive invention and innovation are given by the current/expected boundary conditions, and the evolvable objectives. The current objectives for efficient, safe, secure, and economic electricity production coincide with the ultimate, universal vision for energy production characterised by minimal use of resources and production of waste, while being economically affordable and safe, secure and reliable in operation.

Following these objectives, a new innovative proposal has been shown which is highly sustainable in operation and creates an innovative solution for the P&T request in addition. The proposed nuclear system operation is based on utilising SNF from LWRs as the main fuel, fulfilling the request of sustainability in innovative electric energy production. A proof of feasibility is provided based on a reactor neutronics point of view which demonstrates a solution to the major challenge, establishing a breeding process which generates sufficient fissile material from the inserted SNF for long-term self-sustained operation. For the start up of the system a support of fissile material in the initial core and a transition phase is required. A first optimisation of the design has been performed defining an improved initial composition to reduce the plutonium feeding period.

The result of such a design would be a system which does neither require new resources nor produce additional waste, providing an option to sustainability of a future nuclear system fulfilling the sustainability goals of the United Nations. Consequently, no dedicated reactor and fuel cycle for P&T would be required, the P&T requirements for an innovative waste management strategy would be fulfilled in the power production system as an advantageous side effect. In addition, the system provides enhanced resistance against misuse of Pu arising from a high Pu-240 content, with a very limited possibility for short time insertion fertile material, coupled with Pu remaining in the system without further separations using a demand driven separation technology. The demand would be to separate the fission products which prevent the reactor from long-term operation instead of separating the fissile material. The sustainability of such a system is supported by the significantly
reduced environmental impact of the front end of the fuel cycle. The whole fuel cycle is reduced to 3 steps, i.e. dissolving the SNF, reactor operation, storage of fission products. Eliminating the front end of the fuel cycle results in, eliminating the major source of toxicity in nuclear technology arising from the mining/extraction process.

The design requirement of such an innovative nuclear power system would be safety, reliability, and affordability with limited financial risk in both development and construction. This forms the challenge which should ideally be tackled by an international consortium. The ideal consortium would consist of all countries with an interest in P&T for solving their long-term nuclear waste problem. The development of such a sustainable, and innovative nuclear reactor is a major challenge across almost all disciplines of engineering, but it could have a substantial impact of the future of nuclear technology and thus on the future of the worldwide energy production.

References

3. Achievements: Reactors Designed by Argonne National Laboratory.
10. B. Merk, D. Litskevich: “Transmutation of All German Transuranium under Nuclear Phase Out Conditions – Is This Feasible from Neutronic Point of View?”, PLOS ONE, DOI: 10.1371/journal.pone.0145652.
13. acatech Ed. (2014), “Partitioning and Transmutation of Nuclear Waste: Opportunities and Risks in Research and Application” (acatech position paper), Munich 2014,
www.acatech.de/fileadmin/user_upload/Baumstruktur_nach_Website/Acatech/root/de/Publikationen/Stellungnahmen/acatech_EN__POS_Transmutationsforschung.pdf.


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Some implications of recycling used CANDU fuel in fast reactors

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Abstract

This paper presents the potential implications of adopting an advanced nuclear fuel cycle where used CANDU fuel is reprocessed and supplied to fast reactors designed to burn actinides. The analysis assumes that used CANDU fuel is reprocessed to recover uranium, plutonium and other actinides, which are then used to fabricate fresh fuel for the fast reactors. Once in operation, the used fast reactor fuel is reprocessed and recycled together with makeup from used CANDU fuel to produce energy. Pyroprocessing is assumed for reprocessing of CANDU and fast reactor used fuels.

Deployment of fast reactors in the nuclear energy system is assumed as a method for waste management and for electricity production. With respect to waste management, mass-flow calculations estimate that the reduction in mass of transuranics for disposal would be accompanied by a larger increase in the mass of fission products. The time required to consume most of the transuranics in the used CANDU fuel is also estimated.

Understanding the long-term hazard of the wastes from an advanced fuel cycle (AFC) is important to assess options for their long-term management. Estimates of the radioactivity, radiotoxicity, thermal power and unshielded dose from a reprocessing/fast reactor wasteform and from used CANDU fuel are presented.

The long-term safety of the fast reactor waste is also addressed. Two options are considered, placement in a deep geological repository, and placement after 300 years decay in a near-surface landfill. The analysis estimates that the dose consequences as a result of surface disposal of reprocessing wastes could exceed regulatory limits over long periods of time. That is, even after a few hundred years of decay, the fast reactor wasteform is sufficiently radioactive that it would require appropriate long-term management, such as in a deep geological repository.

Introduction

Research and development studies of advanced nuclear fuel cycles are being pursued by a number of countries through national and international collaborative projects to examine the waste management, resource use, economics and proliferation resistance of different types of fuel cycles using thermal reactors in combination with fast spectrum reactors or accelerator-driven systems (ADS). Typically these studies focus on recycling used fuel from the light water reactors (LWRs). Recently, some assessments on recycling CANDU used fuel in fast reactors have been published [1, 2]. This paper presents a high-level analysis of some of the implications of adopting an AFC where used CANDU fuel is reprocessed and supplied to fast reactors designed to burn actinides [3, 4].
Nuclear fuel cycle

A closed nuclear fuel cycle is considered using fast reactors designed to burn actinides. These reactors require a continuous external source of fissile material in addition to uranium, and the external fuel could include transuranic (TRU) elements from the used fuel of a thermal reactor. Fast reactors operating in this mode have been proposed for waste management purposes. This analysis assumes that used CANDU fuel is reprocessed to recover the uranium, plutonium and minor actinides such as neptunium, americium, and curium, which are then used to fabricate the fresh fuel required for starting and operating a fleet of burner fast reactors (see Figure 1). Once in operation, the used fast reactor fuel is reprocessed and recycled continuously together with make-up from used CANDU fuel to produce energy.

Reprocessing and fuel fabrication

Reprocessing is a key process in a closed nuclear fuel cycle. Various technologies are presently being considered, such as aqueous, pyro, and fluoride volatility processes. They are at various stages of development, ranging from conceptual phase to industrial-scale operation. The technology considered in this analysis for reprocessing both CANDU and fast reactor used fuel is pyrometallurgy reprocessing (“pyroprocessing”). This technology has been implemented on a small scale for the Experimental Breeder Reactor II fast reactor spent fuel [5]. Laboratory scale tests of reprocessing used LWRs fuel, an enriched-uranium oxide fuel, have been completed [6].

The present analysis assumes that pyroprocessing can be successfully adapted to reprocess CANDU as well as fast reactor used fuel on a commercial scale. The pyroprocessing rate is assumed to be sufficient to meet the fuelling requirements of the fast reactors.

A range of recovery efficiencies have been reported or estimated for pyroprocessing. For this study, during reprocessing, a 99.5 wt% efficiency is assumed for recovery of TRU elements, 100 wt% for the separation of fission products, and 99.0 wt% for the recovery of uranium [3]. A further 99.9 wt% overall fabrication efficiency is assumed for the TRU elements and uranium during fuel fabrication [3].
Fast reactor

The fast reactor considered in this analysis is based on the preliminary design of advanced burner reactor developed at the Argonne National Laboratory (ANL), which is based on the 380 MWe SuperPRISM (S-PRISM) reactor designed by GE Hitachi Nuclear Energy, with a 38% thermal efficiency [7].

The reactor is assumed to operate with different core configurations, each with the same power output but with different conversion ratios\(^1\) (CRs) less than one. From a waste management perspective, a fast reactor with a very low conversion ratio, for example as low as 0.25, would be advantageous in a nuclear fuel cycle as it would require a larger amount of external fuel, therefore maximizing the burnup of TRUs from the wastes. A low conversion ratio however would require a high TRU enrichment, beyond current irradiation experience with fast reactor fuels. Based on current technology, the conversion ratio would be more likely in the range of 0.5-0.6 [9]. This paper assumes a range of 0.25-0.75 to estimate the overall system inventory, and further discusses the nature of hazard and long-term safety of the fast reactor waste for a system employing fast reactors with a favourable (for actinide burning) very low conversion ratio of 0.25.

The reactor is assumed to operate with a capacity factor of 85% for all core configurations. The capacity factor would ultimately depend on the conversion ratio as the refuelling frequency, number of assemblies replaced per outage, and unplanned outages related to fuel failures, will differ from one core configuration to another.

Mass-flow assessment

Mass-flow calculations are performed for two scenarios considering the deployment of fast reactors only (i.e. no further CANDUs) in the nuclear energy system as a method for waste management and for electricity production. One scenario considers operation of a block of two S-PRISM type fast reactors (0.76 GWe) as a deliberate method for long-term waste management of all of the used CANDU fuel in Canada’s reactors. This generates some electricity in parallel, but that is not the primary intent. The second scenario considers the current CANDU nuclear fleet would be replaced with fast reactors producing the same amount of electricity. In this case, 36 S-PRISM type fast reactors would be required to operate producing about 13.7 GWe annually. This is an energy supply scenario; however the analysis in this paper is with respect to the implications of this scenario for waste management. All reactors are assumed to start operation simultaneously in year 1 and operate continuously for 60 years. The current analysis provides an overall mass balance perspective, however, it does not comment on the practicality of these fast reactor related technologies, the deployment of fast reactors, or the specific isotopic and reactor physics implications of these fuel cycles.

In all cases, it is assumed that the used CANDU fuel is available to be reprocessed into TRU and uranium (U) streams to support operation of the fast reactors. The amount of used CANDU fuel considered to be available is 103 000 tonnes of (initial) heavy metal. This is the estimate for a high

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1. “Conversion (breeding) ratio” is defined as the number of fissionable atoms produced to the number of fissionable atoms consumed in a reactor. If the ratio is less than 1, it is referred to as “conversion ratio”. If it is greater or equal to 1, it is referred to as “breeding ratio”. [8]
scenario of projected nuclear fuel waste from the existing reactors, based on approximately 5.2 million bundles, assuming that most of the current reactors are refurbished [10].

The fuel characteristics are derived from reference data or estimates from literature. The used CANDU fuel composition is based on the radionuclide inventory for actinides and fission products in CANDU fuel at a reference burnup of 220 MWh/kgU [11]. The fissile plutonium content (Pu-239 and Pu-241) is about 66 wt% of the total TRU content in the used CANDU fuel, where the TRUs include plutonium, neptunium, americium, and curium. The compositions of the fresh and used fast reactor fuel are estimated from the ANL data [7]. The present mass-flow assessment focuses on the TRU content as an aggregate, and does not specifically consider the isotopic content. In practice, a certain amount of more fissile isotopes would be required in the core. The fissile plutonium content derived for the fast reactor fresh fuel is between about 39 wt% (CR=0.25) to 54 wt% (for CR=0.75) of the total TRU content [7]. Since the fissile plutonium content in TRU from used CANDU fuel is higher than this, it is assumed that the CANDU TRU would be suitable for direct use in these fast reactors. However, it may be that some adjustment of the fuel may be needed, in order to make the reactor physics work.

The total amounts of TRUs and fission products that would constitute high-level waste (HLW) from the advanced nuclear fuel cycle are estimated for both scenarios, i.e. the waste management scenario with two fast reactors, and energy supply scenario with 36 fast reactors, for different conversion ratios.

An illustration of the results obtained assuming the favourable very low conversion ratio of 0.25, starting from 103 000 tonnes of used CANDU fuel, is presented in Figure 2. With one power block of two S-PRISM type fast reactors (0.76 GWe) in operation, it is noted that the yearly consumption of the TRUs is very small (a similar trend was observed for higher conversion ratios of 0.5 or 0.75). Extending the analysis from Figure 2 for two fast reactors, it is estimated that it would take almost 1,000 years to consume the entire amount of TRUs in 103 000 tonnes of used CANDU fuel. However, as the operating life of a fast reactor is assumed to be 60 years, this means that approximately 15 generations of 2 fast reactors (or 30 fast reactors) would be needed to burn the entire amount of TRUs in the used CANDU fuel. During this period, there would also be continuous production of fission products.

If the current nuclear fleet is replaced with fast reactors, it would require 36 S-PRISM type fast reactors to produce 13.7 GWe annually. If all these reactors were assumed to start operation simultaneously at year 1 with a favourably low conversion ratio of 0.25, it would take about 50 years to burn the TRUs in the 103 000 tonnes of used CANDU fuel. Although there would be insufficient TRU amounts in the remaining unreprocessed used CANDU fuel to continue operation of all the fast reactors after slightly over 40 years, as noted in Figure 2, there would still be significant TRU amounts in the fast reactor cores. This could be consumed by continued longer operation of one or two fast reactors. For the same scenario (36 fast reactors) and considering more practical conversion ratios (0.5 or 0.75), the CANDU TRU inventory would not be consumed within the assumed 60 years life of these fast reactors.
For both scenarios and for all conversion ratios, the total amount of TRU plus fission products that would have to be managed as HLW in Canada increases with time, reflecting the steady production of fission products resulting from the fission of TRUs and uranium at a rate that is faster than the TRU consumption. The nature of the HLW will change over time as the TRU fraction decreases, and becomes more fission product based. The preliminary implications of this on long-term safety and heat generation are presented in the following sections of this paper.

With respect to uranium, the results indicate that there is little reduction in the total amount of uranium in the system inventory that would need to be managed, for any of the conversion ratios considered. A large amount of uranium would be recovered during the reprocessing of the used CANDU fuel, which could be either stored for future re-use as make-up in the fast reactor or sent for disposal.

System inventory and waste form

The total amounts of uranium (U), TRUs, and fission products (FP) in the used CANDU fuel available in the system initially, at startup of the fast reactors, as well as their remaining quantities after operating the fast reactors for a number of years, are provided in Table 1. The results are presented for the scenario with 36 fast reactors and 103 000 tonnes of used CANDU fuel, for all conversion ratios.

Table 1 shows that reprocessing of the used CANDU fuel and the fast reactor fuel will generate radioactive waste products. Assuming the used fuels are pyroprocessed, waste salt from the electorefining process will contain significant quantities of radionuclides for disposal. A ceramic waste process [5], in which the waste salts are blended with zeolite and borosilicate glass and then consolidated into glass-bonded zeolite ingots, produces a stable wasteform.
Table 1: System inventory assuming 36 fast reactors and 103,000 tonnes of used CANDU fuel

<table>
<thead>
<tr>
<th></th>
<th>Initial</th>
<th>Final CR=0.25 (41 yrs)</th>
<th>Final CR=0.5 (60 yrs)</th>
<th>Final CR=0.75 (60 yrs)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Unprocessed used CANDU fuel [tonnes]</td>
<td>U</td>
<td>101,524</td>
<td>714.4</td>
<td>6,786.8</td>
</tr>
<tr>
<td></td>
<td>TRU</td>
<td>454</td>
<td>3.2</td>
<td>30.4</td>
</tr>
<tr>
<td></td>
<td>FP</td>
<td>1,022</td>
<td>7.2</td>
<td>68.3</td>
</tr>
<tr>
<td>Fast reactors core(s) [tonnes]</td>
<td>U</td>
<td>0</td>
<td>93.3</td>
<td>216.9</td>
</tr>
<tr>
<td></td>
<td>TRU</td>
<td>0</td>
<td>112.8</td>
<td>105.4</td>
</tr>
<tr>
<td></td>
<td>FP</td>
<td>0</td>
<td>16</td>
<td>17.9</td>
</tr>
<tr>
<td>Reprocessing and fuel fabrication:</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Waste sent for disposal [tonnes]</td>
<td>U (3)</td>
<td>0</td>
<td>1,018.5</td>
<td>980.4</td>
</tr>
<tr>
<td></td>
<td>TRU (4)</td>
<td>0</td>
<td>8.7</td>
<td>10.7</td>
</tr>
<tr>
<td></td>
<td>FP</td>
<td>0</td>
<td>1,485.8</td>
<td>1,655.4</td>
</tr>
<tr>
<td>Recovered uranium [tonnes]</td>
<td>U (5)</td>
<td>0</td>
<td>99,540.2</td>
<td>93,128</td>
</tr>
<tr>
<td>Total U [tonnes]</td>
<td></td>
<td>101,524</td>
<td>101,366</td>
<td>101,112</td>
</tr>
<tr>
<td>Total TRU [tonnes]</td>
<td></td>
<td>454</td>
<td>125</td>
<td>146</td>
</tr>
<tr>
<td>Total FP [tonnes]</td>
<td></td>
<td>1,022</td>
<td>1,509</td>
<td>1,742</td>
</tr>
</tbody>
</table>

Notes:
1. At the end of year 41, there will be insufficient TRU in the unprocessed used CANDU fuel to continue operation of 36 fast reactors with CR=0.25.
2. It is assumed that the operating lifetime of the fast reactors is 60 years.
3. Uranium losses from reprocessing of CANDU and fast reactor used fuel, plus losses from fuel fabrication.
4. TRU losses from reprocessing of CANDU and fast reactor used fuel, plus losses from fuel fabrication.
5. Uranium recovered during reprocessing of used CANDU fuel which could be stored for future use or sent for disposal.

**Nature of the hazard**

This section compares the potential hazard posed by waste forms resulting from a once-through CANDU fuel cycle (i.e. used CANDU fuel bundles) with waste forms produced as a result of adopting a fast reactor based fuel cycle (i.e. glass-bonded zeolite waste forms). For a conversion ratio of 0.25, roughly 1,486 tonnes of fission products, 1,019 tonnes of uranium and 8.7 tonnes of TRUs will be sent for disposal. Based on trial fabrication reported in the literature, the fast reactor waste forms are assumed to have a mass of 400 kg [5] and contain 8 wt% waste products [12], resulting in 78,553 waste packages. This would mean a total of 31,421 tonnes of waste, about one third of the initial 103,000 tonnes of used CANDU fuel. Assuming the fission products, uranium and TRU wastes are evenly distributed among all the waste packages means each package will contain roughly 18.9 kg of fission products, 13 kg of U and 0.11 kg of TRU.

Used CANDU fuel and fast reactor fuel will also contain small amounts of light element activation products from impurities in the fuel [11], most notably C-14. Assuming the total inventory of radioactive light element activation products from reprocessed CANDU fuel and fast reactor fuel is evenly distributed among the waste packages results in each waste package containing approximately 0.0096 kg of light element activation products.

To estimate the potential hazard posed by the two waste forms, one has to consider the radionuclide makeup of the fission products, uranium, TRUs and light element activation products. For the CANDU fuel bundles these data are taken from Reference [11], assuming a burnup of...
220 MWh/kgU. For the fast reactor wasteform, the radionuclide makeup is assumed to be the same as used CANDU fuel with a burnup of 220 MWh/kgU [11]. That is, the amount of a radionuclide per kg of fission products would be the same as in CANDU fuel. This is a reasonable assumption because roughly two thirds of fission products and nearly all the uranium, which make up the bulk of the radionuclides in the waste stream, are from the reprocessing of the used CANDU fuel. The radionuclide profile in fast reactor used fuel will be different because of the different initial fuel composition, different neutron spectrum, and the higher fuel burnup. However, a relevant fission product inventory data for the fast reactor used fuel for this scenario is not currently available.

It should be noted that the 99 540 tonnes of uranium recovered from reprocessing as listed in Table 1 are not included in the fast reactor wasteform described above. The potential hazard of this surplus uranium is compared to that of the fast reactor wasteform and used CANDU fuel below.

Figure 3 compares the total radioactivity (in Bq) of the entire inventory of the base case of 103 000 tonnes of used CANDU fuel bundles with that resulting from reprocessing and re-use of this CANDU fuel to produce more electricity – about 31 000 tonnes of fast reactor waste forms and 99 540 tonnes of surplus uranium. The radioactivity of the fast reactor wasteform is dominated by fission products for all times after discharge while the radioactivity of the CANDU fuel bundles is initially controlled by fission products but beyond a few hundred years the total radioactivity is controlled by the actinides. Initially the radioactivity of the fast reactor waste forms is higher than the CANDU fuel bundles due to the larger inventory of shorter lived fission products.

Figure 4 compares the radiotoxicity (in Sv) of the total inventory of used CANDU fuel, fast reactor waste forms and surplus uranium. The radiotoxicity is calculated by multiplying the radioactivity of each radionuclide by that radionuclide’s corresponding ingestion dose coefficient [13]. Figure 5 shows the thermal power (in W) for the total inventory of used CANDU fuel, fast reactor waste forms and surplus uranium. Both the radiotoxicity and thermal power of the fast reactor wasteform and CANDU fuel follow similar trends. For times less than a few hundred years fission products are dominant, after which the actinides control the radiotoxicity and thermal power. Due to the larger inventory of actinides, the used CANDU fuel has a higher radiotoxicity and thermal power than the fast reactor wasteform beyond a few hundred years. Figure 4 and Figure 5 also show that the long-term management of the surplus uranium is a factor that needs to be considered in evaluation of future scenarios.

Figure 6 presents the calculated annual dose rate per kg of wasteform to an unshielded person at 10 m distance from a fast reactor wasteform and a CANDU used fuel bundle. Initially the dose rate (per kg) from the fast reactor wasteform is about six times higher than the CANDU fuel bundle; however, after 1,000 years decay, the fast reactor wasteform is comparable to a CANDU fuel bundle. Beyond 100 000 years, the dose rate for the fast reactor wasteform remains dominated by fission products but starts to decrease as the dominant species (Sn-126) decays. The CANDU used fuel dose rate remains relatively flat at long times due to the ingrowth of uranium decay chain daughters.
Long-term management of reprocessing waste

The previous section shows that the fast reactor wasteform from reprocessing and the CANDU fuel are broadly similar especially initially in terms of radioactivity, radiotoxicity, thermal power and unshielded dose rate. After several hundred years the fast reactor wasteform is less hazardous than the comparable amount of CANDU used fuel. Two potential long-term management options for disposal of the fast reactor wasteform are discussed below, that is: disposal in a deep geological repository and disposal in a near-surface landfill site.

Deep geological repository

The purpose of a deep geological repository is to safely isolate used nuclear fuel or nuclear wastes from the surface environment through a number of passive barriers. In the case of used CANDU fuel, these barriers consist of the used fuel bundle, a steel and copper container, bentonite clay, and hundreds of metres of low permeability rock. It is intended that these barriers will remain intact.
essentially indefinitely. Post closure safety assessments [14, 15] show such a facility could meet regulatory requirements for the protection of people and the environment. In these assessments, the significant dose contributors are either fission products or light element activation products. The most influential radionuclide is I-129, a mobile (soluble and non-sorbing), long-lived fission product. Despite uranium and TRUs having long half-lives and making up the bulk of the used CANDU fuel, these species do not contribute significantly to the total dose due to the insoluble and immobile (insoluble and high sorbing) nature of these species.

If a parallel is drawn between a repository containing used CANDU fuel and one containing fast reactor waste from reprocessing, one would expect higher dose consequences from the fast reactor waste forms (in a similar container and geological setting) given that it would have a higher fission product inventory. Obviously, the dose consequences would depend on a number of factors including the degradation rate of the wasteform. However, it is likely that a deep geological repository could be designed to safely store the glass-zeolite waste forms from reprocessing.

**Surface disposal**

The radioactivity and radiotoxicity of the fast reactor waste forms drop significantly over the first 300 years after discharge. It is therefore sometimes implied or inferred that beyond 300 years, this material need no longer be considered (long-lived) nuclear waste. If that were correct, then by implication the post-300 year material could be disposed in a near-surface landfill facility. In reality a number of nuclear regulations would apply to the fast reactor wasteform given the residual level of radioactivity. However, in this assessment these requirements are not considered and the potential implications of near-surface disposal are evaluated.

After the 300 year cooling period, the entire inventory of fast reactor waste forms are assumed to be placed in a modern near-surface (but non-nuclear) landfill. The near-surface landfill is assumed to consist of two liners made of high density polyethylene and compacted clay each separated by a gravel leachate collection system. An attenuation layer of low permeability soils separates the landfill from a nearby aquifer (see Figure 7).

**Figure 7: Surface landfill design**

![Surface landfill design](image-url)
The dose consequence of a near-surface landfill is assessed using a compartment modelling software AMBER v5.7.1 [16]. In this model the waste, liner layers, leachate collection systems, attenuation layer, and aquifer are represented by a series of compartments and radioactive decay and ingrowth are considered throughout the model. The conceptual model is shown in Figure 8.

**Figure 8: Conceptual AMBER model**

Over time, the wasteform degrades and radionuclides are able to start migrating through the landfill. The model considers three separate wasteform degradation modes. The first wasteform degradation mode only considers leaching and radionuclides are released from the wasteform via an element dependent leaching rate until the wasteform is depleted of radionuclides. The second degradation mode assumes that the leaching phase is unsustainable and ceases after 10 years; however, radionuclides will continue to be released via congruent dissolution of the wasteform. The third degradation mode considers both the short-term leaching phase and long-term congruent release from the wasteform but also considers precipitation of insoluble species once released from the wasteform.

Once free from the wasteform matrix, the radionuclides are able to migrate via advection and diffusion through the landfill liners, leachate collection systems, attenuation layer and into the aquifer. It is assumed that the primary and secondary leachate collection systems are able to remove 90% of the leachate while they remain intact (lifetimes of 500 and 100 years respectively). Beyond 1000 years 100% of the leachate is assumed to migrate into the attenuation layer and subsequently into the aquifer.

100 m from the landfill a domestic water well is assumed to intercept the aquifer and capture a fraction of the radionuclides in the aquifer. The ingestion dose rate for a person drinking contaminated well water is estimated on a per annum basis. Figure 9 shows the ingestion dose results for the three wasteform degradation modes described above.

Figure 10 examines the model sensitivity to a few key assumptions. The first case examines the effect of extending the leaching phase duration from 10 years to 100 years. The second case studies the effect of increasing the congruent dissolution rate by a factor of 10. The third case assumes the leachate collection system do not capture any leachate migrating through the landfill and the final case examines the influence of reducing the well capture fraction to 1%. In all sensitivity cases the third wasteform degradation mode was used.
The results in Figure 9 and Figure 10 show that the dose consequences are quite dependent on some uncertain model parameters and assumptions, but within these uncertainties the dose consequences from disposal of reprocessing waste in a near-surface facility after 300 years could exceed regulatory limits. In cases considering solubility-limited release from the wasteform, the doses are dominated by fission products and light element activation products (I-129, C-14 and Cl-36). In other cases the dose rate is controlled by the uranium daughters (notably Ra-226).

Discussion

This paper documents a high-level analysis of an advanced nuclear fuel cycle where the TRUs from the used CANDU fuel are assumed to be burnt in a S-PRISM type fast reactor. Mass-flow calculations and a preliminary hazard assessment have been performed to estimate the impact of such a nuclear fuel cycle from a waste management perspective. Scenarios considered the deployment of fast reactors only (i.e. no further CANDUs) as a method for waste management and for electricity production, with all reactors assumed to start operation simultaneously.

With one power block of two S-PRISM type fast reactors (0.76 GWe) in operation, the yearly consumption of the TRUs is very small, regardless of whether the fast reactors are operating with a conversion ratio of 0.25, 0.5 or 0.75. For two fast reactors operating with a favourable very low conversion ratio of 0.25, it was estimated that it would take almost 1,000 years to consume the entire amount of TRUs in 103,000 tonnes of used CANDU fuel. However, as the operating life of a fast reactor was assumed to be 60 years, this means that about 15 generations of 2 fast reactors (or 30 fast reactors) would be needed to burn the entire amount of TRUs in the used CANDU fuel. During this period, there would also be continuous production of fission products.

If the current nuclear fleet is replaced with fast reactors, 36 S-PRISM type fast reactors would be required to produce 13.7 GWe annually. If all these 36 reactors are assumed to start operation simultaneously at year 1 with a favourably low conversion ratio of 0.25, it would take about 50 years to burn the TRUs in the 103,000 tonnes of used CANDU fuel. For the same scenario (36 fast reactors) and considering more practical conversion ratios such as 0.5 or 0.75, the CANDU TRU inventory would not be consumed within the assumed 60 year lifetime of these reactors.

In all cases, it was observed that the total amount of TRUs plus fission products from the advanced nuclear fuel cycle increases with time, reflecting the steady production of fission products resulting from the fission of TRUs and uranium at a rate that is faster than the TRU consumption.
That is, neither of these fast reactor scenarios results in a net loss of the hazardous components of used fuel. For clarity, as more power has been generated from the recycled used CANDU fuel, there is less total waste per amount of power produced, but more total waste.

The nature of the resulting HLW would change over time as the TRU fraction decreases, and becomes more fission product based. Therefore the hazard associated with waste produced by reprocessing used CANDU fuel and burning this in fast reactors was also estimated. The results of this assessment indicate that the reprocessing and fast reactor wastes from 103 000 tonnes of used CANDU fuel could be stabilised in about 31 000 tonnes of a glass-zeolite fast reactor wasteform (and about 99 000 tonnes of uranium). These waste forms could have a level of radioactivity, radiotoxicity, thermal power and unshielded dose rate broadly similar to that of a CANDU fuel bundle – higher initially, lower after about 300 years with the differences quantified in this paper.

A simplified post closure assessment assumed disposal in a modern surface landfill of the fast reactor wasteform after a 300 year decay period. The results show that the dose to a person drinking water from a well that intercepts an aquifer near the landfill could exceed public dose limits. It is recognised that this analysis has made several simplifying assumptions; however, it is likely that wastes from the reprocessing of CANDU and fast reactor fuel would still be sufficiently radioactive to be considered as long-lived nuclear waste. Such wastes would remain hazardous for very long times, would need to be carefully managed and would ultimately need to be disposed of in a safe manner similar to the existing inventories of used CANDU fuel.

References


Plenary Session VI: Advanced Nuclear Fuel Recycling

Chair: C. Ekberg and G. Lumetta
Sigma team for advanced actinide recycle: Highlights and directions

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Oak Ridge National Laboratory, United States

Abstract
Separation of minor actinides has been an active research area worldwide towards achieving closed nuclear fuel cycles that reduce the heat and radiotoxicity burden on geologic repositories, reduce the consequences of potential disruptive events involving repositories, and increase the utilisation of nuclear fuel. Simple and efficient americium and curium separations, from lanthanides and from each other, are regarded as major technical challenges requiring new technology. While research so far has produced demonstrated technologies for such separations, they have tended to be complex and difficult to control. Needs for simpler chemistry, greater robustness to aggressive conditions, reduced waste production, and increased inefficiency are widely recognised. The Sigma Team for Advanced Actinide Recycle (STAAR) in the USDOE Fuel Cycle Technology Programme has been developing new science and technology to achieve these goals, with particular focus on the problem of separating trivalent actinides from lanthanides. Accordingly, integrated hypothesis-driven research mainly within the paradigm of aqueous reprocessing of used oxide nuclear fuel dissolved in nitric acid is being pursued. Questions being addressed deal with principles of selectivity through various means including aqueous-phase complexation, soft-donor extractants, high oxidation states of americium, and ion exchange. Explicit attention is also paid to the question of robustness of chemical systems under conditions relevant to future technology implementation. This paper reviews the goals of the STAAR, significance of achieving these goals, STMAS scientific thrusts to address the above aims and questions, recent highlights of achievements, and future directions.
The EXAm process: Towards the reprocessing of a concentrated PUREX raffinate

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Abstract

Recycling americium (Am) alone from the spent fuel is an important option studied for the future nuclear cycle (Generation IV systems) since Am is one of the main contributors to the long-term radiotoxicity and heat power of ultimate waste. The liquid-liquid extraction process called EXAm has been developed by the CEA[1,2] to allow the recovery of Am alone from a PUREX raffinate (already cleared from U, Np and Pu). In the extraction step, a mixture of malonamide and HDEHP (di-2-ethylhexylphosphoric acid) in TPH is used as the solvent and the Am/Cm selectivity is improved using TEDGA (tetraethylidiglycolamide) as a selective water soluble complexing agent to maintain Cm and heavier lanthanides in the acidic aqueous phase. A specific molybdenum scrubbing, with a polyaminocarboxylic acid (DTPA), was introduced before the americium recovery. In order to make the process and the future plant associated more compact, the EXAm process has been recently adapted to a concentrated raffinate.

With such conditions, the process operates close to saturation both for the solvent and the complexing agent TEDGA during the Am extraction step. Consequently the three main steps to validate the process in such conditions were:

- batch experiments with higher concentrations of cations;
- a continuous pilot tests with spiked solutions of Am and Cm;
- a test with genuine concentrated PUREX raffinate in the CBP hot cell of the ATALANTE facility.

The main conclusions of each step are described, including the design of the flowsheet.

The results obtained during this hot test are presented in terms of americium recovery and decontamination factor.

Although the results are very good, some additional batch tests will be performed to explain some unexpected results and to consolidate the EXAM process. The 2.5 grams of americium obtained will be manufactured to obtain a pellet for a future transmutation in an experimental reactor.

References


Technology readiness levels (TRL) assessment of separation processes for current and advanced fuel cycles

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Abstract

Under the mandate of the Nuclear Science Committee (NSC), the Expert Group has conducted a comprehensive review of the progress of separation technology and current achievement, considering mechanical, aqueous and dry pyroprocesses. This state-of-the-art report describes the different steps of the Spent Fuel separation processes from the preparation of the used nuclear fuel (head-end processes) to the extraction of the different elements. Each process was described according to its purpose: U recovery, U/Pu extraction, co-processing of actinides, TRU extraction using new solvents. Various options have been investigated for the extraction of actinide elements as a group, for example the GANEX process or to separate a target element such as Am-Cm. The separation of other elements, such as Cs and Sr, is also covered. Processes using not only a nitric media, but exploring other media, such as carbonates, are described.

An important part of this report is dedicated to the assessment of the TRLs. The technical maturity and state of R&D of the major aqueous and pyrochemical processes was compared. These assessments of the status of the various processes were performed by the adoption of the well-established technology readiness level (TRL) methodology, which is widely used and recognised internationally. It should be noted that only the technology readiness of each separation process step was assessed and not the TRL of the integrated process plant. An additional tool, “Scientific readiness levels” (SRL), was also used to evaluate how well the processes are underpinned by scientific understanding as opposed to empirically based process testing.

The processes with higher maturity (higher TRLs) are the ones based on the commercially operated PUREX process, which allows the separation of U/Pu with or without Np (COEX™, simplified PUREX). A few processes are close to industrial implementation. Innovative processes for the separation of U/Pu/Np and/or the separation of other elements are at TRL 4-5. Pyroprocesses, such as dry oxidation, zirconium recovery from cladding, fluoride volatility, molten salt precipitations, and electrochemical separations are less advanced than some aqueous separation processes, and fewer routes have been developed so far. However, the implementation of advanced fuel cycles (AFC) has led to a renewed interest for the various types of pyroprocessing, in particular for the treatment of advanced fuels such as metallic and nitride fuels.
Actinide separation process development at the
UK National Nuclear Laboratory

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Abstract

Although the current policy in the United Kingdom (UK) is to stop reprocessing of spent nuclear fuel around 2020 (i.e. transition to an open fuel cycle), the option to return to a closed fuel cycle later this century is being kept open. Specifically, the UK’s nuclear energy roadmap identifies scenarios in which an expanded use of nuclear energy, significantly above the 16 GW planned for nuclear new build, would be necessary to meet energy security and carbon emissions targets by 2050. In these scenarios, it is expected that the introduction of fast reactors with spent fuel recycling would be required to optimise the use of fuel resources and minimise high-level waste (HLW) for disposal in a repository. Therefore, competitive, safe, secure, industrially deployable technology options for future spent fuel recycling are needed, including separation flowsheets for advanced reprocessing and minor actinide partitioning. At the UK’s National Nuclear Laboratory (NNL), in collaboration with other partners through national and international projects such as “SACSESS”, we are interested in the development of potential flowsheets for thermal and fast reactor reprocessing following both heterogeneous and homogeneous routes for actinide recycle. That is, for heterogeneous recycle, an advanced PUREX process followed by an innovative (i) selective actinide extraction (SANEX) or extraction of americium alone (EXAm) process and the grouped actinide extraction (GANEX) process for homogeneous recycling. Our recent efforts have been placed on the development of the GANEX process, neptunium extraction in Advanced PUREX and americium recovery in i-SANEX/EXAm processes as well as process safety studies including initial efforts regarding the integration between the solvent extraction and conversion flowsheets. There have also been spin-out benefits in applying the separations chemistry being developed for advanced reprocessing to other actinide materials recovery programmes, for residues management, isotope production or space power applications. This paper consequently provides an overview of our recent progress related to the development of actinide separation processes together with a perspective on potential future directions for recycle R&D in the UK.
Recent progress on R&D of the separation process for minor actinides using new extractants

T. Matsumura, H. Suzuki, Y. Ban, S. Hotoku, Y. Tsubata
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Abstract
The separation process for minor actinides (MA = Am, Cm and Np) has been developed at Japan Atomic Energy Agency using new innovative extractants to improve the partitioning process from the viewpoints of the economy and the reduction of secondary wastes. Phosphorus-free compounds consisting of carbon, hydrogen, oxygen and nitrogen (CHON principle) were applied to the separation steps for MA.

In this study, the MA separation process consists of three steps. At the first step, MA and lanthanide elements (Ln) are recovered from high-level liquid waste by solvent extraction with N,N,N',N'-tetradodecyldiglycolamide (TDdDGA). Trivalent actinides, Am and Cm, are separated from RE at the next step by solvent extraction using new hybrid type extractant such as N, N, N', N', N'', N''-hexaoctylnitrilotriacetamide (HONTA) or soft-donor extractant such as N,N,N',N'-tetrakis(pyridin-2-ylmethyl)- decane-1,2-diamine (TPDN). At the last step, Am is extracted by new extractant selectively. This paper presents the recent progress of the research and development programme for the MA separation process.

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SACSESS: Three years of optimisation of safe actinide separation processes

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Abstract
Nuclear power plays a key role in limiting EU’s greenhouse gas emissions, and makes an important contribution towards improving European Union’s independence, security and diversity of energy supply. However, its social acceptance is closely linked to an enhanced
safety in the management of long-lived radioactive waste. Among the different strategies, partitioning and transmutation allows a reduction of the amount, the radiotoxicity and the thermal power of these wastes, leading to an optimal use of geological repository sites.

As a continuation of the FP7 ACSEPT project, SACSESS, started in March 2013, has provided a structured framework to improve existing fuel cycles by addressing the safety issues of the actinide separation processes selected within ACSEPT. These processes involve new chemical systems. To be developed further, these processes required a comprehensive study of the multiform safety issues that any chemical process requires under operation conditions or maloperation. Important experimental work was dedicated to the improvement of the process flowsheets, focusing not only on the key performances of the process but also on numerous side parameters linked to kinetics, radiolytic stability, downstream effects. A new process, dedicated to recovery of the sole americium from a PUREX raffinate has also been developed and tested, in order to optimise the composition of the nuclear waste to be vitrified.

In addition, for the first time, with the help of TSOs and with feedback from safety analyses, methodologies have been developed and applied to these processes in order to identify safety issues and then to optimise these processes. Also, all the results have been integrated to optimise the flowsheets, to perform system studies and ensure the link with other projects and initiatives to ensure the relevance of the SACSESS research programme.

This paper presents the main achievements of SACSESS; a few weeks before its end.
Status of R&D on pyrochemical processes for spent nuclear fuel treatment in Europe within the SACSESS Project

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Abstract

Future advanced nuclear energy systems aim at effective fuel utilisation for sustainability and waste minimisation through the recycling of all actinides. It requires the development of efficient separation processes for recovery of all actinides from the spent fuel. Two types of processes can be applied, a hydrochemical one, based on aqueous solvents and a pyrochemical one, using non-aqueous chemicals, mainly molten salts. The most significant feature of pyrochemical techniques is the potential to process the new fuel types which are being developed for metallic fuel cycles or advanced P&T schemes. Typical examples are metallic An-Zr based alloy fuels or inert matrix fuels/targets for transmutation of Pu and/or minor actinides, based on cermet or cercer refractory fuels with high Pu content.

In Europe, two promising pyrochemical separation processes were identified, developed and assessed within the EC/EURATOM Framework Programme projects PYROREP (FP5), EUROPART (FP6) and ACSEPT (FP7). They are electrorefining in molten chloride for recovery of all actinides on a solid aluminium cathode from spent metallic nuclear fuel and liquid-liquid reductive extraction in molten fluoride/liquid aluminium dedicated to the treatment of oxide or carbide fuels.

This work summarises the most important results achieved within the follow-up European FP7 project SACSESS, where a pyrochemical domain was devoted to the safety issues of pyrochemical processes through 4 work-packages. One part was focused on the safety aspects of pyrochemical systems. The physico-chemical behaviour of actinides and fission products that impacts chemical safety, e.g. solubility, volatility, influence of oxygen ingress, viscosity, both in chloride and in fluoride melts was studied. A second work package was dedicated to safety related to metallic fuel treatment, where the electrochemical techniques were investigated. The third section dealt with safety related to refractory cermet and cercer fuels treatment, where the specific safety factors arising from treating these fuels by the liquid-liquid reductive extraction were addressed. The last work package investigated the conditioning of used chloride and fluoride molten salts into waste streams suitable for safe storage or disposal in a deep geological repository.
Engineering and materials considerations for industrialised LKE systems

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Abstract
NNL recently re-commissioned their Molten Salts Dynamics Rig to investigate the industrialisation of a lithium chloride-potassium chloride eutectic system for the reprocessing of spent nuclear fuel. Shortly after re-commissioning, a significant leak of molten salt was noted and the rig declared as no longer operational.

From inspecting and analysing the rig, a number of engineering and materials considerations can be found including:

- Improvement of design and operation of freeze valves through branching of pipes or forced cooling
- The use of butt welds under an inert blanket as the preferred welding method to prevent corrosion
- The need for suitable instrumentation to be investigated due to multiple failures of radar, pneumercators and sight glasses.
- Standard mineral wool insulation soaks and wicks LKE, leading to criticality, inventory control and accident recovery concerns
- 304 stainless steel is not suitable for a LKE system due to its susceptibility to corrosion in this environment. 316 stainless is suitable in the absence of air or moisture.

Introduction

Pyroprocessing and REFINE

Pyroprocessing is an alternative method to aqueous techniques for the reprocessing of spent fuel which does not require the use of solvent extraction and associated chemicals. In particular, molten salt (MS) reprocessing relies on the dissolution of spent fuel into an MS media, e.g. LiCl-KCl eutectic (LKE), from which U can be electrochemically separated using a solid cathode and the Pu and transuranics (TRU) using a liquid Cd cathode (LCC). Advantages of MS reprocessing compared to aqueous reprocessing include, but are not limited to:

- Lower quantities of waste produced
- Inherent actinide partitioning
- Good non-proliferation characteristics
- Low off-gas flows

The REFINE consortium was a group of UK universities led by the University of Edinburgh, which also included the National Nuclear Laboratory, formed as part of a 4 year Engineering and Physical Science Research Council (EPSRC) funded programme to investigate and develop molten salt
reprocessing technology. One of the aims of the REFINE project was to begin work on industrialisation of an LKE system to show it could be considered against any current or future aqueous processes. It was identified that defining fluidisation characteristics and transporting molten salt on an engineering scale would represent major progress towards the industrialisation of molten salt technology, as moving salt would be a key requirement of any production scale process. To this end, NNL would re-commission and conduct work on its existing Molten Salts Dynamics Rig (MSDR).

**Molten salts dynamics rig**

The MSDR was designed and built in 2004 for the testing of three different pumping technologies for the transport of molten salts: a gas lift, a centrifugal pump and a reverse flow diverter (RFD). It was operated until 2006, with work conducted on the pumping technologies and freeze valves, after which it was placed in a state of care and maintenance. As part of the REFINE consortium, NNL agreed to re-commission the MSDR and conduct a pumping and freeze valve testing programme on the rig to investigate industrialisation of LKE reprocessing technology.

The MSDR consisted of three vessels: V2001, V2002 and V2003. V2001 was originally used to charge the rig with new salt, but was not utilised during re-commissioning. The different pumping technologies were to be utilised to transport the LKE inventory of approximately 120kg (~70L when molten) from the pumping vessel, V2002, to the receipt vessel, V2003, with a gravity drain from V2003 to V2002 for return of the LKE.

LKE has a melt temperature of 352°C \(^1\). Melting was achieved using three coil heaters capable of >500°C positioned at the top, middle and bottom of each vessel. The heaters surrounded, but did not make contact, with the outer wall of the vessel, and were then encased with a thin sheet of stainless steel. A layer of Rockwool\(^\circ\) insulation was then wrapped around the inner case, with a second stainless-steel case then placed around the insulation. The piping was heated using heating tapes, also capable of >500°C, which were wound around the pipes and covered with layers of Rockwool\(^\circ\) insulation to prevent the heat from dissipating and the salt from freezing. Temperature readings were taken externally using thermocouples, providing temperature inputs for the associated heater controllers.

The rig was also used to test freeze valves. This is a non-mechanical “valve” where a section of pipe is left un-lagged and is wrapped by a heating tape which is controlled independently from the rest of the line. To stop flow, the heating tape is turned off which freezes the salt this particular section, imitating a valve. Sections of the gravity drain and gas lift lines were left un-lagged for this purpose.

![Figure 1: Simplified MSDR diagram](image)
Unfortunately, prior to the commencement of the gas lift pumping trials, molten LKE was noticed leaking out of the bottom pumping vessel V2002 (Figure 3). As the source of the leak was unknown and it appeared a large volume of LKE was lost, a decision was made to stop all further work on the MSDR and conduct an “autopsy” to investigate the failure of the MSDR and learn as much as possible on LKE in industrial settings.

This paper details the findings from previous operation of the rig through re-commissioning to the autopsy investigation and its findings.

**Engineering considerations**

**Freeze valves**

The two freeze valves on the MSDR were trialled in the original campaign, both from a static baseline, (i.e. the salt was frozen in the vessels and then melted with the freeze valve “closed”), and from a dynamic baseline (i.e. the salt was being pumped when an attempt was made to “close” the valve).

In a representative static test, the freeze valve on the gravity drain was successful in preventing flow from V2003 to V2002. The freeze valve was then “opened”, a process which took 3.5 hours and a melt temperature of 393°C.

Although the freeze valves were successful in a static LKE situation, they were not in dynamic tests. The gas lift freeze valve could not close over a period of 3 hours under the following conditions:
Overall rig temperature of 500°C
- Argon gas lift pressure of 1.15barg and flowrate of 0.2m3/h
- Corresponding to a molten LKE flowrate of 2L/min

The temperature recorded at the gas lift freeze valve was 347°C, which is only 5°C under the bulk melt temperature of 352°C. This small temperature difference over such a small length was not enough to cool the LKE by any appreciable amount.

In this test, the gravity drain was set to be “closed”, reaching a steady-state temperature of 282°C. It failed to stop flow, however it did restrict flow, causing a 3mm decrease in the LKE height in V2002 as the LKE could not transfer from V2003 to V2002 as quickly as normal. The decrease in height is noteworthy when it has been illustrated in the original trials that the gas lift flowrate is strongly dependent on the pressure head generated by LKE height.

The failure of the freeze valves in dynamic situations is likely due to the orifice effect as the LKE freezes. As the salt froze at the walls of the pipe, it created an orifice, increasing salt velocity, and preventing the salt from freezing due to the speed at which it flowed through the freeze valve (see Figure 4).

**Figure 4**: Freeze valve section

Possible ways of increasing the effectiveness of freeze valves include lengthening the valve as a single pipe or branching into multiple pipes to increase surface area or a forced cooling mechanism to increase the rate of heat transfer for the given area.

**Joint and welding techniques**

As part of re-commissioning, a corner of the gas lift line was required to be excised and replaced with a new section due to materials failure during the original campaign period. The original corner section was removed and replaced with a 316 stainless-steel section, with end caps in place for the joins to the original pipe (Figure 5). The join was performed by butting two pipes against one another and a collar, wider than the pipe diameter, welded over the top.
After the work was conducted, re-commissioning commenced on the MSDR. As re-commissioning operations progressed, the gas lift line was heated to allow for commissioning of the gas lift itself. During one heating operation, it was noted that one of the thermocouples on the heating tapes was showing that no rise in temperature was occurring, leading to the rig being shut down and allowed to cool to ambient temperatures to investigate the failure. After the rig had cooled to ambient temperatures, the lagging around the rig was removed, which revealed a breach in the pipework where the end cap was placed during replacement of the excised corner, with significant “ballooning” visible (Figure 6).

It was determined that the salt had leaked from the rupture and damaged a large section of the heating tape, which then led to its failure. It was also noted that the salt travelled up the outside of the pipe against gravity (Figure 7), although it was thought at the time if this was due to surface tension and the effect of the insulation and pressure “pushing” the LKE up the pipe.
To investigate this further, the end cap was removed, bagged in argon to minimise corrosion and contamination and sent for analysis. A longitudinal cross section of the cap end (Figure 8) revealed that the collar walls had thinned significantly. Frozen LKE salt was located inside the collared section which discoloured around where the 2 pipes almost met. This green discolouring was similar to that observed in the excised corner and was assumed to be a result of a build-up of corrosion product in the salt. No gap should have been present between these 2 pipes and the mistake should have been identified through a post-weld analysis. As shown in Figure 8, LKE salt had entered the collared section when molten and significant corrosion had taken place. Figure 9 illustrates how the metal pipework had rapidly thinned through corrosion and how the corrosion products had built up in the LKE salt. In order for this corrosion to happen, air must have been present inside the pipework. This may have been due to the pipework not being purged with argon after the weld was performed, or air entering the MSDR through another path.

**Figure 7: Salt travelling against gravity**

**Figure 8: Internal contents of the gas lift line after being sectioned**
The principle cause of the failure of the end cap was LKE passing through the poor join on the main pipe and into the collar. The LKE then corroded both the collar and pipe in the presence of air and moisture, which may have ingressed at the time of work, weakening the steel. In addition, as the collar underwent heating cycles, the LKE would expand and contract, placing stresses on the internal face of the collar. It is thought that this, in combination with the corrosion weakening the steel, is what caused the “ballooning” (see Figure 6) and the eventual rupture of the end cap.

From the failure of the end cap, it is recommended that all modification operations take place under a complete inert (argon or nitrogen) blanket to minimise air and moisture ingress. It is also recommended that all piping joint welds be done as butt welds to prevent the possibility of “microclimates” of corrosion to form, as was the case with the end cap.

**Instrumentation**

When first constructed, the MSDR had a number of sensor systems present in both vessels including closed circuit television (CCTV), sight glasses and radar level measurement. However, within 2-3 months of first operations, all three had failed. Although the cause was not determined at the time, it is thought that LKE vapour damaged the glass in the sight glasses and the lenses of the radar and CCTV instruments (Figure 10).
An online gas analyser was also present to perform analysis of the argon blanket that provided an inert atmosphere within the MSDR. However, this failed to function as LKE vapour crystallised in the capillary piping, blocking gas flow and rendering the analyser redundant.

Instrumentation in LKE systems will be a requirement for safety and control of any industrialised system. It is recommended that further research take place to identify and test various instrumentation techniques, particularly stand-off techniques, to allow for industrialisation to progress.

Materials considerations

**Insulation materials**

The vessels were constructed in three layers: the primary vessel containment, an intermediate casing and an outer casing. The heaters used to provide the temperature were placed between then primary vessel and the intermediate casing, with the Rockwool insulation placed between the intermediate and outer casing.

![Vessel section illustrating vessel construction](image)

The insulation material used on the rig for both the vessels and pipework was Rockwool®, a standard mineral wool industrial lagging material. During the leak investigation, one of the difficulties in gaining access to the rig was due to the insulation having soaked up the LKE (Figure 12). This resulted in the Rockwool® layers compressing together and being saturated by the frozen LKE and the material becoming very tough and difficult to remove.

![LKE-soaked Rockwool insulation](image)
The LKE in the Rockwool® deliquesces at a very slow rate based on anecdotal inspection and the insulation still remains hard today, approximately 18 months after the initial leak (April 2015). The LKE also causes the insulation to stick to the material it was covering, further complicating the removal process. In removing the insulation from V2002, dedicated cutting tools (angle grinders in this case) were required to remove the bulk of the LKE-soaked insulation, with the small remainder removed manually with a hammer and chisel.

A laboratory experiment was performed to understand how the molten LKE salt interacted with the Rockwool® and to determine the saturation point of the material. This was done by placing 17.5g of anhydrous LKE salt in a carbon crucible within a dry glove box and suspending a strip of Rockwool® insulation (5g) in a vertical orientation so only the bottom surface was in contact with the LKE salt. The carbon crucible was then placed in a quartz vessel and heated to 500°C for 12 hours before being allowed to cool. As shown in Figure 13, all of the LKE salt had been absorbed by the Rockwool® and had vertically travelled up the test piece against the force of gravity by capillary action (wicking effect). This experiment showed that Rockwool® is an excellent material for absorbing molten LKE salt and is able to absorb over 3 times its own weight.

In a nuclear context, the wicking effect of Rockwool®, a type of mineral wool insulation standard in industry, presents two major obstacles to be overcome. Firstly, it can delay diagnosis of leaks, as was the case in the MSDR as it is unknown for what duration the vessel was losing LKE, which would be an unacceptable loss of inventory control in an active system. Once identified, human access to leak sites may not be possible due to radiation dose, therefore suitable strategies must be identified for recovery from fault operations, especially considering the corrosion potential of LKE accelerating plant degradation. In addition, the hardening and adhesion of the Rockwool to the vessel complicates recovery operations, especially in the absence of human operators.

Secondly, if the LKE was loaded with nuclear materials, e.g. U, Pu, minor actinides, then there could be criticality concerns if the insulation was not safe-by-shape. In addition to the criticality concerns raised by the Rockwool, there are much further consequences. The loss of inventory to the Rockwool would be unacceptable in a nuclear reprocessing plant, especially if the LKE was not recoverable, leading to the need to dispose of a highly-radioactive solid waste that was not planned.
and likely has no identified waste route. As such, it is recommended that different insulation materials or techniques, e.g. vacuum insulation, be investigated for future molten salt systems.

**Steel grades**

During the MSDR’s first operations (2005-2006), a leak had occurred at a flange on the corner of the gas lift line (Figure 14) but repairs were not able to be made before the rig was put into C&M. As part of re-commissioning operations, the flange was cut out from the rig and bagged in argon to minimise further contamination and corrosion of the flange. The flange was then sent for analysis to determine the failure mode, with pipework put in place to form a corner piece on the rig (see Joint and Welding Techniques).

**Figure 14**: Images showing a) the leak from the gas lift flange and b) a close-up of the corroded flange

![Figure 14](image)

Note: flange clearly stamped 316L grade stainless steel.

Figure 15 shows the flanged-leg section of the gas lift line that was removed from the MSDR, which was subsequently bagged in argon to prevent further corrosion and transported for analysis. As shown in Figure 16, significant corrosion was observed around the bolts, nuts and washers on the flange end, particularly around bolts 2 and 3. The graphite O-ring between the flange and the flange plate was also shown to be severely degraded as illustrated in Figure 16b. The flanged end was disassembled in an argon dry box to inspect the interior faces. Bolts 2 and 3 were found to be significantly harder to remove, possibly due to the higher degree of corrosion. The graphite O-ring material crumbled as the flanged end was disassembled. Analysis of the flanged leg was performed using optical emission spectroscopy (OES) and showed that the corner pipe section including the flange was fabricated from 316L grade stainless steel as specified. However, the flange plate, nuts, bolts and washers were made from 304 grade stainless steel (despite the material stamp) which was not suitable for this application. Due to the significant increase in corrosion experienced by this flange compared to the rest of the rig, it is recommended that 304 grade stainless steel is discounted as a material for any system that may come into contact with an LKE salt. It is likely that 304 grade stainless steel would also suffer from the same corrosion in other chloride and fluoride salt systems. Therefore, it is recommended that it also be discounted as a material for these salt systems.
Conclusions

A number of materials and engineering recommendations can be made from the NNL experience with the MSDR:

1. Freeze valve design should consider the use of branching and forced cooling to ensure successful operation.
2. Butt welds should be used where possible to minimise the possibility of “microclimates” for corrosion.

3. Suitable instrumentation that can withstand the harsh climate of molten salt processes will need to be identified for process control and safety in an industrial setting.

4. As standard mineral wool insulation wicks molten salt, leading to criticality and inventory concerns, other materials and techniques should be investigated.

5. 304 stainless steel is not suitable in an LKE environment due to its susceptibility to corrosion in this environment. 316 stainless steel is suitable as long as air and moisture are controlled to minimal levels.

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References

Recent activities on pyroprocessing of spent nuclear fuel in CRIEPI

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Abstract

Pyroprocessing has been attracting attentions worldwide as a promising method to process spent nuclear fuels, recover minor actinides and separate them from fission products, in the partitioning and transmutation scenario because of its intrinsically high proliferation resistance, high stability against radiation, compact and flexible batch process and so on. Central Research Institute of Electric Power Industry (CRIEPI) has been developing the technology of pyroprocessing from basic to engineering scale; fundamental properties of actinides and lanthanides in the molten chloride salts/liquid metals systems, basic feasibility confirmed from results using non-irradiated fuels and simulants, equipment design data accumulated from engineering scale tests with unirradiated uranium fuels (5 kg-U/batch scale), waste treatment such as actinides recovery from the fuel residues and fission products absorption in zeolite.

Recently, the tests using the irradiated metallic fuels were carried out to obtain detailed material distribution data of actinides (U, Pu, Am, Np and Cm) and fission products (rare earth, alkali, alkaline earth and noble metal): Electrochemical behaviours of the irradiated metallic fuels in molten chloride salts were investigated in detail and actinides recovery from the irradiated metallic fuels were successfully demonstrated. Concerning waste treatment, glass-bonded sodalite, stable form of zeolite absorbing fission products, was fabricated under several conditions and supplied to the long-term leaching tests.

CRIEPI is proposing a novel pyroprocessing concept as an option. The concept using a liquid Ga as electrode material is expected to achieve both a recovery of actinides and a separation from the fission products with a high separation efficiency which are required especially in the partitioning and transmutation scenario.

Introduction

Metal fuel cycle composing of metal fuel fast reactor, fuel fabrication by injection casting and pyroprocessing has been attracting attentions as a promising concept for partitioning and transmutation (P&T) scenario (Figure 1) [1-2]. An effective burning of minor actinides (MAs) is expected due to the high heavy metal density and high-energy neutron spectrum of the metal fuel fast reactor. The injection casting suitable for a remote operation accepts active materials with a higher MAs concentration from a shorter cooling period. Pyroprocessing has a lot of advantages for processing spent metallic fuels because of its intrinsically high proliferation resistance, high stability against radiation, compact and flexible batch process and so on. The main step of the pyroprocessing is electorefining where MAs are recovered in liquid Cd with uranium and plutonium. It is noticed that the enclosure of MAs is inherently fulfilled in the metallic fuel cycle without additional recovery step.
Central Research Institute of Electric Power Industry (CRIEPI) has been developing the technology of pyroprocessing from basic to engineering scale [3]. Fundamental properties of actinides and lanthanides in the molten chloride salts / liquid metals systems were investigated, based on which demonstrative tests were performed using non-irradiated fuels and simulants to confirm the basic feasibility of the pyroprocessing. The engineering-scale tests were carried out with unirradiated uranium fuels (5 kg-U/batch scale) to accumulate equipment design data. Much effort was devoted to the studies for waste treatment, such as actinides recovery from the fuel residues, fission products absorption in zeolite to recycle spent molten salts, glass-bonded sodalite fabrication using the zeolite containing fission products and so on. Detailed material distribution data of actinides (U, Pu, Am, Np and Cm) and fission products (rare earth, alkali, alkaline earth and noble metal) were obtained from the tests using the irradiated metallic and oxide fuels and genuine high-level liquid waste: Electrorefining of the irradiated metallic fuels and the electrochemically reduced MOX fuels was successfully demonstrated. Actinides in the genuine high-level liquid waste were recovered through pyro-partitioning technology, de-nitration, chlorination and reductive extraction. CRIEPI is proposing a novel pyroprocessing concept as an option. In the concept, both a recovery of actinides and a separation from the fission products with a high separation efficiency are expected, which are required especially in the partitioning and transmutation scenario. The concept uses liquid Ga as a key electrode material. Electrochemical behaviours of the liquid Ga electrode in LiCl-KCl melts were investigated to confirm the feasibility of the concept.

The recent activities are selected from the above-mentioned topics to be presented in detail in the following sections; the actinides recovery from irradiated metallic fuels, glass-bonded sodalite fabrication and novel concept of pyroprocessing using a liquid Ga electrode.

**Actinides recovery from irradiated metallic fuels [4-5]**

Spent metallic fuel contains actinides with fission products, such as rare earths, alkalis, alkaline earths and noble metals. At electrorefining, the main step of the pyroprocessing, actinides are recovered from the spent fuel separating from fission products. A schematic drawing of its concept is shown in Figure 2.
Figure 2: Concept of electrorefining. MA: minor actinides, FP: fission products.

The spent metallic fuel pins are loaded in a metallic basket which is used as the anode in LiCl-KCl melts at 500 °C. Then, the actinides in the fuels are anodically dissolved in the melts,

\[ \text{An} \rightarrow \text{An}^{3+} + 3e^- \quad \text{(An: U, Pu, minor actinides (MA))} \]  

(1)

The U\(^{3+}\) ion is selectively reduced to U metal on an inert solid cathode,

\[ \text{U}^{3+} + 3e^- \rightarrow \text{U} \]  

(2)

Pu and minor actinides are recovered in a liquid Cd cathode together with U,

\[ (\text{U}^{3+}, \text{Pu}^{3+} \text{or MA}^{3+}) + 3e^- \rightarrow (\text{U, Pu or MA})_{\text{in Cd}} \]  

(3)

Alkalis and alkaline earths fission products dissolve and accumulate in the melts. Some portions of the dissolved rare earth fission products are deposited in the liquid Cd cathode according to their distribution behaviours.

Above-mentioned process, electrorening, was demonstrated using irradiated metallic fuel in an Ar atmosphere hot cell with a biological lead shielding installed in European Commission-Joint Research Centre (JRC) Karlsruhe. U-19wt%Pu-10wt%Zr and U-19wt%Pu-10wt%Zr-2wt%MA-2wt%RE alloys irradiated at PHENIX reactor in France with approximate maximum burn-up of 2.5 at% was cut into pieces. The length and weight of each piece were ~5 mm and ~2 g, respectively. A stainless-steel basket loading the pieces was used as the anode. It was confirmed that almost all of actinides in the pieces were dissolved into the melts (a high dissolution ratio of actinides (U> 99%, Pu> 99%, Np> 99%, Am: 99%, Cm: 99%)) leaving most of Zr and noble metal fission products in the anode residues.

It was found that the actinides and rare earths fission products dissolved with the progress of the electrolysis, while almost all of alkalis and alkaline earths fission products dissolved at the initial stage of the electrolysis, suggesting that alkalis (AL) and alkaline earths (ALE) fission products dissolved by chemical reaction with U\(^{3+}\) ion in the melts,

\[ 3\text{AL} + \text{U}^{3+} + \text{AL}^+ + \text{U} \]  

(4)

\[ 3\text{ALE} + 2\text{U}^{3+} + \text{ALE}^{2+} + 2\text{U} \]  

(5)

SEM-EDX observations showed that the anode residue was composed of three distinct concentric areas. Porous Zr structures remained at the centre \((r/r_0 = 0 \sim 0.1, r: \text{distance from the centre of the fuel residue, } r_0: \text{radius of the fuel residue})\) and outer \((r/r_0 = 0.5 \sim 1)\) areas, and small Zr particles were
aggregated at the intermediate area ($r/r_0 = 0.1 \sim 0.5$). During irradiation, the re-distribution of the fuel components were induced by the radial temperature gradient in the fuel (~670°C at the centre to ~550°C at the periphery), resulting in three concentric areas with different alloy compositions; higher Zr concentration at the centre (27 wt%-Zr) and outer (14 wt%-Zr) areas, and lower Zr concentration at the intermediate area (2 wt%-Zr) [6]. The three distinct concentric areas found in the anode residue were considered to be strongly correlated with the fuel components re-distribution during irradiation.

The dissolved U was selectively recovered on the solid cathode with a high current efficiency. The grouped recovery of actinides in the liquid Cd cathode was confirmed. Separation factors were similar to those obtained at experiments using non-irradiated materials (see Table 1).

$$SF_M = \frac{X_{M \text{ in melts}}}{X_{U \text{ in melts}}} \times \frac{X_{M \text{ in Cd}}}{X_{U \text{ in Cd}}}$$  \(6\)

Table 1: Separation factor of the representative elements based on uranium

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<tbody>
<tr>
<td>U</td>
<td>1(base)</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Np</td>
<td>2.3 (237Np)</td>
<td>2.15</td>
<td>2.07</td>
</tr>
<tr>
<td>Pu</td>
<td>2.4 (239Pu)</td>
<td>2.04</td>
<td>2.22</td>
</tr>
<tr>
<td>Am</td>
<td>3.1 (243Am)</td>
<td>3.42</td>
<td>-</td>
</tr>
<tr>
<td>Cm</td>
<td>5.2 (244Cm)</td>
<td>3.82</td>
<td>-</td>
</tr>
<tr>
<td>La</td>
<td>141 (139La)</td>
<td>81.2</td>
<td>150</td>
</tr>
<tr>
<td>Ce</td>
<td>45 (140Ce)</td>
<td>51.9</td>
<td>55.6</td>
</tr>
<tr>
<td>Nd</td>
<td>43 (144Nd)</td>
<td>32.3</td>
<td>53.3</td>
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All samples, bulk salts, salts at the bottom of crucibles, cathode deposits, and anode residues were analysed by ICP-MS, confirming the detailed mass distributions of important elements including minor actinides at electrorefining of irradiated metallic fuels.

Electrorefining of irradiated metallic fuels with a higher burn-up, 7 at%, was carried out, where actinides were successfully recovered at cathodes similarly with the case of irradiated metallic fuels with 2.3 at% burn-up. The detailed mass distributions of actinides and fission products and the effects of the burn-up on the anodic behaviour will be discussed in a report in near future.

Sodalite solidification of spent salt [9]

As described in the previous section, fission products such as rare earths, alkalis and ALE accumulate in the melts in the form of their cations at electrorefining (see Figure 2). The accumulated fission products are removed from the melts by absorbing them in the structure of zeolite. Type-A zeolite, a synthetic zeolite Na$_{122}[(AlO_2)_{122}(SiO_2)_{122}]$, is one of the suitable zeolites for this use. The cations of fission products exchange with those in the zeolite according to their affinities. The zeolite containing fission products is then mixed with glass and heated to form a glass-bonded sodalite, which is stable and suitable for disposal.
Since it is clear that the heating condition affects the quality of the product, the transformation of the mixture of zeolite and glass to glass-bonded sodalite was performed under various heating conditions, the maximum temperature of the heating, the heating duration at the maximum temperature, the ratio of glass to initial loadings and the weight load for pressing (see Table 2).

Table 2: Glass-bonded sodalite fabrication condition surveyed [9]

<table>
<thead>
<tr>
<th>Maximum temperature for heating (°C)</th>
<th>Heating duration at maximum temperature (hours)</th>
<th>Glass ratio to initial loading (wt%)</th>
<th>Weight load for pressing (g cm⁻³)</th>
</tr>
</thead>
<tbody>
<tr>
<td>770, 820, 915, 1000</td>
<td>5, 10, 20</td>
<td>13, 25, 33</td>
<td>2, 36, 70, 200</td>
</tr>
</tbody>
</table>

As the results, the following tendency was found: The amounts of volatiles during the heating and free salts in the product were reduced by lowering the maximum temperature. The higher apparent density of the product was obtained at longer heating duration and higher weight load. Then, an optimised condition was proposed, maximum temperature: 820°C, heating duration at maximum temperature: 5 hours, ratio of glass to initial loading: 25 wt% and load weight for pressing: 200 g cm⁻³. The glass-bonded sodalite prepared under the proposed heating condition has been supplied to the long-term leaching test, of which results such as normalised release rate of the components will be reported in near future.

Novel concept of pyroprocessing using a liquid Ga electrode [10]

Pyroprocessing with a higher minor actinides (MA)/lanthanides (Ln) separation efficiency is required in the case of the fuel fabricated from a higher Ln/MA ratio source and the fuel with a higher content of MAs to be fabricated. As seen in Table 1, in the case of actinides recovery in a liquid Cd cathode from irradiated metallic fuel, the separation factors of MA based on Ce are calculated to be 20 for Np, 15 for Am and 8.7 for Cm. On the other hand, a liquid Ga is reported to have higher separation factor of Am, 112, than that of Cd at equilibrium state [11]. This indicates that liquid Ga has high potentiality as the electrode material in pyroprocessing for recovering MA with a high separation efficiency from Ln. However, basic knowledge on the liquid Ga electrode in LiCl-KCl melts is very limited. Thus, CRIEPI first measured the following basic properties of the liquid Ga electrode to investigate the feasibility of a liquid Ga as the electrode material.

1. Cyclic voltammetry using a liquid Ga electrode was measured in LiCl-KCl melts containing an alloying element chloride (1.6wt%UCl₃, 2wt%PuCl₃, 2wt%CеТCl₃ or 2wt%NdCl₃) to determine the potential for binary alloy formation. The obtained alloy formation potential is summarised in Table 3. This showed a sufficiently large difference between the formation potentials of actinides-Ga and lanthanides-Ga alloys.

2. Based on the results (1), potentiostatic electrolysis for forming alloys, U-Ga, Pu-Ga, Ce-Ga and Nd-Ga, and their de-alloying were carried out. The obtained alloys were analysed by ICP-AES, which showed that the current efficiency for the binary alloys formation and their de-alloying was calculated to be ~100 %.
Table 3: Binary alloy formation potential at 773 K [10]

<table>
<thead>
<tr>
<th></th>
<th>U-Ga alloy</th>
<th>Pu-Ga alloy</th>
<th>Ce-Ga alloy</th>
<th>Nd-Ga alloy</th>
</tr>
</thead>
<tbody>
<tr>
<td>Binary alloy formation</td>
<td>1.55</td>
<td>1.4</td>
<td>1.3</td>
<td>1.3</td>
</tr>
<tr>
<td>potential (V vs. Li⁺/Li)</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

The above results confirmed that a process using the alloy formation and de-alloying on the liquid Ga electrode had high separation efficiencies of actinides from lanthanides. The concept of the process is shown in Figure 3.

Figure 3: Concept of pyroprocessing using a liquid Ga electrode. An: actinides, FP: fission products.

The process consists of two steps which utilises electrochemical alloy formation and de-alloying reactions in molten chlorides. In the first step, spent nuclear fuel pins are loaded in a metal basket, which is used as the anode in molten chlorides. The actinides (An) dissolve in the melts electrochemically,

\[ \text{An} \rightarrow \text{An}^{3+} + 3e^- \]  \hspace{1cm} (7)

At the same time, lanthanides in the fuels dissolve in the melts to form their cations. At the liquid Ga cathode, actinides are preferentially deposited to form An-Ga alloy leaving lanthanides in the melts,

\[ \text{An}^{3+} + 3e^- \rightarrow \text{An}_{\text{in Ga}} \]  \hspace{1cm} (8)

In the second step, the An-Ga alloy formed at the first step is used as the anode to dissolve the actinides (de-alloying),

\[ \text{An}_{\text{in Ga}} \rightarrow \text{An}^{3+} + 3e^- \]  \hspace{1cm} (9)

The dissolved actinides are deposited in a liquid Cd cathode,

\[ \text{An}^{3+} + 3e^- \rightarrow \text{An}_{\text{in Cd}} \]  \hspace{1cm} (10)
The actinides are finally recovered by distilling the Cd [12]. Concerning fission products, such as rare earth, alkaline and alkaline earth elements, they accumulate in the melts of the first step after repeating the process. The salt treatment process [13], fission products absorption in the structure of zeolite, is applied to remove them from the melts. As the future study, the separation factors of minor actinides at non-equilibrium state are measured and the operational condition taking into account the low solubility of actinides in liquid Ga is optimised.

Acknowledgements

Parts of the results presented in this paper are the results of “Novel pyroprocess with a high separation efficiency of actinides from lanthanides” entrusted to Central Research Institute of Electric Power Industry (CRIEPI) by the Japanese Ministry of Education, Culture, Sports, Science and Technology (MEXT). The authors would like to express their appreciation to Mr H. Tanaka for his great efforts on the presented works.

References

Poster Session

Chair: YJ. Choi
Preliminary analysis of the uncertainty propagation using a Monte Carlo method in PATEROS fuel cycle scenarios

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Abstract
Nowadays, the analysis of fuel cycle scenarios include uncertainty propagation for only a small set of linearly correlated input and output parameters. In order to make a general uncertainty analysis in this field, the TR_EVOL code (developed at CIEMAT) has been upgraded to include a Monte Carlo method to deal with the simultaneous impact of several magnitudes uncertainties in the final results, no matter the relationship between them.

A significant number of perturbed samples of the same reference scenario can now be performed in the TR_EVOL code with the aim of addressing a better estimation of the global uncertainty when non-linear relations do not allow such approach in a sensitivity analysis. The main objective of this work is to apply this methodology to a series of relevant scenarios and to explore the largest uncertainties arising for future studies.

This methodology has been applied to the study of the scenarios developed in the EU PATEROS project of the 6th FP, which are of particular interest at European level to draft a roadmap for partitioning and transmutation. The results are presented in terms of mass according to their total accumulated value, final value or maximum reached value, as defined by the user. These results are given as mean values and their uncertainties as the standard deviation of the samples. Non-linear effects can be seen such as biases that affect the Gaussian curves shape of the results.

Introduction
Fuel cycle scenario assessments simulate the whole fuel cycle, from mining to waste management, over a period of time. Although previous studies were focused on equilibrium modes scenarios, nowadays transition scenarios for the comparison of different reactors fleets, reprocessing and waste management strategies, etc., are evaluated. Additionally, the propagation of uncertainties due to the assumptions made for the scenario definition is also receiving attention because of its importance to quantify which of these assumptions have significant impacts in the output parameters of the scenario.

This effort has been recently started by the Nuclear Energy Agency (NEA), by means of the Expert Group on Advanced Fuel Cycle Scenarios. As part of the activities of this group, a special task devoted to study the effects of the uncertainty of input parameters on nuclear fuel cycle indicators is being finalised [1]. In that work the general approach was to identify sources of uncertainty and report their impacts on the total system by means of parametric analyses.

In order to make a general uncertainty analysis in this field, the TR_EVOL code (developed at CIEMAT) [2] has been upgraded to include a Monte Carlo (MC) method to deal with the simultaneous
impact of several magnitudes uncertainties in the final results, no matter the relationship between them (linear or non-linear).

A significant number of perturbed samples of the same reference scenario can now be performed in the TR_EVOL code with the aim of addressing a better estimation of the global uncertainty even when non-linear relations do not allow such approach in a sensitivity or parametric analysis. The main objective of this work is to apply this methodology to a series of relevant scenarios and to explore the largest uncertainties arising for future studies.

Scenarios description

This methodology has been applied to the study of the first two scenarios defined in the EU PATEROS project of the 6th FP [3] [4] which are of particular interest at European level to draft a roadmap for partitioning and transmutation. These scenarios consider two different groups of countries: one at a standstill for nuclear energy that has to manage its own spent fuel (group A) and another in a continuation scenario that optimises its Pu management for the future deployment of fast reactors. Both of them participate in the deployment of a group of shared accelerator-driven subcritical systems (ADS) that uses the Pu of group B and transmutes the entire Minor Actinide (MA) inventory. Pu can be either mono-recycled (Scenario 1) or multi-recycled (Scenario 2) (see Figure 1 for more details).

The energy produced by the different fuels and reactors is drawn in Figure 2, and a brief description of the most important aspects of each scenario is given in the following paragraphs. A detailed description of the scenarios can be found on References [3] [4]. On the other hand, TR_EVOL needs some specifications for the fuel cycles not available in bibliography, so extra arbitrary assumptions have been made to complete the scenario definition. However this is of low importance in this work since the main objective is the study of uncertainty propagation and not the fuel cycle itself.

Figure 1: Scheme of reference scenarios
Scenario 1

Each pressurised water reactor (PWR) unit has an output of 1,000 MWe while for the ADS is 154 MWe. The PWR are fuelled with UOX or MOX (8.5% of Pu) and ADS’ fuel contains a mixture of 45% Pu and 55% MA. The reprocessing plants of the group B start their operation in 2010 with a capacity of 1,700 t/y for UOX and 120 t/y for MOX. Group A starts its reprocessing strategy in 2040 (850 t/y) and the ADS in 2050 (1,700 t/y). After 60 years of operation, some ADS units have to be decommissioned until the stabilisation of the MA amount is achieved.

Scenario 2

This scenario is similar to Scenario 1 for Group A. For Group B, all the spent fuel (SF) of this group is reprocessed in the same reprocessing plant, which starts in 2005 with a capacity of 1,600 t/y. After year 2025, the UOX and MOX fuel are substituted by MOX-RMA, a special mixture consisting of a 4.2% multi-recycled Pu and natural uranium. Again, after 60 years of operation, some ADS units have to be decommissioned (smaller amount than in Scenario 1) for the stabilisation of the MA amount.

Uncertainty methodology: Monte Carlo method

The uncertainty propagation of all the relevant parameters along the nuclear fuel cycle can be made with a Monte Carlo perturbation approach. This consists in the simultaneous random variation of all the parameters involved in the cycle and the posterior analysis of the multiple outputs in terms of frequency. Even though this method provides useful information like global uncertainties, the identification of the most relevant parameters contributing to that uncertainty is still required and has to be provided by other methodology in the future.
The methodology used in the MC approach and implemented in TR_EVOL code is described as follows. All numerical parameters in the cycle have been sampled with the exception of the energy generated by the nuclear plants of group B. The variation follows a uniform distribution with upper and lower boundary limits of 5% from the central value (taken as the value of the reference scenario), except for the reprocessing efficiency that is only of 0.05% to guarantee the number being less than unity. Then, a total number of 10,000 different executions have been run (so 10,000 different sets of input parameters and output files have been generated).

For temporal parameters, power plants of group B are assumed to be fixed on time, but for ADS the beginning of the commissioning can be delayed up to two years and the ending either delayed or advanced (always considering 60 years of lifetime for the reactors).

Depending on the nature of each output indicator, and following the considerations by [1], the study of the accumulated value, its maximum or final value has been considered. In all the cases, the uncertainty is statistically calculated as the standard deviation of the results.

The principles of Sensitivity/Uncertainty analysis (SA) are well known and documented and have been extensively applied to study the impact of the nuclear data uncertainties on reactor core performance parameters such as the irradiated fuel isotopic composition [5] [6] or the \( k_{\text{eff}} \). The objective of this analysis is the quantification of the variation of an output indicator because of the variation of an input parameter, given a linear dependence between them. This method complements the MC approach since it makes possible, through the sensitivity coefficients, the quantification of the most important sources of uncertainty for a particular output parameter. The results from this methodology have also been included here for comparison purposes. It has to be considered as well that the temporal parameters have been included in the calculations but not in the results of SA.

Results

The uncertainty results for the selected output parameters obtained with the MC method are summarised in Tables 1 and 2 for Scenario 1 and 2 respectively. The results obtained with the sensitivity method are also included in the tables. In the case of the MC methodology, the best-estimate value has been taken as the mean value of all samples’ output, while for SA this value is the result of the reference scenario. Also, in the following figures, the histograms from the MC calculations showing the uncertainty in terms of frequency are displayed. Three different probability distributions (uniform, Gaussian and asymmetric) have been found depending on the relationship between the randomised input parameters and the selected output indicator, as will be explained later. For the sake of convenience, a Gaussian fit has been drawn in all the histograms so deviations from this distribution can be easily identified.

Depending on the results of the random sampling applied to the input parameters, it is possible to build a ‘broken scenario’, i.e. a scenario for which it is impossible to finish the simulations for the whole period of 200 years. In the two scenarios studied here, this ‘broken scenario’ issue is produced by the lack of MA for the ADS fuel fabrication. Also, broken scenarios will be translated into a loss of statistics.

Scenario 1

Table 1 shows that the results of the reference scenario (sensitivity analysis) and the average value obtained by the Monte Carlo calculations are similar for the selected set of output parameters, although the uncertainties obtained are quite different. From the SA it has been found that the
parameters that can break a scenario are the initial spent fuel inventories, the parameters of the ADS fleet, the burn-up of the PWR reactors and the composition of MA in the ADS fuel. In case that the random sampling of one of these parameters makes its value to exceed a certain limit, then the scenario will break. A region of the phase space formed by the possible sets of values for the input parameters will then be lost as broken scenarios and the uncertainty will be underestimated since it does not consider all the possible cases. This issue is of special importance for the sensitivity analyses. However, for the MC calculations, all the parameters are randomly varied at the same time and the probability of a broken scenario is smaller than for SA since the impact of some input parameters can be compensated by others.

In general, the relative uncertainties are similar to those chosen (arbitrarily) for the input parameters meaning that they are caused by the combination of a few number of input parameters’ uncertainties and the possible correlations (positive or negative) between them (the analysis of the existence and quantification of these possible correlations is foreseen as future work). However, in certain cases like the maximum amount of separated Pu from UOX or of MA from ADS these uncertainties are much larger, which is a result from the complex behaviour produced by the non-linear dependences (in the second output parameter). Future work will be made on this line, distinguishing each individual contribution to the global uncertainty of each parameter.

The results for Scenario 1 are presented in Table 1.

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Monte Carlo</th>
<th>Sensitivity Analyses</th>
</tr>
</thead>
<tbody>
<tr>
<td>Total amount of depleted U [tHM]</td>
<td>$(1.47 \pm 0.07) \times 10^6$</td>
<td>4.8% $(1.5 \pm 0.2) \times 10^6$</td>
</tr>
<tr>
<td>Total amount of gr. B MOX SF reprocessed [tHM]</td>
<td>$(2.09 \pm 0.06) \times 10^4$</td>
<td>2.9% $(2.1 \pm 0.3) \times 10^4$</td>
</tr>
<tr>
<td>Max. amount of separated Pu from gr. B MOX [tHM]</td>
<td>860 ± 50</td>
<td>5.8% 848 ± 2</td>
</tr>
<tr>
<td>Max. amount of separated Pu from gr. B UOX [tHM]</td>
<td>300 ± 80</td>
<td>26.7% 317 ± 2</td>
</tr>
<tr>
<td>Max. amount of separated Pu from gr. A [tHM]</td>
<td>201 ± 7</td>
<td>3.5% 199 ± 4</td>
</tr>
<tr>
<td>Max. amount of separated MA from gr. A &amp; B [tHM]</td>
<td>128 ± 4</td>
<td>3.1% 122.7 ± 1.1</td>
</tr>
<tr>
<td>Max. amount of separated Pu from ADS [tHM]</td>
<td>89 ± 6</td>
<td>6.7% 94 ± 2</td>
</tr>
<tr>
<td>Max. amount of separated MA from ADS [tHM]</td>
<td>16 ± 5</td>
<td>31.2% 16 ± 3</td>
</tr>
</tbody>
</table>

As can be seen in Figure 3, most selected output indicators (excepting Figure 3b and Figure 3f) follow a Gaussian distribution. Given that the number of samples is large enough, the Monte Carlo technique ensures that the final results will be mainly distributed around a central value in the form of a Gaussian curve when the dependence between parameters is linear: the multiple perturbations will cancel each other in the less probable regions and pile up symmetrically around the best-estimate value forming this distribution.

For the MA reprocessed from the ADS an asymmetric shape (Figure 3f) is clearly inferred so non-linear effects appear in this case. The pile up takes place around another value so a certain bias in the shape of the normal distribution appears. However, sometimes the effect of the asymmetry can be
small compared to other uncertainty sources so that the shape of the curve can be approximated to a Gaussian curve as well.

Finally, the uniform distribution (Figure 3.b) indicates that the output parameter has a very strong dependence in only one input. In the case of the total amount of group B MOX SF reprocessed, this is dominated by the initial spent fuel inventories of MOX from France. This assumption can also be validated with the fact that the range of variation of the output parameter is almost equal to the 5% variation of the input parameter.

**Figure 3**: Total Monte Carlo results for Scenario 1

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**Scenario 2**

The use of multi-recycled Pu in the form of MOX-RMA fuel in PWRs favour the generation of MA, and the scenario will be less likely to break. From the SA, it has been found that the only causes of scenario breaking are now the ADS fleet parameters and the weight of MA in the ADS fuel, so a better agreement between the uncertainties obtained with MC and SA are found, as shown in Table 2.
Table 2: Results for Scenario 2

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Monte Carlo</th>
<th>Sensitivity analyses</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Value</td>
<td>Value</td>
</tr>
<tr>
<td>Total amount of depleted U [tHM]</td>
<td>$(1.16 \pm 0.06) \cdot 10^6$</td>
<td>$(1.17 \pm 0.06) \cdot 10^6$</td>
</tr>
<tr>
<td>Total amount of gr. B SF reprocessed [tHM]</td>
<td>$(2.11 \pm 0.04) \cdot 10^5$</td>
<td>$(2.11 \pm 0.07) \cdot 10^5$</td>
</tr>
<tr>
<td>Max. amount of separated Pu from gr. B [tHM]</td>
<td>$450 \pm 40$</td>
<td>$440 \pm 40$</td>
</tr>
<tr>
<td>Max. amount of separated Pu from gr. A [tHM]</td>
<td>$193 \pm 8$</td>
<td>$192 \pm 5$</td>
</tr>
<tr>
<td>Max. amount of separated MA from gr. A &amp; B [tHM]</td>
<td>$131 \pm 4$</td>
<td>$126 \pm 2$</td>
</tr>
<tr>
<td>Max. amount of separated Pu from ADS [tHM]</td>
<td>$73 \pm 8$</td>
<td>$77 \pm 4$</td>
</tr>
<tr>
<td>Max. amount of separated MA from ADS [tHM]</td>
<td>$3 \pm 4$</td>
<td>$2 \pm 3$</td>
</tr>
</tbody>
</table>

Regarding the histograms (Figure 4), a likely Gaussian distribution can be found in most of them even small deviations caused by non-linear effects may occur. As in this scenario the MOX and the UOX fuels are reprocessed together, the uniform distribution obtained for the extracted mass for reprocessed SF in Figure 3.b is lost (see Figure 4.b as an almost Gaussian curve) since more parameters play a fundamental role now (all the initial spent fuel inventories are varying independently). Also, the Pu recovered from reprocess (Figure 4.c) shows an asymmetric shape caused by the irradiation of multi-recycled Pu (providing non-linear effects).

Finally, for the maximum amount of MA recovered from the ADS, a shift towards zero appears on the distribution compared with the results of Scenario 1. This is produced because of the scenario definition. In this case, a fewer amount of ADS are decommissioned. When the last cores of these ADS are reprocessed, a certain amount of MAs are generated (that ultimately provides the value of the maximum amount of MA separated). However, the ADS fleet is sufficiently large so that all the MAs can be used for ADS fuel fabrication in the same year, providing the final value of zero for the total amount of separated MA from ADS. Although the uncertainty in this magnitude is similar to that obtained for Scenario 1 (4 tHM instead of 5 tHM), the best-estimate value is so small that the relative uncertainty becomes very large (133%).
Conclusions

This work includes the analysis of the uncertainty propagation in the fuel cycle scenario parameters using the Monte Carlo methodology. This method allows estimating the propagation of the uncertainties from the input parameters to the output ones, no matter the kind of dependence between them (linear or not). On the contrary, the sensitivity analysis, in spite of being a first order approximation, provides the information of which input parameter is more relevant, which will be used in future works.

This methodology has been applied to two PATEROS-based fuel cycle scenarios, where the main objective is the reduction of the spent fuel inventories by the partitioning and transmutation strategy.

By means of the MC methodology, the uncertainty of a selected group of output parameters has been obtained, in terms of histograms of frequency. Three different curves have been obtained for these histograms depending on the relation between the input and the output parameters:

- Gaussian distribution: The multiple perturbations will cancel each other in the less probable regions and pile up symmetrically around the best-estimate value when the dependence between parameters is linear. The detection and quantification of the possible correlations between parameters will be carried out in future works.
Uniform distribution: A linear dependence is also found in this case, and the output parameter has a very strong dependence in only one input parameter.

Asymmetric distribution: The pile up takes place around a value different to the best-estimate value so a certain bias in the shape of the normal distribution appears. It is characteristic of a non-linear dependence.

The uncertainties calculated by both methodologies have been obtained, although the sensitivity results should be carefully considered since they are based on a first order approximation theory and some non-linear behaviours have been found (with the MC analysis). Additionally, the existence of broken scenarios may cause an underestimation of the uncertainty calculated with this methodology, since a certain region of the phase space would not be considered.

Acknowledgements

This work has been partially supported by ENRESA in the frame of the CIEMAT-ENRESA collaboration on transmutation applied to high-level waste.

References

2. F. Álvarez-Velarde et al. (2010), “TR_EVOL, upgrading of EVOLCODE2 for transition scenario studies”, First Workshop on Technology and Components of the ADS (TCADS-1) OECD/NEA, Karlsruhe, Germany.
Analysis of transition to fast reactor U/Pu continuous recycle

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Abstract
This paper contains an assessment of the key considerations that affect performance and challenges of transition from the current US nuclear energy system to a future nuclear energy system in which U/Pu is continuously recycled in a fleet of fast reactors (FRs) and enriched uranium is no longer needed (referred to as “EG23”). The key considerations with the highest impacts on transition behaviour were identified as: 1) fast reactor startup pathways (i.e. source of fissile materials for deployment of first fast reactors), 2) the scale of the future energy system (i.e. the average rate of growth and eventual size of the fleet), and 3) the recycle time (i.e. total time from reactor discharge to recharge as fresh fuel). There are also many other considerations of secondary importance such as fuel cycle technology choices (reactor, fuel fabrication, recycling, etc.), and various system issues and constraints (technological, material, systematic, economic, and policy-related). These considerations and their impacts are briefly described. Lastly, a small number of specific transition scenarios were analysed to demonstrate the potentially large quantitative impacts of key decisions and to develop an understanding of transition under a broad range of scenarios that are expected if EG23 were implemented.

Some of the key findings from this work are: 1) For all scenarios envisioned, the fast reactor technology is needed as soon as possible in order to maximise the benefits of transition; 2) Life extension that keeps existing light water reactors (LWRs) operational until after the fast reactor technology is available will lead to better transition performance; 3) The use of LEU to fuel the fast reactors during initial deployment allows decoupling the development, demonstration, and deployment of the recycle technologies from the deployment of the fast reactors; 4) Shorter recycle time for the fast reactor fuel makes the fissile material produced in the fast reactors available sooner and reduces material held up in processes outside of the reactor. This allows for a much more rapid expansion of the fast reactor fleet before Pu availability constrains the transition; 5). For Pu constrained transitions, using LEU in FRs to support transition may result in lower cumulative front-end requirements during transition including less natural uranium and enrichment required.

Introduction

As identified in a recent evaluation and screening (E&S) study \cite{1} of a comprehensive set of technology-neutral nuclear fuel cycle options, a fuel cycle in which U/Pu is continuously recycled in fast critical reactors with only natural uranium make-up (designated as “EG23”) is one of the most promising since it offers substantial improvements in almost all of the established benefit
performance metrics, including resource utilisation and waste management, compared to the current US fuel cycle. Although the E&S results considered transition to such fuel cycles in terms of high-level challenge metrics (e.g. development time, compatibility with existing infrastructure, etc.), the quantitative results were mainly assessed based on fully developed and deployed (i.e. steady-state) fleets. Therefore, in order to assess the performance metrics of the fuel cycle system before it reaches steady state, a follow-on effort was undertaken to determine the time-dependent behaviour for example scenarios of transition to EG23. This paper highlights the findings from this spin-off effort that is based on the E&S results and assumptions, with the main goal of identifying the major considerations (scenario assumptions or strategic choices) and quantifying their impacts on transition.

It must be emphasised that this effort was not intended to identify a specific transition scenario that is preferred, but to inform on the broad range of likely transition scenarios to the EG23 system. A “better” transition will ultimately be based on the value judgements of decision makers regarding the trade-offs in timing, benefits, and challenges identified in this study, as well as other priorities outside of the scope of this effort. The paper describes these trade-offs as a function of the key considerations for transition (fast reactor startup pathways, scale of energy system and growth rate, and the recycle time). For each of these key considerations and under different future conditions, the important factors, impacts on performance, differences in challenges, and any other information of potential interest to decision makers were identified and assessed with the intent to provide information highlighting the key research, development and deployment needs/options, key decision points, and major factors that affect transition.

The primary motivation and challenges of transition to EG23 were identified by the nuclear fuel cycle evaluation and screening (E&S) report [ES-1]. These motivating factors are the result of performance improvements relative to the current fuel cycle. The E&S also identified challenges to deploying this alternative which are potential impediments to the deployment of EG23. The benefits of transition to EG23 include: reduced mass and improved properties of spent nuclear fuel (SNF) and high-level waste (HLW), reduced mass of depleted uranium, reduced mass of natural uranium required, and other environmental impacts (e.g. less CO$_2$, less land use) which are generally tied to reductions in natural uranium required. There are many areas in which challenges were identified. These challenges were considered in two broad categories in this study: technology development (issues related cost, timing, complexity of integration, and level of technology challenge) and transition economics (issues related largely to underutilisation and/or reduced scale of facilities). The assumption is that if they are important for the end state, then they should be important for consideration during the analysis of the transition to that end state. There are also additional transition-specific considerations, such as the transitional material inventories in storage, which may be important to decision makers. These factors form the basis for informing on the primary impacts, issues, challenges, trade-offs, etc. for transition to EG23.

Additionally, this study does not consider any of the other many end-state possibilities. The assumptions are that EG23 will be the future fuel cycle, EG23 will be deployed as soon as practical, and transition to and operation of EG23 will continue uninterrupted for centuries into the future. Benefits related to supporting other alternative fuel cycles, mitigating challenges, major system disruptions, etc., are not considered beyond some brief discussion of the overlap in technology development needed for other alternative fuel cycle options. These technology development overlaps are significant and may allow postponing many key decisions regarding the exact technology (i.e. specific reactor and reprocessing plant designs) until these technologies are more mature.
Key considerations

Fast reactor startup pathways

The first key consideration for transition to EG23 is the fast reactor (FR) startup pathway. The FR startup pathway is defined by the source of the fissile material used to initially deploy the FR fleet before the fleet is self-sustaining by recycling its own fuel. Initial deployment is the first few commercial FRs and supporting facilities. After successful deployment, the FR fleet will go through a period of rapid (above the long-term average) growth rate as the FR fleet expands from just a few reactors to 100 or more FRs. During this build out phase, an external fissile source (not from recycled FR fuel) will be required to supplement what is available from the FRs. Over the course of the build out, the fraction that comes from the external source will decrease and by the end of transition, the external source will no longer be required because the fleet has become self-sufficient, i.e. all new fissile material required will be produced by the FR fleet.

In this study, several likely pathways were assessed and at least one scenario for each pathway was modelled using fuel cycle simulation codes such as DYMOND [2], ORION [3], and VISION [4] to compare their different impacts on the duration and performance during transition. These codes required technology-specific data for the reactors, so the sodium-cooled fast reactor (SFR) was modelled as representative of FRs in the simulations. Only the two main pathways are discussed in this paper. The first pathway is the “base case” pathway which uses only Pu and U recovered from light water reactor (LWR) used nuclear fuel (UNF), which is referred to as “LWR RU/Pu”, to start up the first FRs. This was the startup pathway assumed in the example scenario of transition to EG23 in which a 1% annual growth rate in nuclear power must be sustained during and after the transition while only recycling UNF generated after 2015. The electricity generation profile and annual reactor capacity started up are shown in Figures 1 and 2, respectively. The time from fuel discharge until its recovered U/Pu is reloaded into another core (the recycle time) is 7 years. This pathway runs into SFR fuel shortages if SFRs were the only reactors built after 2050, so LWRs using LEU fuel need to continue to be deployed after 2050, thereby extending the transition period; transition was not considered complete until the entire system consisted of fast reactors, as defined by the EG23 system. Under different (lower) growth rates, different technology assumptions (e.g. using FRs with lower Pu requirements and higher production rates or shorter recycle times), etc. these shortages may not occur, hence, this scenario must only be considered as an “example scenario”.

The second pathway is the same as the base case except that it uses low-enriched uranium (LEU) during initial deployment and SFRs were assumed to have the flexibility to switch fuel types during refuelling outages. The LWR RU/Pu is utilised during the build out with any shortfall in fissile material being made up by loading SFRs with LEU. This pathway is designated as the “LEU Support” pathway, and the electricity generation profile is shown in Figure 3. Notice how the last LWR, built at around 2050, retires at around 2130 instead of 2160 as in the base case. This results in completing transition 30 years earlier and significant savings in cumulative front-end requirements, as shown in Figure 4, since the energy generated by LWRs and LEU (blue area in Figure 3) is dramatically reduced.
The LEU Support pathway provides significant performance benefits for transitions in which there are significant Pu availability constraints, largely because of the ability to switch to using recycled fuel when the system becomes self-sustainable. Otherwise, as in the base case, LWRs using LEU would have to be built and operate for 80 years before they are retired, even if the system can already be self-sustaining on U/Pu. However, if LWRs were to have a similar ability to switch from
LEU to MOX fuels, then similar benefits could result, but the presence of LWRs burning U/Pu fuel along with FRs producing U/Pu renders the system into another fuel cycle category or evaluation group, which is EG29 from the E&S. However, having an intermediary stage of EG29 during the transition from the current cycle to EG23 is another possible pathway, which was not studied. The main challenges of the LEU support pathway are the rapid, but temporary rise in annual demand in enrichment and mining requirements due to the much higher enrichment (up to 20 wt%) required for LEU fuel in fast reactors. Figure 5 shows the annual SWU requirements; the annual U mining rate shows a similar trend. This may represent practical and economic challenges to meet this rapid, temporary rise. FR LEU fuel fabrication facilities will need to be started up and may only be operated for a short period of time and at relatively low capacity factors. These two example pathways demonstrate how important and impactful these decisions are to transition performance, as well as any practical constraints. For example, it may be difficult to achieve the rapid build rate of the enrichment facilities required to handle the maximum throughputs shown in Figure 5.

Figure 4: Comparison of cumulative NU requirements for base case and LEU support case

Figure 5: Comparison of annual SWU requirements for base case and LEU support case

Scale of energy system and growth rate
The future demand for nuclear energy is usually modelled as a constraint on the system. However, there are large uncertainties in how much nuclear energy will be deployed over time far into the future. Depending on the assumption of how much nuclear energy is desirable, the design of the system may look very different. As the amount of nuclear energy produced in the future increases, so
do the benefits from transitioning. Also the faster nuclear generation grows, the more challenging transition becomes. These are two competing effects. The investment in technology development and initial deployment is relatively insensitive to the future energy generation. This makes the decisions about what research is needed and the value of that research highly sensitive to the assumed future nuclear energy generation. At some extreme low value, the future benefits would not be sufficiently large to justify investment. At higher levels, there are far more benefits to justify the investment. At still higher levels on energy generation, the current system may not be sustainable, which results in the transition being necessary.

There are a number of ongoing efforts within the US Department of Energy that make predictions about future energy demand under a range of different assumptions about the future. These predictions are not intended to give exact predictions, but general trends that would be anticipated if the assumed conditions were to hold. The Annual Energy Outlook [5] published by the US Energy Information Administration (EIA) puts the growth over the next 25 years at a low of near 0% to a high of around 0.6% per year. These are more “business as usual” scenarios. For the range of scenarios considered by the EIA, there are no significant carbon reductions in any of the scenarios. These seem likely to represent the low and mid-range of likely scenarios. There are a wide range of longer-term analyses that look at carbon reduction scenarios. These are very sensitive to the assumption of the relative cost of alternative low-carbon technologies and the level of reduction targeted. These generally predict nuclear growth at slightly over or greatly over 1% per year. Figure 6 shows the relative difference in scale of this broad range of possible future energy systems.

**Figure 6:** Schematic of range of future energy growth scenarios

Three different scenarios are considered as far as the future nuclear energy system. The low range assumes replacement only, which means 0% growth in energy generated (slightly below the low EIA estimate) with a future energy system that is around 100 GWe of capacity deployed in about a century. The mid-range is roughly a constant market share of electricity with an average annual growth rate of around 1% per year growth in energy generated (a little above the upper EIA estimate) resulting in approximately 300 GWe in about a century. The high-range is for a low-carbon future with greater nuclear contribution to overall energy. This will require growth rates of well above 1% per year sustained for a significant period and an eventual nuclear energy system that is likely in excess of 500 GWe-equivalent, and possibly over 1 TWe-equivalent within a century.
Recycle time

The average inventory required to deploy additional capacity consists of the initial core load prior to the start of operation and then, on average, all fuel reloads until the 1st batch of discharge fuel is recycled back into a reactor. To better explain this, Figure 7 shows an idealised schematic of a core that is assumed to discharge 1/4 of the fuel each cycle (4-batch) with an average recycle time equivalent to 5 cycle lengths. The startup inventory in this example is 4x for the initial core load, where 1x is the average batch size. Then, 5 additional batches of fuel must be set aside to load the reactor until the first batch discharged is cooled, separated, re-fabricated, and loaded into the core again (which takes a total time equivalent to 5 cycle lengths). From then on, no additional fissile material is needed to sustain operation in the idealised model. Therefore, for this idealised example, all new capacity will require 9x the average batch size.

If the recycle time were reduced from an average of 5 cycles to an average of 2 cycles, the total inventory would drop from 9x (4x in core and 5x outside the core) to 6x (4x in the core and 2x outside the core). Reducing the needed startup inventory from 9x to 6x would allow for the deployment of 50% more capacity for a given quantity of fissile material. This shows how sensitive the startup inventory is to recycle time. However, it should be noted that there are a number of technical reasons why the cooling time has typically been longer, e.g. centralised location of reprocessing plant, shielding, heat loads during transport, limitations on radiation exposure and heat for chemicals involved in reprocessing, etc.

There are other factors that will impact the startup inventory, but this in general is the one that is most sensitive to technology choices and system configuration. The base case assumes a 7-year recycle time (~5 cycles) typical for off-site recycle of LWR UOX. On-site recycle is more typically assumed to have a recycle time of around 2-3 years (~1.5 to 2 cycles). There are many approaches that can shorten or lengthen the recycle time. These require a detailed look at the specific technologies and the specific constraints, costs, and tradeoffs that come from varying the recycle time. This level of detail is beyond the scope of this work, but the impact on material availability and its impact on system performance are considered because it is a key variant for all scenarios. Any combination of technologies that enable the reduced recycle time of the fast reactor fuel will see the benefits of making the fissile material produced in the fast reactors available sooner.

Shortening the recycle time makes the fissile material produced in the fast reactors available sooner which allow a more rapid build out of the fast reactor fleet by simultaneously allowing more capacity to be deployed from the available inventory of LWR UNF and by increasing the rate at which the fast reactor fleet can fuel new capacity. For high sustain growth rate scenarios (>~1.5%), it will be necessary to reduce the cooling time below 7 years to sustain the expansion after the technology transition is complete. The amount of fissile material required to sustain the operation of a FR is the total amount of material that is in the reactor as well as all material that is in storage or held up in processes outside of the reactor including decay storage, separations, fabrication, transportation, etc. This total time from reactor discharge to recharge as fresh fuel is referred to as the recycle time. The use of technologies or deployment of those technologies in a way that reduces recycle time will significantly reduce the fissile material required to start up and sustain the operation of a FR. However, there are no significant benefits from reduced recycling time of the LWR UNF, since there is already a large inventory of LWR UNF that exists when the FR deployment begins.
Secondary considerations

There is a broad set of other secondary consideration that will have impacts on transition. For most of these considerations, what is beneficial for the technology performance is also beneficial for transition performance, e.g. greater thermal efficiency and higher average discharge burnups for the FR will both improve the economics of the FR and make transition more efficient. For most of the potentially important secondary considerations, the technology and transition performance are aligned. The notable exception is LWR burnup. Higher burnup is better for LWR performance but will result in less Pu available in the LWR UNF inventory. The amount of Pu produced per unit of energy produced by the LWRs is reduced by 20% if average discharge burnups are increased from 50 MWd/kg to 75 MWd/kg which may reduce transition performance for Pu constrained transitions.

Technology choices

The level of detail of this analysis does not provide insight into the differences in transition performance for specific technologies (e.g. sodium vs. lead FR or oxide vs metallic fuel). While there are certainly important trade-offs to be considered, this requires a level of design detail and understanding that is not currently available with respect to a fully developed commercial fleet. As long as the technology is able to perform the necessary function and can do that when needed, this analysis will not identify any differences in performance between different technologies.

A range of functions were identified for the different pathways. For the technology functions that are part of a particular scenario, the technology must be taken from its current state of development through commercial deployment on the time frame necessary for a particular scenario. EG23 prescribes a certain set of functions that must be available once transition is complete. Each specific transition scenario includes descriptions of any additional technologies that are required to facilitate transition (e.g. LWR UNF reprocessing) and the timing and scale of all technologies over the transition. The availability of technology is a key constraint on the system. Different scenarios will have different constraints and different challenges associated with deploying the technologies in the time that they are needed.

For all scenarios envisioned, the fast reactor technology is needed as soon as possible in order to maximise the benefits of transition. Most of the benefits of transition are realised by minimising the energy produced by LWRs using LEU fuel and maximising the energy from FRs using Pu-based
fuels. In certain scenarios, this may be achieved by fuelling the FRs with LEU when there is insufficient Pu available. As long as the overall effect is a reduction in the cumulative mining and enrichment requirements for LEU fuel in both LWRs and SFRs, then that transition strategy is likely more beneficial.

LWRs are deployed as needed to meet energy demand when FRs cannot because the FRs are either not yet technologically available or there is insufficient fissile material to fuel them. This results in the lifetime of the existing LWRs and the technological availability of FRs to be intertwined. Existing LWRs shut down after FRs are available may be replaced by FRs, but those shut down before the introduction of FRs must be replaced by new LWRs that are assumed to operate for another 80 years, thereby increasing the cumulative amount of energy generated with LEU fuel and delaying the end of transition. The more of the existing LWRs that remain operational until after the FRs is available, the more benefits that will result from transition. Life extension that keeps existing LWRs operational until after the fast reactor technology is available will lead to better transition performance.

The use of LEU fuel for the FRs is assumed to be the least technologically constrained approach for initial deployment of the FRs. While this technology will still require development, deployment, and demonstration, this is thought to be the easier path compared to using recycled fuel and possibly even the necessary path for initial deployment based on past experience and the relative challenges of producing qualified fuel in the time and quantities necessary for initial deployment. This also has another significant advantage in overcoming the technology challenges by temporarily circumventing recycling technology challenges. The LWR RU/Pu pathway requires well-integrated technology development of the FR, LWR UNF separations, and RU/Pu fuel fabrication. The development, demonstration, and deployment of all three of these major technologies are on or close to the critical path.

The use of LEU to fuel the fast reactors during initial deployment allows decoupling of the development, demonstration, and deployment of the recycle technologies from the deployment of the fast reactors. By decoupling the recycle technologies from the deployment of the FRs, significant investments in these technologies can be delayed until after the FR technology is demonstrated. This avoids the need to fund multiple demonstration projects in the same time frame.

Other secondary considerations

There are many other potential constraints (e.g. economic considerations), but many of them are not clearly defined at this time. Better understanding of practical limits and the constraints that may be imposed needs to be developed because these may have significant impact on transition. For example, are there any practical limits on the annual expenditures for technology development and demonstration? The cost of facilities during initial deployment, the costs as a function of facility size, and the cost of processing materials over a wide range of properties are all unknowns that are essential factors for developing an economically efficient transition. The tools exist to evaluate the impacts, but the appropriate assumptions and/or necessary data do not currently exist.

This study provided insight on EG23 based on the assumptions that it will be the end state and a smooth transition will occur. This may need to be considered within a broader context as part of an integrated system that includes other possible end states, risk management, and other national priorities. Even if EG23 is the ultimate end state, the decisions will have to include all of these and other considerations which were beyond the scope of this analysis.
Conclusions

The biggest factor that likely affects the performance of transition to EG23 is the ability to deploy fast reactors as quickly as practical. The development and demonstration of a fast reactor technology should proceed promptly since it is the critical path technology for the transition to EG23.

If LWR RU/Pu is going to be used initially in the transition, the material recovery and fuel and fuel fabrication technologies to utilise the LWR UNF need to proceed in parallel with the fast reactor technology development and demonstration since all three technologies must be demonstrated on the same time scale.

If the decision to use LEU to fuel the fast reactors during initial deployment is made, the development and demonstration of the separation and RU/Pu fuel and fabrication technologies is no longer in the initial critical path and provides a lot of flexibility in the overall technology development and demonstration.

Enabling the existing LWRs to operate until after it is possible to replace them with fast reactors, even LEU fuel fast reactors, will offer significant benefit to the transition to EG23. For very low or zero growth scenarios, it is essentially impossible to transition to EG23 in a reasonable time period if a large fraction of the existing fleet of LWRs is replaced by new LWRs.

The scale of the future nuclear energy system is a key factor in the issues, challenges, requirements, etc. for transition. Very different conclusions about the value of transition and the technologies required are likely to result over the span of likely nuclear futures. The larger the future nuclear energy system, the more valuable transition becomes. However, it also becomes more constrained in terms of having the material to build out the fleet of fast reactors. The investment and technology challenges are, to a first order, independent of the size of the future energy system.

The base case assumed that LWR RU/Pu is the transition pathway. This works well until the growth rate reaches a value for which growth becomes material-constrained. At that point, either shorter recycle times and/or LEU support becomes necessary. Both have their pros and cons.

There are a lot of complex interactions and important factors that need to be understood during the early part of the transition and build out of the fast reactor fleet. Many of these are driven by the complex economic relationship, funding issues, and related factors that occur during the demonstration and initial deployment.

Lastly, there is not a single definition of what makes a “good” transition. This study has only discussed the differences that result from different considerations. Some approaches to performing quantitative comparisons based on defined sets of potential value judgements and the resulting weight factors and functions would be challenging, but is plausible. It is a capability that would seem to be of eventual interest to decision makers in order to explore the vast design space that exists for transition not just to EG23, but all other options as well.

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References


Progressive deployment of plutonium multi-recycling in SFRs in the French nuclear power fleet feasibility analysis

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Abstract

Since 2013 the CEA, together with its industrial partners EDF and AREVA, has undertaken a programme of scenario studies concerning the future of the French nuclear reactor fleet and the integration of fast reactors into this fleet. Indeed, the sodium fast reactor technology seems to be the most logical solution for using the plutonium contained in MOX fuels (currently not used) and more generally for ensuring the recurrent recycling of recoverable materials.

Rather than suddenly switching over to the large-scale deployment of fast reactors, it is deemed preferable to ensure the progressive implementation of this technology through successive phases: each phase corresponds to a specific technical objective.

- Phase A: This phase falls in line with the current French situation which involves recycling U and Pu in PWR ERU and MOX fuels. Spent MOX and ERU fuels are placed in interim storage. This phase allows the spent UOX inventory to stabilise.

- Phase B: This involves launching the industrial operation of a limited number of SFRs and starting the industrial-scale reprocessing of PWR-MOX fuel. This phase allows the stabilisation of the spent MOX inventory. It also provides an outlet for the plutonium produced by the current PWR fleet. However, this phase does not provide an outlet for the plutonium contained in spent SFR fuels.

- Phase C: This phase involves deploying a symbiotic fleet comprising PWR UOX, PWR MOX and SFRs so it is possible to multi-recycle the plutonium and to stabilise its inventory. This phase is characterised by the industrial reprocessing of spent SFR fuels and the recycling of Pu in SFRs and PWR-MOX reactors. SFRs provide an outlet for Pu contents extracted from spent MOX fuel while enabling isotopic correction to recycle Pu in PWR-MOX reactors.

- Phase D: The aim of this phase is to deploy a fleet of reactors that no longer consumes natural uranium. It may involve the full deployment of SFRs or their partial deployment in a symbiotic fleet comprising breeder SFRs and PWR MOX. This fleet is based on the assumption of equilibrium between the production and the consumption of Pu, consistent with a constant installed capacity.

The criteria used to analyse these scenarios should consider technical aspects and requirements for potential industrialisation, for example, inventories and characterisation of materials and waste, impact on the waste repository (footprint), impact on the industrial facilities (fabrication plant, reprocessing plant, interim storage), transportation needs and economic impact. The impact of americium transmutation loaded in the SFR reactor is also studied.

This paper presents an overview of the main outcomes of this study and gives some perspectives for the future programme. A part of these results are reported in the “Dossier 2015” (Report 2015) written by CEA within the framework of the 2006 Waste Management French Act.
Comparison of transition to U/Pu and U/TRU recycle in fast reactors

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Abstract

This study compares the features of transition from a current fuel cycle state to a self-sustaining fuel cycle using fast reactors that will either recycle uranium and plutonium (discard the minor actinides) or all uranium and transuranic elements. The US currently has no commercial nuclear fuel recycling facilities, but has a fleet of LWRs producing about 2 000 tonnes of used nuclear fuel (UNF) per year which depending on assumptions about plant life extensions will be retired over the next 20 to 40 years. The demand for nuclear energy in the future is uncertain with estimates that range from no growth (replacing existing capacity) to low-carbon scenarios that would result in a fleet many times larger than currently exists.

Much of transition is driven by getting technologies to a commercially deployable level. Another key driver is the performance aspects that are most important to the decision makers.

Recycle of the uranium and plutonium is a primary driver in achieving significant natural resource benefits. There are also potentially significant waste management benefits that result from disposing HLW consisting of primarily the fission products and minor actinides instead of UNF. Extending recycle to include the minor actinides is primarily driven by the potential for significant additional waste management benefits with only marginal improvements in resource utilisation. The study does not compare the merits of these benefits, but assumes that if the decision is made to transition to a particular fuel cycle option, these are the motivating factors. The goal of transition would be to maximise these potential benefits while minimising other issues and challenges.

The largest difference in transition to either of the two options is the relative challenge of recycle of U/Pu versus U/TRU (or U/Pu/MA). Significantly less time and investment may be required for the U/Pu system. If true, this could allow for a more rapid transition to the U/Pu system.

The U/TRU system can be deployed either homogeneously (all elements recycled together) or heterogeneously (MA recycled in special targets). There is a potential for the system to transition more quickly if initially only U/Pu fuel is used and the introduction of U/TRU or MA-bearing targets is delayed. This offers some interesting tradeoffs that are unique to transition. These and other related issues are analysed for the two transition alternatives.

Introduction

There are many fuel cycle options that when fully implemented will provide significant benefits over the once-through fuel cycle utilising existing technology. This number is particular large when the
different technology options are considered (e.g. sodium-cooled fast reactors versus lead-cooled fast reactors, aqueous versus electrochemical separation, and metallic versus oxide fuel). In order to achieve those benefits, it is necessary to transform the entire US nuclear energy system as it exists today to that eventual alternative system. During that transition, the system must satisfy all constraints imposed at all times. These constraints include funding for research and development (R&D), future demand for nuclear energy, constraints on shipping materials, and a wide range of technical and non-technical constraints. Transition analysis must inform on these broad and uncertain futures to inform decision makers today so they can make informed decisions and balance the costs and risks associated with their decisions.

This paper focuses on the transition to an alternative fuel cycle option based on continuous recycle of uranium/transuranic elements (U/TRU) in fast reactors without the use of uranium enrichment. This alternative fuel cycle option was identified as a “most promising” option by the evaluation and screening study (E&S) where it is identified as Evaluation Group 24 (EG24) [1]. The E&S identified a number of positive attributes relative to the performance of a once-through thermal reactor operation with uranium enrichment (referred to as EG01 in the E&S) which is representative of the performance of the current fleet of LWRs operating in the US if the LWR is directly disposed. The E&S did not look at the performance using specific technologies, but only the performance that is achievable based on the physics of these types of systems at steady-state conditions and the challenges of developing and deploying technologies that would achieve the expected performance without considering specific transition scenarios.

This paper is an extension of the earlier work on transition to U/Pu recycle with disposal of the minor actinides (MA) as HLW referred to as “EG23” [2]. The analysis of EG24 built on the knowledge developed in that analysis of EG23. The comparison with the results for EG23 allows for informing on the impact that recycle of the MA has on transition, since that is the only difference between these two alternatives. Those differences are described here.

**Key considerations**

**Motivation and challenge**

A key part of the transition analysis is to understand what motivates the transition to this alternative fuel cycle and what minimum set of challenges must be overcome. There will not be one single factor to be considered in either case. It will be a range of factors that will be of varying importance between different decision makers. The motivation is the areas where there is the potential for significant improvement. The challenges are those things that must be overcome to achieve those improvements.

The potential motivating factors (performance metrics) for transition from the current system to the alternative fuel cycle option were identified in the E&S [1] based on the differences between EG01 (representative of the current fuel cycle) and EG24 (representative of the end state of the transition being analysed). They are the same factors that motivate transition to EG23. These same motivating factors (metrics analysed by the E&S) are assumed to be equally valid during transition. If a decision maker is motivated to transition to EG24 in order to improve the spent nuclear fuel (SNF) and high-level waste (HLW) performance (i.e. less mass, better properties and no SNF disposed), then that decision maker is likely to consider that same factor when measuring the effectiveness of transition. If it is better to have less waste produced by the end state system, then a transition alternative that produces less waste during transition might be considered better than alternatives that produce more. This is significantly more complex for the dynamic system during transition than
for the steady-state system because differences in timing are also likely to be considered important (i.e. benefits that occur sooner are considered more valuable than the same benefits accruing later).

A summary of the motivation for transition to EG23 or to EG24 is provided in Table 1. The potential additional benefits of EG24 relative to EG23 were discussed and considered by the E&S as well and considered in greater detail by Heidet [3]. These additional potential benefits result from transmutation of the MA component of the TRU in the UNF in addition the Pu component. The MA component of the TRU in the LWR UNF is about 10% of the TRU and significantly smaller fractions of the TRU in the discharged fast reactor UNF at steady state (varies around a few percent depending on the specific design when only MA is sent to the HLW stream).

<table>
<thead>
<tr>
<th>E&amp;S metric</th>
<th>Benefits of transition to EG23 or EG24</th>
<th>Additional benefits of transition to EG24</th>
</tr>
</thead>
<tbody>
<tr>
<td>Mass of SNF&amp;HLW</td>
<td>Yes</td>
<td>No (very small)</td>
</tr>
<tr>
<td>Activity of HLW*</td>
<td>Yes</td>
<td>No (small)</td>
</tr>
<tr>
<td>Radiotoxicity of HLW*</td>
<td>Yes</td>
<td>Yes</td>
</tr>
<tr>
<td>Decay Heat of HLW*</td>
<td>Yes</td>
<td>Yes</td>
</tr>
<tr>
<td>Natural Uranium Required</td>
<td>Yes</td>
<td>No (very small)</td>
</tr>
<tr>
<td>Environmental</td>
<td>Yes</td>
<td>No (except for possible repository benefits)</td>
</tr>
</tbody>
</table>

The benefits in mass of material (HM & FP) sent to the HLW stream are small. The difference in natural uranium required will really be determined by the speed at which the system can transition, with the difference at steady state being quite small. The only potentially significant performance difference results from impact on the properties of the HLW that result from removal of the MA from the waste stream. Specifically, these are decay heat and radiotoxicity of the HLW after the period dominated by fission products. The benefits will be larger for the LWR UNF because of the higher concentration of MA, which will make the benefits a little larger than is implied from the steady state, but not sufficient to lead to different conclusions at the current level of understanding of the importance of these benefits to the HLW system.

The E&S also identified challenges in deploying the alternative fuel cycle. The challenges of deploying EG24 identified by the E&S are: 1) development cost and time; 2) deployment cost prototypic validation to FOAK commercial; 3) compatibility with the existing infrastructure; 4) existence of regulations / familiarity with licensing; 5) existence of market incentives and/or barriers; and 6) financial and economic risk.

The first four apply to the transition period and are the focus of this report. Once commercially deployed, no more technology development is required, but a continual improvement of performance would be expected. After transition, the system will be deployed beyond FOAK, the infrastructure will have transitioned, and the regulations and licensing will exist. Resolution of these challenges will have occurred during transition. The remaining two are challenges that may exist both during transition and could remain after a steady state is reached. The market barriers and financial and economic risk will certainly be far more challenging during transition, but may persist in steady state.

A summary of the challenges of transition to EG23 or EG24 are provided in Table 2. If the MA are kept out of the waste and therefore incorporated in the recycle stream, there are potentially
significant additional challenges in almost every area being considered. They stem from two aspects. The first is that technologies for recycle of MA are not as well developed as the technologies for recycle of plutonium. This will represents greater technology risk and requires more time and resources to advance them to the point of commercial deployment. The second is that recycle of MA will result in handling materials that will have much higher decay heat and radiation emissions which will require remote operations and other design or technology alternatives that are likely to increase operational challenges (e.g. remote maintenance) even after the technologies are fully developed.

Table 2: Challenges for transition to EG23 or EG24 from EG01

<table>
<thead>
<tr>
<th>E&amp;S metric</th>
<th>Challenge of transition to EG23 or EG24</th>
<th>Additional challenge of transition to EG24</th>
</tr>
</thead>
<tbody>
<tr>
<td>Development Cost</td>
<td>Yes</td>
<td>Yes</td>
</tr>
<tr>
<td>Development Time</td>
<td>Yes</td>
<td>Yes</td>
</tr>
<tr>
<td>Deployment Cost</td>
<td>Yes</td>
<td>No</td>
</tr>
<tr>
<td>Compatibility</td>
<td>Yes</td>
<td>No</td>
</tr>
<tr>
<td>Regulations</td>
<td>Yes</td>
<td>Yes</td>
</tr>
<tr>
<td>Market Incentive</td>
<td>Yes</td>
<td>Yes</td>
</tr>
<tr>
<td>Capital At Risk</td>
<td>Yes</td>
<td>No</td>
</tr>
<tr>
<td>Levelised Cost At Equilibrium</td>
<td>Yes</td>
<td>Yes</td>
</tr>
</tbody>
</table>

The fundamental tradeoff at steady state is whether it is better to handle these high decay heat/radiologically challenging elements in the HLW stream or the recycle streams because they represent a challenge in whichever stream they are ultimately sent. The benefit of removing them from the HLW provides a challenge to the recycle system. This will ultimately determine whether transition to EG23 (handle the MA in the waste stream) or EG24 (handle the MA in the recycle stream) is the preferred approach between these two alternatives. By the purpose of this study, the assumption is that they are recycled and then to assess the impact of transition on the increased challenges that exist.

To be clear, additional challenge does not mean that it will not do better ultimately. It only means that there is a larger hurdle to overcome for successful deployment. For example, consider the levelised cost at equilibrium. The cost of recycle of U/Pu using aqueous separation and glove-box fabrication technologies is more advanced with greater experience. A likely technology of electrochemical separation and remote metal fuel fabrication is less advanced so there is greater uncertainty. Given that they are very different technologies, while today’s understanding looks as if they are likely to be more expensive, there is more than enough uncertainty that it may ultimately prove more economical to utilise the electrochemical technology. This could be particularly true for high growth rate scenarios where the system is tightly constrained by Pu making the Pu far more valuable (expensive) resulting in the ability to do onsite recycle with very short recycle times a much more economically favoured approach. The purpose here is only to show where there is additional challenge and not imply what the performance of future technologies will be or the economic conditions at that time. It is overcoming this additional challenge that is the largest difference in transition to the U/Pu and the U/TRU system. While this focuses on single-stage system, the same would be true for two-stage systems. It is the tradeoff between greater challenge of recycle of the MA and the benefit of recycling that MA. In the next section, a range of startup pathways are described many of which are explicit tradeoffs of more MA in the waste earlier in order to allow more time to overcome the challenge versus more benefits by overcoming more challenge sooner.
Fast reactor startup pathways

The first key consideration for transition to EG24 is the fast reactor (FR) startup pathway. The FR startup pathway is defined by the source of the fissile material used to initially deploy the FR fleet before the fleet is self-sustaining by recycling its own fuel. Initial deployment is the first few commercial FRs and supporting facilities. After successful deployment, the FR fleet will go through a period of rapid (above the long-term average) growth rate as the FR fleet expands from just a few reactors to 100 or more FRs. During this build out phase, an external fissile source (not from recycled FR fuel) will be required to supplement what is available from the FRs. Over the course of the build out, the fraction that comes from external sources will decrease and by the end of transition, the external sources will no longer be required because the fleet has become self-sufficient, i.e. all new fissile material required will be produced by the FR fleet.

For transition to EG23, three primary pathways were identified [2] for transition to the U/Pu system. For EG24, there are many additional pathways that can be envisioned in the transition to the final end state system that recycles all U, Pu, and MA. There are now even two different end states that can be envisioned: homogeneous (U/TRU recycled together) and heterogeneous (some or all of the MAs recycled separately from the Pu typically as MA-bearing targets or blankets). Table 3 shows this much broad set of transition pathways. The transition can proceed directly to the end state or can proceed with an interim step where U/Pu fuel is used in the FR with the MA either being separated and stored for later integration or disposed as HLW for an interim period. In the case of proceeding directly to the end state, it is assumed that when the TRU is recovered and utilised from the LWR UNF, it can be done either homogeneously or heterogeneously and it will continue that way for the FR UNF. No motivation for transitioning from homogeneous to heterogeneous or vice versa was identified, so it was not considered.

The pathways included in Table 3 are split into four groups. Group 1 is RU/TRU recovered from the LWR UNF. Group 2 is the same as Group 1 with LEU supporting the transition. Group 3 is only LEU utilised during transition. These are all consistent with the pathways used for transition to EG23 [2], but now with the MA included with the Pu as applicable because the system is transitioning to U/TRU recycle and not U/Pu recycle. Group 4 is a new set of pathways that involves using only the RU/Pu recovered from the LWR UNF as the primary fissile source for at least the initial phase of transition.

Table 3: Fast reactor startup pathways

<table>
<thead>
<tr>
<th>Transition pathway #</th>
<th>Primary transition fissile source</th>
<th>MA at initiation of transition</th>
<th>LEU</th>
<th>End state recycle (homogeneous or heterogeneous)</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>LWR RU/TRU</td>
<td>Irradiated</td>
<td>None</td>
<td>Same as initial</td>
</tr>
<tr>
<td>2</td>
<td>LWR RU/TRU</td>
<td>Irradiated</td>
<td>LEU Support</td>
<td>Same as initial</td>
</tr>
<tr>
<td>3</td>
<td>LEU</td>
<td>Irradiated</td>
<td>LEU Only</td>
<td>Same as initial</td>
</tr>
<tr>
<td>4a</td>
<td>LWR RU/Pu</td>
<td>Dispose</td>
<td>None</td>
<td>Heterogeneous</td>
</tr>
<tr>
<td>4b</td>
<td>LWR RU/Pu</td>
<td>Dispose</td>
<td>LEU Support</td>
<td>Heterogeneous</td>
</tr>
<tr>
<td>4c</td>
<td>LWR RU/Pu</td>
<td>Store</td>
<td>None</td>
<td>Heterogeneous</td>
</tr>
<tr>
<td>4d</td>
<td>LWR RU/Pu</td>
<td>Store</td>
<td>LEU Support</td>
<td>Heterogeneous</td>
</tr>
<tr>
<td>4e</td>
<td>LWR RU/Pu</td>
<td>Dispose</td>
<td>None</td>
<td>Homogeneous</td>
</tr>
<tr>
<td>4f</td>
<td>LWR RU/Pu</td>
<td>Dispose</td>
<td>LEU Support</td>
<td>Homogeneous</td>
</tr>
<tr>
<td>4g</td>
<td>LWR RU/Pu</td>
<td>Store</td>
<td>None</td>
<td>Homogeneous</td>
</tr>
<tr>
<td>4h</td>
<td>LWR RU/Pu</td>
<td>Store</td>
<td>LEU Support</td>
<td>Homogeneous</td>
</tr>
</tbody>
</table>
Group 4 is an approach unique for transition to recycle of U/TRU relative to what was analysed for recycle of U/Pu. An intermediate step could be added where initially, the use of MA-bearing fuels is delayed, and only U/Pu recycled fuels are utilised. There is significant challenge associated with deployment of the U/Pu recycled fuel technologies. However it is likely less than with the U/TRU or U/Pu/MA technologies [1]. There are things (e.g. annual funding levels, more fuel development and/or irradiation experience) that could increase the time it take to be able to commercially deploy MA-bearing fuels relative to U/Pu fuels. Using an intermediate step allows transition being initiated with the recycled fuel technologies that may require significantly less time and investment to be developed. Regardless of the reason, the possibility was assumed that recycle of MA could delay commercial deployment relative to the time to do the same with only the recycle of U/Pu.

Group 4 offers a number of potential benefits for an assumed quicker initiation of transition relative to U/TRU recycle. It is expected to reduce the amount of front-end resources (e.g. NU and enrichment) required relative to using in a longer term the RU/TRU from the LWR because the FR can be deployed sooner. When LEU support is used to fuel FRs, it may also reduce front-end requirements if the transition from using LEU to recycled fuels is quicker. It may require some additional investment because of the added steps in transition and the associated technologies required.

There are two approaches for dealing with the Pu plus MA. If the MA is sent to waste, this will clearly be degradation in transition performance because of more MA being sent to waste than if it were not. The other approach is heterogeneous recycle where both a U/Pu and MA-bearing stream are recovered, but initially only the U/Pu stream is used to manufacture FR fuel delaying the more challenging fabrication of MA-bearing fuel or targets. This approach requires more costly material recovery processes, but does not send any additional MA to the HLW system. It does require some extended storage of the MA-bearing material until the technology is developed that allows MA to be incorporated into fuel or targets.

This approach of a partial intermediate step can be utilised with and without LEU as variations of the pathways. Such utilisation of LEU would add another level of complexity and when considered in combination with the other compatible approaches adds many possible permutations in the transition pathway. It provides a way to balance the benefits that come from utilising recycled fuel as quickly as possible with the challenges that come from utilising MA-bearing fuels. It provides for a more complex set of tradeoffs because of the greater flexibility in transition.

The use of an intermediate Pu option will impact how much MA is either sent to waste or accumulated in storage for future use. The total amount of material will be determined by the magnitude and duration that the approach is utilised. The goal will be to minimise its duration. There are many ways for implementation and each will have different behaviour with different practical and technical constraints.

The recovery of just the RU/Pu initially from the LWR (MA sent to waste) will require the further development of the separation technology to recover the MA elements. This can be done either heterogeneously or homogeneously. The approach will affect what fuel technologies must be developed. Regardless of the approach an additional fuel technology is required. This could either be the U/TRU fuel (U/Pu fuel becomes obsolete) or a MA-bearing targets (U/Pu fuel remains at steady state).

When recovery of both the RU/Pu and MA-bearing material is considered with the initial interim storage of the MA-bearing material, this separation technology can be the final steady-state separation technology. The MA-bearing target fuel technology must be developed for the complete
suite of technologies to be developed. If transition initially involves heterogeneous separation, transition to a heterogeneous end state seems to be the more natural transition. However, given the large uncertainty, the possibility of transition to a homogeneous recycle system is included.

**Technology transition**

There are two constraints that must be satisfied at all times. These are the availability of material and the availability of technology. Material availability is constrained by technology (i.e. reprocessing and fabrication technologies must be available to make material available), by the available infrastructure (i.e. assumptions of the scale of deployed facilities), and by physical constraints (i.e. Pu produced in LWR UNF and material requirements and production in FR). The material constraints not related to technology availability are driven by the fissile inventory required and available and these change very little between EG23 and EG24. Therefore, the difference is driven by the availability of technology and its transition. The technology will transition from the current state to the final end state with any interim technologies required for a particular transition scenario envisioned.

The many different fast reactor startup pathways described imply an equally large set of technology transition pathways. These technology transitions must be understood and a plausible timeline developed for that technology transition that corresponds to that pathway. If unlimited budgets and development of all technologies proceeding on the schedule envisioned, there would be no reason to consider anything but the most straightforward pathway of either Group 1 through 3 and all Group 4 would not be considered. However, it is anticipated that there may be budget constraints or a desire to not so tightly couple all technologies during the develop, demonstration, and initial deployment phases, but to stage them to avoid both the large annual funding requirements and having all technologies on or near critical path adding to the schedule risk for individual demonstration and first few commercial facilities during this period.

To understand and inform on this very complex part of transition, the first step is to identify all important technologies that are required over the course of transition. This was developed for all fast reactor startup pathways described in Table 3 as well as for EG23. Examples are included for EG24 Group 4b and the LEU Support approach for EG23. Table 4 shows the technologies that will be required over the course of transition for the given example for EG23 and Table 5 for EG24. The columns provide a simple representation of the evolution. Those that exist, those that will come in during transition and in the example for EG24 those that come in earliest in transition and those later in transition, and then the technologies that remain once transition has completed.

There are two insights. The first is that many technologies will need to be developed and incorporated into any planning. The second is that there is a lot of commonality between them, even between EG23 and EG24 which will not preclude alternatives in the foreseeable future for most pathways envisioned. These are just two examples of many alternatives specifically chosen because of the large overlap in technologies during initial transition. Other alternatives will have less overlap, and is easily inferred from the tables.
Table 4: EG23 Technology requirements Using U/Pu from LWR UNF and LEU support

<table>
<thead>
<tr>
<th>Fuel supply</th>
<th>Exist</th>
<th>Transition</th>
<th>Final</th>
</tr>
</thead>
<tbody>
<tr>
<td>NU</td>
<td>x</td>
<td>x</td>
<td>x(^1)</td>
</tr>
<tr>
<td>Uranium enrichment</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>&lt; 5 wt. %</td>
<td>x</td>
<td>x</td>
<td></td>
</tr>
<tr>
<td>&gt; 5 wt. %</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Fuel fabrication</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>LWR: LEU</td>
<td>x</td>
<td></td>
<td></td>
</tr>
<tr>
<td>FR: LEU</td>
<td></td>
<td>x</td>
<td></td>
</tr>
<tr>
<td>FR: U/Pu</td>
<td></td>
<td></td>
<td>x</td>
</tr>
<tr>
<td>FR: MA-bearing targets</td>
<td></td>
<td></td>
<td>x</td>
</tr>
<tr>
<td>Reactors</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>LWR</td>
<td>x</td>
<td></td>
<td></td>
</tr>
<tr>
<td>FR</td>
<td></td>
<td>x</td>
<td></td>
</tr>
<tr>
<td>Reprocessing</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>LWR UNF (RU/Pu)</td>
<td></td>
<td></td>
<td>x</td>
</tr>
<tr>
<td>FR UNF (RU/Pu)</td>
<td></td>
<td></td>
<td>x</td>
</tr>
<tr>
<td>LWR UNF (RU/Pu/MA)</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>FR UNF (RU/Pu/MA)</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Disposal</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>DU</td>
<td></td>
<td>x(^2)</td>
<td></td>
</tr>
<tr>
<td>RU</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>LWR SNF</td>
<td>x(^2)</td>
<td></td>
<td></td>
</tr>
<tr>
<td>HLW (LWR UNF Rep. w/ MA)</td>
<td></td>
<td></td>
<td>x</td>
</tr>
<tr>
<td>HLW (FR UNF Rep. w/ MA)</td>
<td></td>
<td></td>
<td>x</td>
</tr>
<tr>
<td>HLW (LWR UNF Rep. w/o MA)</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>HLW (FR UNF Rep. w/o MA)</td>
<td></td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

1. Inventory of DU could effectively displace the need for NU for several centuries.
2. Assumes existing inventories of LWR UNF and DU are disposed.

Note: Storage and transport for a wide range of materials that will vary over time is required. The nature of those materials will vary significantly with the specific transition pathway being considered.
Table 5: EG24 technology requirements using U/Pu from LWR UNF, interim disposal of MA, and LEU support transitioning to a heterogeneous recycle system

<table>
<thead>
<tr>
<th></th>
<th>Exist</th>
<th>Transition</th>
<th>Final</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Initial</td>
<td>Final</td>
<td>Final</td>
</tr>
<tr>
<td>Fuel supply</td>
<td>NU</td>
<td>x</td>
<td>x</td>
</tr>
<tr>
<td></td>
<td>x</td>
<td>x</td>
<td>x</td>
</tr>
<tr>
<td>Uranium enrichment</td>
<td>&lt; 5 wt. %</td>
<td>x</td>
<td>x</td>
</tr>
<tr>
<td></td>
<td>&gt; 5 wt. %</td>
<td>x</td>
<td>x</td>
</tr>
<tr>
<td>Fuel fabrication</td>
<td>LWR: LEU</td>
<td>x</td>
<td>x</td>
</tr>
<tr>
<td></td>
<td>FR: LEU</td>
<td>x</td>
<td>x</td>
</tr>
<tr>
<td></td>
<td>FR: U/Pu</td>
<td>x</td>
<td>x</td>
</tr>
<tr>
<td></td>
<td>FR: MA-bearing targets</td>
<td>x</td>
<td>x</td>
</tr>
<tr>
<td>Reactors</td>
<td>LWR</td>
<td>x</td>
<td>x</td>
</tr>
<tr>
<td></td>
<td>FR</td>
<td>x</td>
<td>x</td>
</tr>
<tr>
<td>Reprocessing</td>
<td>LWR UNF (RU/Pu)</td>
<td>x</td>
<td>x</td>
</tr>
<tr>
<td></td>
<td>FR UNF (RU/Pu)</td>
<td>x</td>
<td>x</td>
</tr>
<tr>
<td></td>
<td>LWR UNF (RU/Pu/MA)</td>
<td>x</td>
<td>x</td>
</tr>
<tr>
<td></td>
<td>FR UNF (RU/Pu/MA)</td>
<td>x</td>
<td>x</td>
</tr>
<tr>
<td>Disposal</td>
<td>DU</td>
<td>x(^2)</td>
<td>x(^2)</td>
</tr>
<tr>
<td></td>
<td>RU</td>
<td>x(^2)</td>
<td>x(^2)</td>
</tr>
<tr>
<td></td>
<td>LWR SNF</td>
<td>x</td>
<td>x</td>
</tr>
<tr>
<td></td>
<td>HLW (LWR UNF Rep. w/ MA)</td>
<td>x</td>
<td>x</td>
</tr>
<tr>
<td></td>
<td>HLW (FR UNF Rep. w/ MA)</td>
<td>x</td>
<td>x</td>
</tr>
<tr>
<td></td>
<td>HLW (LWR UNF Rep. w/o MA)</td>
<td>x</td>
<td>x</td>
</tr>
<tr>
<td></td>
<td>HLW (FR UNF Rep. w/o MA)</td>
<td>x</td>
<td>x</td>
</tr>
</tbody>
</table>

1. Inventory of DU could effectively displace the need for NU for several centuries.
2. Assumes existing inventories of LWR UNF and DU are disposed.

Note: Storage and transport for a wide range of materials that will vary over time is required. The nature of those materials will vary significantly with the specific transition pathway being considered.

Table 6 shows estimated time and costs associated with technology development, demonstration, and deployment of the technologies to perform the functions required for different transition pathways from the E&S [1]. There are likely significant costs savings above the simple summation for particular combinations of transition scenarios since much of the costs will apply to more than one technology in a particular area if they are done in an integrated approach.
Table 6: Estimated development, demonstration, and deployment time and cost

<table>
<thead>
<tr>
<th>Fuel supply</th>
<th>NU</th>
<th>FOAK deployment Cost (USD billions)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Development Cost</td>
<td>0</td>
<td>0</td>
</tr>
<tr>
<td>Development Time</td>
<td>0</td>
<td>0</td>
</tr>
<tr>
<td>Fuel supply</td>
<td>FOAK</td>
<td>0</td>
</tr>
<tr>
<td>Uranium enrichment</td>
<td>&lt; 5 wt. %</td>
<td>0</td>
</tr>
<tr>
<td></td>
<td>&gt; 5 wt. %</td>
<td>0</td>
</tr>
<tr>
<td>Fuel fabrication</td>
<td>LEU LWR</td>
<td>0</td>
</tr>
<tr>
<td></td>
<td>LEU FR</td>
<td>0</td>
</tr>
<tr>
<td></td>
<td>RU/Pu FR</td>
<td>&lt;0.2</td>
</tr>
<tr>
<td></td>
<td>RU/TRU FR</td>
<td>2-10</td>
</tr>
<tr>
<td></td>
<td>MA Targets</td>
<td>2-10</td>
</tr>
<tr>
<td></td>
<td>RU, DU, NU – Blankets</td>
<td>0</td>
</tr>
<tr>
<td>Reactors</td>
<td>LWR</td>
<td>0</td>
</tr>
<tr>
<td></td>
<td>FR</td>
<td>2-10</td>
</tr>
<tr>
<td></td>
<td>Reactors</td>
<td>2-10</td>
</tr>
<tr>
<td></td>
<td>LWR UNF – RU/Pu</td>
<td>0.2-2</td>
</tr>
<tr>
<td></td>
<td>FR UNF – RU/Pu</td>
<td>0.2-2</td>
</tr>
<tr>
<td></td>
<td>LWR UNF – RU/Pu/MA</td>
<td>2-10</td>
</tr>
<tr>
<td></td>
<td>FR UNF – RU/Pu/MA</td>
<td>2-10</td>
</tr>
<tr>
<td>Storage and transport of fuel cycle materials</td>
<td>0</td>
<td>0</td>
</tr>
<tr>
<td>Interim Storage of MA</td>
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<td>0</td>
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<tr>
<td>Disposal</td>
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<td>0</td>
</tr>
<tr>
<td></td>
<td>RU</td>
<td>0</td>
</tr>
<tr>
<td></td>
<td>LWR SNF</td>
<td>0</td>
</tr>
<tr>
<td></td>
<td>MA HLW (LWR UNF Rep.)</td>
<td>&lt;0.2</td>
</tr>
<tr>
<td></td>
<td>MA HLW (FR UNF Rep.)</td>
<td>&lt;0.2</td>
</tr>
<tr>
<td></td>
<td>HLW (LWR UNF Rep.)</td>
<td>&lt;0.2</td>
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<tr>
<td></td>
<td>HLW (FR UNF Rep.)</td>
<td>&lt;0.2</td>
</tr>
</tbody>
</table>

1. A value of 0 means that the technology is considered to be beyond FOAK.
2. The cost of the disposal facility is not included. This is only for waste forms.
3. There is cost associated with developing the specific specifications and processes, but deployment will not incur additional FOAK costs.

Conclusions

This analysis of transition to fast reactor (FRs) with continuous recycle of U/TRU (EG24) found that the understanding of the transition to FRs with continuous recycle of U/Pu (EG23) [2] is applicable, and that the two alternatives face many of the same issues and challenges. However, there are certain areas where the challenges are greater for EG24 as a result of recycling minor actinides (MA).

Recycling the MA, either homogenously (grouped with the Pu) or heterogeneously (separately from the Pu), results in additional challenges that need to be overcome. These include greater challenge (likely greater cost and time required) to develop the recycle technologies and as well as the ongoing challenge of recycling fuels that will have higher decay heats and radiation emission rates. Both of these challenges have potentially significant impact on transition. It does however offer additional potential benefits associated with avoiding the need for disposal of MA.
As far as fissile material constraints, these are driven by the Pu component which does not show first order differences. This leads to transition behaviour for EG23 and EG24 being very similar in this regard.

EG24 can be deployed using an intermediate transition step where initially only the less challenging U/Pu fuel is utilised. This presents a unique transition pathway where tradeoffs can be made between cost, risk, and benefits by delaying some or all of the challenges associated with recycle of MA. The integral “challenge” may be greater, but the challenge to be overcome by a certain date (e.g. 2050) can be reduced substantially in this manner. This could potentially lead to significant benefits during transition if the additional cost or technical challenge of recycling the MA would in fact require more time to deploy recycled fuel in the FRs (the primary source of many benefits of transition).

This intermediate pathway has two variants. There is the possibly less challenging (to meet a certain date to begin utilising recycled fuel), but less beneficial, approach of initially only recycling U/Pu and sending the MA to waste. There is also the possibly more challenging, but more beneficial, approach of separating both the U/Pu and a MA-bearing stream while only initially using U/Pu fuel and interim storage of the MA for future use. In either case, the system will transition to continuous recycle of all TRU elements (Pu and MA).

Acknowledgements

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References

Ameriicum transmutation in a scenario of progressive SFR deployment

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Abstract

After the studies carried out within the framework of the 2006 French Waste Management Act, CEA, EDF and AREVA decided to work together on potential progressive transition scenarios from the current French nuclear fleet to a SFR fleet which does not require natural uranium to operate. This paper describes one of these scenarios.

The first part of the paper focuses on the presentation and results of this transition scenario, with no minor actinides (MA) transmutation taken into account. The scenario starts with the current French fleet and its replacement by an EPR reactor fleet in which plutonium keeps on being mono-recycled. MOX PWR spent fuel reprocessing begins in 2040 in order to feed three breakeven SFR, commissioned between 2050 and 2059. To prepare for a larger SFR deployment, three more SFRs are commissioned between 2075 and 2085 and small-scale SFR spent fuel reprocessing starts in 2060. In 2090, additional SFR are deployed in order to stabilise total plutonium and used fuel inventories, with a fleet composed of 16 breeder SFR and 22 EPR reactors. Finally, between 2150 and 2185, this EPR/SFR reactor fleet is progressively renewed by a new one composed of 41 breakeven SFR. At the end of the scenario, the total Pu inventory is stabilised at 1 260 tonnes but the MA inventory in the waste is still increasing at a rate of 2.5 tHM/y.

The following parts of the paper focus on the impact of americium (Am) transmutation in Am-bearing blankets (AmBB) loaded in the SFR commissioned in the scenario. Two cases are described, one considering the Am transmutation in the fleet ultimately composed only of SFR, and the other considering the Am transmutation starting with the mixed SFR/EPR fleet. Finally, one row of AmBB containing 10 wt% of Am is enough to stabilise the Am inventory (in both cycle and waste) if the fleet is composed only of SFR. In the case of a mixed fleet composed of 16 SFRs and 22 mixed EPRs, two rows of AmBB containing 15 wt% of Am are required in each SFR core in order to recycle all of the separated Am. However, in that case the total Am inventory is not stabilised as it is still increasing in the spent fuels and blankets. It is noteworthy that, compared to a scenario with Pu multi-recycling but without transmutation, Am transmutation in AmBB can lead to a 30% reduction of the radiotoxicity of the waste accumulated all over the scenario.
Corrosion behaviour of pressurised austenitic cladding tubes in oxygen containing PbBi at 550°C

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Abstract

Pb-alloys as coolant media are foreseen in the lead fast reactor (LFR) and the MYRRHA reactor in Belgium. For both 15-15 Ti-stabilised steel (1.4970) should be used as fuel cladding material due to its low swelling at high doses. Its compatibility with lead (Pb) and lead-bismuth eutectic (LBE) has been studied in the past focusing on its behaviour at different temperature, oxygen concentrations and different flow velocities of the liquid metal. During operation, swelling of the fuel by irradiation or fuel cracking will increase the stresses on the cladding wall, which can influence the corrosion behaviour. To perform tests in Pb/PbBi on cladding tubes under stress, a new test device was constructed in which an internal pressure can be applied to tubes. The first tests with pressurised 1.4970 tubes at 550°C in PbBi with an oxygen concentration of 10⁻⁶ wt% were conducted. The outer diameter of the reference samples measured after exposure is within the fabrication tolerance of the tubes. All other samples showed a clear increase in diameter between 17 and 23 µm.

As expected, a thin, not visible Fe-Cr-Mn spinel with Si was formed at the surface of the reference sample. The localised scale is interrupted by oxides nodes consisting of magnetite at the top with a spinel layer underneath and an inner oxidation zone (IOZ). At 550bar internal pressure the number of oxide nodes at the surface increases and the grain boundary oxidation is much more pronounced. The oxide roots went up to 11 µm, while that on the reference sample only reached around 5-6 µm deep into the steel.

The results after 300 h and 600 h at 600 bars are comparable and differ from previously described samples. The induced stress enhances the oxidation. Nearly the whole surface is covered by a multi-layered oxide consisting of magnetite, spinel and IOZ. Sometimes the magnetite is spalled off. After 300 h, the IOZ increases to 15 µm deep into the steel. However, a small IOZ can occur in the area with a thin Fe-Cr-Mn oxide. Cracks at the surface were not observed.
Flexible operation of nuclear reactors in advanced fuel cycles

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Abstract

Light water reactors (LWRs) that are currently being deployed and operated in the United States are under economic stress in an evolving energy market with the low prices of natural gas and with the large fraction of intermittent sources of energy that is penetrating the power grid. Fast reactors associated with continuous recycling of Plutonium or Transuranics along with the recovered uranium will provide benefit in terms of fuel utilisation and nuclear waste generation. However, the development of such advanced fuel cycles (AFC) requires demonstrating they can be used in an economically competitive way in the future US energy market. Load following operation is investigated as a possible solution to improve the competitiveness of nuclear power in an energy environment disrupted by a large penetration of intermittent sources, by avoiding for instance power generation when electricity prices are low or negative. In this context, the capability of sodium-cooled fast reactors (SFR) for following load variations was compared with that of pressurised water reactors (PWR). Fast reactors such as SFRs are found to be intrinsically better suited than thermal reactors for load following. SFRs should allow much more flexible operation, allowing power variations several times per day during all their operating cycle. Their flexible operation is not a reactor physics issue since fast reactors are not significantly constrained by fission product poisoning and are very stable. However, SFR control system would need to allow performing synchronised control actions to limit coolant temperature variations during reactor power transients to limit mechanical stresses and to allow faster transients.

Such load following operation could improve the competitiveness of SFRs with AFCs by avoiding power generation when electricity prices are low during periods of low demand or high renewable generation, as illustrated based on possible future scenarios applied to the California power grid.

Introduction

In the United States, only LWR are currently being deployed in a once-through fuel cycle and are operated as base-load units. Those LWRs are currently under economic stress in the current US energy market, which is evolving due to the low prices of natural gas and to the large fraction of intermittent sources of energy that is penetrating the power grid. The state of California, for instance, targets 50% of electricity sold from renewables by 2030 [1]. An evaluation and screening (E&S) of fuel cycle options has been conducted recently [2] by the Fuel Cycle Technologies Office of US Department of Energy (DOE), Office of Nuclear Energy, to identify potential benefits and challenges of nuclear fuel cycle options together with a relatively small number of promising fuel cycles for the United States. The E&S study identified continuous recycle of plutonium (Pu) or Transuranics (TRU)
along with the recovered uranium (U) in fast critical reactors as a very promising fuel cycle option to improve natural resource utilisation and reduce nuclear waste generation. Even if fast reactors associated with an AFC might provide these benefits, the economic market challenges that nuclear systems face might compromise their deployment. Consequently, studies are currently being conducted to evaluate possible approaches to improve the competitiveness of nuclear systems. The focus of this study is to evaluate what advantage load following power might have on the competitiveness of nuclear reactors in AFCs for instance by avoiding power generation when electricity prices are low or negative during periods of low demand or high renewable generation.

Load following involves reactor operation in a turbine leading mode, where the nuclear power follows the power demand from the turbine. Different types of grid management operations are considered, any of which requiring variation of the power level sent to the grid: frequency control of the grid (primary or secondary), load following mode with daily power variations, and rapid transient operations to disconnect/reconnect a reactor from the grid. Different reactor concepts have different challenges to varying the power level and this paper compares the flexible operation potential of fast reactors in an AFC, the sodium-cooled fast reactors (SFRs), to that of a pressurised water reactor (PWR), for which load following operation is already implemented in a few countries [3]. This study based on past and current experience, literature review, and expert judgements, focuses on what limits the manoeuvrability of the reactor systems, how fast and often those can sustain large power variations, and how flexible operation could favour future grid management. Due to its highly ambitious renewable development target, the California power grid is chosen as a test-case in this paper to analyse the impact nuclear load following could have on the grid management.

Comparison of flexible operation potential between PWRs and SFRs

Various phenomena (neutronics, thermal hydraulic...) affect the behaviour of the reactor during a power transient and need to be taken into account from a reactor operation standpoint. They are different from one technology to another and those differences are critical and need to be understood. The objective of this section is not to fully detail every phenomenon, but instead to introduce the key differences that could impact the load following operation of the PWR and SFR concepts.

 Difference in physics

The xenon effect in thermal reactors is due to the variation in equilibrium concentration of 135I depending on the power level of a PWR, and subsequently in the concentration of its decay daughter 135Xe, which displays a large thermal absorption cross section. This effect is traditionally managed during load following operation in PWRs though Boron dilution/insertion, which is costly in terms of effluent generation, and unavailable in the final 5-20% of the operating cycles for most technologies [3]. On the other hand, cross sections of fission products (FPs) are much lower in a fast spectrum and so the FPs do not play a significant role in the reactivity control of an SFR. For PWRs, limiting the changes in axial power distributions (axial offset) due to control rod movements is the principal challenge associated with load following operation [3]. Inserting or withdrawing the control rods affects the axial power distribution, especially if those rods are very efficient such as the “black” rods used in early PWR designs. A change in axial power distribution affects the xenon level in the different axial regions of the core, with risks of causing a xenon oscillation. Those axial oscillations should be avoided and they are the main reason the axial offset must be monitored during operation and remain within the operational domain defined in the specifications of the reactor design. The length of the driver fuel in SFR is typically only one metre and the axial power distribution is very
stable, especially since the mean free path of fast neutrons is relatively long with respect to the length of the fuel column, and there is no risk of xenon oscillation. In addition, the control rods need small movement to control the power level in SFR, limiting the changes in power distribution.

During power transient, the change in electrical power output produced on the turbine does not immediately propagate to the nuclear output because of different time constants. Differences between electrical and nuclear power should be avoided to minimise thermal imbalances between the different loops in the system. Consequently, the thermal inertia of the system can potentially limit the speed of transients available during the load following operation. In a PWR, the slowest dynamics responsible for thermal imbalances comes from the thermal inertia in the U-tube steam generator. For same power extraction, larger steam generators (containing larger mass inventory of water) are needed in PWRs compared with SFRs due to the difference between the saturated cycle (in PWR) and super-heated cycle (in SFR). The slowest dynamics of an SFR is not found in the once-through steam generator, but in the primary and intermediate systems due to the large amounts of sodium they contain.

This benefits the inherent safety of the SFR since the coolant temperature takes time to rise following loss of heat sink scenario. However, for load following operation, the larger thermal inertia of primary and intermediate circuits of the SFR might lead to longer transition time for a temperature perturbation in the primary system to be transmitted to the power conversion system [4]. As discussed in the following section, the control system of the SFR should limit changes in coolant temperatures to allow fast power variations. During load following operation, the fuel undergoes significant changes in power and temperatures that can result in pellet-cladding chemical and mechanical interactions. For these reasons, PWRs in France do not operate in the load following mode during the first two weeks of the operational cycle for the conditioning of the oxide fuel [3].

For SFRs, the metallic fuel option was found to display excellent transient capabilities for load following application [5], as demonstrated in EBR-II with a series of low- and high-ramp-rate tests. The absence of fracturing of the metallic slug reduces risks of relocation of fragments in the bond region that could generate clad failure. At BOC, metallic fuel is separated from the cladding by a large gap allowing its swelling and expansion; it is then linked to the cladding early in the cycle and follows its thermal expansion. Its good plasticity should not generate much stress on the cladding during load following transients. For all these reasons, metallic fuel in SFR might allow load following operation from the beginning of the cycle [6]. Metallic fuel also displays good capability of running beyond cladding breach, including during transients [7], which might allow continuous load following operation even after cladding failure [6].

The variations in coolant temperature in the different components lead to cycling changes in the mechanical load in some parts of the components, which can potentially induce localised structural damage [3]. Continuous monitoring of the equipment fatigue is performed in nuclear reactors. For PWR, it was shown that the operation regimes used for European Utilities’ Requirements (EUR) should allow up to 200 cycles per year during plant lifetime of 60 years [3]. For SFR, sufficient experience and analyses are unavailable to allow judgement on this point. However, it should be noted that different operating modes can be employed and the one minimising changes in coolant temperatures should be favoured to minimise the impact of load following on structural components. The core of a PWR is designed to maximise the reactivity in nominal operation, which means that a loss-of-coolant or fuel melting will result in-core shutdown. This is not the case for an SFR where the core is not in its highest reactivity condition at full power. Consequently, the SFRs must demonstrate inherent safety capability to sustain large transients, such as a loss of flow or a loss of heat sink, that
are unprotected by control rods. During load following operation, the initial rated power (Pr) being decreased from 100%, the consequences of such transient will be lower than what it would be at full power. The only concern might be for an unprotected transient over power in the case where SFR is being operated by control rod movements, with potential increase in risks of control rod ejection.

**Conclusions on load following operations in PWRs and SFRs**

SFRs are intrinsically better suited than thermal reactors for load following application as summarised in Table 1. Load following operation in PWRs can be limited by a reactor physics issue, mainly due to xenon poisoning, which can be managed by design changes, extra monitoring, and definition of appropriate specifications. Consequently, in some countries load following operation in PWRs is used to adapt the grid to scheduled daily power variation and to help with frequency management. PWRs allow relatively fast power variations for daily variations typically between 2.5%Pr/min and 5%Pr/min [3].

SFRs should allow much more flexible operation, allowing power variations several times per day during all their operating cycle. Their flexible operation is not a reactor physics issue since fast reactors are not significantly constrained by fission product poisoning and are very stable, which facilitates their operation. However, SFRs might not necessarily allow quicker load following operation than current PWRs, since the speed of transients in SFR is limited by the high thermal inertia of the primary and intermediate systems (delaying the nuclear power adjustment to the electric output requested), and by the stress produced on pipes and structure components by coolant temperature changes. Consequently, a control system developed for load following in SFR should minimise coolant temperatures variations with power changes by adjusting the control rods and flow rates with nuclear and electric power levels. SFRs can be operated with autonomous or active control systems, depending on if action is taken to assist the power variations of the core. Autonomous control is advantageous in terms of operation simplicity since it relies only on the neutronics feedback coefficients of the core to follow the power level requested by the grid. This type of operation was demonstrated with the Plant Inherent Control Testing (PICT) experiments in EBR-II in November 1987 [8].

However, the autonomous control relies on inlet and outlet primary coolant temperatures that take time to propagate and would increase the fatigue on structural components. An active reactor control system consists in actively controlling the reactivity of the core with control rods, and maintaining coolant temperatures by means of flow rates control. A model-based active control system [9] could be developed with the objective of anticipatory adjustment of the core power and flow rate at the same time to limit temperatures changes. However, this idealised approach is difficult to attain as it requires a very good model with accurate knowledge of the reactor neutronics and thermal-hydraulic behaviour at different power levels. This will be challenging to obtain without preliminary feedback experience using SFRs. Such control system, however, might allow at least similar speed of power variations as compared with advanced PWRs [10].
Table 1: Summary of potential load following operating capabilities in SFRs and PWRs

<table>
<thead>
<tr>
<th></th>
<th>PWR</th>
<th>SFR</th>
</tr>
</thead>
<tbody>
<tr>
<td>Operation at EOC</td>
<td>Limited to first 80-95% portion of the operating cycle.</td>
<td>Can be operated throughout the cycle.</td>
</tr>
<tr>
<td>Operation at BOC</td>
<td>Avoided in the first 2 weeks of operating cycle.</td>
<td>Metallic fuel should allow operation at beginning of cycle.</td>
</tr>
<tr>
<td>Operation during cladding failure</td>
<td>Avoided.</td>
<td>Metallic fuel and sodium coolant should allow flexible operation with cladding breach.</td>
</tr>
<tr>
<td>Daily maneuverability</td>
<td>Large variations in power are usually limited to one cycle per day.</td>
<td>No limitation.</td>
</tr>
<tr>
<td>Lower range of power operation</td>
<td>Some advanced PWRs can be operated down to 15% Pr (Pr).</td>
<td>- With steam cycle, operation should be similar to that in PWRs.</td>
</tr>
<tr>
<td></td>
<td></td>
<td>- With S-CO2 cycle, possible operation between 0%Pr and 100%Pr.</td>
</tr>
<tr>
<td>Speed of transients</td>
<td>Can be up to 5%Pr/min (lower at EOC and at low power level).</td>
<td>Advanced control system should allow at least similar speed of power variations as compared with advanced PWRs.</td>
</tr>
<tr>
<td>Number of transients over the lifetime of the plant</td>
<td>The European Utilities Requirements (EUR) allow 200 duty cycles per year over 60 years.</td>
<td>Advanced control systems for SFR that avoid variations in coolant temperatures might be required to limit the fatigue on components and allow faster transients.</td>
</tr>
</tbody>
</table>

Continuous load following operation to low-power level might be required for the nuclear power plant to avoid sending electricity to the grid when prices are low or negative due to low demand or to a peak in intermittent sources production. Advanced PWRs such as the AP1000 allow continuous load following operations down to 15%Pr [11]. Load following in SFR could be done in a large range of power from 100% to 0%Pr with an S-CO2 power conversion cycle because of the higher level of flexibility to reduce the pressure (or inventory) of S-CO2 [4, 12]. With steam generator, the SFR is foreseen to allow similar continuous operation with low power as advanced PWR concepts, provided optimised SFR control and power conversion systems are implemented [10].

Load following operation in PWR is only available for scheduled power operation to adapt the grid to daily power variations. PWRs have also the ability to perform frequency regulation and rapid transients. Load following in SFR allow additional flexibility as it can be operated at any time since it is always capable of manoeuvring. As a consequence, frequency regulation and rapid transients should be available in SFR without additional issues. This additional level of flexibility is especially appreciated in an electrical grid with large penetration of intermittent renewable energy like wind and solar, for which electrical output cannot always be predicted ahead with sufficient accuracy and might require several large power transients during the day [13].

Application to the State of California

In 2011, the Renewables Portfolio Standard (RPS) [1] was codified in the state of California, which requires that “all retail sellers of electricity shall serve 33% of their load with renewable energy by2020” [Gov. A. Schwarzenegger, Executive Order S-14-08]. Already, ~25% of electricity retail sales in California were produced from renewable energy in 2014, which is ahead of schedule for meeting the RPS requirements. The RPS is being re-instituted to propose the ambitious objective to reach 50% by 2030. The challenge brought by this policy on the grid management is assessed together with the potential benefit from a flexible nuclear energy. This scoping study is based on publicly available data [14] for several days selected in 2015, but only those of November 4, representing an average day in
terms of temperatures, wind and electricity consumption, are reported here. This study uses simple models assessing how to meet the daily energy demand in California, without exceeding it, based on the different sources of energy installed and on the hourly production of renewables. A detailed economic analysis with grid management optimisation should be conducted in the future for confirming and extending the conclusions of this study.

Renewable energy considered by the RPS includes wind, solar photovoltaic, solar thermal, small hydro, geothermal, biomass and biogas [1]. Large hydroelectric facilities (>30MW) are not eligible for the RPS and are reported separately. California’s operating renewable capacity can be located both within or outside the state, but only in-state capacity will be considered in this work, which represents more than 90% of the installed capacity considered. California imports a significant fraction of its electricity from neighbouring states, while it has sufficient installed natural gas capacity to sustain its needs. For simplicity purposes, only the domestic production of California is considered here. The imports are neglected and the natural gas and coal generation (mentioned at “Thermal” in Figure 1) available in California is supposed to make up for the difference between the consumed electricity and the generation from renewables, large hydro, and nuclear.

Figure 1: Cumulative electricity generated per different energy sources in California in November 4, 2015

The cumulative electricity generated per different energy sources is displayed in Figure 1 assuming three different scenarios:

- In the reference case, the fraction of energy generated from renewable energy is 20% and 10% comes from nuclear energy. The demand can be met with domestic production without overcapacity generation.
- In the second scenario, the solar and wind capacities are increased by factors of 4 and 2 respectively (compared to 2015 capacities), as examples, to see how a significant increase in renewable affects the electricity production. The total capacity of installed wind and
solar would be 12 and 40 GWe, respectively. In this scenario, the natural gas and coal generations are reduced during peak hours to avoid exceeding the demand. However, over-generation of electricity is foreseen in the absence of renewable energy curtailment due to the large generation of solar energy during day-time. This is especially concerning in the case of nuclear energy when operating on a base-load capacity, since this electricity might have to be sold at low or even negative prices at peak hours. In this situation, nuclear cannot compete with the other non-renewable energies (thermal or hydraulic), those can be shut down during low demand or high-renewable production hours.

- In the last scenario, nuclear energy is supposed capable of flexible operation, which prevents over-generation in the increased renewable capacity scenario. For this analysis, one assumes nuclear systems capable of doing at least one load cycle per day with power down to 0%Pr with ramping speed at least of 2%Pr/min. In this and the previous scenarios, the fraction of electricity generated coming from renewable energy is 43%.

**Figure 2:** Impact of increased nuclear energy on scenarios for 4 November 2015

The increase in renewable capacity modelled in the wind and solar increase scenario with data from 4 November 2015, would allow the generation of 57% of electricity from low CO\(_2\) emitting sources, while it is 33% with the current capacity. The results displayed in Figure 2 indicate the impact of an increase or decrease of flexible or base-load nuclear energy capacity. Based on the energy production and consumption in California from 4 November 2015, an increase in nuclear capacity with a factor of, for example, 7.5 would lead to ~95% electricity generation with low CO\(_2\) emitting sources, and this, independently of an increased capacity from renewable energy. As seen in Figure 1, the load following capacity would prevent over-capacity generation of electricity during peak solar generation. However, the reactor utilisation, which is defined as the energy generated during the day over energy potentially generated at 100%Pr, is seen to decrease. This will lead to
lower fuel utilisation (and potential operating cycle length extension), but also might be associated with lower revenue. However, it is noted that the power reduction is achieved on purpose in the high-renewable generation scenario, when there is low demand or large renewable generation, which would be associated with low or negative revenues.

Conclusions

Fast reactors associated with continuous recycling of Plutonium or Transuranics along with the recovered uranium will provide benefit in terms of fuel utilisation and nuclear waste generation. However, the development of AFCs requires demonstrating they can be used in an economically competitive way in the future US energy market. Load following operation is investigated as a possible solution to improve the competitiveness of nuclear energy in a power grid disrupted by a large penetration of intermittent sources, for instance by avoiding power generation when electricity prices are low or negative. Consequently, the expected capability of sodium-cooled fast reactors (SFR) for following load variations was compared in this paper with that of pressurised water reactors (PWR).

This study based on past and current experience, expert judgement and literature reviews, shows that SFRs allow improved flexibility when compared with current PWRs, but not necessarily quicker transient operation. Flexible operation is not a reactor physics issue for SFRs since fast reactors are not significantly affected by fission product poisoning and are very stable, which facilitates their operation. However, SFR load following operation requires a control system capable of synchronising the control actions to the electrical power variations to limit coolant temperature changes in the system for reducing stresses on structure components and allowing faster transients. Such load following operation could improve the competitiveness of SFRs with AFCs by avoiding power generation when electricity prices are low during periods of low demand or high renewable generation, as illustrated based on possible future scenarios applied to the California power grid.

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The authors acknowledge the help of different reactor and fuels experts in the conduct of this analysis. For the fuel behaviour analysis, Dr G. Hofman provided invaluable expertise. The comments and reviews from Dr R. Vilim was also much appreciated. Argonne National Laboratory’s work was supported by US Department of Energy (DOE) under Contract number DE-AC02-06CH11357. The direction and support of the US DOE Office of Nuclear Energy is greatly appreciated.

References


Concepts of waste transmutation purpose fusion-driven subcritical system

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Abstract

Feasibility of the fusion-fission hybrid reactor (FFHR) for the incineration of long-lived TRU and fission product has been continuously studied. There have been many concepts proposed depending on the sizes, specific goals and material choices. Recently, a concept of dual goal FFHR has been proposed in order to add the energy multiplication (EM) function to the waste transmutation (WT). By adding fertile zone of thorium or depleted uranium, reduction of reactivity at the end of cycle can be compensated by conversion resulting in reduction of plasma power requirement.

This concept has been applied to the FFHR design for transmutation (Hyb-WT) [1]. A new concept has three zones in the blanket. The 1st zone is designed for WT. The choice of (U-TRU)Zr fuel and Pb-Bi coolant aimed for neutron economy. The 2nd zone is dedicated for energy multiplication using the concept of Direct Reuse of Used PWR (DRUP) fuel instead of thorium or depleted uranium. DRUP fuel is produced by dry fabrication process after decladding of used PWR fuel. It is the same concept with DUPIC (Direct Use of spent PWR fuel Into CANDU reactor) [2]. The use of DRUP gives two effects; one is fissile production during power production and the other is reduction of PWR spent fuel to be deposited. EM zone is cooled with light water for the spectrum softening. The 3rd zone is designed for tritium breeding to satisfy self-sufficient tritium breeding ratio. The two zones just after the plasma facing surface do not have any tritium breeding material including lithium. The third zone far away from plasma has neutrons already softened. The choice of Li_4SiO_4 pebble and He coolant is ideal for tritium extraction.

Neutronics calculations are performed with MCNPX 2.6.0 with ENDF/B-Ⅶ.0 neutron cross section library. Variation of k_eff value is reduced by fissile supply from DRUP fuel, as a result, energy multiplication factor is increased and plasma power is reduced.

Introduction

Feasibility of the fusion-fission hybrid reactor (FFHR) for the incineration of long-lived TRU and fission product has been studied by authors. Many different design concepts have been developed for the choices of reactor size, material and specific design goal. Design concept of a hybrid reactor for waste transmutation (Hyb-WT) showed very good transmutation performance [1].

However, one of the drawbacks of Hyb-WT is the large reactivity swing from BOC to EOC. Reduction of k-eff comes from the fast burning of TRU, especially Pu-fissiles. As a result, fusion power should be increased during the burnup cycle by more than 6 times.

By increasing fusion power, a large amount of tritium should be produced to satisfying self-sufficiency TBR. Therefore, loaded amount of Li is increased, however loaded amount of nuclear fuel for WT is reduced relatively in the blanket. In addition, integrity of structure material is weakened by
neutron irradiation, because increment of fusion power means that number of 14.1 MeV neutrons is increased.

Therefore, dual goal FFHR that WT and energy multiplication (EM) are combined is suggested for solving this problem. Mainly Th and depleted uranium are loaded in FFHR for EM, but DRUP fuel is loaded in this paper. DRUP fuel is produced by dry fabrication process without reprocessing after decladding of used PWR fuel. It is the same concept with DUPIC (Direct Use of spent PWR fuel into CANDU reactor) [2]. The use of DRUP gives two effects; one is fissile production during power production and the other is reduction of PWR spent fuel to be deposited. As the final outcome, WT and EM goals are satisfied by using of used PWR fuel.

Feasibility of DRUP fuel is already checked through previous research [3]. In this paper, (U-TRU)Zr fuel zone for WT and DRUP fuel zone for EM are separated in order to use the DRUP fuel more suitable through thermal system. Neutronics parameter, mass variation and waste transmutation ratio (WTR) are analysed.

Calculation model

Neutronics calculations are performed with MCNPX 2.6.0 with ENDF/B-VII.0 neutron cross section library. There are 2 design models based on Hyb-WT [1]. MCNPX modeling of Hyb-WT without DRUP (reference model) and with DRUP (modified model) are shown in Figure 1. Design parameters are listed in Table 1.

Hyb-WT without DRUP is dedicated for transmutation of TRU and long-lived fission products, and has two zones only; (U-TRU)Zr zone and FP zone. They have pin type target zone with LiPb coolant. On the other hands, there are 3 zones in modified model. WT zone with fast neutron system is designed to locate closed to plasma which emits 14.1 MeV neutron. TRU is enriched to 30 wt% in pin type (U-TRU)Zr fuel [4]. Pb-Bi coolant aimed for neutron economy. DRUP fuel of pebble type is designed in EM zone. Pebble is surrounded with graphite to spectrum softening effect. H2O coolant aimed also for spectrum softening effect. There are 2 types coolants; PbBi and H2O, therefore, coolant channels are separated on coolant types. In addition, H2O coolant affects integrity of structure material thus H2O coolant should be pressurised by low vapour pressure. As a result, DRUP fuel is cooled indirectly through H2O coolant pipe. TBZ (Tritium Breeding Zone) is designed behind of the EM zone. Li4SiO4 which has high Li number density as pebble type is loaded. Li breeder also cooled by H2O coolant with same way of EM zone. In the last region, fission product (FP) zone is designed and only Tc99, I129 and Cs135 that have long half-life and high radio-toxicity are loaded. FP zone design is same with reference model.
Neutronics analysis

Neutronic performance is listed in Table 2. Cycle length is 1 100 days as same as cycle length of Hyb-WT [1]. \( k_{\text{eff}} \) level variation of modified design is 4 948 pcm during operation period. \( k_{\text{eff}} \) value of modified design is 6 000 pcm higher compared to reference model at EOC. As shown in Figure 2, \( k_{\text{eff}} \) level is increased until 400 days by fissile production from DRUP fuel. \( k_{\text{eff}} \) level is reduced after 400 days because amount of fissile consumption is higher than amount of fissile production. Required fusion power with modified design is reduced 43 MW compared to reference model because of small \( k_{\text{eff}} \) level variation. Required fusion power is not exceed 53 MW. EM factor of modified design is also improved during operation period due to maintain high \( k_{\text{eff}} \) level. Modified design has high EM factor and low fusion power. In other words, Using DRUP fuel is beneficial for EM.

Table 1: Design parameters

<table>
<thead>
<tr>
<th>Region</th>
<th>Model</th>
<th>Thickness (cm)</th>
<th>Volume Fraction (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>(TRU-U)Zr Fuel Zone</td>
<td>Reference</td>
<td>45</td>
<td>(U-30TRU)Zr: 23.84; LiPb: 38.98; SiC: 5.94; Clad: 3.54; Bonding: 17.7</td>
</tr>
<tr>
<td>(TRU-U)Zr Fuel Zone</td>
<td>Modified</td>
<td>21.8</td>
<td>(U-30TRU)Zr: 29.92; Pb-Bi 35.85; SiC: 6.11; Clad: 13.9; Bonding: 14.22</td>
</tr>
<tr>
<td>DRUP Fuel Zone</td>
<td></td>
<td>6</td>
<td>DRUP: 27.34; C: 42.66; He: 30</td>
</tr>
<tr>
<td>Tritium Breeding Zone</td>
<td></td>
<td>3.7</td>
<td>Li4SiO4 (Si: 3.4, Li6:8.69, Li7:4.97; O: 13.65); C: 29.28, He-gas:40</td>
</tr>
<tr>
<td>Structure Wall</td>
<td></td>
<td>5</td>
<td>ODS steel(MA957):70; He-gas:30</td>
</tr>
<tr>
<td>FP Zone</td>
<td></td>
<td>29</td>
<td>Cei (129I: 0.42; 135Cs: 1.76); 99Tc: 0.82; SiC: 2.5; C: 78; He-gas:16.5</td>
</tr>
<tr>
<td>Tungsten Shield</td>
<td></td>
<td>10</td>
<td>W</td>
</tr>
<tr>
<td>B4C Shield</td>
<td></td>
<td>5</td>
<td>B: 80; C: 20</td>
</tr>
<tr>
<td>Superconductor Toroidal MF Coil</td>
<td>20</td>
<td>Nb:93.70; Sn:30; He</td>
<td></td>
</tr>
<tr>
<td>Reflector</td>
<td></td>
<td>20</td>
<td>C: 80; He-gas: 20</td>
</tr>
</tbody>
</table>
Table 2: Neutronics performances

<table>
<thead>
<tr>
<th>Model</th>
<th>Reference</th>
<th>Modified</th>
</tr>
</thead>
<tbody>
<tr>
<td>Cycle length</td>
<td>1 100 days</td>
<td></td>
</tr>
<tr>
<td>Keff [BOC / EOC]</td>
<td>0.96641/0.84</td>
<td>0.95151/0.90203</td>
</tr>
<tr>
<td>Required Fusion Power (MWth)</td>
<td>13.92 ~ 95.3</td>
<td>11.59 ~ 52.28</td>
</tr>
<tr>
<td>Average TBR</td>
<td>1.25</td>
<td>1.77</td>
</tr>
<tr>
<td>EM (Energy Multiplication factor)</td>
<td>26.19 ~ 143.51</td>
<td>47.24 ~ 172.28</td>
</tr>
</tbody>
</table>

Figure 2: Keff value and required fusion power

Isotopic mass variation of actinides during operation period is listed in Table 3. Actinides are classified into 3 types; total actinide, (U-TRU)Zr and DRUP in modified design. Because mass variation between (U-TRU)Zr fuel and DRUP fuel was significantly different. Modified design has higher fission to capture ratio than reference design. Fission reactions are dominant in thermal energy region. Because of Li6, absorber for thermal neutron is not generated from PbBi coolant.

Total mass reduction with modified design is much higher compared to reference model. Especially, uranium is reduced more on modified design. However, TRU mass variation of modified design is a half of reference model, Pu, especially fissile Pu is produced. This mass variation is different on fuel types. Mass reduction ratio of (U-TRU)Zr with modified design much higher than reference design in spite of same (U-TRU)Zr fuel. There are 2 causes, one is coolant. Coolant of reference design is LiPb, Li6 absorbs thermal neutrons. On the other hand, U238 absorbs most of thermal neutrons in modified design because of PbBi coolant. The other one is loaded DRUP fuel into core. DRUP fuel is reutilised fuel without reprocessing, it has some impurities such as FP compared to general fuel. Therefore, core spectrum is softening by DRUP fuel compared to reference design. Uranium mass is significantly reduced because of high U238 capture reaction. In addition, Pu239 produced from U238 capture reaction has high reaction rate with thermal neutron, mass of Pu and total TRU is reduced.

Uranium mass is reduced 24% of initial loading in DRUP fuel. Because U238 is 85 wt% of DRUP fuel and designed for thermal system by water coolant, most of Pu is produced from U238 capture
reaction. However, amount of SNF is reduced because amount of consumed uranium is higher than amount of produced TRU. Therefore, reutilisation of SNF could be possible through only DRUP without reprocessing.

### Table 3: Mass variation during operation period

<table>
<thead>
<tr>
<th>Model</th>
<th>Reference</th>
<th>Modified</th>
</tr>
</thead>
<tbody>
<tr>
<td>Fission to Capture Ratio</td>
<td>1.48</td>
<td>1.79</td>
</tr>
<tr>
<td>Fuel Type</td>
<td>Total</td>
<td>Total (U-TRU)/Zr DRUP</td>
</tr>
</tbody>
</table>

Mass variation (kg)

<table>
<thead>
<tr>
<th>Initial Loading</th>
<th>56 900</th>
<th>75 600</th>
<th>51 400</th>
<th>24 200</th>
</tr>
</thead>
<tbody>
<tr>
<td>Total Actinide</td>
<td>-2080</td>
<td>-9360</td>
<td>-5540</td>
<td>-3820</td>
</tr>
<tr>
<td></td>
<td>(-3.67%)</td>
<td>(-12.4%)</td>
<td>(-10.79%)</td>
<td>(-15.82%)</td>
</tr>
<tr>
<td>Total U</td>
<td>-1 050</td>
<td>-9220</td>
<td>-3390</td>
<td>-5830</td>
</tr>
<tr>
<td></td>
<td>(-2.63%)</td>
<td>(-15.4%)</td>
<td>(-9.42%)</td>
<td>(-24.37%)</td>
</tr>
<tr>
<td>Fissile Pu</td>
<td>-920</td>
<td>472</td>
<td>-979</td>
<td>1 450</td>
</tr>
<tr>
<td></td>
<td>(-10.1%)</td>
<td>(5.63%)</td>
<td>(-11.85%)</td>
<td></td>
</tr>
<tr>
<td>Total Pu</td>
<td>-926</td>
<td>-175</td>
<td>-2100</td>
<td>1930</td>
</tr>
<tr>
<td></td>
<td>(-6.36%)</td>
<td>(-1.3%)</td>
<td>(-16%)</td>
<td></td>
</tr>
<tr>
<td>Total TRU</td>
<td>-1010</td>
<td>-358</td>
<td>-2330</td>
<td>1970</td>
</tr>
<tr>
<td></td>
<td>(-5.89%)</td>
<td>(-2.31%)</td>
<td>(-15.23%)</td>
<td></td>
</tr>
<tr>
<td>Total MA</td>
<td>-117</td>
<td>-183</td>
<td>-229</td>
<td>46.6</td>
</tr>
<tr>
<td></td>
<td>(-8.05%)</td>
<td>(-8.4%)</td>
<td>(-10.59%)</td>
<td></td>
</tr>
<tr>
<td>Total FP</td>
<td>-190</td>
<td>-1040</td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>(-7.6%)</td>
<td>(-37.4%)</td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

Mass variation on LL TRU (Long-Lived: half – life ≥ 100 years) and SL TRU (Short-Lived: 100 years > half – life ≥ 10 years) is shown in Figure 3. Reduction ratio with LL TRU is significantly high. On the other hand SL TRU is produced in modified design. 70 ~ 80 % of produced SL TRU is Pu241, it is produced from Pu239 capture reaction. Although cross section with neutron of Pu241 is high, it is smaller than Pu239 and Pu240. Thus Pu241 mass is significantly increased.

**Figure 3: Mass variation on LL TRU and SL TRU**
Figure 4: Waste transmutation ratio on each nuclide

Waste transmutation ratio (WTR) on nuclides is shown in Figure 4. Mass reduction of uranium with modified design is much higher than reference design by DRUP fuel and thermalizing spectrum. Mass reduction of Pu239 and Pu240 is also high. However, most of the reaction is captured, as a result mass of Pu241 is increased to 0.8 times of initial mass. Although WTR of Am243 and Cm244 are high, amount is negligible in total TRU.

Conclusions

In this paper, when FFHR is designed, decreasing keff level is compensated by using DRUP fuel. As a result, feasibility of plasma operation is improved and operation period could be extended within design constraint condition because fusion power is reduced.

Additionally, dual goals; WT and EM are satisfied by reutilisation of SNF. Unfortunately, TRU transmutation performance is not beneficial compared to reference model. Uranium is most of DRUP and core spectrum is softening by DRUP fuel. As a result, mass of U-238, Pu239 and Pu240 are reduced, on the other hand mass of Pu241 is increased.

However, reutilisation of SNF without reprocessing could be possible through DRUP fuel in this study,because the amount of consumed uranium is higher than the amount of produced TRU. In addition, it produces energy. Pu241 production problem could be solved through operation period expansion and overcome of core spectrum thermalisation.

Acknowledgements

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References

Current status of the sodium fast reactor advanced fuel project within the Generation IV International Forum

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Abstract

The goal of the Sodium Fast Reactor Advanced Fuel Project (SFR-AF) is the investigation of Minor Actinide (Np, Am, Cm) bearing, high burn-up fuel for sodium-cooled fast reactors (SFR) to satisfy the Generation IV criteria regarding safety, economy, sustainability, proliferation resistance and physical protection. The project was started in 2007 for an initial period of 10 years within the frame of the Generation IV Sodium-Cooled Fast Reactor Programme. The original project arrangement was signed by the five partners to the SFR system arrangement; EURATOM, France, Japan, Korea and US. In the year 2015 China and Russia joined the SFR AF project as full members. After the extension of the Framework Agreement for International Collaboration on Research and Development of Generation IV Nuclear Energy Systems in February 2015 the Advanced Fuel project will be prolonged for another 10 years.

The SFR AF project aims at investigating different fuel options (oxide, metal, nitride, carbide, also inert matrix targets) for Minor Actinide incorporation and high burn-ups as well as cladding and wrapper materials withstanding high neutron doses (consequence of high burn-up) and temperatures. High burn-ups will allow uninterrupted reactor operations over longer periods of time and higher temperatures can enable higher plant efficiency. However, all of these present a challenge to the performance of fuels and cladding materials.

The project was structured in three steps: evaluation of advanced fuels and materials options, Minor-Actinide bearing fuels evaluation, and assessment of high burn-up capability of advanced fuel(s) and materials. During the first stage, fuels under consideration were mixed uranium-plutonium based driver fuels with a Minor Actinide content of a few percent in accordance with the so-called homogeneous recycling path. A first milestone, the identification of advanced fuels and materials options, was reached in 2009. During the second stage (Minor Actinide fuels evaluation) the scope was enlarged to also include the heterogeneous recycling path, with higher Minor Actinide concentrations in dedicated fuels located at the core periphery. A second project milestone, the primary selection of advanced fuels, was reached in 2015. While the final selection of fuels will be dependent upon multiple domestic factors for each country, the evaluation pointed out that experience in oxide and metal fuels is highest and adequate performance was demonstrated, while nitride and carbides are still at an early stage of development. Ferritic/martensitic materials were recommended as a starting option with the aim of transition to other advanced alloys, such as oxide dispersion strengthened steels, in the longer term. The SFR AF project has now entered into its third stage, the high burn-up fuel behaviour evaluation. With the extension of the project for another 10 years, beginning in 2017, a new project plan is under development. The paper gives an overview of the progress status and outlook of the project.
Novel nano-engineered materials boosts the fuel cycle and nuclear power applications

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Los Alamos Academy of Sciences, United States

Abstract

The actual fuel cycle is specific to an immature stage of development of nuclear technology based on intensive use of fissile actinides, high dilution of fission and radioactive by-products in the auxiliary materials, generating huge amounts of nuclear waste, and sometimes followed by hazardous reprocessing technologies, based on reactive acids, expensive solvents and complex P&T processes. The breed and burn technologies using 232Th and 238U are not really operational due to neutron economy, and these isotopes require 1.8-2 neutrons to release by fission 200+ MeV and the excess neutrons, therefore large masses of enrichment waste are added to the fission usage waste that immobilises high amounts of potentially usable actinides, adding one dimension of complexity to nuclear fuel cycle. The novel technologies based on micro-nano engineered hetero-materials have the potential to drastically improve the fuel cycle, if appropriately developed and used in advanced nuclear reactor generations, solving important actual problems. Fuel reprocessing and re-cladding is done much easier when micro-hetero structures are used that use the fission process kinematics to self-separate fission products from the fuel, preventing fuel damage due to Bragg peak, and accumulating the FP on the pellet’s cladding extremes. Using self-repairing radiation damage fractal materials, the advanced burn-up is made possible without re-cladding, assuring a near-perfect burning. In order to obtain super grade transmutation products, for special applications, using nano-cluster enhanced separation processes may use nano-hetero-clustered structures to bust strategic isotope production, and ease the separation process. These novel materials may be packed in various nuclear fuel elements, compatible with the actual nuclear reactors structures, and may gradually make the transition from the actual homogeneous fuel pellets to micro-nano-engineered hetero-materials with similar characteristics, but with superior performances. The above-mentioned materials usage will increase usability of natural uranium and thorium in B&B processes, using encapsulated process generated and burnt 239Pu and 233U, the only ones able to generate the needed neutron excess, with minimal proliferation risk. The present technologies are now in TRL-3 having a time to maturation of about 10-20 years, if appropriate R&D is done. The future nuclear reactors based on these developments will be more robust to LOCA, with higher power density admitted for the fuel rods, having near perfect burning, and reduce the need for enrichment for power production. The easy separation and partitioning will facilitate the controlled transmutation of fission products in more usable materials, transforming the actual nuclear waste in a future ore. The fuel cycle will be improved by reducing the nuclear waste, and the need for fuel extraction, nuclear power alone being able to assure Earth’s energy for the next 10 000 years based on fission only, it being the most ecological power source.
Pre-design of a target assembly for minor actinides transmutation

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² CEA Cadarache, DEN/DEC/SESC, France

Abstract

Minor actinides (MA) transmutation options for critical fast reactors are divided in two different approaches, the homogeneous one in which MA are diluted in the driver fuel and the heterogeneous one in which Ma are concentrated in UO₂ based fuel in sub-assemblies located at the periphery of the core. This latter option, named minor actinides-bearing blankets has a small impact on core behaviour, at the expense of lower transmutation performances due to the lower flux level experienced by the targets.

As such, there is an incentive to maximise the volume fraction of minor actinides loaded in the target assemblies in order to achieve optimal transmutation performances. However, a high MA fraction leads to an increase in gas – especially helium – production and negatively impacts fuel swelling and pressurisation of the fuel pin along with its thermo-mechanical behaviour. Consequently, an iterative optimisation process must be carried out during the pre-design step of such an assembly to optimise both the neutronic and mechanical performances. Using a conservative approach, we assumed that all produced gases were released in the pin free space. This production was evaluated using depletion calculations and the corresponding pin internal pressure and resistance criterion were computed. Centreline temperature of the hottest pin was also evaluated. Sensitivities to technological constraints considered into the model were also computed.

Several options were evaluated to develop a suitable assembly: smear density decrease, plenum size increase, cladding thickness increase and modification of the minor actinides volume fraction. We found that an optimum existed at 47% of fuel volume fraction, corresponding to a situation with wider pins, thicker cladding and increased gas expansion plenum height compared to standard fuel assemblies. The associated transmutation performances were in the range of -8.2 kg/TWhe, or a 33% increase in the minor actinides consumption compared to standard fuel assembly design. A pressure drop model was also implemented and it was verified that the pressure drop remained below the one of a standard fuel assembly. The new assembly design impacts on decay heat and neutron source were also assessed and it was shown that a design margin existed for optimisation with regards to these impacts.

Introduction

In the heterogeneous approach of minor actinides transmutation, the nuclei to be transmuted are concentrated in UO₂ based fuels in sub-assemblies located at the periphery of the core. This approach exhibits several advantages. It leads to a physical separation of the regular core operation from the transmutation process and more importantly allows the use of two different fuel cycles. It
also only slightly impacts core operations due to the peripheral location of minor actinides in the core. However, this specific location means that the transmutation performances are decreased compared to a case where MA are directly loaded into the core as the flux level in the targets area is lower [1].

The limited impact on core behaviour allow for compensation of the loss in performances by increasing the minor actinides mass loaded in the blankets, which counterbalance the lower efficiency of the heterogeneous approach. However, loading of minor actinides has several adverse potential effects on the fuel pins: decrease of thermal conductivity and margin to melting point, increase in gases production, especially helium due to alpha decay of short-lived isotopes such as $^{242}$Cm and a possible increase in swelling. The increase in gases production and release can lead to over-pressurisation of the pin and clad rupture. Consequently, the increase in fuel volume fraction in the assembly is limited by pin resistance. Additionally, for a given mass to be loaded, there is an incentive to increase the fuel volume fraction in the assembly in order to decrease the Am content in the fuel and thus limit the specific activity of the fuel.

An optimisation of the assembly pre-design with regards to the loaded fuel fraction and the pin thermo-mechanical design is carried out here. After presenting the tools and methods used here, an optimal design is presented and commented in terms of assembly behaviour and transmutation performances.

**Tools and methodology**

We considered heterogeneous transmutation of minor actinides using uranium oxide as support matrix, as proposed in [2]. The minor actinides isotopic vector used is given in Table 1. It is deemed representative of the vector available in France in 2035. Core calculations were carried using the homogeneous SFR V2b core designed by CEA, EDF and AREVA [3]. Assumptions used on the core management and fuel cycle can be found in [4] and are detailed below. The innermost reflector ring was replaced by target assemblies for minor actinides transmutation, as shown in Figure 1. Neutronic calculations were performed using the ERANOS deterministic code package [5] along with the DARWIN code package for depletion calculations [6]. The JEFF 3.1 nuclear data library [7] with a 33 group energy mesh was used. The analysis of target behaviour was carried out at an equilibrium situation in which the transmutation performances are identical over two consecutive cycles. To achieve this equilibrium, it was considered that plutonium produced in the targets was reloaded in the core and minor actinides production of the core loaded in the blankets, which were then topped up to 20% vol using the initial minor actinides feed.

<table>
<thead>
<tr>
<th>Table 1: Isotopic vector used for minor actinides at initial loading</th>
</tr>
</thead>
<tbody>
<tr>
<td>Element</td>
</tr>
<tr>
<td>Fraction (%mass)</td>
</tr>
</tbody>
</table>
Figure 1: 2D-RZ representation of the SFR-V2B core with minor actinides-bearing blankets and fuel assembly specifications

<table>
<thead>
<tr>
<th>Dimension</th>
<th>Length (mm)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Inner flat to flat</td>
<td>197.3</td>
</tr>
<tr>
<td>Wrapper thickness</td>
<td>4.5</td>
</tr>
<tr>
<td>Sodium thickness between assemblies</td>
<td>4.5</td>
</tr>
<tr>
<td>Cladding thickness</td>
<td>0.5</td>
</tr>
<tr>
<td>Spacing wire thickness</td>
<td>1</td>
</tr>
<tr>
<td>Gap</td>
<td>0.15</td>
</tr>
<tr>
<td>Plenum height</td>
<td>989</td>
</tr>
<tr>
<td>Fissile column height</td>
<td>1 000</td>
</tr>
</tbody>
</table>

Considering the fuel assembly specifications of Figure 2, we can plot the relationship between pin diameter and fuel volume fraction in the assembly, as shown in Figure 1. With regards to the historical data from past SFRs, pin diameters between 5.8 and 15.8 mm were considered, which corresponds respectively to the fuel pins of the FFTF in the USA and of the blanket pins of SuperPhénix in France [8]. The highest achievable fraction is then 55.6 vol% for the assembly parameters of the considered core.

Figure 2: Fuel fraction vs pin diameter in a SFR V2B assembly

Following what was done in [4], we considered a total irradiation time of 4 100 EPFD for a blanket assembly, which is twice the one of a regular fuel assembly. This is done in order to compensate for the low flux level in the periphery of core. At 2 050 EPFD, we considered that the assemblies were rotated by 180°C in order to smooth the irradiation profile in the assembly and thus the gas production profile, as shown in Figure 3. This leads to a reduction of 16% of the maximal gas production, which decreases the dimensioning constraint by a same margin.
Several parameters must be taken into account during the pre-design phase, namely the cladding resistance, the margin to fuel melting and the assembly pressure drop. All the evaluations were carried out at the end of irradiation when the gas production is maximal and in hot conditions. For neutronic calculations, the radial blanket was divided in nine zones with equal volumes and rotation occurred along the fifth one. The fuel volume fraction was calculated considering a fuel density equal to 88% of the theoretical density.

Evaluation of the cladding resistance was done using the Von-Mises stress for non-damaged material and thick tube, where the constraint is defined as \( P_m = \sqrt{3} \frac{r_{\text{ext}}r_{\text{int}}}{r_{\text{ext}}^2 - r_{\text{int}}^2} \Delta p \) with \( r_{\text{int}} \), \( r_{\text{ext}} \) the internal and external radius of the clad and \( \Delta p \) the pressure difference on the cladding. The pressure inside the pin was calculated using the DARWIN code system and the conservative hypothesis that all the fission gases and helium produced was released in the free volume of the pin. This hypothesis is consistent with the results observed in the SUPERFACT experiment [9]. Pressure level in the pin was evaluated at the end of target irradiation and under hot conditions, which is the most penalising case. As no solid swelling model for minor actinides fuels is available, solid swelling considerations were not included in this work. The innermost blanket ring was used as it is the dimensioning zone for pin design here. Fifty days refuelling outages were taken into account to factor in the helium production due to the decay of short-lived isotopes such as \(^{242}\)Cm. Gas temperature was taken at 428.7°C. The maximum admissible strain was taken as the one of ODS steel cladding and the strain criterion was defined as the ratio of \( P_m \) over this value.

![Figure 3: Effect of rotation on gas production distribution](image)

Fuel centreline temperature was calculated using the relation derived by Nishi in [10] for thermal conductivity of the UAmO\(_2\) and defined as \( \lambda (W.m^{-1}.K^{-1}) = (0.1006 + 1.664 \times 10^{-4}T(K))^{-1} \) and an inner clad temperature of 620 °C. Gap thermal conductivity was taken as 0.280 W.m.K\(^{-1}\), which is the one of pure helium at 700K taken from [11]. It should be noted that according to [10], the thermal conductivity of a sub-stoichiometric americium oxide is lower than the one of UAmO\(_2\). A minimum value of 1 W.m\(^{-1}\).K\(^{-1}\) has alternatively been considered in this paper for comparison purpose.
The maximum fuel centreline temperature was taken arbitrarily taken at 1800 °C, considering a maximal peaking factor of 1.26. Looking at Figure 5, the temperature limit is no longer a dimensioning constraint above 1800 °C. However, considering the lower melting temperature for minor actinides dioxides [12], it may be necessary to lower this limit depending on the effective melting point of the fuel and the requested margin with regards to detailed accident scenarios. However, precise calculations of the melting point of a U-MA dioxide are not available as of now. The pressure drop was computed using the Novendstern model for friction factor taken from [13] and the average power in the assembly was used. The mixing effects from the wrapper were not accounted for. The limiting value was taken as 134 kPa, which is the pressure drop for a regular fuel assembly.

Pre-design of the assembly

Thermomechanical behaviour

A preliminary calculation was done using a standard fuel assembly design as given in Figure 1 and a fuel volume density equal to 43.67%. Various geometrical designs, given in Table 2, can be considered to achieve this density. As one can see here, the pressure drop is limited in the target assemblies due to their low power and large hydraulic diameters. The main dimensioning constraint is cladding resistance. For low diameter pin, a margin exists to increase the fuel centreline temperature.

<table>
<thead>
<tr>
<th>Number of rings</th>
<th>Pin diameter (mm)</th>
<th>Fuel centreline temperature (°C)</th>
<th>Pressure drop (kPa)</th>
<th>Strain criterion</th>
</tr>
</thead>
<tbody>
<tr>
<td>8</td>
<td>10.59</td>
<td>1343</td>
<td>14.9</td>
<td>2.25</td>
</tr>
<tr>
<td>7</td>
<td>11.99</td>
<td>1488</td>
<td>11.8</td>
<td>2.55</td>
</tr>
<tr>
<td>6</td>
<td>13.83</td>
<td>1703</td>
<td>8.9</td>
<td>2.96</td>
</tr>
</tbody>
</table>

A second optimisation process based solely on pin diameter was carried out. In this case, the gap, cladding and plenum dimensions are the ones reported in Figure 1. In order to accommodate the increase in gas production, the pin size must be decreased which limits the gases production but also the maximal achievable volume fraction. The best situation is obtained for minimal-size pin with a 5.8 mm diameter and 13 rings. The corresponding fuel volume fraction is 33 % with a pressure drop of 42 kPa. It was also observed that it was not possible to obtain a design with broader pins due to the necessity of respecting the strain criterion.

Several other options can be chosen to reduce the pin pressurisation. The following were investigated here:

- Increase in the gap size in order to increase the pin free volume. This solution is limited by the fuel centreline temperature limit.
- Increase in the cladding size, which increases the pin resistance to pressurisation.
- Increase in the plenum height up to 70 cm higher by extending the expansion volume into the sodium plenum. This solution increases the pressure drop but to a small extent.

When considering only pin diameter and plenum height, an optimum situation was found at a volume fraction of 41 % with 11 rings of 7.61 mm diameter, as shown on Figure 4. The limiting factor
here is the cladding resistance due to pin pressurisation. It was also not possible to obtain a design with fewer than 10 rings due to the strain criterion.

A last optimisation process was carried out with regards to the four geometrical parameters, namely pin diameter, gap thickness, clad thickness and plenum height, the objective being to obtain the maximal fuel volume fraction in the target assembly in order to increase the transmutation performances in terms of mass consumption of minor actinides. Regarding those performances, it should be noted the relative efficiency of the transmutation process actually decreases when the fuel fraction increases due to spectral effects.

Looking at Figure 4, one can see that there is an optimum around 47 % for a 6 rings assembly. The competition between pin radius and pin pressurisation explains this maximum. For low pin diameters, the gas production is limited but the volume fraction is also limited as shown in Figure 2. On the other hand, big pin leads to increased fuel fraction but their diameter is restricted by the pin pressurisation limit, which leads to a sub-optimal filling of the assembly. The optimum on Figure 4 corresponds to a situation with 169 pins of 14.33 mm of diameter with a gap thickness of 0.15 mm and a plenum height of 168.9 cm. The cladding thickness is 0.94 mm, or 85 % higher than for standard fuel assembly. This corresponds to a 47% fuel volume fraction. Increasing the gap thickness does not prove to be an effective solution as it leads to a severe increase in fuel centreline temperature. The pressure drop for this optimised assembly design is 14 kPa, below the limiting value set at 134 kPa. The various data from each case are synthetised in Table 3.

We can see that it is possible to achieve a 12 points gain in absolute value in terms of fuel volume fraction, which translates both into an increase in the transmutation performances and a potential decrease in the assembly fabrication costs as fewer pins must be manufactured. The associated pressure drop is also smaller due to a larger hydraulic diameter in the assembly. The high linear power rate associated with big pins will also facilitate fuel restructuring and gas release, thus possibly validating a posteriori the total release hypothesis.
Table 3: Comparison of the various assembly designs

<table>
<thead>
<tr>
<th></th>
<th>Radius optimisation</th>
<th>Radius and plenum optimisation</th>
<th>Optimised pin</th>
</tr>
</thead>
<tbody>
<tr>
<td>Pin diameter (mm)</td>
<td>5.8</td>
<td>7.55</td>
<td>14.30</td>
</tr>
<tr>
<td>Number of rings</td>
<td>13</td>
<td>11</td>
<td>6</td>
</tr>
<tr>
<td>Cladding thickness (mm)</td>
<td>0.5</td>
<td>0.5</td>
<td>0.916</td>
</tr>
<tr>
<td>Gap thickness (mm)</td>
<td>0.15</td>
<td>0.15</td>
<td>0.15</td>
</tr>
<tr>
<td>Plenum height (mm)</td>
<td>98.9</td>
<td>168.9</td>
<td>168.9</td>
</tr>
<tr>
<td>Fuel volume fraction (%)</td>
<td>33</td>
<td>41</td>
<td>46.6</td>
</tr>
</tbody>
</table>

The sensitivity of the model to the constraints taken on fuel centreline temperature and on cladding strain was also evaluated, as shown in Figure 1. The fuel centreline constraint has a smaller impact than the strain constraint. This is explained by the fact that a lower maximal temperature requires a decrease in the pin radius while a lower acceptable constraint requires both a reduction of the pin radius and an increase in the cladding thickness.

Figure 5: Impacts of the constraints on the optimal fuel volume fraction

If the 1 W.m\(^{-1}\).K\(^{-1}\) minimal thermal conductivity is used as discussed before, the optimum case is slightly shifted towards smaller pins due to an increase in the fuel centreline temperature and an optimal fuel fraction of 43.5 % can be reached with 9 rings of pins with a diameter of 9.46 mm and a cladding thickness of 0.61 mm. The corresponding results are shown in Figure 6.

Figure 6: Optimal fuel volume fraction versus number of rings for a fuel thermal conductivity of 1 W.m\(^{-1}\).K\(^{-1}\)
Transmutation performances

In terms of transmutation performances, as one can see in Table 4, the increase in the fuel volume fraction leads to an amelioration of the transmutation performances by 33% in terms of specific consumption. The efficiency of the process, expressed as the total transmutation rate, is two points lower in the optimised case due to spectral effects but this is compensated by the increase in the total mass loaded.

Table 4: Comparison of the transmutation performances of the standard and optimised case

<table>
<thead>
<tr>
<th></th>
<th>Standard case, fuel volume fraction = 33 %</th>
<th>Optimised case, fuel volume fraction = 46.6 %</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Loaded mass (kg)</td>
<td>Unloaded mass (kg)</td>
</tr>
<tr>
<td>Np</td>
<td>312</td>
<td>185</td>
</tr>
<tr>
<td>Am</td>
<td>1 512</td>
<td>794</td>
</tr>
<tr>
<td>Cm</td>
<td>238</td>
<td>204</td>
</tr>
<tr>
<td>Total</td>
<td>2 062</td>
<td>1 183</td>
</tr>
</tbody>
</table>

The impacts on the target assembly decay heat and neutron source were also assessed. When considering the heterogeneous transmutation strategy, it is necessary to take into account the added constraints on cooling time, transportation, reprocessing and manufacturing due to the high activity of the target assembly. The above-mentioned parameters were evaluated at 30 days and 5 years, which are times representative of the in-core handling and fuel transportation to the reprocessing plant. The results are given below in Table 5. However, considering Figure 4, one can see that tuning the fuel volume fraction in order to comply with fuel cycle constraints is achievable while keeping higher volume fraction than with the standard assembly design.

Table 5: Impacts on assembly decay heat and neutron source

<table>
<thead>
<tr>
<th></th>
<th>Standard case, fuel volume fraction = 33 %</th>
<th>Optimised case, fuel volume fraction = 46.6 %</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Decay heat (kw/assembly)</td>
<td>30 days</td>
</tr>
<tr>
<td>Np</td>
<td>31.3</td>
<td>7.5</td>
</tr>
<tr>
<td>Am</td>
<td>41.4</td>
<td>9.9</td>
</tr>
<tr>
<td>Cm</td>
<td>44.3</td>
<td>6.71</td>
</tr>
<tr>
<td>Total</td>
<td>39.9</td>
<td>8.22</td>
</tr>
</tbody>
</table>
Conclusion and perspectives

A pre-design of a target assembly for heterogeneous minor actinides transmutation in a SFR V2b core [14] was done using the hypothesis of complete gas release. Considering the set of assumptions used and a MABB cycle of 4 100 EFPD, an optimum for the volume fraction was found around 47%, which corresponds to an hexagonal assembly with an increased plenum height and 12.31 mm of diameter pins with thicker cladding, +45% compared to standard fuel pins used in the driver assemblies. The transmutation performances of the optimised assembly were computed along with the impacts on the fuel cycle and it was shown a corresponding 25% increase in the consumption of minor actinides can be obtained. A continuation of this work will be done by using the GERMINAL code to extend the number of parameters taken into account and to validate the design. Other possible issues will also be investigated, such as swelling, fuel-cladding interaction or the important variation of power in the assembly during irradiation.

References

Fabrication and short-term irradiation behaviour of Am-bearing MOX fuels

Japan Atomic Energy Agency, Japan

Abstract
In order to investigate the effect of the addition of americium to MOX fuels on the irradiation behaviour, the “Am-1” programme is being conducted at JAEA. The Am-1 programme consists of two short-term irradiation tests of 10-min and 24-h irradiation periods, and a steady-state irradiation test. Am-bearing MOX (Am-MOX) fuels were fabricated by remote handling technique using manipulators. The short-term irradiation tests and their post-irradiation examinations (PIEs) have been successfully completed. To date, the data for PIE of the Am-MOX fuels focused on the microstructural evolution and redistribution behaviour of Am at the initial stage of irradiation have been obtained and reported. In this paper, the results obtained from the Am-1 programme are reviewed and detailed descriptions of the fabrication and inspection techniques for the Am-MOX fuels prepared for the programme are provided. PIE data for the Am-MOX fuels at the initial stage of irradiation have been accumulated. In this paper, unpublished PIE data for the Am-MOX fuels are also presented.

Introduction
Uranium (U) and plutonium (Pu) mixed-oxide (MOX) fuels containing several percent of minor actinides (MAs), such as americium (Am), neptunium (Np) and curium (Cm), are promising candidates for a closed nuclear fuel cycle system based on a fast reactor [1]. The fuels are also considered to be effective in reducing the degree of hazard and the amount of high-level radioactive waste. Although the content of MAs in MOX fuels varies according to the required mass balance for MAs, the maximum total content of MAs is considered to be 5 % [2]. Americium (Am), which is a representative of MAs, should be considered important in view of its high generation yield in spent fuel, high and lasting radiotoxicity, and expected impacts on various properties of MOX fuels. Am-bearing MOX (Am-MOX) fuels, therefore, are being studied as a first step in the promotion of research and development (R&D) of the MA-bearing MOX (MA-MOX) fuels. As a part of the research and development of MA-MOX fuels, an irradiation programme, “Am-1,” has been underway [3-5] at the experimental fast reactor Joyo of the Japan Atomic Energy Agency (JAEA). Am-1 consists of two short-term irradiation tests of 10-min and 24-h irradiation periods, and a steady-state irradiation test. Both short-term irradiation tests and their post-irradiation examinations (PIEs) were successfully completed. In this paper, the results obtained from Am-1 are reviewed and detailed descriptions of the fabrication and inspection techniques of the Am-MOX fuels prepared for the programme are provided. PIE data of the Am-MOX fuels at the initial stage of irradiation to date have been obtained and accumulated. In this paper, unpublished PIE data for the Am-MOX fuels are also presented.
Outline of Am-1 programme

The objectives of the short-term irradiation tests are to confirm whether or not fuel melts at a higher linear heating rate and to evaluate the redistribution behaviour during the initial burn-up. The steady-state irradiation test is expected to evaluate the behaviour of the fuel cladding chemical interaction (FCCI), helium (He) release behaviour, and transmutation behaviour of MAs up to medium or high burn-up. Two fuel types were prepared for the Am-1 irradiation tests. One was MOX fuel containing 3 or 5 wt% of Am (Am-MOX) and the other was MOX fuel containing 2 wt% of Np and 2 wt% of Am ((Am,Np)-MOX). Am-MOX fuels were fabricated using a remote handling technique in the shielded air-tight hot cell of the Alpha-Gamma Facility (AGF) at JAEA’s Oarai Research and Development Centre. (Am,Np)-MOX fuels were fabricated in a glove-box unit of JAEA’s Plutonium Fuel Production Facility (PFPF) at the Tokai Research and Development Centre. Two values of the oxygen to metal molar fraction ratio (O/M) of the fuel pellets were used as a test parameter for the Am-1. This paper reports the results of Am-MOX fuels.

Fabrication of Am-MOX fuels

Sintering of fuel pellets

As Am has strong gamma-ray radioactivity, the Am-MOX fuel must be fabricated in a shielded air-tight cell by remote handling technique in order to protect workers from gamma-ray exposure. The AGF was originally constructed as a PIE facility for irradiated Pu-bearing fuels. It was later equipped with a small-scale fuel fabrication unit in the hot cell for the purpose of developing a remote fuel fabrication technology for MA-MOX fuel [6]. The apparatuses which were installed in the AGF were systematically placed in the hot cell to ensure a smooth fabrication flow. Figure 1 shows the flow of the fuel pellet fabrication process. The fuel pellet fabrication procedure was composed of powder treatment by rotation milling and mixing of raw powders, compaction by uni-axial pressing, and then followed by sintering in a reducing atmosphere. This fabrication procedure, which is conventional powder metallurgy, is relatively simple and compatible with remote operation.
Since characteristics of Am-MOX fuel fabrication were not known at the beginning, fundamental fabrication tests were carried out [6]. Through trial and error, optimal sintering atmospheres, together with suitable temperature control patterns, were established for the fabrication of Am-MOX fuel pellets.

\text{UO}_2\text{PuO}_2\text{PuO}_2\text{Powder and two types of Am-bearing PuO}_2\text{powder were used as raw materials. Detailed characteristic of the raw powders has been reported in Reference [6]. Before the sintering step, these three powder types were annealed at 903 K for 2 h under a flowing gas atmosphere of dry Ar-5 \% H}_2\text{. In the furnace with a kantal heater to remove their absorbed moisture. After heat treatment, each of the powders was weighed within an error of 1 mg using an electronic balance to adjust the amount to the predetermined weight ratio. This was followed by mixing in a ball mill for 5 h at a rotating speed of 120 rpm. The ball mill pot (200 ml capacity), which can blend and pulverise powder simultaneously, was made of aluminium. The inside surface of the ball mill was lined with silicone rubber. The alumina balls (10 mm in diameter) were inserted into the ball mill together with the powder. During the ball mill, swing motion was inserted every 1.5 h. The number of swing motion cycle was 14 /min, the rotating speed and duration during swing motion were 40 rpm and 5 min, respectively. Zinc stearate was added as binder (additive rate: 0.7 wt \%) and the powder was further mixed for 30 min by swing motion. The powder was subjected to cold-pressing to obtain compacts of green pellets under a pressure of 3.84 t/cm\textsuperscript{2} for 10 s. The green pellets were pre-sintered at 1 073 K for 2.5 h under a flowing gas atmosphere of dry Ar-5 \% H}_2\text{ in the furnace with the kantal heater to remove the binder. The heating rate was 200 K/h and cooling rate was non-programmed (furnace cooling). The pellets were sintered at 1973 K for 3 h under a flowing gas atmosphere of Ar-5 \% H}_2\text{ with 2000 ppm moisture in a furnace with a tungsten mesh heater. The heating rate from room temperature to 1 073 K was 400 K/h and from 1 073 K to 1973 K was 200 K/h. The cooling rate was 200 K/h. During the cooling step, the addition of moisture was stopped below 1 073 K. The pellets were then heat treated to adjust the O/M ratio to the targeted values.}

**Adjustment of fuel pellet O/M ratio**

The pellet O/M ratio was confirmed gravimetrically at room temperature. All heat treatments were carried out under a flowing gas atmosphere of Ar-0.05 \% H}_2\text{ or Ar-5 \% H}_2\text{ by adding an appropriate amount of moisture. The sintered fuel pellets were heat treated at 1 123 K for 7 h under a flowing gas atmosphere of Ar-0.05 \% H}_2\text{ with 2 000 ppm moisture to adjust the O/M ratio to 2.00. The heating rate was 1 123 K/h and the cooling rate was approximately 1 000 K/h. After this process, the fuel pellets were heat treated at 1 473 K for 7 h under a flowing gas atmosphere of Ar-5 \% H}_2\text{ with 450 ppm moisture to adjust the O/M ratio to 1.98. For adjustment of the O/M ratio to 1.95, the pellets were heat treated at 1 573 K for 7 h under a flowing gas atmosphere of Ar-5 \% H}_2\text{ with 100 ppm moisture. The heating rate and cooling rate for the heat treatment for adjustment of O/M ratio to 1.95 and 1.98 were 1 073 K/h and approximately 1 000 K/h, respectively. By adapting the heat treatment, the O/M ratio of Am-MOX fuel pellets were appropriately adjusted within ±0.01.}
Figure 2: Change of O/M ratio in the heat treatment for 5 wt.% Am-MOX pellet to adjust O/M ratio to 1.95

Figure 2 shows O/M change during the heat treatment to adjust O/M ratio to 1.95, which was calculated from oxygen potential and chemical diffusion coefficient [7]. Pellets of the 5 wt.% Am-MOX were heated in an atmosphere of Ar-5 % H₂ with 100 ppm moisture. The calculation result shows that the O/M ratio of the pellet approached to O/M=1.95, but higher than target O/M ratio. Amount of moisture in the atmosphere was measured at inlet of the electrical furnace. So, it is considered that the moisture content inside the furnace was lower than the measured moisture content.

Material inspection of the fuel pellets

As a small amount of impurities can degrade thermal properties of the fuel, degradation such as the lowering of the melting temperature and thermal conductivity, impurities in fabricated fuel pellets must be kept at a very low level. Therefore, inspection technology for the Am-MOX fuel was also established based on that applied to standard MOX fuel. Table 1 lists material inspection items for sintered Am-MOX fuel pellets. Actinide compositions such as fissile content, Pu enrichment, ²³⁵U enrichment, and Am content were determined by a combination of alpha, gamma and mass spectrometry. Volatile, moisture and metal impurities were quantified using pressure measurement, coulometry, ICP-AES, and HPLC (high performance liquid chromatography). Alpha-autoradiograph was used to investigate the homogeneity of the fuel constituent elements. Pellet density was calculated using the measured weights, diameters and heights of sintered pellets. Pellet diameters and heights were measured using laser metrology.
### Table 1: Material inspection items and methods

<table>
<thead>
<tr>
<th>Inspection items</th>
<th>Method</th>
</tr>
</thead>
<tbody>
<tr>
<td>Fissile content</td>
<td>Combination of - alpha spectrometry</td>
</tr>
<tr>
<td></td>
<td>- gamma spectrometry, and</td>
</tr>
<tr>
<td></td>
<td>- mass spectrometry</td>
</tr>
<tr>
<td>Pu enrichment</td>
<td>Weight measurement</td>
</tr>
<tr>
<td>$^{235}$U enrichment</td>
<td>Pressure measurement</td>
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<tr>
<td>Am content</td>
<td>Coulometry</td>
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<td>O/M ratio</td>
<td>ICP-AES</td>
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<td>Volatile impurities</td>
<td>HPLC</td>
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<td>Moisture impurities</td>
<td>Dimension and weight measurement</td>
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<tr>
<td>Metal impurities</td>
<td>Alpha-autoradiograph</td>
</tr>
<tr>
<td>Halogen/carbon impurity</td>
<td>Alpha-autoradiograph</td>
</tr>
</tbody>
</table>

### Fuel pin fabrication

The sintered and inspected Am-MOX fuel pellets were inserted into a cladding tube made of austenitic stainless steel, together with UO$_2$ thermal insulator pellets and reflector components. The Am-MOX fuel pellets containing 5 wt % Am were loaded at the middle of the fuel column and 3 wt % Am-bearing MOX fuel pellets were placed at the lower and upper ends of the fuel column [4]. The fuel pins were sealed by TIG welding and then inspected by He-leak detector to confirm their airtightness/soundness. After the visual, metrological, welding inspections, the fabricated fuel pins were assembled into a fuel subassembly. Three fuel pins were fabricated in Am-1 short-term irradiation tests. Two were fuels having an O/M ratio of 1.98 and the other one was that of 1.95. The entire process of Am-MOX fuel pin fabrication was also performed by remote handling technique using a manipulator.

### Irradiation conditions

All three Am-MOX fuel pins were subjected to the 1$^{st}$ irradiation test. The reactor power was raised continuously to the targeted peak linear heating rate of 43 kW/m and kept constant for 10 minutes, and then manually shut down. After irradiation, three fuel pins were discharged and subjected to non-destructive PIEs. After the non-destructive PIEs, destructive PIEs were performed for one fuel pin with an O/M ratio of 1.98. The remaining two fuel pins were subjected to the 2$^{nd}$ irradiation test. The second irradiation test was performed at almost the same power history up to the targeted peak linear heating rate of approximately 45 kW/m and held for 24 hours. After maintaining the power at this level for 24 hours, it was decreased following the normal reactor operation procedure [8, 9]. The two fuel pins were subjected to non-destructive and destructive PIEs.
Results of post-irradiation examinations

Non-destructive examinations

After the irradiation tests at Joyo, fuel pins were transported to the Fuel Monitoring Facility (FMF) adjacent to Joyo for non-destructive examinations (NDEs), including X-ray CT scans [10], X-ray radiography, weight measurements, visual inspections, cladding outer diameter profilometry, and gamma scanning. NDE results showed that the Am-MOX fuel pins were irradiated without any failure, as was expected.

Figure 3 shows enlarged X-ray CT images of the 5 wt.% Am-MOX fuels irradiated for 10 min and 24 h on the transverse cross section at the core mid plane. The formation of central voids was found in the fuels irradiated for 10-min, and cracks were observed along the radial directions on the Am-MOX fuels. After 24-h irradiation, a central void had developed. Qualitative features of restructuring in the fuels with O/M ratios of both of 1.95 and 1.98 resemble each other. To quantitatively investigate the behaviour of fuel restructuring at the initial stage of irradiation, image analyses were conducted on the X-ray CT images to estimate the size of the central void. Figure 4 shows the axial distribution of the size of the central void obtained by analysis of X-ray CT images of 5 wt.% Am-MOX fuels irradiated for 24-h. As can be seen in this figure, there was little difference in microstructures and central void sizes between the specimens of the fuel having an O/M ratio of 1.95 and those of 1.98. This is in agreement with the data obtained from ceramography [5]. Although many studies have reported that a decrease in O/M ratio decreases the thermal conductivity of the fuel, which results in a difference in the restructuring progress, no effect of O/M ratio on fuel restructuring was found in the Am-MOX fuels at the initial stage of irradiation.

Figure 3: X-ray CT images of the 5 wt.% Am-MOX fuels irradiated in B11 for 10 min and 24 h

<table>
<thead>
<tr>
<th>Pin ID (O/M ratio)</th>
<th>10 min</th>
<th>24 h</th>
</tr>
</thead>
<tbody>
<tr>
<td>P502 (1.98)</td>
<td>![Image]</td>
<td>![Image]</td>
</tr>
<tr>
<td>P603 (1.98)</td>
<td>![Image]</td>
<td>![Image]</td>
</tr>
<tr>
<td>P601 (1.95)</td>
<td>![Image]</td>
<td>![Image]</td>
</tr>
</tbody>
</table>
**Destructive examinations**

After the NDEs, the fuel pin was punctured and sectioned to cut off the upper and lower ends, and then transported to the AGF for destructive PIEs. The Am-MOX fuel pins were cut at different axial positions into several segments. Several cross-sectional specimens were prepared by the following processes: each segment (about 20 mm in length) was impregnated with epoxy resin in vacuum and cut transversely into discs of about 5 mm in thickness; these discs were then mounted into holders using epoxy resin; and they were then ground and mirror-polished with anhydrous lubricant. The microstructure was observed using an optical microscope. Electron Probe MicroAnalysis (EPMA) of U, Pu and Am was performed using an SX-100R model (Cameca) which was specially shielded with lead and tungsten to permit the analysis of irradiated nuclear fuels. Electron acceleration potential was 25 kV and beam current was 50 nA. Three wavelength dispersive spectrometers with pentaerythritol (PET) diffracting crystals were used for the analysis simultaneously. Selected characteristic X-rays were M-alpha lines for U and Am, and the M-beta line for Pu. The secondary electron image was used to obtain information on the fuel microstructure at the locations selected for analysis and to position the electron beam. Immediately prior to EPMA of the samples a thin film of carbon was applied to their surface by vacuum evaporation to avoid electron charging effects.

According to the ceramographs obtained in this study, some cracks were observed along the radial and circumferential directions on both specimens. Plano-convex-shape lenticular pores were observed around the central void of the specimen irradiated for 10 min. Lenticular pores were also found in the vicinity of cracks which developed in this fuel pellet due to thermal stress. Crack healing occurred in some parts of both fuel pellets after the lenticular pores moved to the fuel centre along the temperature gradient. The configuration of the lenticular pores was similar to that of conventional oxide fuels irradiated for a short time at the high linear heating rate condition. No significant difference in the restructuring feature was observed between the 5% Am-bearing and 3% Am-bearing MOX fuel pellets. After the 24-h irradiation, the central void had developed to the size of approximately 1 mm in diameter. No lenticular pores were observed, and highly dense columnar grains appeared on the specimen taken from around the axial mid position of the fuel pin. In order to confirm whether or not the fuel melted at the high linear heating rate, careful observation was carried out. No sign of fuel melt was found in any of the specimens taken from the Am-MOX fuel pins irradiated for 10 min and 24 h. This implies that the thermal design for this test had a sufficient safety margin. All of the ceramographs obtained in this study are found in References [4, 5].
Figure 5 illustrates the effect of the initial O/M ratio on the radial distributions of Am and Pu after the 24-hour irradiation. The relative concentrations of Am and Pu indicated in the vertical axis were determined from the concentrations measured by EPMA divided by the as-fabricated concentrations. A relative concentration of 1 means the concentration did not change during irradiation. The data were obtained on specimens taken from the top of the axial position (X/L=0.98 or 0.99) which were located at the upper 3 wt.% Am-MOX fuel columns. As can be seen in this figure, the relative concentrations of Am and Pu increase at a position closer to the edge of the central void; and it is evident from the figure that the increase in the Am relative concentration at this position is more significant and that the maximum relative concentration is higher than that of Pu. This implied that Am accumulates more rapidly than Pu in the centre of the fuel pellet during the initial stage of irradiation. According to past studies on vapour pressure, it is reasonable to assume that the magnitude of the vapour pressures in an Am-MOX fuel of slightly hypo stoichiometric composition decreases in the order of U-bearing, Pu-bearing, and Am-bearing gaseous species. The EPMA results can be explained by assuming that the vapour pressures of the gaseous species decrease in the order given above.

The radial profiles of Am and Pu concentrations from 0.15 of relative radius to the pellet surface in the fuel having an initial O/M ratio of 1.95 were similar to those measured in the fuel with an initial O/M ratio of 1.98. The degree of increase of Am and Pu in the vicinity of the central void, however, is larger in the fuel pellets having an initial O/M ratio of 1.98. This tendency was found in the measurement results for 5wt% Am-MOX fuel specimens [5]. The axial distributions of the relative Am and Pu concentrations measured in the vicinity of the central voids reported in Reference [5] indicated that the degree of increase of Am and Pu was larger in the fuel pellets having an initial O/M ratio of 1.98 at all axial positions. This is probably due to the lower vapour transport effect of UO3 as a consequence of the low oxygen potential in the fuel with an initial O/M ratio of 1.95. However, UO3 is still considered to be the dominant gaseous species at the O/M ratio of 1.95.

Figure 5: The effect of initial O/M ratio on the radial distributions of Am and Pu after the 24 h irradiation

Temperature profile in radial direction of 3wt % Am-MOX was calculated at 43 kW/m, assuming that restructuring of the pellet did not start [7]. Figure 6 (a) shows that the pellet with O/M=1.95 was irradiated at higher temperatures, because having lower thermal conductivity as compared with the pellet with O/M=1.98. Vapour pressure at the local position in radial direction of the pellet was evaluated by the expanded Markin’s model as shown in Figure 6(b) [7]. The total vapour pressure was high in low O/M pellet. The higher vapour pressure prompted larger pore migration and central void formation, which are consistent with measurement result of the central void diameter shown in Figure 4.
Figure 6: (a) Temperature profile and (b) vapour pressure change in the radial direction of the 3wt % Am-MOX pellet irradiated at 43kW/m

Summary

Remote Am-MOX fuel fabrication technology was established at the JAEA AGF. The experience and findings obtained through the development of fabrication and inspection techniques for Am-MOX fuels will be useful as basic data for the realisation of partitioning and transmutation technologies.

Short-term irradiation tests were conducted on Am-MOX fuel. The results of non-destructive PIEs showed that all Am-MOX fuel pins were irradiated without any failure, as expected. Microstructural evolution was observed by optical microscopy, and the redistribution behaviour of constituent elements was determined by EPMA. The ceramography showed that structural changes, such as the formation of lenticular voids and the central void, occurred quickly and early in 10-min and 24-h irradiation. EPMA revealed that Am migrated to the radial centre of the fuel pellet along the temperature gradient during the initial stage of irradiation. This tendency is similar to that of Pu. The increase in the concentrations of Am and Pu in the centre of the fuel pellets is attributed to the vapour transport of fuel constituents by an evaporation-condensation mechanism in the pores. The extent of redistribution of Am and Pu was higher in the fuel with an O/M ratio of 1.98 than in that with an O/M ratio of 1.95. The irradiation behaviour at the high linear heating rate obtained in Am-1 will be useful for the modelling and design study of Am-MOX fuels.
Acknowledgements

The authors would like to express their appreciation to Messrs Takashi Hirosawa, Hiroshi Obayashi, Hiroaki Kubo and Ms Yoko Akutsu (JAEA) for fruitful discussions. The authors also greatly appreciate Messrs. Shin-ichi Sekine (E&E Techno Service), and Mitsuo Suto and Susumu Misawa (Inspection Development Company) for their technical support in the experiments, as well as all of the members of the Alpha-Gamma Section (AGS) and Fuel Monitoring Section (FMS) at the JAEA Oarai Research and Development Centre.

References

Technological investigation of the simplified MA-bearing fuel fabrication process for fast reactors and accelerator-driven systems

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Abstract

Researchers at Japan Atomic Energy Agency (JAEA) have proposed the transmutation of minor actinides (MAs) by both fast reactors (FRs) and accelerator-driven systems (ADSSs) as a way to significantly contribute to the reduction of the volume and the potential radiotoxicity of radioactive waste. Pu-U mixed oxide with a small amount of MA is adopted as fuel of FRs, while U-free nitride and mixed oxide of MA, Pu and inert element are candidates for fuel of ADSSs. JAEA researchers have also studied the simplified MOX fuel fabrication process to decrease the fabrication cost. This report describes a technological investigation of the simplified MA-bearing fuel fabrication process that prepares fuel suitable for both FRs and ADSSs. Furthermore, this report includes an analysis of the feasibility of fabricating MA macro-dispersion pellets and MA micro-dispersion pellets for ADSSs by using the simplified MOX fabrication process.

A preliminary test using Ce as a substitute for Pu and Am was done, and two target composites were fabricated: micro-dispersed composites of CeO$_2$-MgO and macro-dispersed composites of CeO$_2$ inclusions 0.2 mm diameter in an inert matrix of ZrO$_2$. The microwave heating denitration method, which is the method used in Japan’s nuclear reprocessing plants, was used to convert a mixed solution of Ce(NO$_3$)$_3$6H$_2$O and Mg(NO$_3$)$_2$6H$_2$O into a mixed-oxide (CeO$_2$-MgO) raw micro-dispersion powder, and to convert a nitrate solution of Ce(NO$_3$)$_3$6H$_2$O to CeO$_2$ raw macro-dispersion powder. For the CeO$_2$ raw macro-dispersion powder, the inert matrix of ZrO$_2$ was mixed and blended using a ball mill device without balls for crushing. The respective obtained powders were processed by a tumbling granulator for subsequent pelletising. The granulated powders were pressed into green pellets directly and smoothly using the die wall lubrication method. After sintering, metallographic examination of the two pellet types, micro-dispersion and macro-dispersion were carried out by SEM and EDX in order to evaluate the distribution homogeneity of both composites.

The flow abilities of both de-nitrated raw powders were improved with the tumbling granulator, and the Carr flow ability index was more than 70 for both. The green pellets had no defects such as cracks in their appearance. The CeO$_2$ particles were dispersed homogeneously in both macro-dispersion and micro-dispersion pellets.

As a result, it was judged that the simplified MOX fuel fabrication process is applicable to fabrication of MOX pellets for ADSSs as well as FRs.
Progress of the Czech Republic deep geological repository programme

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Abstract

Through the Radioactive Waste Repository Authority (SÚRAO) established in 1997, the Czech state fulfils its responsibilities for the disposal of all radioactive waste including spent nuclear fuel. According to the Updated Concept of Radioactive Waste and Spent Nuclear Fuel Management (hereinafter as Concept) official notice as well as to the Update of the State Energy Policy by Government taken on 15th December 2014 the deep geological repository (DGR) is to be considered a final disposal of spent fuel (the selection of candidate sites should be ready in year 2020 and the selection of a final one in 2025; the beginning of the construction is planned for 2050; commissioning of a DGR is stated for 2065).

The initial stages of DGR development involved the screening of geological conditions of the whole territory. Granites would provide the most suitable rock environment for DGR siting purposes. At the end of 2012 SÚRAO proposed seven sites for the potential geological surveys and the first stage of site characterisation work. Licences for geological survey for this stage have been in force since October 2015. The main goal of the RD&D programme is to support the siting programme for the selection of the candidate sites, to adapt a DGR design on suitable sites and to support the safety case. The construction of the Bukov Underground Research Facility in the southern section of the Rožná uranium mine at a depth of 600 m below the surface in crystalline rock environment started in 2013. The concept states that the nuclear power station operator should focus on the direct disposal of SNF in a DGR, however, it does not exclude the option of fuel reprocessing, particularly in view of the advanced fuel cycles (AFCs) of IV Generation reactors the adoption of which would reduce both the potential hazard level and volume of the waste disposed of.

The rock environment is made up of gneiss and migmatite. Two horizontal boreholes, 150 metres and 100 metres long have been drilled which will allow for the characterisation of the geological environment of the area surrounding the Bukov URL as well as for the testing of various geophysical methods, the conducting of hydraulic tests and general monitoring. It is planned that a further borehole will be drilled from the surface to the URL which will, among other things, provide information on the development of the geological, hydrogeological and geotechnical parameters of the rock massif with depth.

Without the involvement and concentration of the research capacities available in the process and the support of the whole of Czech society it will simply not be possible to fulfil this task. SÚRAO and the senior state authorities involved are well aware of this fact and will make every effort to achieve the various objectives in an atmosphere of mutual co-operation including the public, which should be described in the special Law.
Evaluating the risk of retrieving high-level waste repository during post-closure phase

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Abstract

This paper focuses on the retrievability of high-level waste (HLW). Several scenarios exist which require retrieving HLW. Nevertheless, the geological disposal for HLW is designed to have conservative safety, up to a management time of thousands years, at most inevitably entails uncertainty. Expanding cracks of host rock due to natural disaster, climate change and change of future society are some of the examples.

This work simulates the retrieval process of damaged waste packages and evaluates the associated risks. First, nuclides leakage data were studied, taking into account changing time in a given environment, in which waste packages lose their function because of irreparable damage. Second, calculations of radiation exposure to workers and ecological system during a series of retrieval processes containing excavation surface were performed, pulling out damaged waste packages.
Evaluation of minimum critical mass to inform performance requirement of partitioning and transmutation to eliminate criticality risk in a geological repository

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Abstract

Partitioning and transmutation (P&T) has the potential to eliminate the criticality risk for a deep geologic repository for high-level nuclear waste by reducing the mass of fissile material in the waste. Though criticality safety is already an integral part of the design of a geologic repository, after the repository is sealed, subsequent events are out of human control. When waste canisters fail, the nuclides that comprise the spent fuel can be rearranged by dissolution, transport and deposition by groundwater. If fissile material from multiple canisters accumulates at a single location in or away from the repository, a critical mass could be formed. This could increase the radiological dose to the biosphere and compromise the integrity of the repository.

The goal of this study is to determine the extent to which fissile material should be removed from the waste by P&T to eliminate the far-field criticality risk. P&T of spent fuel could render the criticality risk negligible, such that a deposition containing the total repository fissile inventory does not attain a critical mass. To achieve this, the study evaluates the minimum critical masses of spherical depositions, each containing uranium from a different light water reactor (LWR) spent fuel. The different spent fuels are defined by reactor type, initial enrichment, and burnup in the reactor for both uranium-oxide and mixed-oxide fuels. The minimum critical mass represents a configuration of uranium, water, and rock most favourable for criticality and implies the allowable limit of fissile material in a hypothetical repository. The minimum critical mass can be lower than 1MT for depositions from LWR spent fuel. Initial enrichment, burnup, and reactor type have a significantly greater effect on the minimum critical mass than do fuel assembly type and capacity factor. In general, the minimum critical mass is can be approximated as a linear combination of initial enrichment and burnup. This result provides a quantitative basis for the reduction of fissile material by P&T for different LWR spent fuels, ultimately informing the performance requirement for future fuel cycle technologies that aim to reduce far-field criticality risk.

† Deceased
Introduction

Any nuclear waste repository containing fissile material requires a criticality safety analysis (CSA) in order to guarantee that the repository system, including surrounding geologic formations, remains subcritical over the licensing period. Typically, the canister environment is designed to ensure subcriticality of the waste, but over time geologic processes can rearrange repository contents, potentially resulting in the accumulation of fissile material away from the repository [1,2]. Although the analysis of such scenarios requires site-specific information, a scoping analysis over environment parameters can provide a scientific basis to inform future design decisions. Such analysis focuses on determining the minimum critical mass of a fissile accumulation under conservative conditions [3] with the assumption that no critical accumulation is possible if the entire fissile inventory of the repository cannot become critical under conservative conditions.

Partitioning and transmutation (P&T) of spent nuclear fuel is often proposed as a way to ease the requirements and burden (i.e. inventory, waste toxicity) on a geologic repository [4–7]. Although the impact of P&T on overall repository performance is unclear due to the relative immobility of actinides relative to long-lived fission products [8], decreasing the mass of fissile material emplaced in the repository reduces, but does not eliminate, the risk of waste criticality.

The purpose of this study is to assess the extent to which P&T must reduce the mass of fissile material from LWR spent fuel – currently destined for direct disposal in a geologic repository – to eliminate the risk of far-field criticality. Under the assumed conditions, the difference between the minimum critical mass and the repository inventory represents the reduction in fissile material in the repository that must be achieved to eliminate the risk of criticality. In this work, the minimum critical masses of depositions containing fissile materials from various types of LWR spent fuel are calculated using neutronic analysis.

Model and methodology

This study assumes a fictitious model to represent an underground deposition outside of a geologic repository. The repository and surrounding geologic formations are assumed to be water-saturated. Over time, groundwater will corrode the canisters, dissolve the waste and transport radionuclides away from the repository. Due to different subsurface transport properties, different elements will travel at different rates. Uranium is more mobile than other actinides and may become separated from them and other neutron poisons as it travels with groundwater [9]. Over time, higher actinides in the waste will decay to uranium isotopes, contributing to the plume. In particular, Pu-239 decays to U-235 ($t_{1/2} = 24,100$ years), adding to the fissile material in the plume. Assuming no criticality or other disrupting event takes place in the repository environment or along the transport pathway, the uranium could encounter geologic conditions that cause precipitation of the plume.

The temperature of the deposition is assumed to be 20°C, although the ambient temperature will most certainly be elevated due to the geothermal gradient and the decay heat from radionuclides in the groundwater. Complex simulations involving coupled heat and mass transfer calculations and site-specific information are required to accurately determine the temperature. These are outside the scope of this work, and assuming a lower temperature is expected to be conservative as the resonance Doppler effect with increasing temperature reduces reactivity [10].

Conservatively, it is assumed that the entire inventory of uranium in the repository feeds into a single spherical deposition; further, any plutonium that has not decayed to uranium at the time of analysis is lumped into the deposition as well. The uranium and plutonium are assumed to
precipitate uniformly as oxides. Together, the uranium and plutonium will be referred to as heavy metal (HM).

A schematic of the deposition employed in the numerical model is shown in Figure 1. The deposition is assumed to be a homogeneous mixture of host rock, water and heavy metal. A 1-m thick reflector made of host rock surrounds the deposition. The host rock of the deposition is assumed to be sandstone (density 2.71 g/cm$^3$) with composition given in Table 1 [10]. The composition of heavy metal is determined using ORIGEN-ARP [11] as a function of fuel type (uranium dioxide and MOX LWR spent fuels), initial fissile enrichment, burnup, assembly design and reactor type. U-234 and U-236 concentrations in fresh enriched uranium fuel are set as 0.0089 and 0.0046 times U-235 enrichment, respectively [12]. The initial enrichment of MOX fuel is varied by the weight fraction of plutonium of which the initial vector is shown in Table 2 [13]. Fresh MOX fuel also includes americium-241 with 1% loading relative to the plutonium weight fraction. The remainder is natural uranium with U-235 enrichment of 0.71%. Depletion calculations assume three cycles for PWR fuel [14] at specific power of 38 MW/tHM [15] and four cycles for BWR fuel [16] at specific power of 26 MW/tHM [17]. The moderator density of the BWR is fixed at a length-average value of 0.438 g/cm$^3$ [18]. The capacity factor is set at 70% corresponding to the downtime between full power cycles. This value is chosen to reflect that (pre-Fukushima) Japanese capacity factors are in the range of approximately 60-80% [19]. After irradiation, decay takes place over 200 050 years: 50 years of cooling before emplacement and 200 000 years in the repository and in transport to the deposition.

The deposition model was simulated with MCNP v.6.1 [20] using ENDF/B-VII.0 cross section library at 300K for uranium and plutonium isotopes. The cross sections used for other materials are taken from a combination of ENDF/B-VII.0, ENDF/B-VI.8, and ENDL92 cross section libraries.

The amounts of rock, water and heavy metal in the deposition are described using two free variables: (1) void volume fraction (VVF) that represents the porosity of the host rock and the amount of space in the deposition filled with water and heavy metal—rock fills the remaining volume (1-VVF) in the deposition; (2) heavy metal volume fraction (HMVF) that describes the fraction of the void space (porosity) that is filled by heavy metal—the amount of water is given by VVF-HMVF. An iterative search was conducted varying VVF and HMVF to determine the minimum mass of heavy metal required to reach criticality—the criticality criterion was conservatively set as $k_{\text{eff}} = 0.98$. The minimum critical mass is calculated for combinations of fuel type, initial enrichment, burnup and reactor types as listed in Table 3. The total HM inventory in the repository is assumed to be 32 000 t, the amount of Japanese spent fuel that will either be reprocessed or emplaced in a geologic repository [21]. This provides an upper bound for critical mass in this model.

**Figure 1:** Schematic of the deposition model simulated in MCNP [10]
### Table 2: Composition of sandstone host rock in the far-field deposition [10]

<table>
<thead>
<tr>
<th>Composition</th>
<th>Weight fraction (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>SiO$_2$</td>
<td>78.70</td>
</tr>
<tr>
<td>TiO$_2$</td>
<td>0.25</td>
</tr>
<tr>
<td>Al$_2$O$_3$</td>
<td>4.80</td>
</tr>
<tr>
<td>Fe$_2$O$_3$</td>
<td>1.10</td>
</tr>
<tr>
<td>FeO</td>
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</tr>
<tr>
<td>MnO</td>
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</tr>
<tr>
<td>MgO</td>
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</tr>
<tr>
<td>CaO</td>
<td>5.50</td>
</tr>
<tr>
<td>Na$_2$O</td>
<td>0.45</td>
</tr>
<tr>
<td>K$_2$O</td>
<td>1.30</td>
</tr>
<tr>
<td>H$_2$O</td>
<td>1.30</td>
</tr>
<tr>
<td>P$_2$O$_5$</td>
<td>0.08</td>
</tr>
<tr>
<td>CO$_2$</td>
<td>5.00</td>
</tr>
</tbody>
</table>

### Table 3: Isotopic fractionation of plutonium in MOX fuel [13]

<table>
<thead>
<tr>
<th>Isotope</th>
<th>Weight fraction (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Pu-238</td>
<td>1.5</td>
</tr>
<tr>
<td>Pu-239</td>
<td>60.1</td>
</tr>
<tr>
<td>Pu-240</td>
<td>24.5</td>
</tr>
<tr>
<td>Pu-241</td>
<td>8.8</td>
</tr>
<tr>
<td>Pu-242</td>
<td>5.0</td>
</tr>
</tbody>
</table>

### Table 4: Summary table of cases for parametric burnup study

<table>
<thead>
<tr>
<th>Fuel type</th>
<th>UO$_2$</th>
<th>UO$_2$</th>
<th>UO$_2$</th>
<th>UO$_2$</th>
<th>UO$_2$</th>
<th>MOX</th>
<th>MOX</th>
</tr>
</thead>
<tbody>
<tr>
<td>Reactor type</td>
<td>PWR</td>
<td>PWR</td>
<td>BWR</td>
<td>BWR</td>
<td>BWR</td>
<td>PWR</td>
<td>BWR</td>
</tr>
<tr>
<td>Fuel assembly type</td>
<td>W17x17</td>
<td>CE14x14</td>
<td>GE8x8</td>
<td>GE9x9</td>
<td>ATRIUM-9</td>
<td>17x17</td>
<td>8x8</td>
</tr>
<tr>
<td>Moderator density (g/cm$^3$)</td>
<td>0.756</td>
<td>0.756</td>
<td>0.438</td>
<td>0.438</td>
<td>0.438</td>
<td>0.756</td>
<td>0.438</td>
</tr>
<tr>
<td>Specific power (MW/tHM)</td>
<td>38</td>
<td>38</td>
<td>26</td>
<td>26</td>
<td>26</td>
<td>38</td>
<td>26</td>
</tr>
<tr>
<td>Cycles</td>
<td>3</td>
<td>3</td>
<td>4</td>
<td>4</td>
<td>4</td>
<td>3</td>
<td>4</td>
</tr>
</tbody>
</table>
Results

The minimum critical mass for the far-field deposition of fissile material stemming from LWR spent fuel is calculated in the parameter space of initial enrichment and discharge burnup. Figure 2 shows the base-10 logarithm of minimum critical mass for heavy metal from UO$_2$-fuelled PWR spent fuel. The contour lines are generated using the OriginPro plotting software [22] and represent constant minimum critical mass. The contours corresponding to minimum critical masses of 0.1, 1.0, 10, and 100 MTU are marked with bold dashed lines and labelled with the base-10 logarithm of the minimum critical mass value. The black dots mark points for which MCNP calculations were performed. The colour band at right describes the colours on the plot. The plot boundary and empty space in the low-initial enrichment, high-burnup region represents the fact that the fissile material from these spent fuels cannot form a critical mass even with the entire inventory of the repository.

Figure 2: Minimum critical mass for deposition containing heavy metal UO$_2$-fuelled PWR (W17x17)

The contours for 0.1, 1.0, and 10.0 MT are labelled and highlighted.

The minimum critical mass is smaller when spent fuel initial enrichment is higher and burnup is lower; such spent fuels may exist due to accidents or early fuel failures. This general tendency can be explained by considering the amount of fissile material left in the spent fuel after reactor operation. The minimum critical mass contours are linear in this parameter space because they follow the contours for constant fissile enrichment (sum of weight fractions of U-233, U-235, and Pu-239) of the heavy metal, as shown in Figure 3. This indicates that fissile enrichment of the heavy metal is the dominant factor affecting the minimum critical mass. The linearity of the contours in Figure 3 reflects the fact that LWR fuel reactivity is linear in the same parameter space [12,14].
Figure 3: Fissile enrichment of the heavy metal in the deposition in the space of initial enrichment and burnup for UO$_2$-fuelled PWR (W17x17)

The dependence of minimum critical mass on the reactor type and fuel type can be discerned by focusing on a specific initial enrichment. Figure 4 shows the minimum critical mass for depositions from various types of spent fuel initially enriched to 4.0 wt.% (fissile). The points can be arranged into four groups considering both fuel type (UO$_2$ vs. MOX) and reactor type (PWR vs. BWR); assembly type is not significant. The heavy metal from PWR fuel yields a smaller minimum critical mass than that from BWR fuel because it is more reactive. The increased reactivity is due to increased conversion of U-238 to Pu-239 in the ORIGEN calculations for PWR, a result of the cross sections calculated for the problem.

Figure 4: Minimum critical mass plotted against discharge burnup for 4.0% initially enriched UO$_2$- and MOX-fuelled PWR and BWR
In Figure 4, at low discharge burnup, the heavy metal from MOX spent fuel has greater minimum critical mass than that from the UO$_2$ spent fuel. However, at higher burnup, the heavy metal from MOX spent fuel has a smaller minimum critical mass than that from the UO$_2$ spent fuel for both PWR and BWR. As burnup increases, the minimum critical mass of the MOX difference in minimum critical mass between heavy metals from UO$_2$ and MOX spent fuels increases. The cause of this difference is also explained by a difference in the fissile enrichment; the MOX heavy metal contains more fissile material at higher burnup. As burnup is increased, the fissile enrichment of the heavy metal from MOX spent fuel becomes even greater than that from UO$_2$ spent fuel. Figure 5 shows the difference in fissile enrichment between heavy metals from MOX and UO$_2$ spent fuels as a function of the initial fissile enrichments for different values of burnup. The plot includes configurations that could not become critical (in particular, heavy metal from UO$_2$ spent fuel with initial enrichment below 3% and burnup greater than 40%). As burnup increases, the heavy metal from MOX spent fuel becomes increasingly more reactive relative to the heavy metal from UO$_2$ spent fuel. At low burnup with high initial enrichment, there is more fissile material in the heavy metal from UO$_2$ spent fuel than in that from MOX spent fuel.

Figure 5: Difference in fissile enrichment (wt.% fissile) as a function of initial enrichment between heavy metals from UO$_2$- and MOX-fuelled PWR for different burnup

Fissile enrichment is the primary factor that determines minimum critical mass. However, other variables have a smaller effect. For example, the isotopic composition of the heavy metal will affect neutronics. Two heavy metals that have equal fissile enrichment but different initial enrichment and discharge burnup will have differences in composition that are burnup-dependent. The non-fissile affects are primarily from neutron absorbers, which act as poisons in the heavy metal and increase the minimum critical mass. Over the course of burnup, certain neutron-absorbing isotopes of uranium and plutonium are accumulated by neutron capture on precursors. In particular, Uranium-236 in the heavy metal is generated in appreciable quantities from the absorption of neutrons on U-235 in the reactor and, in the time between emplacement and criticality, from the alpha decay of Plutonium-240 (t$_{1/2}$ = 6 561 years).

The minimum critical mass of heavy metal from uranium-PWR SNF with equal fissile enrichment is plotted against discharge burnup in Figure 6. To generate heavy metal compositions with equal
fissile enrichment, the initial enrichment is determined by iterative ORIGEN calculations to find the value that yields the desired fissile enrichment 200,000 years after emplacement.

**Figure 6:** Minimum critical mass versus discharge burnup of depositions with equal heavy metal fissile enrichment (from UO$_2$-fuelled PWR, W17x17)

![Graph showing the trend of minimum critical mass versus burnup](image)

The trend is very nearly linear. The discrepancies (most notable where burnup = 30 and 50 GWd/MTU) are due to error in the determination of critical mass by Monte Carlo iterations. As burnup is increased, the minimum critical mass increases as well due to the buildup of neutron poison uranium and plutonium isotopes, primarily U-236.

**Discussions**

These results have many implications for P&T from the perspective of repository criticality safety. First are the values of the minimum critical masses themselves. LWR spent fuel can be characterised as having burnup [GWd/MTU] to initial enrichment [wt.% U-235] ratio of between 9 and 12. Within those bounds (on Figure 2), this model demonstrates that minimum critical masses of less than 1 MT could be possible. For the 32,000 MTU estimated Japanese SNF inventory potentially destined for a repository, this means that more than 99.997% of fissile material (1/32000) would need to be removed by P&T to eliminate the criticality risk under these conditions.

Which spent fuels are most in need of treatment by P&T from the perspective of criticality safety? Those with high initial enrichment and low burnup are of primary concern. Fortunately, these will also be attractive to P&T because of their high fissile content. The relationships between the minimum critical mass behaviours of heavy metals from different types of spent fuels can provide important insights. Of particular relevance to potential P&T fuel cycles is that heavy metal from MOX fuel has a smaller minimum critical mass than that from UO$_2$-fuel and that this difference between the minimum critical masses of heavy metals from MOX and UO$_2$ heavy metals becomes greater as burnup increases. MOX fuel is envisioned to be a high-burnup fuel, so for fuel cycles that involve once-through burning of MOX fuel in LWRs, this indicates an elevated repository criticality risk. From the criticality safety standpoint, MOX spent fuel should warrant a higher priority for P&T (than UO$_2$-spent fuels).
It should be noted that even if all LWR spent fuel is treated by P&T, fissile material will still be sent to the repository because separation technology is not 100% efficient. Further, depending on the fuel cycle employed, fissile material may become too contaminated with neutron poison isotopes to be recycled, requiring more advanced technology for further transmutation or disposal of that material in the repository. Current separations efficiencies are in the range of 99.98% for uranium and plutonium isotopes. However, because P&T will drastically increase the burnup of fuel over the course of many cycles, it is possible that buildup of non-fissile and absorbing isotopes in the uranium and plutonium could eliminate the risk of criticality from fissile material in waste streams. In addition, reactor technology included in a P&T fuel cycle may improve to the point where sufficient burnup can be achieved to significantly reduce or eliminate the risk of criticality for even once-through spent fuel.

Conclusions and future works

P&T can significantly reduce the amount of fissile material in nuclear waste destined for a geologic repository. This has significant implications on repository criticality safety, which is an important part of the performance assessment. In order to study the potential effect of P&T on criticality risk, the minimum critical mass of heavy metal from the once-through LWR fuel cycle is evaluated for different types of spent fuel in the parameter space of initial enrichment and discharge burnup. This provides a measure for the extent to which P&T would have to remove fissile material from the waste emplaced in a repository in order to eliminate the risk of criticality. Minimum critical masses of heavy metal stemming from LWR spent fuel direct disposition can be as low as 1 MT for average spent fuels. Heavy metal from MOX spent fuels has a higher criticality risk than that from uranium spent fuels. Though the minimum critical mass contours (e.g., Figure 2) are generated for only one type of spent fuel, the repository will be filled with many different kinds of waste that could contribute uranium and plutonium to the deposition as well. The linear tendency of these contours implies that mass averaging could account for the blending of different types of spent fuel and nuclear waste.

The calculation of the minimum critical mass relies on many assumptions, many of which were meant to be conservative. As such, these results should be kept in perspective with the conditions under which they were generated and the assumptions should be considered critically. First, the configuration of heavy metal, host rock and groundwater is important. In this work, these materials are assumed homogeneously distributed. However, other studies have shown that different arrangements – in particular, heterogeneous deposition in parallel fractures – can lead to smaller minimum critical masses [10]. In reality, the heavy metal will precipitate randomly and unevenly in the complex network of pore space available in the host rock. Whether a deposition in random fractures is more or less reactive than one in a homogeneous or parallel-fractured system is unclear. The host rock itself may not be sandstone, which has low concentrations of neutron-absorbing minerals. A rock with elevated iron content could decrease deposition reactivity, resulting in greater minimum critical masses.

Perhaps the most conservative assumption in the model was the range of possible porosities (VVF), which was considered from 0–80%. A geologic formation with porosity of ~80% should not be considered rock but rather a highly fractured zone in which large amounts of space have been isolated. Alternatively, a deposition with VVF=80% could arise if the incoming plume removes and replaces material in the formation. In general, as VVF increases, the critical mass decreases due to increased moderation by water relative to the host rock. Numerically, the minimum critical mass always occurs at the maximum porosity because water is a stronger moderator than host rock and
the maximum porosity leaves the most room for optimum organisation of water and heavy metal. If the maximum porosity is decreased, the minimum critical mass increases significantly, and the critical region in Figure 2 decreases in size.

The primary thrust of future work will be to quantify the uncertainties associated with minimum critical mass model. With robust uncertainty analysis, this aspect of a CSA could then be utilised to generate results and compare them with regulation criteria. In addition, further analysis into the implications of advanced reactors and fuel cycles on criticality safety can and should be done.

References


Feasibility of experiments in the TAPIRO reactor to improve minor actinides nuclear data

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Abstract

The main challenge associated with the R&D on nuclear waste management is the radiotoxicity reduction of the nuclear spent fuel, which is mainly due to some elements, i.e. plutonium and other minor actinides (MAs) such as neptunium, americium and curium. By fissioning these elements in appropriate nuclear reactors, they can be eliminated. A deeper knowledge in the nuclear data of these MAs is of primary importance in order to deal with their transmutation.

Focusing on this goal, the activation of OSMOSE sample in TAPIRO (AOSTA) campaign, under the aegis of the NEA Expert Group on Integral Experiments for Minor Actinide Management, is oriented to analyse the feasibility of a MAs irradiation campaign in the TAPIRO Italian fast source research reactor.

The feasibility studies were carried out by calculations modeling the irradiation, in different TAPIRO irradiation channels, of some samples, supplied by CEA and coming from the French experimental campaign OSMOSE, containing different types and contents of MAs, to improve the knowledge on MAs capture cross sections.

The OSMOSE sample series are enriched by AmO\textsubscript{2} samples from IRMM (EC-JRC Institute for Reference Materials and Measurements), currently used in the AMSTRAMGRAM programme in MINERVE, as well as a complementary study of miniature fission chambers loaded with MA deposits for spectral indices measurements in tangential channels and in the thermal column.

Another open issue is the need to figure out how the response of the MAs reaction rates, essentially capture, is linked to the TAPIRO copper reflector nuclear data.

In this work, the sensitivity coefficients of some MAs capture rates to Cu cross section variations have been preliminary estimated by means of both generalised perturbation theory (GPT) and direct approach by deterministic calculations.

Introduction

The main issue of R&D activities for a sustainable use of nuclear fission energy is certainly the development of a reliable fuel cycle and a safe management of radioactive wastes. The main contribution to nuclear wastes is given by the spent fuel of the nuclear power plants, and the hazard from the spent fuel is mainly due to a few chemical elements, in particular plutonium and some minor actinides (MAs) such as neptunium, americium and curium, plus some long-lived fission
products such as iodine and technetium. In this framework, R&D activities have shown that one of the promising approaches to the appropriate management of MAxs in order to reduce their risks is to transmute them in nuclear systems. However, due to a lack of experimental data, mainly regarding the accuracy of MAxs nuclear data, it is not easy to establish a detailed design of transmutation systems with reliable accuracy. In the frame of the NEA Expert Group on Integral Experiments for Minor Actinide Management [1] a joint collaboration between ENEA (Italian National Agency for New Technologies, Energy and Sustainable Economic Development) and CEA (French Alternative Energies and Atomic Energy Commission) was established with the aim to study the feasibility of a MAxs irradiation campaign in the TAPIRO fast neutron source research reactor located at the ENEA Casaccia centre [2]. On the basis of neutron transport calculation results, obtained by both deterministic and Monte Carlo methods, an estimate of the irradiated samples counting levels from the activation of OSMOSE sample in TAPIRO (AOSTA) experimental campaign has been evaluated in previous works [3].

This paper provides results obtained by calculations modeling the irradiation, in different TAPIRO irradiation channels, of some CEA samples from IRMM coming from the French AMSTRAMGRAM programme in MINERVE [4]; each sample was loaded with different contents of MAxs. On the basis of neutron transport calculations results, obtained by both the deterministic ERANOS [5] and the Monte Carlo SERPENT [6] codes, an estimate of these irradiated samples counting levels, taking into account both geometry and efficiency of the counting system, is provided.

Another open issue to carry out the experimental campaign is the need to figure out how the response of the MAxs reaction rates, essentially capture, is linked to copper reflector nuclear data uncertainties. To preliminary address this issue, in this work an analysis of some MAxs capture reaction rates sensitivity due to a 2% copper density reduction in the whole TAPIRO reflector is carried out. The 2% variation is chosen to be representative of an uncertainty on the nuclear data for copper. Calculations are performed by means of generalised perturbation theory (GPT) [7], implemented in the deterministic code ERANOS. The study has involved two different MAxs isotopes: Am$^{241}$ and Np$^{237}$. GPT results have been compared with those coming from a direct approach.

The TAPIRO fast neutron source research reactor

TAPIRO (TAratura PIna Rapida a potenza zERo — Fast Pile Calibration at 0 Power) is a fast neutrons source research reactor located at C.R ENEA CASACCIA (Italy). The project, entirely developed by ENEA staff, is based on the general concept of AFSR (Argonne Fast Source Reactor – Idaho Falls). It was built to support an experimental programme on fast reactors and it is in operation since 1971. TAPIRO is currently used, in addition to education and training, for experimental programmes in support of different research fields such as nuclear data, nuclear fusion, aerospace industry.

The reactor has a maximum power of 5 kW with a neutron flux around $4 \times 10^{12}$ n/(cm$^2$·s) in the centre of the core. The core is cylindrical with a diameter of about 12 cm and a similar height. It is made by metallic uranium (98.5 % uranium and 1.5 % molybdenum) with an enrichment of 93.5 % in U$^{235}$. It consists of 2 parts: the upper part is fixed while the lower one is movable. The core is surrounded by a double layer of a copper reflector and by an external borate concrete biological shield. The core is cooled by helium. The reactor is equipped with 2 shim rods, 2 safety rods and a regulating rod. These rods are made of the same material of the reflector, i.e. copper, and the reactor is controlled increasing or reducing the neutron leakage. The system has different experimental channels with various diameters. A horizontal section of the reactor is shown in Figure 1.
Energy averaged capture cross sections evaluation

Neutron transport calculations have been performed by two different methodologies, deterministic by the ERANOS code and stochastic by the Monte Carlo SERPENT code. In Figure 2 the calculation models are shown.

![Figure 1: TAPIRO horizontal section](image)

**Figure 2:** ERANOS and SERPENT TAPIRO calculation models

(a) - RZ ERANOS model  
(b) - SERPENT model

Neutron fluxes, energy spectra and reaction rates have been evaluated for several minor actinides in correspondence of different radial positions in the diametral experimental channel (cf. Figure 1). Then, position dependent average microscopic capture cross sections have been calculated to predict the impact on their values provided by the spectral variations across the system. Average capture microscopic cross sections \( \bar{\sigma}_c(r) \) are defined as:

\[
\bar{\sigma}_c(r) = \frac{\int \sigma_c(r, E)\phi(r, E) dE}{\int \phi(r, E) dE}
\]  \hspace{1cm} (1)
where $\varphi$ is the neutron flux. As an example, the behaviour of the average capture microscopic cross sections for the minor actinide Am$^{241}$ across the system, calculated with both ERANOS and SERPENT codes, is shown in Figure 3 [3].

**Figure 3:** Am$^{241}$ microscopic capture cross section

A satisfactory agreement can be observed, also for the other samples, between ERANOS and SERPENT codes results.

**Evaluation of the counting rates after irradiation of the IRMM samples**

The IRMM samples, shown in Figure 4, contain Am$^{241}$ only in a single aluminium sheath. The matrix for all the samples is composed by AmO$_2$.

**Figure 4:** IRMM AmO$_2$ samples for the AMSTRAMGRAM programme

Due to the samples diameter, their irradiation can only be performed in the TAPIRO tangential channel. The calculated counting rates on Am$^{242m}$ are provided in Table 2. Only one sample position has been considered along the diametral channel: $r=12.07$ cm (near the core), see Figure 2 (a). In correspondence of such position the activity values for each IRMM sample have been calculated by the FISPACT code [8]. The amount of Am in each sample is high enough to consider only a daily irradiation characterised by 5 hours of irradiation and 2 hours of cooling. Activity values have been evaluated after these last 2 hours of cooling.
The results, obtained using the ERANOS neutron flux results for this preliminary analysis, are shown in Table 1.

### Table 1: Energy averaged cross sections and activity levels for each IRMM sample in TAPIRO

<table>
<thead>
<tr>
<th>AMSTRAMGRAM samples</th>
<th>Position</th>
<th>$r = 12.07 \text{ cm}$</th>
<th>$\phi [\text{n/cm}^2 \cdot \text{s}]$</th>
<th>$\sigma [\text{barn}]$</th>
<th>Activity level [A [Bq]]</th>
</tr>
</thead>
<tbody>
<tr>
<td>IRMM_1</td>
<td></td>
<td>7.44E+11</td>
<td>1.162</td>
<td>1.096E+07</td>
<td></td>
</tr>
<tr>
<td>IRMM_2</td>
<td></td>
<td>1.433E+07</td>
<td>1.371E+07</td>
<td></td>
<td></td>
</tr>
<tr>
<td>IRMM_3</td>
<td></td>
<td>1.393E+07</td>
<td>1.371E+07</td>
<td></td>
<td></td>
</tr>
<tr>
<td>IRMM_4</td>
<td></td>
<td>1.401E+07</td>
<td>1.393E+07</td>
<td></td>
<td></td>
</tr>
<tr>
<td>IRMM_5</td>
<td></td>
<td>1.409E+07</td>
<td>1.401E+07</td>
<td></td>
<td></td>
</tr>
<tr>
<td>IRMM_6</td>
<td></td>
<td>1.431E+07</td>
<td>1.409E+07</td>
<td></td>
<td></td>
</tr>
<tr>
<td>IRMM_7</td>
<td></td>
<td>1.431E+07</td>
<td>1.431E+07</td>
<td></td>
<td></td>
</tr>
<tr>
<td>IRMM_8</td>
<td></td>
<td>1.441E+07</td>
<td>1.431E+07</td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

The counting rate [counts/s] for each detector have been evaluated by the relationship:

$$C = A \cdot I_\gamma \cdot \varepsilon$$  \hfill (1)

where: $A$ activity level [Bq], $I_\gamma$ intensity of the $\gamma$ or X emission [%], $\varepsilon$ geometric efficiency of the detector [%].

The geometric efficiency $\varepsilon$ depends by the detector type and by the overall experimental geometrical arrangement. In our case a n-type coaxial HPGe detector made of high-purity germanium, showing high precision and efficiency for both $\gamma$ and X-rays in the energy range 3 keV÷10 MeV, was used. The geometric efficiency has been evaluated by Monte Carlo MCNP code [9] modeling the arrangement of the counting system (Figure 5).
Table 2 summarises the obtained results. In particular, for each sample are shown the γ or X-rays characteristics (energy and intensity), the geometric efficiency and the counting level obtained by relationship (1).

Table 2: Counting rates for the selected AMSTRAMGRAM samples in TAPIRO (ERANOS neutron fluxes)

<table>
<thead>
<tr>
<th>AMSTRAMGRAM Samples</th>
<th>Position</th>
<th>$\phi \ [n\cdot cm^{-2}\cdot s^{-1}]$</th>
<th>$\gamma_{242}$ $E_{X_{\alpha1}} \ [keV]$</th>
<th>$X_{\alpha1}$ Intensity [%]</th>
<th>$\epsilon$ Detection [%]</th>
</tr>
</thead>
<tbody>
<tr>
<td>IRMM_1</td>
<td>C [cps]</td>
<td>668</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>IRMM_2</td>
<td>C [cps]</td>
<td>874</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>IRMM_3</td>
<td>C [cps]</td>
<td>836</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>IRMM_4</td>
<td>C [cps]</td>
<td>850</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>IRMM_5</td>
<td>C [cps]</td>
<td>854</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>IRMM_6</td>
<td>C [cps]</td>
<td>873</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>IRMM_7</td>
<td>C [cps]</td>
<td>867</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>IRMM_8</td>
<td>C [cps]</td>
<td>859</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>IRMM_9</td>
<td>C [cps]</td>
<td>873</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>IRMM_11</td>
<td>C [cps]</td>
<td>879</td>
<td></td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

The counting rate levels shown in Table 2 seem to predict the feasibility of the AOSTA experimental campaign although, as evident, a more detailed analysis is needed in the next future to confirm these promising preliminary results. In particular, actual experimental problems could be the
discrimination of interesting peaks (like Kα1 peak of Am$^{242m}$) among a lot of peaks including the background coming from Am$^{241}$. In this connection, in the future it will be interesting to simulate the whole γ spectrum in the 100 keV range, or at least calculate the count rate coming from Am$^{241}$ peaks, especially the 102.98 γ ray.

**Sensitivity coefficients for copper density reduction**

The TAPIRO research reactor is well suited for minor actinides studies due to its particular spectrum variety starting from a hard spectrum in the central core, near to fission one, an intermediate spectrum in the copper reflector up to a moderated spectrum in the biological shielding.

The reliability and significance of the measurements carried out in TAPIRO are also influenced by the presence of a copper reflector, a peculiarity of this reactor leading to the variety of spectral zones discussed above. Therefore, to assess the influence of the uncertainty on the reflector properties on the MA measures, the capture reaction rates sensitivities for two different MAs, Am$^{241}$ and Np$^{237}$, after a 2% copper density reduction in the whole reflector, have been evaluated. Results for both the MAs are in correspondence of four different positions along one of the TAPIRO experimental channels, the diametral channel (at axial reactor mid-plane).

The four different positions analysed along the diametral channel are: r=0.5 cm near the core centre (P1), r=12.07 cm near the core boundary (P2), r=24.58 cm at about the reflector centre (P3) and r=45.5 cm at the entrance of the thermal column (P4).

The energy integrated capture rates variations (total sensitivity), obtained by means of generalised perturbation theory (GPT) [7] using a 49 energy groups grid for the calculations, for each actinide and position considered, are shown in Table 3.

<table>
<thead>
<tr>
<th>Isotope</th>
<th>Position</th>
<th>0.5 cm</th>
<th>12.07 cm</th>
<th>24.58 cm</th>
<th>45.5 cm</th>
</tr>
</thead>
<tbody>
<tr>
<td>Am$^{241}$</td>
<td>-3.24E-03</td>
<td>-1.29E-02</td>
<td>9.04E-03</td>
<td>5.34E-02</td>
<td></td>
</tr>
<tr>
<td>Np$^{237}$</td>
<td>-3.59E-03</td>
<td>-1.41E-02</td>
<td>8.04E-03</td>
<td>5.34E-02</td>
<td></td>
</tr>
</tbody>
</table>

The results show that, depending on the measurement position in TAPIRO, the effects on the MAs capture rates are different, and for the considered Cu cross sections perturbation less than 6% in absolute value.

The sensitivity coefficients are shown in Figures 6 through 9.
The sensitivity coefficients in the central core position for both nuclides, Figure 6, show that the main contribution to the capture rate reduction is due to the copper elastic scattering reactions, being the influence of capture and inelastic scattering negligible.
Figure 7: Am$^{241}$ (a) and Np$^{237}$ (b) capture reaction rate sensitivity coefficients in position P2

The positive influence of the capture reactions start to be evident in the copper reflector at 12.07 cm from the core centre, as shown in Figure 7 for both nuclides, even if the main contribution to the variation is due to the elastic scattering reactions.
Figure 8: Am$^{241}$ (a) and Np$^{237}$ (b) capture reaction rate sensitivity coefficients in position P3

![Am$^{241}$ capture reaction rate sensitivity coefficients for -2% copper reduction (P3)](image)

![Np$^{237}$ capture reaction rate sensitivity coefficients for -2% copper reduction (P3)](image)

In the reflector central position there is a change in the total variation that is now positive (cf. Table 3), and mainly due to the capture reactions even if the elastic scattering contribution is not negligible and in the opposite direction (i.e. negative).
At the entrance of the thermal column there is a positive contribution for all the reactions, being the main contribution to the total sensitivity due to elastic scattering reactions.

In all the cases taken into account the variation is mainly influenced by elastic scattering and capture reactions in the energy range from 20 eV to 800 keV.

By means of a direct approach (direct calculations of unperturbed and perturbed systems) the capture reaction rates for both Am$^{241}$ and Np$^{237}$ have been evaluated for all the different positions along the diametral channel, and have been compared with the total sensitivities evaluated by means of GPT, Tables 4 and 5.
Table 4: GPT and direct approach results comparison for Am\textsuperscript{241}

<table>
<thead>
<tr>
<th>Position (cm)</th>
<th>Isotope</th>
<th>GPT</th>
<th>Direct</th>
</tr>
</thead>
<tbody>
<tr>
<td>0.5</td>
<td></td>
<td>-3.24E-03</td>
<td>-3.14E-03</td>
</tr>
<tr>
<td>12.07</td>
<td></td>
<td>-1.29E-02</td>
<td>-1.33E-02</td>
</tr>
<tr>
<td>24.58</td>
<td></td>
<td>9.04E-03</td>
<td>8.76E-03</td>
</tr>
<tr>
<td>45.5</td>
<td></td>
<td>5.34E-02</td>
<td>5.58E-02</td>
</tr>
</tbody>
</table>

Table 5: GPT and direct approach results comparison for Np\textsuperscript{237}

<table>
<thead>
<tr>
<th>Position (cm)</th>
<th>Isotope</th>
<th>GPT</th>
<th>Direct</th>
</tr>
</thead>
<tbody>
<tr>
<td>0.5</td>
<td></td>
<td>-3.59E-03</td>
<td>-3.51E-03</td>
</tr>
<tr>
<td>12.07</td>
<td></td>
<td>-1.41E-02</td>
<td>-1.48E-02</td>
</tr>
<tr>
<td>24.58</td>
<td></td>
<td>8.04E-03</td>
<td>7.48E-03</td>
</tr>
<tr>
<td>45.5</td>
<td></td>
<td>5.34E-02</td>
<td>5.59E-02</td>
</tr>
</tbody>
</table>

The results show a very good reconstruction of the integral effects by the GPT methodology.

Conclusions

In the frame of the NEA Expert Group on Integral Experiments for Minor Actinide Management, a joint collaboration between ENEA and CEA was established with the aim to study the feasibility of a MAs irradiation campaign, named activation of OSMOSE sample in TAPIRO (AOSTA) in the TAPIRO fast neutron source research reactor located at the ENEA Casaccia centre near Rome. Results from previous irradiation campaign simulations indicate the feasibility of the AOSTA experimental campaign.

This work focuses on a preliminary analysis of the sensitivity of the response of the MAs reaction rates, essentially capture, to the TAPIRO copper reflector nuclear data. Experimental uncertainties on Cu nuclear data have been simulated, in this preliminary approach, by a uniform change on the Cu reflector density. The sensitivity coefficients of the MAs reaction rates to Cu cross section variations have been estimated by means of both generalised perturbation theory (GPT) and direct approach by deterministic calculations.

The results show that, depending on the measurement position in TAPIRO, uncertainties on the Cu nuclear data may play different roles, even as order of magnitude, on the experimental results. Of course further investigations, by means of Cu variance-covariance nuclear data matrices, are needed to find optimal measurement positions with regard to both neutron spectrum entering the samples and influence of Cu cross sections uncertainties on the measured MAs reaction rates.

References


Removal of Cs and Sr from aqueous solutions by microcapsules enclosing different zeolites

C.-P. Lee, T.-L. Tsai, M.-C. Wu, S.-C. Tsai

Abstract

$^{137}$Cs and $^{90}$Sr, both having a relatively long half-life of about 30 years, exhibit high radioactivity and heat generation, and large amounts of Cs are contained in the high-level liquid radio-waste (HLLWs) or spent nuclear fuel (SNF). Therefore, the selective separation of $^{137}$Cs and $^{90}$Sr from HLLWs is a very important environmental issue for nuclear waste management. It has been proved that geological minerals presented a favourable environment for sorption of Cs and Sr, such as A-type zeolite, Y-type zeolite, and mordenite. Zeolite, a natural mineral ($\text{Na}_8\text{(AlO}_2\text{)}\text{8(SiO}_2\text{)}\text{40-2H}_2\text{O}$), exchanger having high selectivity towards Cs and Sr were encapsulated in the biopolymer matrices (calcium alginate, CaALG). The characterisation of microcapsules (MCs) was examined by SEM-EDS, and the uptake of Cs and Sr were investigated by batch methods. Spherical and elastic granules were obtained, and the particle size of microcapsules was approximately 700 μm. A relatively high uptake (%) of Cs and Sr above 90% for MCs enclosing A-type zeolite, Y-type zeolite, and mordenite was obtained in the presence of 0.001-1 M NaCl solution, and the uptake equilibrium was attained within 5 hours.
Accelerated deactivation of reactor waste by controlled transmutation to stable isotopes in growing microbiological cultures

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\textsuperscript{2}Moscow State Univ., Moscow, Russia

Abstract

This report discusses the results of accelerated deactivation of water solutions of radioactive isotopes in growing microbe syntrophin associations. The process of deactivation was connected with accelerated transmutation of long-lived radionuclides to different stable isotopes during growth and metabolism of microbiological syntrophin associations MCT \cite{1-3}. The physical mechanism of suppression of the Coulomb barrier action is connected with self-similar formation of coherent correlated states of interacting particles in the dynamic nanowells \cite{4,5} which are generated in zones of growth of microcultures. We observed speeded up decay of Cs\textsuperscript{137} isotope by accelerated transmutation Cs\textsuperscript{137}+p=\textbf{Ba}\textsuperscript{138} in all experiments with MCT and with the presence of different additional salts in active water (see Figure 1). The most speeded up decay of Cs\textsuperscript{137} isotope with duration of accelerated deactivation $t^* \approx 310$ days (accelerated by 35 times) was observed in the presence of Ca salt.

These results can answer the question about the reasons of abnormal accelerated decrease of environmental radioactivity (natural environmental remediation) in some isolated areas inside the Chernobyl accident zone with initial high-level of radiation pollution. This method of reactor water deactivation can also be used for clearing reactor water and in the case of accidents at nuclear power stations (e.g. Chernobyl and Fukushima Daiichi).

Figure 1: Accelerated deactivation (accelerated rates of decay) of Cs\textsuperscript{137} isotope in “biological cells” in the presence of different chemical elements.
References


A parametric study on near-field criticality safety for the disposal of spent fuels

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Abstract

Criticality safety has been one of the important concerns for the direct disposal of spent nuclear fuels in a deep geologic repository. Partitioning and transmutation (P&T) technology provides fundamental solution to reduce or eliminate the risk of criticality in a geologic repository by reducing the mass and concentration of fissile nuclides in the waste stream. The positive role of P&T in enhancing the long-term safety of waste disposal can be justified by understanding the criticality risk in a geologic repository. Numerical results over a broad range of input parameters including lattice parameters and different spent fuels have been calculated using a numerical scheme based on SERPENT code. The numerical results indicate that, for UO$_2$ and MOX spent fuels with common enrichment and discharge burnup, the single canister model will be sub-critical. However, for spent fuels with relative low burnup, the system can be super-critical even if it was subcritical at the time of emplacement in the repository.

Introduction

Criticality safety has been one of the important concerns for the direct disposal of spent nuclear fuels in a deep geologic repository. Partitioning and transmutation (P&T) technology provides fundamental solution to reduce or eliminate the risk of criticality in a geologic repository by reducing the mass and concentration of fissile nuclides in the waste stream. The positive role of P&T in enhancing the long-term safety of waste disposal can be justified by understanding the criticality risk in a geologic repository.

Three major stages can be identified to evaluate the criticality safety for the disposal of spent nuclear fuels: (1) the stage before package failure, (2) the stage after package failure, while fissile nuclides remain within the engineered barriers, and (3) the stage in which fissile nuclides originated from multiple packages deposit in far-field host rocks. The present work focuses on the neutronics analysis for the second stage, in which Monte Carlo simulations are made to calculate the neutron multiplicity for a waste canister containing spent fuels after assembly structures and claddings have failed. Because the canister designs commonly require the canister being sub-critical in both dry and water filled conditions, the primary concern for the criticality safety is the time period after all assembly structures and claddings fail, until the bare fuel pellets in the canister have completely dissolved. The conditions in the failed spent fuel canister could not be known exactly, and spent fuels with different designs and burnup histories may behave differently. To obtain sufficient understanding about the system, the present work includes two primary tasks. The first task is to explore under what conditions the neutron multiplicity of a spent fuel canister can be maximised, so that the uncertainty of the result can be bounded. The second task is to perform a parametric study to examine spent fuels with different designs and burnup histories.
Background and assumptions

The repository is assumed to be in a water-saturated reducing environment. The neutronics model consists of a canister containing spent fuel and the buffer material surrounding the canister. The composition and dimension of the canister and the buffer for BWR and PWR spent fuels are assumed according to the design developed by JAEA [1]. The BWR canister contains seven assemblies, and the PWR canister contains two assemblies. The schematic layout of the canister-buffer system has been shown in Figure 1.

The spent fuels are assumed to be emplaced in the repository after 50 years of cooling, the canister failure time is assumed to be 1,000 years after emplacement. After its failure, the canister is assumed to be filled with groundwater. The failure of assembly structures and claddings is assumed to be a transient event in a time scale of thousand years. Therefore, we assume the spent fuels exist in the form of pellets immediately after the canister failure. In a reducing environment, the fuel pellets dissolve and are released from the canister over the next hundreds of thousands of years. The BWR assembly design is assumed to be STEP-III B type [2], and the PWR assembly is assumed to be 17$	imes$17 type. Although these assembly designs are for UO$_2$ fuels, same conditions are assumed for MOX fuels. Numerical results for specific MOX fuel design can be generated in the future. Because the present work only consider the situation when assembly structures and claddings have failed, in our model, the assembly designs would only affect the mass of the spent fuel to be contained per canister, and the size of the fuel particles. Spent fuels with different initial enrichments and burnups are considered. They are described in details in later discussions. For the system, including the spent fuel pellets, the canister, and the buffer remains subcritical during the fuel dissolution time, we need to explore under what conditions the neutron multiplicity of the system can be maximised.

Method and numerical scheme

The present work aims at calculating numerical results over a broad range of input parameters including lattice parameters and different spent fuels. The numerical scheme needs to be carefully designed to reduce the computation time. The criticality calculations have been carried out by a Monte Carlo code called SERPENT [3].

The first step of the numerical scheme is to identify the fuel particle diameter and the pitch distance to give maximum $k_{inf}$ for the infinite HCP lattice for different spent fuels at different decay times. To make the parameter search more efficiently, a broad survey has been firstly made in a two-dimensional parameter space (diameter and pitch distance) for BWR and PWR UO$_2$ spent fuels with different values for the initial enrichment (IE), burnup (BU), and decay time.

The compositions for spent fuels are calculated in ref. [4], for UO$_2$ and MOX fuels from BWRs and PWRs with different burnups and initial enrichments. The burnup calculations have been carried out by ORIGEN-ARP using an input generator developed in ref. [4]. For most LWR spent fuels, the ratio of discharge burnup [MWd/t] to initial enrichment [wt. %] is typically between 9 and 12 [5]. Therefore, three combinations of initial enrichment and burnup (IE = 2.0%, BU = 20 GWd/t; IE = 4.0%, BU = 40 GWd/t, IE= 6.0 %) have been considered, which crudely cover the parameter range of common spent fuels. Combining two reactor types (BWR/PWR), three initial enrichment and burnup cases, and three decay times (1,050, 10,050, and 100,050 years), 18 cases in total have been calculated.
Figure 1: Schematic layout of the canister-buffer system. The carbon-steel canister is filled with spent fuel particles in HCP lattice and is surrounded by bentonite buffer.

Based on the numerical results from the two-dimensional parametric search, the parameter search has been reduced to one-dimensional. A second round of the parametric search has been made for more cases of burnup and initial enrichment for UO$_2$ and MOX fuels. For UO$_2$ spent fuels, 20 cases of burnup and initial enrichment have been calculated for both BWR and PWR spent fuels. The cases roughly covered the burnup to initial enrichment ratio range between 9 and 12. Due to the relative lack of industry experience with MOX fuels, 42 cases have been defined by combining the burnup ranging from 10 GWd/t to 60 GWd/t and the plutonium weight fraction ranging from 4.0% to 10.0%. The isotopic fractionations of plutonium isotopes in the MOX fuels are shown in Table 1.

<table>
<thead>
<tr>
<th>Isotope</th>
<th>wt.%</th>
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<tr>
<td>Pu-238</td>
<td>1.5</td>
</tr>
<tr>
<td>Pu-239</td>
<td>60.1</td>
</tr>
<tr>
<td>Pu-240</td>
<td>24.5</td>
</tr>
<tr>
<td>Pu-241</td>
<td>8.8</td>
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<tr>
<td>Pu-242</td>
<td>5.0</td>
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</table>

Table 1: Isotopic fractionation of plutonium isotopes in the simulated MOX fuels

The combination of the fuel particle diameter and the pitch distance to give maximum $k_{inf}$ has been identified for each case. Then, we have calculated the $k_{eff}$ values using the diameters and pitch distances in the canister-buffer model to examine whether the canister could be critical.

The input parameters for the BWR and PWR canisters are shown in Table 2. The total volume inside the canister $V_0$ can be calculated by $V_0 = \pi r^2 L$, where $r$ is the canister inner radius, and $L$ is the canister inner length. The total volume of spent fuel per canister ($V$) is defined as $V = N \times n \times \frac{1}{4} \pi d^2 l$, where $N$ is the number of assembly per canister, $n$ the number of fuel rods per assembly, $d$ the pellet diameter, and $l$ the fuel length. The fuel particle diameter $D$ in the present...
model has been determined to give the same volume of a cylindrical fuel pellet. The length of a fuel pellet is assumed to be 1 cm.

Note that same fuel design parameters for UO$_2$ fuels are assumed for MOX fuels. Numerical results for specific MOX fuel design can be generated in the future. Because the present work only considers the situation when assembly structures and claddings have failed, the fuel designs would only affect the mass of the spent fuel to be contained per canister, and the size of the fuel particles.

Table 2: Summary of input parameters for BWR and PWR spent fuel canisters

<table>
<thead>
<tr>
<th>Canister design parameters$^1$</th>
<th>BWR</th>
<th>PWR</th>
</tr>
</thead>
<tbody>
<tr>
<td>Canister inner radius $r$ [cm]</td>
<td>34.40</td>
<td>27.95</td>
</tr>
<tr>
<td>Canister inner length $L$ [cm]</td>
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<td>450</td>
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<td>Canister thickness $\theta_1$ [cm]</td>
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<td>14</td>
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<td>Buffer thickness $\theta_2$ [cm]</td>
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<td>70</td>
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<td>Number of assemblies per canister $N$</td>
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<td>2</td>
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<tr>
<td>Fuel design parameters</td>
<td>BWR</td>
<td>PWR</td>
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<tr>
<td>Fuel type</td>
<td>BWR STEP-III B type</td>
<td>PWR 17x17 type</td>
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<td>Fuel length $l$ [m]</td>
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<td>Number of fuel rods per assembly $n$</td>
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<td>264</td>
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<tr>
<td>Fuel density</td>
<td>97% of theoretical density</td>
<td>95% of theoretical density</td>
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<td>Pellet diameter [cm]</td>
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<tr>
<td>Calculated model parameters</td>
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<td>PWR</td>
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<td>Total spent fuels volume $V$ [m$^3$]</td>
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<tr>
<td>Fuel particle diameter $D$ [cm]</td>
<td>1.114</td>
<td>1.003</td>
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Results and discussions

Two-dimensional parametric study for BWR and PWR spent fuels in infinite HCP lattice

The parametric study on $k_{inf}$ has been performed for the particle diameters D ranging from 0.4 cm to 1.4 cm, and P/D ranging from 1.0 to 1.7. To understand the general behaviour, totally six combinations of burnups, enrichments and fuel types have been calculated. These cases cover the low burnup, low enrichment fuels (BU = 20 GWd/t, IE = 2.0%); mid burnup, mid enrichment fuels (BU = 40 GWd/t, IE = 4.0%); and high burnup, high enrichment fuels (BU = 60 GWd/t, IE = 6.0%) for BWRs and PWRs. For example, the $k_{inf}$ contours for BWR (STEP-III-B) type UO$_2$ spent fuels with burnup of 40 GWd/t and initial enrichment of 4.0% are plotted for decay times of 1 050, 10 050, and 100 050 years, in Figures 2-4, respectively. Because the results for other cases show similar tendency, we hereafter discuss the results shown in Figures 2-4 as examples, so that other results could be similarly understood.
**Figure 2**: $K_{\text{inf}}$ contour plot for BWR UO$_2$ spent fuel in infinite HCP lattice, after 1,050 years decay. The spent fuel has burnup of 40 GWd/t and initial enrichment of 4.0%.

![Contour plot](image1.png)

**Figure 3**: $K_{\text{inf}}$ contour plot for BWR UO$_2$ spent fuel in infinite HCP lattice, after 10,050 years decay. The spent fuel has burnup of 40 GWd/t and initial enrichment of 4.0%.

![Contour plot](image2.png)
Figure 4: $k_{\text{inf}}$ contour plot for BWR UO$_2$ spent fuel in infinite HCP lattice, after 100 050 years decay
The spent fuel has burnup of 40 GWd/t and initial enrichment of 4.0%

The first observation is that, the neutron multiplicity changes drastically as P/D changes. For a given diameter D, the $k_{\text{inf}}$ gain significant increases as P/D increases, and then gradually decreases when the system is over-moderated. The diameter D of the spherical fuel particle that has same volume of a BWR fuel pellet (D = 1.11 cm) is indicated by the vertical dashed line in the contour plots. Because we assumed the spent fuel particles are only shrinking during dissolution, the diameters should be equal or smaller than 1.11 cm. The parameter pair that maximises the $k_{\text{inf}}$ value should always be found on the dashed line, because for the left side of the dashed line, the contour lines always have a convex shape towards smaller diameter values. This is a general observation for all calculated cases, which cover the range of different spent fuels. Therefore, we can simplify the parameter search from two-dimensional to one-dimensional, i.e. to fix the diameter at its maximum value and search for pitch distances to maximise the $k_{\text{inf}}$. With this simplification, we can perform the one-dimensional parametric search for the pitch distance with a smaller step length. The results are discussed in the next section.

One-dimensional parametric search on pitch distance

One-dimensional parametric search on the pitch distance was made for different spent fuel types with three different decay times to search for the maximum $k_{\text{inf}}$. For UO$_2$ spent fuels, 20 cases of burnup and initial enrichment have been calculated for both BWR and PWR spent fuels. For all cases, the maximum $k_{\text{inf}}$ values and corresponding P/D ratios are tabulated in Table 3 for BWR spent fuels and in Table 4 for PWR spent fuels. If we fix the same fuel type, burnup, and initial enrichment/plutonium weight fraction, and compare the results for different decay times, the maximum $k_{\text{inf}}$ is found at the 10 050 years decay. It has also been observed that the pitch distance that maximises the $k_{\text{inf}}$ tends to decrease with increasing decay time. The reason is probably that, for Pu-239, due to the resonance neutron capture in epithermal energy, Pu-239 favours higher moderation than U-235. As a result, the higher fraction of Pu-239 among the fissile nuclides, the higher moderation (meaning larger pitch distance) the system requires to reach maximum $k_{\text{inf}}$. That
explains two observations from the results. First, as time increase, Pu-239 decays to U-235, resulting in the decrease of the pitch distance for maximum $k_{inf}$. Second, the pitch distance for maximum $k_{inf}$ is always higher for MOX fuels than in UO$_2$ fuels, because the fissile nuclide initially in the MOX spent fuels is mainly Pu-239. Comparing spent fuels with different burnup and initial enrichments, it is obvious that the $k_{inf}$ is higher, for lower burnup or higher initial enrichment, because in either situation, there is greater mass of fissile nuclides remaining in the spent fuels. Another interesting observation is that, for same burnup and initial enrichment, the PWR spent fuels gives slightly higher $k_{inf}$ than the BWR spent fuels. This is actually a net effect of two opposing factors: the PWRs generally breed more fissile nuclides than BWRs and thus more fissile nuclides remain in the spent fuels; while the BWRs have larger pellets than the PWRs, which is more favoured for higher $k_{inf}$.

Table 3: Results for parametric search on pitch distances for the maximum neutron multiplicity in infinite HCP lattice for BWR UO$_2$ spent fuels after different decay times

<table>
<thead>
<tr>
<th>Burnup [GWd/t]</th>
<th>Initial enrichment [wt. %]</th>
<th>After 1 050 years decay</th>
<th>After 10 050 years decay</th>
<th>After 100 050 years decay</th>
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<tr>
<td></td>
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<td>$P/D$ to maximise $k_{inf}$</td>
<td>$k_{inf}$</td>
<td>$P/D$ to maximise $k_{inf}$</td>
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Table 4: Results for parametric search on pitch distances for the maximum neutron multiplicity in infinite HCP lattice for PWR UO$_2$ spent fuels after different decay times

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<tr>
<th>Burnup [GWd/t]</th>
<th>Initial enrichment [wt. %]</th>
<th>After 1 050 years decay</th>
<th>After 10 050 years decay</th>
<th>After 100 050 years decay</th>
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<tr>
<td></td>
<td>P/D to maximise $k_{\text{eff}}$</td>
<td>$k_{\text{eff}}$</td>
<td>P/D to maximise $k_{\text{eff}}$</td>
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</tr>
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</table>

The next step is to apply the results from the parameter search to calculate the $k_{\text{eff}}$ values of the canister-buffer system. The canister design and initial spent fuel loadings have been described in previous sections. The first task is to examine whether or not the fuel particle diameter and the pitch distance identified from the parameter search could be realised for the given canister design and initial loadings. In other words, we need to check if the canister will be over-filled if such lattice parameters are assumed. From Table 2, the total spent volume is 0.1353 m$^3$ for BWR canisters and is 0.1003 m$^3$ for PWR canisters. The packing fraction $f$ (the volume fraction of fuel particles), in the present situation, for the HCP lattice with the particle diameter $D$ and the pitch distance $P$ is given by

$$f = \frac{\pi}{3\sqrt{2}} \left(\frac{D}{P}\right)^3, P \geq D$$

(1)

Among all calculated cases, the maximum $P/D$ value is 1.78 for both BWR and PWR cases. Therefore, the packing fraction $f$ should be larger than 0.1313. Therefore, the maximum volume of the HCP lattice is 0.1353 m$^3$ / 0.1313 = 0.1035 m$^3$ for BWR canisters, and 0.1003 m$^3$ / 0.1313 = 0.7639 m$^3$ for PWR canisters. Apparently, these values are smaller than the canister volumes calculated in Table 2, which is 1.672 m$^3$ for BWR canisters, and 1.104 m$^3$ for PWR canisters. Therefore, based on...
the discussions in the previous section, we assume all initial spent fuels to be included inside the canister.

**Numerical results for the canister-buffer system**

By utilising the results from the parametric search in the infinite lattice, the $k_{eff}$ values are calculated for the canister-buffer system for different combinations of fuel types, burnup, initial enrichment/plutonium weight fraction, and decay times. The $k_{eff}$ contour plots for UO$_2$ BWR, UO$_2$ PWR, MOX BWR, and MOX PWR spent fuels, after 10 050 years decay, are shown in Figures 5-8, respectively.

The first observation is that for all calculated UO$_2$ spent fuel cases, the $k_{eff}$ values of spent fuel canisters are all far below unity. Within the present model assumptions, numerical scheme and parameter ranges (burnup, initial enrichment, and decay time), we could not construct a case to make the canister critical. Comparing BWR and PWR UO$_2$ spent fuels (see Figures 5 and 6), the $k_{eff}$ for the BWR canister is slightly higher than the PWR canister due to higher mass loading. Although most of the spent fuels are within the range of burnup and initial range considered in the present work, there are spent fuels with very low burnup relative to initial enrichment for various reasons, such as early fuel failure. For those special fuels, further study is necessary.

For the MOX fuel results, there are combinations of plutonium weight fractions and burnups that make the canister supercritical. The critical parameter ranges are shown in the contour plots (Figures 7 and 8) for the MOX fuel cases by indicating the contour lines for $k_{eff} = 1.0$ (red lines) and 0.98 (blue lines), which could be considered as nominal criticality safety limits.

**Figure 5**: $k_{eff}$ contour for a spent fuel canister containing BWR UO$_2$ spent fuel after 10 050 years decay for different combinations of spent fuel burnups and initial enrichments
Figure 6: $K_{eff}$ contour for a spent fuel canister containing PWR UO$_2$ spent fuel after 10 050 years decay for different combinations of spent fuel burnup and initial enrichment.

Figure 7: $K_{eff}$ contour for a spent fuel canister containing BWR MOX spent fuel after 10 050 years decay for different combinations of spent fuel burnup and plutonium mass fractions.
Summary

We presented our discussions on the near-field criticality analysis, focusing on two primary tasks. The first task is to explore under what conditions the neutron multiplicity of a spent fuel canister can be maximised, so that the uncertainty of the result can be bounded. The second task is to perform a parametric study to examine spent fuels with different designs and burnup histories.

Because the present work aims at obtaining numerical results over a broad range of input parameters including lattice parameters and different spent fuels, a new numerical scheme based on SERPENT code has been designed to reduce the computation time. A two-dimensional parametric search has firstly been made for a few representative cases of burnup and enrichment. Based on the numerical results, the parametric search has been reduced to a one-dimensional case. The numerical results indicate that, under the conditions assumed, for all UO$_2$ spent fuels and part of the MOX spent fuels, the single canister model will always subcritical.

Future improvements can be made in the following directions: (a) more fuel types should be considered for MOX fuels; (b) the corroded canister, assembly structural and cladding materials should be accounted for in the model of the failed canister; (c) the potential collapsing of the canister and buffer which could significantly alter the canister geometry should be considered; (d) the random distribution of fuel rods or fuel pellets in the canister after assembly and rod failure needs to be studied.

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References


Reducing long-term safety uncertainty of high-level waste disposal: PyroGreen, decontaminating high-level waste into intermediate-level waste

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Abstract
Disposal of HLW from nuclear power plant leaves scientific, technical and social uncertainty which continue over 10,000 years. Many countries have been used up the budget and manpower to reduce this issue, but it is doubtful whether the efforts could draw the consensus from experts with various backgrounds and the acceptance from public. As an innovative solution, PyroGreen aiming to decontaminate spent nuclear fuel into intermediate-level waste without high-level waste (HLW) has been studied by Nuclear Transmutation Energy Research Centre of Korea in Seoul National University. In this study, (1) the review on overall process of PyroGreen is conducted and (2) the long-term safety impact by decontaminating SNF into intermediate-level waste (ILW) was assessed in an aspect of environmental risk.
Investigation of physiochemical and thermodynamic properties of synergistic extraction systems

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Abstract

Solvent extraction is often the technique of choice for separating solutes of interest in processes such as the separation of used nuclear fuel and hydrometallurgical recovery of metals in mining. Such processes utilise extraction reagents which facilitate the transport of the desired solutes between immiscible aqueous and organic phases. Often, these reagents possess surface active properties which can lead to the formation of reverse micelles and micro emulsions under the correct conditions. It is also known that the use of multiple extractants in a single organic phase my lead to a phenomenon known as synergism resulting in enhanced extraction efficiency. It has been suggested in literature that the formation of reverse micellar aggregates and micro emulsions may be related to the synergistic effect. Here we present work exploring several synergistic extraction combinations, HTTA: TBPO, BDA: TERPY and HDNNS: LIX63 and their physiochemical properties as well as selected thermodynamic properties to investigate the connection between the formation of reverse micellar aggregates and micro emulsions and the synergism phenomenon. Results show that while the systems under study display similar metal extraction properties the physiochemical properties differ greatly, suggesting different synergistic mechanisms between systems. Thermodynamic studies on the HDNNS: LIX63 system suggests a definite pre-organisation of the organic phase with estimates of the enthalpy of interaction being possible to obtain from calorimetric measurements.
Speciation of lanthanides and americium with organophosphorus acids and effect of coextracted water

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Abstract

Complexes of the trivalent americium and lanthanides with di-2-ethylhexyl phosphoric acid (HDEHP) dissolved in n-dodecane were probed with UV-Vis, X-ray absorption fine structure (XAFS), and small angle X-ray (SAXS) spectroscopy. Water concentration was determined by Karl Fischer titrations. This work compares the behaviour of Nd, Pr, Ho, and Am in different metal: ligand concentration ratios with HDEHP, diluted with n-dodecane. Results indicate that the effects of extracted water depend on the effective charge of metal cation, and decreases with decreasing ionic radius (increasing atomic number) of lanthanide. The spectral features observed for the electronic transition band in the Nd hypersensitive UV-Vis absorbance region are attributed to the changes in speciation of the extracted metal complex, caused by the amount of extracted water and ionic radius of the extracted f-element. We established that high water content can cause the distortion of the octahedral symmetry of the six-co-ordinated Nd, resembling the reported spectra of the seven-co-ordinated Nd compounds. Extended X-ray Absorption Fine Structure (EXAFS) analysis of the Nd in high concentration HDEHP solutions also indicates an increase in the co-ordination number from 6 to 7. In comparison, HEH[EHP], the phosphonic acid analogue of HDEHP, exhibits a smaller capacity for water extraction and does not appear to affect the electronic absorption spectra of Nd or Am, although speciation does still seem to be a function of the effective charge of the metal.

Introduction

Development of the advanced separation processes has recently been focused on partition trivalent minor actinides from lanthanides by their group co-extraction from PUREX raffinate, followed by a selective stripping. Typically, group extraction into an organic phase is performed using a combination of cation-exchange (chelating) and neutral solvating ligands. [1] Our investigation towards development of the ALSEP separation process involves comparison of two chelating ligands HDEHP (di-2-ethylhexyl phosphoric acid) and HEH[EHP] (di-2-ethylhexyl phosphonic acid) alone and in the mixtures with diglycolamides in n-dodecane diluent. [2-3] HEH[EHP] is a slightly softer phosphonic acid analogue of HDEHP [4], which is the historically employed TALSPEAK ligand, and despite the metal-HDEHP complexes have been subject to several investigations, their speciation in the HDEHP containing organic phases remains somewhat poorly understood. Several explanations for the effects seen in UV-Vis hypersensitive regions for neodymium and holmium were published; hence the present work focuses on the complexation of trivalent americium and selected lanthanides with two studied organophosphorus acids, diluted with n-dodecane (Figure 1A, B).
Figure 1: Organic phase cation-exchange molecules: (A) HDHEP and (B) HEH[HEP]

Experimental

Materials and methods

HDEHP was purified by precipitation of Cu[DEHP]₂ out of diethyl ether. Procedure was carried out 3 times and between each precipitation, the Cu[DEHP]₂ product was washed with acetone, dried for 2 hours with a stream of dry nitrogen, then redissolved in diethyl ether. To ensure all of the precipitate was dissolved, at least one hour was allowed before next step. After converting Cu[DEHP]₂ back to CuSO₄ and HDEHP, the organic phase was washed five times (20 mL per wash) with deionised H₂O to ensure that there was no sulphuric acid remnant in the organic phase. HDEHP was concentrated by removing the solvent in vacuo on a Schlenk line with a 25 mmHg vacuum while the HDEHP-diethyl ether flask was heated to 90°C. Also HEH[HEP] was purified using the same copper precipitation purification method. Purity of both phosphorus acids as confirmed by both ³¹P NMR spectroscopy and acid-base titration in 80% methanol-water mixture [5] was >99.95%.

Lanthanide chlorides were individually prepared by dissolving their oxides in concentrated hydrochloric acid, evaporating and redissolving in 0.001M HCl with the final solution measured to be pH 2.80 +/- 0.05 as checked with a combined pH electrode. Lanthanide concentrations in the stock aqueous solution was determined by EDTA titrations with xylene orange as an indicator [6], while for extraction samples the colorimetric technique with Arsenazo III at pH 9 buffered by triethanolamine [7] was applied.

Americium chloride was prepared similarly through conversion of an in-house $^{243}$Am stock solution purified using a DGA column (Eichrom) by dissolution in concentrated HCl and subsequent evaporation, followed by dissolution in pH 2.80 HCl. Concentrations of $^{243}$Am in both the aqueous and organic extraction phase samples were determined through the LSC alpha/beta discrimination counting method with Perkin Elmer Tricarb 3180 to prevent the interference from the beta-emitting $^{239}$Np daughter radioisotope.

Solvent extraction

Equal volumes of organic and aqueous phases were vortexed for 10 minutes and then allowed to split by centrifugation at 3000rpm for 5 minutes. Aliquots from both phases were taken for determination of equilibrium concentrations and calculation distribution ratios of studied metals. Concentration of extracted water in the organic phases was determined by volumetric Karl Fischer titrations on a Mettler Toledo DL58 auto titrator with each sample measured in duplicate.

UV-Vis and FTIR EXAFS spectroscopy

The organic layers of extracted samples were separated and transferred to quartz cuvettes. UV-Vis spectra of the extracted metals were collected on a Cary 6000i UV-Vis-NIR spectrometer with a jacketed cell held at 20 °C. The reflectance Fourier transform infrared spectra were collected with the Nicolet (ThermoFisher) with diamond plate at room temperature.
EXAFS spectroscopy

For the EXAFS experiment, the organic layer of extracted samples was transferred to a modified polystyrene 4.5mL cuvette with an epoxied Kapton window. The cell window was oriented at a 45° angle with respect to the incident beam, while the detector was nominally positioned at 90° with respect to the incident beam in the horizontal plane. X-ray absorption fine structure (XAFS) spectra were measured on the Materials Research Collaborative Access Team (MRCAT) bending magnet beam line 10-BM at Argonne National Laboratory’s Advanced Photon Source (APS). The incident energy was selected using a double-crystal Si(111) monochromator with the 2nd crystal detuned to 50% of the peak intensity. Neodymium L3 edge spectra were measured in fluorescence mode (Nd Lα emission line) using a Vortex-ME4 4-element silicon drift detector (Hitachi High-Technologies Science America, Inc.). The XAFS data were treated using Athena and Artemis software packages.

Small-angle X-ray Scattering (SAXS) spectroscopy

SAXS data were collected on an Anton-Paar SAXSess instrument utilising Cu Kα radiation (λ =1.54178 Å) and line collimation. Samples of extracted organic phase were measured in 1.5 mm glass capillaries. Pure n-dodecane was used for the background, and scattering was typically measured for 30 min. SAXSQUANT was used for data collection, treatment, and preliminary analysis (normalisation, primary beam removal, background subtraction, desmearing to remove line collimation distortion, and smoothing to remove the noise created by desmearing). The spherical and cylindrical fits of the scattering data were carried out utilising Modeling II in the IRENA macros within IGOR Pro.

Results and discussion

Extraction of metals and water with the HDEHP and HEH[EHP] in n-dodecane

UV-Vis spectra of the Nd-HDEHP/n-dodecane extraction organic phases with the absorption bands of the \( ^1_{b_9/2} \rightarrow ^2_{g_{5/2}} \) electronic transition are shown in Figure 2. At low HDEHP concentrations, the spectra resemble those of a pseudo-octahedron, containing the characteristic 6 absorption bands of such structures with a prominent absorption band at 570 nm (\( A_{570} \)).[8] This type of absorbance has been previously identified as representative of polymeric speciation in the organic phase.[8, 9] With an increase in HDEHP concentration however, \( A_{570} \) is seen to decrease while the absorption band at 583 nm (\( A_{583} \)) is seen to increase. This is consistent with an increase in the co-ordination number of the metal complex to a seven co-ordinate species. [10] Dilution of Nd loaded organic phases of higher HDEHP concentration with non-preequilibrated solutions of the same HDEHP concentration resulted in a substantial shift of the UV-Vis spectra, whereas dilution with preequilibrated organic phases resulted in no spectrum change, which confirms an effect of coextracted water. Within the studied metal concentration region, the amount of extracted water was found independent on the Nd concentration and increasing with the HDEHP concentration in solvent.

Figure 2: UV-Vis spectra of three samples of 0.034M Nd extracted from 0.001M HCl with 0.5M, 1M and 1.5M HDEHP in n-dodecane
Association of water with HDEHP was evident also in the FTIR spectra (Figure 3). On the contrary, the UV-Vis spectra of the Nd-HEH[EHP]/n-dodecane samples are constant. The absence of one oxygen atom in the HEH[EHP] molecule (Figure 1B) explains about 6-fold lower concentration of extracted water and much weaker effect of water on the speciation in organic phase.

![Figure 3: FTIR spectra of three samples of 0.034M Nd extracted from 0.001M HCl with 0.5M, 1M and 1.5M HDEHP in n-dodecane](image)

While analysis of the HDEHP complexes with Pr and Am showed significant spectral changes for different HDEHP concentration in n-dodecane (Figure 4), the spectral shift of Sm is much subtler, and the spectra for Ho and Er remain constant. Furthermore, with an increasing concentration of HDEHP and water in organic phase the absorbance in the Pr and Am spectra increases, while Sm decreases. These spectral shifts in the light lanthanide spectra indicate metal speciation changes, while the constant spectra for the hypersensitive regions of holmium and erbium suggest that water might be less able to co-ordinate with metal centre due to lanthanide contraction.

Praseodymium showed a similar increase in absorbance intensity for HEH[EHP]/n-dodecane, possibly indicating a change in the metal complex symmetry. However, all other metals show no shifts in spectra, regardless of the HEH[EHP] concentration. Furthermore, americium shows much lower absorbance intensity when bonded with HEH[EHP] than when bonded with HDEHP, possibly indicating greater complex symmetry. This data suggests that while the lightest lanthanides may have different metal complexation environments based on HEH[EHP] concentration, lanthanides above neodymium (as well as americium) are not affected by the concentration of ligand and water.
Co-ordination structure of neodymium in HDEHP complexes

The Fourier-transformed spectra of the 0.01M Nd with 2M HDEHP and 0.2M HDEHP were comprised of two major peaks that correspond to the nearby oxygen and phosphorous scattering paths at 1.9Å and 3.4Å (uncorrected for phase-shift), respectively. To resolve their differences, all parameters were fixed including the amplitude reduction factor $S_0^2$ (a generally accepted value of 0.9 was employed based on previous studies that probed Nd in identical or very similar solvents [11, 12], pre- and post-edge reduction, $k$-range (2-11 Å$^{-1}$), $R$-range (1-6 Å), and the input FEFF8.00 file. The input file was comprised of a two-shell fit (O: 2.3Å and P: 3.9Å). The variables allowed to refine were the oxygen co-ordination numbers (CN), bond distances ($R$), Debye-Waller factors ($\sigma^2$) and change in the energy shift $\Delta E_0$. The results are consistent with conventional X-ray absorption interpretations of lanthanide co-ordinate organophosphorus ligands which have thus far accepted a general co-ordination chemistry of 6×O and 6×P at approximately 2.3Å and 3.8Å, respectively.[13-15] The inherent uncertainties in resolving CNs by EXAFS are known [14] and the CN for the P-shell was fixed at 6.0, considering only a three-dimer HDEHP co-ordinated complex. The results of analysis yielded a slightly larger CN for the 2.0M HDEHP data. Targeting the extra source of oxygen scatter by XAFS proved to be inconclusive, considering that only one or two water molecules may be sufficient to disrupt the symmetry of three HDEHP dimers co-ordinated to the metal. However, simultaneously varying all other parameters such as $S_0^2$, the input files always resulted in a more oxygen populated environment for the 2.0M HDEHP solution than for 0.2M. The most revealing feature of the spectra lies within $k$-space; the 2M HDEHP spectrum is slightly shifted relative to the 0.2M HDEHP spectrum. This correlates to a longer Nd-O bond in 2M HDEHP. Our surmise is twofold: either the XAFS is picking up the alleged water molecules co-ordinated to the outer-sphere of the Nd (assumingly 2.49Å based on aqueous Nd(III) XAFS analysis) [15] or the water is co-ordinated to the HDEHP dimers which is stretching the HDEHP Nd-O path lengths. Though we acknowledge that the XAFS results are not entirely sufficient in pin-pointing the components responsible for altering the electronic spectra of 2.0M HDEHP/Nd, the statistically larger Nd-O bond lengths and the higher oxygen CN support the claims that an additional source of oxygen – alongside the six HDEHP molecules – is co-ordinated to the extracted metal.
Aggregation of the Nd-HDEHP complexes in n-dodecane

The pilot SAXS experiment of the extracted samples of 0.5M HDEHP/n-dodecane-0.001M HCl confirmed small aggregates in organic phase (~3.8 Å without Nd and 4.7 Å with Nd present). Figure 5 shows the effect of a higher concentration of HDEHP (1.0M and 1.5M in n-dodecane) on the size, shape and interaction between micelles in this organic phase.

Figure 5: The pilot SAXS experiment; ■ 1.0M HDEHP/n-dodecane, 0.001M HCl; □ 1.0M HDEHP/n-dodecane, 0.001M HCl+0.34 M NdCl3 ; ▲ 1.5M HDEHP/n-dodecane, 0.001M HCl ; ▼ 1.5M HDEHP/n-dodecane, 0.001M HCl+0.034 M NdCl3

All micelles are of similar size, as indicated by the similar regions of scattering intensity decent (Guinier to Porod region), between q=0.5-0.8 Å⁻¹. However, the exact size is difficult to fit, due to the large Coulombic peak at ~0.4 Å⁻¹. For both HDEHP concentrations (1.0 and 1.5 M), the intensity of the Coulombic peak increases with the incorporation of Nd, indicating more repulsion between the micelles with Nd compared to the ones without Nd. However, the size and shape of the micelles remains very similar with incorporation of Nd, and this can be seen by comparing the aforementioned region between q=0.5-0.8 Å⁻¹. These experiments are in progress.

Conclusions

A deeper insight into speciation of complexes of several f-elements with two organophosphorus acids (HDEHP and HEH[EHP]) was attempted by application of multiple speciation methods to their organic extraction samples in n-dodecane diluent. Karl Fischer titrations, UV-Vis, FTIR, XAFS and SAXS spectroscopies revealed that the neodymium speciation in the HDEHP/n-dodecane solvent is quite affected by the concentration of the ligand and extracted water. The observed increase in the co-ordination number of the metal complex indicates that water is present in the inner co-ordination sphere of Nd.. The effect of water on the electronic spectra of the studied Am³⁺, Pr³⁺, Ho³⁺ and Sm³⁺ is entirely dominated by the size of their cationic radius: heavier lanthanides with smaller radii do not show the same propensity as Nd³⁺ in the hyperfine region UV-Vis and XAFS spectra. Relative absorbances in the hypersensitive region of the collected UV-Vis spectra of americium and praseodymium rise with increasing HDEHP concentration. While these transitions are not hypersensitive, the increase in absorbance indicates a decrease in complex symmetry and a change in the co-ordination environment. This suggests that for Am and larger Ln as the first Ln tetrads, the presence of water leads to 7-co-ordinate species of Ln(H(EDHP)₂)₃(H₂O) which resembles a distorted
face-capped octahedron, while the holmium spectrum indicates that water does not affect the co-ordination of these species. Once the amount of extracted water in the HEH[EHP]/n-dodecane solvent is much lower, the ability to form the inner co-ordination sphere water complex is possible only with the lightest lanthanides.

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**References**


Selective Kr/Xe separations
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Abstract
Capture of volatile radioactive isotopes released during used nuclear fuel reprocessing will be necessary to meet regulatory emission requirements. Selective physisorption is being investigated as a less costly and more efficient alternative to cryogenic distillation. An experimental test bed at Idaho National Laboratory was designed and constructed to perform single and multi-column sorbent evaluations. With two cryogenic coolers as well as a custom designed cryostat, adsorption and desorption temperature can be tightly controlled from 191 K to 500 K across a wide range of flow conditions.

On-going research has also resulted in a patented method for developing engineered form sorbents. One of these sorbents, AgZ-PAN, revealed a high selectivity for xenon over krypton. Controlled experiments successfully separated xenon from krypton in an air stream at room temperature. Another sorbent, HZ-PAN, was then used to capture krypton at reduced temperatures. Both sorbents have demonstrated high capacities for select species over a wide range of flow, concentration and temperature conditions, along with measured structural integrity after repeated thermal swing cycles.
Microfluidic and millifluidic separations for recovery of plutonium from residues

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Abstract

Operations within the plutonium facility at Los Alamos National Laboratory routinely generate residues that are too rich in plutonium to discard, but not pure enough to be used as feed for the electrorefining purification process. These residues are currently purified for electrorefining using either a nitric acid-based anion exchange process or a hydrochloric acid-based solvent extraction process. While historically successful, these processes were sized for higher throughputs rather than inherent criticality safety, and the flammable solvents and resins require risk management.

A multidisciplinary team at Los Alamos is testing milli- and microfluidic contactors for use in the chemical separation of actinides. These highly efficient separations at short length-scales have the potential to purify plutonium residues with a smaller footprint than conventional aqueous processing. The slow flowrates required for millifluidic processing allows smaller volumes of solution to be used, which makes it easier to design a process that is inherently safe from a nuclear criticality accident. This in turn makes automation feasible. If a need for increased capacity returns, the process can be scaled up by adding parallel systems. By contacting the aqueous phase with a liquid anion exchanger (Aliquat 336) in an ionic liquid carrier, the flammable solvents and resins can be eliminated. Additionally, higher efficiency separations can result in a decreased volume of liquid transuranic waste requiring disposal.

Microfluidic contactors using co-current (slug flow) and counter-current (membrane separated) configurations were fabricated and tested. A higher throughput, commercially available millilitre-scale unit has been purchased for production scale testing. (LA-UR-16-23287)
Separation of technetium and rare earth metals for the co-decontamination process

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Abstract
There are several technologies under consideration for the separation of the useful components of used nuclear fuel in the United States. The co-decontamination process is one of the options being considered to separate U, Np and Pu in a single step to produce a U/ Pu/ Np and U product stream. Although the behaviour of the actinide elements is relatively well defined in this system, the same is not true for all of the fission products; in particular Zr, Mo, Ru and Tc. These elements are a mix of cationic and anionic species, as such, they may interact with each other to extract in a manner not predicted by models such as AMUSE. Recent experimental work has primarily focused on the extraction behaviour of Tc in the presence of a complete and partial metal raffinate simulant in order to define a baseline extraction profile. However, the performance of Tc did not compare well with the predicted AMUSE model and additional testing was developed in order to more closely approximate Tc interactions throughout the co-decontamination process flowsheet. To that end, a series of batch contact tests have been performed to obtain experimentally determined distribution coefficients for these species with the results presented here.

Batch contact flowsheet tests were performed using a used nuclear fuel (UNF) raffinate simulant with one extraction, one scrub, and two strip stages at an organic/aqueous (O/A) ratio equal to 1. The extraction results for all metals present in the raffinate were calculated with DTc and DU found to be 0.46 and 2.0, respectively. Acid dependency experiments showed an unusual trend for Tc compared to AMUSE. Experimentally, Tc remained static at DTc=0.32 ±0.03; however, AMUSE predicted Tc distribution ratios would steadily decrease from a high of DTc= 1.37 at 1M HNO3 to DTc= 0.34 at 4M HNO3. Most of the remaining metals in the acid dependence experiments (U, Zr, and Mo) agreed reasonably with the AMUSE predictions.

Finally a counter-current flowsheet test was performed using conditions predicted to be found in an actual extraction process. The results were compared to the AMUSE model using the same parameters as the experimental data. The experimental results show Tc distribution ratios increased through subsequent extractions and were in reasonable agreement with distributions predicted by AMUSE. Experimental results at an O/A=2 were also in reasonable agreement with the AMUSE model and showed an overall increase in distribution ratios for Tc as compared to the O/A=1 experiments, with the exception of the final extraction stage, which was lower for DTc, but still in agreement with AMUSE.
Selective complexation and separation of high valency actinides for nuclear processes

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Abstract

Increased knowledge of actinide co-ordination chemistry and the development of advanced actinide separation processes are essential to reducing the radiotoxicity of used nuclear fuel. Commercial separation techniques for nuclear fuel have focused on the selective extraction of U(VI) and Pu(IV), however, contemporary efforts are aimed at extracting the minor actinides, e.g. Np and Am. These four mid-actinides (U, Np, Pu, Am) have accessible oxidation states (+V, +VI) at which they exist as linear dioxo actinyl ions [AnO\textsubscript{2}\textsuperscript{n+}], offering the possibility of co-extraction of all 4 actinide cations. The An=O bonds permit ligand co-ordination in the equatorial plane of the metal ion centre, affording a geometry for unique bonding characteristics. Schiff bases can be prepared with synthetic ease and have rather planar structures that chelate around the equatorial plane of actinyl ions through their N\textsubscript{2}O\textsubscript{2} binding site, presenting the possibility that this distinct co-ordination environment may facilitate the selective solvent extraction or aqueous retention of pentavalent and hexavalent actinides.

Preliminary results indicate that these ligands show great promise for group extraction or retention of the mid-actinides in a biphasic solvent extraction process. A lipophilic salen-based ligand prepared in our lab has been shown to form 1:1 complexes with UO\textsubscript{2}\textsuperscript{2+}, NpO\textsubscript{2}\textsuperscript{2+} and PuO\textsubscript{2}\textsuperscript{2+} which have been structurally and spectroscopically characterised. Solvent extraction studies have also demonstrated the ability to extract UO\textsubscript{2}\textsuperscript{2+} from a slightly acidic nitrate media with 90% or higher extraction in one contact and also demonstrated the ability to extract UO\textsubscript{2}\textsuperscript{2+} in the presence of Dy\textsuperscript{3+} and EDTA with a D\textsubscript{U}/D\textsubscript{Dy}=1.63 and a SF\textsubscript{U}/Dy=2037. Preliminary extraction studies have also demonstrated the extraction of NpO\textsubscript{2}\textsuperscript{2+} from slightly acidic media but not NpO\textsubscript{2}\textsuperscript{3+}. Results of the extraction of Ln(III) and An(V/VI) cations from aqueous solution containing a water soluble H\textsubscript{2}salen-SO\textsubscript{3} by bis(2-ethylhexyl)phosphoric acid (HDEHP) in toluene suggests significant retention of UO\textsubscript{2}\textsuperscript{2+} and NpO\textsubscript{2}\textsuperscript{2+} and weak retention of Eu\textsuperscript{3+}. Solvent extraction data indicate that Schiff base ligands may be useful for group separation of mid actinides from trivalent lanthanides. The stoichiometry, acid dependency, kinetics and thermodynamics of the solvent extraction studies and other on-going studies are presented and discussed.
Continuing studies of the effects of gamma irradiation on solvent extraction systems

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Abstract

In addition to adequate solvent extraction performance any solvent system used in the processing of dissolved nuclear fuel must exhibit a substantial resistance to radiolytic degradation. The radiolysis/hydrolysis test loop, located at the Idaho National Laboratory (INL), was designed to allow the investigation of how radiolytic and hydrolytic degradation processes may impact the efficacy of a solvent extraction process flowsheet. The INL radiolysis and hydrolysis test loop has been used to evaluate the effects of hydrolytic and radiolytic degradation upon the efficacy of liquid-liquid extraction process solvents. Aqueous and organic phases corresponding to PUREX/UREX, TRUEX, TALSPEAK, Advanced TALSPEAK, ALSEP, and other separations processes have been studied using the INL test loop. The impact of gamma radiolysis upon the efficacy of a particular process was evaluated using radiotracer techniques. In addition, a suite of analytical techniques were employed to determine the variation in composition as a function of absorbed γ-dose and in some cases identify the radiolytically produced species. The variation in the concentration of solvent components as a function of gamma dose was used to determine g-values or dose constants for the decomposition of solvent components and the production of degradation products. These results are compared with literature values.
Partitioning and transmutation of $^{237}\text{Np}$ to produce $^{238}\text{Pu}$—flowsheet development

Oak Ridge National Laboratory, United States

Abstract

Domestic partitioning-transmutation production of high purity $^{238}\text{Pu}$ by irradiation of $^{237}\text{NpO}_2$ has been restarted in the United States at Oak Ridge National Laboratory (ORNL) and Idaho National Laboratory (INL). A larger scale production project was operated over a 30-year period from the early 1960s to the early 1990s at the Savannah River Plant, but these large scale facilities are no longer available. Existing US Department of Energy research reactors and radiochemical processing facilities are being adapted for the renewed domestic production project.

This paper describes a transmutation path, functional production process steps, chemical processing and kinetic considerations, an example theory on neptunium extraction and partitioning, secondary purification of plutonium product and recycle neptunium, and conversion to oxide of each product fraction. Initial demonstration operations have been performed.

Introduction

Existing research reactors and chemical processing facilities within the US Department of Energy complex will be used in a new programme to produce a domestic supply of $^{238}\text{Pu}$ by partitioning and transmutation of $^{237}\text{Np}$. A flowsheet for production has been developed and demonstrated. Improvements to allow scale-up to production levels have been identified and are being implemented over the next approximately five years to reach a rate of 1500 g/y of Pu containing $>85\%$ $^{238}\text{Pu}$. Previous production at a greater scale was done from the early 1960s to the early 1990s in the large reactors and one of the processing canyons at the Savannah River Plant. However, those facilities have been closed, and only smaller scale research reactors and chemical processing facilities are available today. Thus, the goal is to adapt the existing facilities to re-establish a domestic supply, albeit smaller scale, for use in radioactive thermoelectric generators (RTGs) for outer space applications [1]

Currently, aluminium metal/NpO$_2$ cermet targets are being irradiated in the High Flux Isotope Reactor (HFIR) at ORNL, and other targets will be irradiated in the advanced test reactor (ATR) at Idaho National Laboratory. The irradiations are designed to transmute only 10 to 20% of the $^{237}\text{Np}$ to minimise production of byproduct $^{239}\text{Pu}$ and activation product $^{236}\text{Pu}$. The irradiated targets are being processed at the ORNL Radiochemical Engineering Development Centre (REDC) to separate and dispose of fission products and to partition and purify the $^{238}\text{Pu}$ product and the unburnt $^{237}\text{Np}$, which will be recycled.
Current target design is an aluminium matrix cermet containing $^{237}$NpO$_2$ in the form of pressed pellets. Each target contains ~32 g of NpO$_2$. At this loading, the programme goal will require the fabrication, irradiation, and chemical processing of 400 to 500 targets per year. Reactor physics modeling and post-irradiation examination have indicated that the irradiation time can be increased to decrease the number of targets required to ~300 per year. In addition, studies are under way to evaluate a target design containing only NpO$_2$ pellets contained in a Zircaloy-4 cladding; this target form would further reduce the number of targets required and potentially allow an increased production rate.

The post-irradiation chemical processing is being done in the REDC hot cells and consists of (1) dissolution of the target components; (2) co-decontamination of the Np and Pu from fission product waste and partial-partitioning of the Np and Pu by means of a modified version of a traditional solvent extraction process using tri-n-butylphosphate, which is carried out in three banks of mixer-settler contactors at the REDC; (3) purification of the Pu product and conversion to PuO$_2$ by means of a modified version of the existing REDC cation resin loading/calcination process; (4) purification of the Np by means of ion exchange processes; and (5) conversion of the purified Np nitrate solution to NpO$_2$ powder by means of the ORNL-developed, modified direct denitration process. Improved waste management practices are being evaluated, including solvent and nitric acid recycle, and segregation/potential solidification of the fission products to a calcine waste form for shielded storage and decay of the short-lived fission products.

**Transmutation path**

The primary production path is through $^{238}$Np, as illustrated in Figure 1. However, other reactions must be considered, such as the n-2n and gamma-n reactions on $^{237}$Np to produce $^{236}$Pu which has a long decay chain of alpha-emitters, including $^{208}$Tl which emits a strong 2.7-MeV gamma-ray. This grown-in radiation source would be detrimental to sensitive instruments onboard the spacecraft. Thus, the production of $^{236}$Pu must be minimised. Accordingly, a specification of 2 ppm in the plutonium product has been established.

![Figure 1: Transmutation Path for $^{237}$Np to $^{238}$Pu](image)

Also, the production of $^{239}$Pu by n-gamma reaction on $^{238}$Pu must be minimised to maintain the isotopic composition of the plutonium to >85% (wt) $^{238}$Pu to maximise the heat/weight ratio of the RTG. Thus, the conversion of $^{237}$Np to $^{238}$Pu must be limited to 10-20% per irradiation, and this makes recovery and recycle of most of the $^{237}$Np necessary.
**Functional production process steps**

The functional production path and process steps are shown in Figure 2. Fresh $^{237}$NpO$_2$ must be added after each irradiation cycle to make up for that consumed by production of the plutonium product and fission products. During storage of the inventory of $^{237}$Np and recycled target fabrication scrap, the alpha decay daughter of $^{237}$Np, $^{233}$Pa, grows in, reaches secular equilibrium with a 27-day half-life, and emits a ~0.3-MeV gamma-ray. Thus, it is necessary to remove the in-grown $^{233}$Pa before target fabrication and pre-irradiation handling are done in unshielded facilities. This process step requires dissolution of the $^{237}$Np in nitric acid solution and passage through a glass-bead column to adsorb the $^{233}$Pa. The decontaminated $^{237}$Np nitrate solution can then be combined with freshly recovered and decontaminated recycle $^{237}$Np from irradiated target processing to make feed stock for conversion to oxide and fabrication into new targets.

During the transmutation of $^{237}$Np to $^{238}$Pu, fission products are generated from the $^{238}$Np and $^{239}$Pu. Decontamination of both the plutonium product and the residual neptunium from the fission products is required, followed by partitioning of the plutonium and neptunium from each other. Specifications require <0.5% neptunium and other actinides in the $^{238}$Pu product, and facility specifications require a maximum of 40 ppm $^{238}$Pu in the neptunium fed to target fabrication.

**Chemical processing**

The chemical processing of irradiated NpO$_2$ targets for removal of fission products and partitioning of the $^{238}$Pu product from the residual $^{237}$Np is being adapted to existing equipment within the REDC. The existing equipment was designed initially for other isotope transuranium element production and for experiments with used nuclear fuel reprocessing. The centrepiece is a solvent extraction facility (Figure 3) which provides three 16-stage mixer-settler contactors and enables a processing rate of ~4 g Pu/h. In addition to the solvent extraction facility, the REDC hot cells contain accessory tanks, evaporators, ion exchange columns, liquid transfer systems, and analytical sample preparation facilities.
The flow sheet process steps being developed for processing irradiated NpO₂ targets is compared to the steps used for processing irradiated UO₂ nuclear fuel in Figure 4. An initial preferential dissolution of the aluminium cladding and cermet matrix in caustic-nitrate solution is required for the irradiated NpO₂ targets, whereas the Zircaloy-clad UO₂ fuel rods are cut into small pieces for leaching and dissolution of the oxide fuel components into nitric acid. Similarly, the oxide fuel components of the irradiated NpO₂ targets are dissolved in nitric acid. Both nitric acid dissolver solutions are then processed by means of solvent extraction to remove the un-extracted fission products and to partition the primary resource components (uranium, plutonium, and neptunium from spent fuels and plutonium and neptunium from the NpO₂ targets).

Component concentrations for the irradiated UO₂ used fuel and the irradiated NpO₂ targets are compared in Table 1. Both contain similar plutonium concentrations and radioactivity; however, the radioactivity in the irradiated UO₂ spent fuel is primarily beta emission from decay of ²⁴¹Pu, whereas...
the radioactivity from irradiated NpO₂ targets is more energetic alpha emission from decay of ²³⁸Pu, which produces significantly more decay heat. The fission product concentrations are also similar, but the radioactivity emission from the irradiated used fuel is primarily from intermediate- and long-lived radionuclides, such as cesium and strontium, while that from the irradiated NpO₂ targets is primarily from short-lived zirconium-niobium, ruthenium, and cerium. The greatest differences are the presence of a large concentration of uranium in the used fuel (absent in the irradiated NpO₂ targets), and the much larger concentration of neptunium in the irradiated targets.

Table 1: Typical component concentrations in solvent extraction feed solutions

<table>
<thead>
<tr>
<th>Components</th>
<th>Irradiated UO₂ Fuels (a)</th>
<th>Irradiated Np-Pu Targets (b)</th>
</tr>
</thead>
<tbody>
<tr>
<td>U</td>
<td>300 g/L</td>
<td>—</td>
</tr>
<tr>
<td>Pu</td>
<td>3.5 g/L</td>
<td>3-5 g/L</td>
</tr>
<tr>
<td>Np</td>
<td>0.18 g/L</td>
<td>25-45 g/L</td>
</tr>
<tr>
<td>Am</td>
<td>0.16 g/L</td>
<td>—</td>
</tr>
<tr>
<td>Cm</td>
<td>0.01 g/L</td>
<td>—</td>
</tr>
<tr>
<td>FP Activity</td>
<td>10¹² Bq/L</td>
<td>10¹² Bq/L</td>
</tr>
<tr>
<td>Pu Activity</td>
<td>10¹² Bq/L</td>
<td>10¹² Bq/L</td>
</tr>
<tr>
<td>Pu Decay Heat</td>
<td>0.05 W/L</td>
<td>2.4 W/L</td>
</tr>
</tbody>
</table>

(a) 40 GWd/MT, 5-y decay
(b) 50-75 d irradiation in HFIR Reflector, (~15-25 GWd/MT) 0.5-1 y decay

For the desired co-extraction of neptunium, previous studies [2,3] have shown that increasing the nitric acid concentration from 3M, as currently practised in commercial reprocessing plants, to 4.5 to 5M, is necessary to obtain complete neptunium extraction. Current development studies showed this to be similar for processing the irradiated NpO₂ targets, even with the much greater Np concentration.

The extractability of the neptunium and partitioning from plutonium is dependent on control of their chemical valences. Np(V) and Pu(III) are the prevalent inextractible species, whereas Pu(IV) and either Np(VI) or Np(IV) are the prevalent extractible species. Control of the valences in the desired species is dependent on acid concentration, added oxidants or reductants, and radiolytically generated oxidants/reductants, such as HNO₂ (from HNO₃) and H₂O₂ (from water). Both HNO₂ and H₂O₂ can be oxidising or reducing agents, depending on the solution acidity. Kinetics of the oxidation/reduction reactions are very important and can be different in aqueous and organic solvent phases.

Example theory on Np extraction

Numerous studies have been made on neptunium chemistry and the theory of its behaviour in nitric acid solutions and in solvent extraction operations [1,2]. These studies have shown that Np dissolved in nitric acid is present as both Np(V) and Np(VI), and the relative concentrations are dependent on the aqueous acidity. The aqueous phase equilibrium equation has been shown to be:

\[
2\text{NpO}_2^+ + 3\text{H}^+ + \text{NO}_3^- \rightleftharpoons 2\text{NpO}_2^{2+} + \text{HNO}_2 + \text{H}_2\text{O}
\]
In the two-phase system, both Np(VI) and NO$_2^-$ have a higher affinity for extraction into the organic phase. In addition, the NO$_2^-$ is volatile and can be vented to the atmosphere. As Np(VI) and NO$_2^-$ are extracted into the organic phase, the aqueous-phase equilibrium is shifted to the right, thus making additional Np(VI) available for extraction.

The distribution coefficient of Np(VI), $K_d = [\text{Np(VI)}_{\text{org}}]/[\text{Np(VI)}_{\text{aq}}]$, increases at higher acidity and decreases at lower aqueous acidity. Experience has shown that about 70% of the Np(VI) is extracted at an aqueous acidity of 3M HNO$_3$, and advanced studies have shown that essentially all Np(VI) is extracted at aqueous acidities of 4.5–5M HNO$_3$. Therefore, the aqueous feed acidity for the $^{237}$Np-$^{238}$Pu process was chosen to be 4.5M HNO$_3$.

At lower acidity, $K_d$ is lower, and NO$_2^-$ can be added to shift the equilibrium equation to the left, thus increasing the amount of Np removed from the TBP organic solvent and transferred to the aqueous phase. The added NO$_2^-$ maintains Pu(IV) in the organic phase, thereby partitioning the Np from the Pu.

Kinetics of the oxidation/reduction reactions and the phase transfers of extractible species can influence the success of the desired separations. The existing mixer-settler contactors being used for the solvent extraction process have relatively long contact times compared to other types of contactors such as centrifugal contactors and pulsed columns. A goal of the development project is to enable the use of the existing contactors to provide sufficient separation efficiencies of the fission product wastes and the Np and Pu products. Subsequent polishing separations will be used to produce the final product purity needed.

**Secondary purification and conversion to oxide**

The Pu and Np products from the solvent extraction process require a final purification from each other, as well as from residual fission products and added chemical reductant elements.

**Plutonium product purification and conversion to oxide**

The Pu product from the solvent extraction currently contains excessive amounts of Np and also thorium (Th) impurity which was introduced into the Np target feedstock during operations at the Savannah River Plant. The specification for use of the Pu product in RTGs is <0.5% other actinides. Following purification of the Pu product solution, it must be converted to solid PuO$_2$ for shipment to Los Alamos National Laboratory (LANL). A method in use at the REDC for many years to convert curium (Cm) to oxide beads is cation resin loading/calcination [4] (Figure 5); this method has been chosen for an innovative application to the Pu product for both the removal of the excess Np, and conversion of the Pu product to PuO$_2$ beads. To remove the Np, its valence is adjusted to Np(V), while the Pu valence is adjusted to Pu(IV), thus allowing the Pu(IV) to be adsorbed onto the cation resin while the Np(V) is not adsorbed on the resin. After washing, the Pu-loaded resin is then calcined to produce PuO$_2$. These are batch operations; therefore, the size of the quartz resin loading column is being increased to minimise the number of batches required.
**Neptunium product purification and conversion to oxide**

The Np product from solvent extraction contains sodium ions from the addition of sodium nitrite to reduce the extracted Np(VI) in the loaded solvent to unextractable Np(V), thus transferring Np to the aqueous Np product solution. The Np product also contains trace amounts of fission product zirconium, Pu, and Th. Classical nitrate-based anion exchange was chosen to purify the Np product; however, to enable sorption of the Np, its valence must be reduced to Np(IV). This can be done by adjusting the nitric acid concentration to 7-8 M and then adding hydrogen peroxide as the reducing agent. Again, the loading, washing, and elution of the anion exchange resin is a batch process. Therefore, the size of the anion exchange column is being increased to minimise the number of batches required.

After purification of the Np product solution, it must be converted to NpO₂ for fabrication into recycle targets. Additional NpO₂ must be added from the feedstock produced at the Savannah River Plant to replace the amount converted to Pu and fission products during irradiation. The modified direct denitration (MDD) process [5] (Figure 6), developed at ORNL for conversion of uranium and other actinide elements to an oxide powder, has been adapted for use in converting the purified Np product to NpO₂.
Summary

Domestic partitioning-transmutation production of high-purity $^{238}$Pu by irradiation of $^{237}$NpO$_2$ has been restarted in the United States at ORNL and INL. A larger scale production project was operated over a 30-year period from the early 1960s to the early 1990s at the Savannah River Plant, but these large-scale facilities are no longer available. Existing US Department of Energy research reactors and radiochemical processing facilities are being adapted for the renewed domestic production project.

A chemical processing flowsheet for the $^{238}$Pu production project is being developed and demonstrated using existing radiochemical processing equipment at the REDC. Three 16-stage banks of counter current, mixer-settler contactors are being used for a solvent extraction process in which the primary co-decontamination of Np and Pu from fission products is made, followed by the partitioning of the Pu product solution from the recycle Np product solution. The co-decontamination and partitioning of Np and Pu are complex operations that are dependent on control of variable chemical valences and reaction kinetics. Successful separations have been achieved, but further purification of the Pu and Np product solutions are necessary to meet product specifications and downstream handling requirements in unshielded facilities.

A cation resin loading/calcination process and equipment which was originally developed for curium processing at the REDC, has been adapted for final purification of the Pu product and conversion to PuO$_2$. Classic nitrate-based anion exchange is being used for final purification of the Np product solution, and it is followed by the use of the ORNL-developed MDD process to produce recycle NpO$_2$.

Acknowledgements

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References


Historical capital cost of reprocessing plants

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Abstract

In this work, the historical construction and operational costs of major reprocessing facilities was examined, with special focus on THORP, La Hague and Rokkasho, including comparisons to identify consistencies and discrepancies in reported costs, and with sensitivities to discount rates and operational life. The cost of reprocessing facilities is generally driven by the complexity necessary to ensure adequate safety and security standards, and by the high amount of highly trained personnel. More THORP data are known than for other facilities because of the extended debates about it since its inception.

Only two large commercial facilities are in operation today in the western world, one (La Hague) owned by AREVA in France and the other (THORP) owned by the UK’s Nuclear Decommissioning Authority (NDA). Both facilities have similar reported construction and operational costs.

The Rokkasho reprocessing facility has experienced severe delays and cost overruns: the cost of the Rokkasho facility as provided by JNFL is about twice those of THORP and La Hague/UP3, for the same annual nominal reprocessing capacity, and for essentially the same technology. According to other estimates, the capital costs are even higher. Considerations on how those high cost could be interpreted are proposed in the paper.

Introduction

This paper examines the historical construction costs of major reprocessing facilities around the world, with special focus on Rokkasho, La Hague and THORP, including comparisons to identify consistencies and discrepancies in reported costs, and with proposed considerations on the cost discrepancies between the European and Japanese facilities. Levelised cost calculations are performed to convert capital costs to unit costs.

Historical construction costs of reprocessing plants

Only two large commercial facilities are in operation today in the western world, one (La Hague) owned by AREVA in France and the other (THORP) owned by the Nuclear Decommissioning Authority in the United Kingdom: both plants started operations in the 1990s.

Large-scale reprocessing facilities are also operational in Russia, China, India and Japan. India has a 100 MT/year reprocessing plant operating at Tarapur, a 100 MT/year reprocessing plant at Kalpakkam and a 50 MT/year plant at Mumbai/Trombay. Russia has an operational 400 MT/year reprocessing facility at Ozersk (Chelyaninsk), and a large (3 000 MT/year) facility, planned for completion by 2025 at Zheleznogorsk (Krasnoyarsk) [1]. Japan currently operates the 210 MT/year at...
Tokai facility, which is in the process of stopping operations. The Japan Atomic Energy Agency (JAEA), which owns the Tokai facility shut-down the dissolution part of the plant in 2014 [2], while the treatment part of the facility will continue operating in order to complete the processing of the solutions currently stored at the facility. The 800 MT/year Rokkasho reprocessing facility (discussed later in this paper) is expected to begin operations in 2018 [3].

The 1500 MT/year Magnox reprocessing facility at Sellafield (UK), has reprocessed Magnox-reactors spent fuel for about 50 years, and also oxide fuel from 1969 to 1973. In 1994, the 900 MT/year THORP facility was commissioned in the UK [1]. In France, the 400 MT/year Marcoule facility operated until 1997, and the facility at La Hague has been in operation since 1976.

In the US, while there is considerable experience with defence reprocessing, only three civilian reprocessing plants have been constructed, even though none is currently operating:

- The West Valley, NY, 300 MT/year throughput facility (Western New York Nuclear Service Center) operated successfully between 1966 and 1972, reprocessing about 1000 SNF assemblies. However, because of the regulatory changes applied “retroactively” to the plant, it was deemed un-economical and shut down in September 1976 [4].

- The Morris, IL, 300 MT/year throughput facility (Midwest Fuel Recovery Plant) operated as a pilot scale, but never reached commercial operations due to technical problems and equipment failures, and was declared “inoperable” in 1974. The facility utilised an “Aquafluor” extraction process, and used remotely-operated equipment. It is currently used as an “independent” SNF storage pool [1].

- The Barnwell, SC, 1500 MT/year throughput facility (Barnwell Nuclear Fuel Plant). The PUREX separation facility and the Uranium Hexafluoride facilities were completed and were undergoing pre-operational testing when the NRC terminated all licensing for the facility on December 23, 1977 as part of a US Government policy to cease all commercial reprocessing [1].

Both the THORP and UP-3 (discussed later in this paper), were built with a unique financing arrangement, that provided essentially zero interest financing [5]: the entire construction capital was provided by “pay-ahead” contracts with the utility customers over a “base-load” period of 10 years, without interest nor return to the investors. For this reason, “zero discount rates” are used as reference for the calculations in this paper. The levelised capital costs of the facilities with a “risk free” discount rates of 3% are also reported, since a higher discount rate would likely be more appropriate if such facilities were to be constructed today in the United States.

**Thermal oxide reprocessing plant (THORP) at Sellafield**

The construction of the Thermal Oxide Reprocessing Plant (THORP) at Sellafield, in the UK started in the 1970s and was completed in 1994. Reprocessing operations started in 1997. The current owner is the UK Nuclear Decommissioning Authority and the operator is Sellafield Ltd. The plant is scheduled for closing in 2018, after the exhaustion of all the existing contracts. At its opening, THORP had secured contracts for a total of 5 334 tonnes of LWR spent fuel from utilities in Japan, Germany, Switzerland, Italy, Spain, Sweden, The Netherlands and Canada.

According to BNFL [6] “construction cost of THORP, spread over the ten years 1983-1992, equates to around GBP 1.9Bn. However, taking account of other projects which are directly related to THORP, the overall capital cost of the programme was around GBP 2.85 Bn.”. The average exchange rate over that entire construction time, calculated based on monthly average exchange rates, is
1.6 US Dollars (USD) per British Pound (GBP). Using the CPI inflation estimator between 1988 (in the middle of the construction time) to 2015, a factor of 2.02, this would result in a total "cost of the programme" of USD 9.2 billion in 2015 dollars. Assuming that the plant could operate at the nominal reprocessing capacity of 800 MT/year, and an operational life of 40 years, this would equate to a capital cost of 288 USD\textsubscript{2015}/kgHM at a 0% discount rate, or 532 USD\textsubscript{2015}/kgHM with 3% used for Interest During Construction (IDC) and a 3% discount rate.

Operating costs are not directly known. However, BNFL made publicly available [7] estimates for the operating costs of a "hypothetical" 900 MT/year PWR spent fuel management plant "similar" to THORP, which are reported in Table 1 in millions of 1991 GBP. The average exchange rate for 1991 according to St. Luis Federal Reserve\textsuperscript{1} was 1.76 GBP/USD, and the CPI index between 1991 and 2015 was 1.75. Therefore the annual O&M (operation and maintenance) cost excluding high-level waste (HL vitrification and interim storage, but including intermediate-level wastes (ILW) and low-level wastes (LLW) handling and disposal, would be USD 635 million in 2015 dollars. On a unit basis, this would amount to a total of 705 USD\textsubscript{2015}/kgHM. The total construction and operation cost is therefore estimated at about 1 000 USD\textsubscript{2015}/kgHM with a zero percent discount rate (or 1 237 USD\textsubscript{2015}/kgHM with 3% used for IDC and a 3% discount rate).

However, it could be argued that the actual operating period of the facility is forecasted to be just 21 years (from 1997, beginning of commercial operations, to 2018, forecasted closure time). Amortising the construction cost over a 21 period, even with zero discount rates, would result in capital costs of 550 USD\textsubscript{2015}/kgHM, and a unit cost of 1 250 USD\textsubscript{2015}/kgHM, (or a capital cost of 796 USD\textsubscript{2015}/kgHM, and a unit cost of 1 501 USD\textsubscript{2015}/kgHM with 3% used for IDC and a 3% discount rate).

Reference [7] provides also a useful breakdown of the capital costs for a reprocessing facility "similar to THORP" (shown in Table 2, deduced from the construction experience of THORP by BNFL.

The total cost (minus HLW vitrification and interim storage) is GBP 2.738 billion in 1991 currency, which is only slightly lower than the GBP 2.85 billion reported directly by BNFL for THORP [6]. GBP 2.738 billion in 1991 would result in USD 8.433 billion in 2015 dollars using the exchange and inflation rates described above\textsuperscript{1}. It is noted in [7] that the costs shown in Table 2 does include the associated research and development costs.

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2. Historical exchange rates between the US dollar and the UK pound, provided online by the St. Luis Federal Reserve; source: https://research.stlouisfed.org/fred2/data/EXUSUK.txt.
Table 1: Operating costs for a 900 MT/y reprocessing facility according to a 1991 BNFL estimate provided to Ref. [7]

<table>
<thead>
<tr>
<th>Function</th>
<th>Operating costs (millions of 1991 GBP)</th>
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<tr>
<td>Fuel Receipt and Storage</td>
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<tr>
<td>Reprocessing</td>
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<td>HLW vitrification</td>
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<tr>
<td>ILW encapsulation</td>
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<td>ILW interim storage'</td>
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<tr>
<td>LLW disposal</td>
<td>11</td>
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<td>LLW encapsulation</td>
<td>5</td>
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<td>TOT</td>
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<tr>
<td>HLW handling</td>
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<tr>
<td>Total minus HLW handling</td>
<td>206</td>
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Table 2: Construction costs for a 900 MT/y “spent fuel management” according to a 1991 BNFL estimate provided to [7]

<table>
<thead>
<tr>
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<td>Total minus HLW handling</td>
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</tbody>
</table>

Start up and decommissioning costs should be included in the estimated capital costs. Reference [5] suggests the inclusion of one year of operational costs for the startup costs, to be amortised at the assumed cost of capital over the entire operational life of the facility, and of decommissioning costs of USD 100/kgHM (in 2003 dollars) (or 130 USD_{2015}/kgHM). Under the assumptions of a 40-year life, the added startup costs at 0% discount rates would be about 20 USD_{2015}/kgHM, and 150 USD_{2015}/kgHM including also the decommissioning costs suggested by [5]. Therefore, the total calculated costs of reprocessing based on the total historical cost of THORP would be 1 120 USD_{2015}/kgHM at 0% discount rate, and 1 350 USD_{2015}/kgHM at a 3% discount rate (about 500 USD_{2015}/kgHM for capital repayments, 700 USD_{2015}/kgHM for O&M costs, and 150 USD_{2015}/kgHM for start up and decommissioning).
La Hague

The La Hague plant has 2 lines: UP2-800 and UP-3. Currently owned and managed by AREVA, it features a total annual capacity of 1 700 MT/year. While the La Hague plant was initially built for other purposes, in 1969 it was converted to civilian recycling. From 1990 to 2007 the La Hague site has reprocessed approximately 23 600 metric tonnes of SNF [4].

Costs for the 800 MT/year UP3 plants were reported by COGEMA [8] at 27.8 billion French Francs (FF) in 1992. The average exchange rate between the French Franc in 1992, according to monthly data provided online by the St. Luis Federal Reserve is 5.29 Francs/USD, while the CPI index between 1992 and 2015 increased by a factor of 1.7. The total cost of UP3 in 2015 US dollars (USD) would therefore be USD 8.92 billion, in 2015 dollars, very similar to the similarly sized THORP plant discussed above, also constructed at around the same time. The O&M costs for UP3 were not disclosed by COGEMA, but BNFL estimated those costs to be comparable to those of THORP. Therefore, the analysis conducted above for THORP appears applicable also to UP3.

Rokkasho-mura

The Rokkasho-mura PUREX reprocessing plant has a design annual capacity of 800 MT. It is owned by Japan Nuclear Fuel Limited (JNFL), a partnership led by Tokyo Electric Power Co. (TEPCO). The Rokkasho complex includes (1) the reprocessing facility; (2) a high-level nuclear waste monitoring facility; (3) a MOX fabrication plant; (4) a uranium enrichment plant; and (5) a low-level waste radioactive landfill. Also, it is currently planned to store the high-level vitrified waste on-site at the Rokkasho facility. The Spent Fuel Storage Facility was commissioned in 1999, while chemical testing of the main process facility were performed in 2002, uranium testing in 2004 and active testing in 2006 [9].

Plutonium and uranium will be co-extracted at the Rokkasho reprocessing facility with a modified PUREX process that recombines some uranium with plutonium before denitration, so that the final product is a roughly 50%-50% mix of uranium and plutonium: “To reduce the proliferation risk, the Rokkasho Reprocessing Plant adopted the Plutonium-Uranium Co-extraction Technology that combines recovered uranium with separated plutonium before denitration. Through this process, plutonium is recovered as uranium-plutonium mixed oxide (MOX) at the end of the process, so that the plutonium is never recovered on its own.” [10].

Construction of the facility started in 1993, and completion was initially expected by 1997. However, as of the latest news on the construction progress from Japan Nuclear Fuel Limited, in November 2015, actual reprocessing operations at Rokkasho are forecasted to start in September 2018 [3].

The cost of the facility as provided by the company in March 2015 [10] was of JPY 2 193 billion (Figure 1). At the 2015 exchange rate of about 0.008 USD/JPY, this would amount to USD 17.544 billion in USD2015. This is about twice as high as those of THORP and UP3, for the same annual nominal reprocessing capacity, and for essentially the same technology.

3. Historical exchange rates between the US dollar and the French franc, provided online by the St. Luis Federal Reserve; Source: https://research.stlouisfed.org/fred2/data/EXUSUK.txt.
Figure 1: Cost estimate for the Rokkasho reprocessing site as provided by JNFL (JNFL 2015, page 4)

According to other estimates [11, 12], the capital costs are even higher. For example, according to ref. [11], the Rokkasho plant was originally expected to cost USD 6.9 billion (in 1989 dollars, equal to USD 13.2 billion in 2015 dollars), and its estimated cost had reached USD 20 billion in 2007 (equal to USD 23 billion in 2015 dollars) [11]. However, these estimates do not cite a source, and were presented by “Green Action 2007”. A 2011 estimate [12] puts the total cost at USD 27.5 billion in 2011 dollars, or USD 29.2 billion in 2015, however, from the context of the article, it appears to include the entire cost of developing all the facilities at Rokkasho, including the MOX fabrication, the uranium enrichment plant, nuclear waste facilities, etc. and possibly also the R&D costs. This estimate is about three times as high as those of THORP and UP3.

The multi-decade delays (if startup really happens as expected in 2018, the construction duration would have been 25 years) and large construction costs are mainly attributed to problems with locally designed components, in particular the vitrification equipment, designed by JNFL [12]. The main reprocessing equipment is supplied by AREVA, based on La Hague in France. Few considerations on those large costs can be made with the information available:

- The use of Purchase Power Parity (PPP) for a country like Japan may give a more realistic comparison of costs than simple exchange rates. According to [5], “the use of PPP rates rather than currency exchange rates would have the effect of significantly reducing the very high cost estimates for the Japanese reprocessing plant at Rokkasho-mura". In general, since the construction of a reprocessing plant requires specialised equipment available from a limited number of international vendors, the construction costs of a generic reprocessing plant would be better evaluated using exchange rates. On the other hand, labour costs is better associated with PPP than with the exchange rates, and in the case of Rokkasho, because of the large amount of indigenously-built components, the labour part of the total cost may be larger than it would be with other projects. The PPP ratio between Japan and the US varied between 1.3 and 1.9 in the early-to-mid 1990s.

- The original estimate for the construction of Rokkasho in 1989 was about USD 13.2 billion, in 2015 dollars. By using PPP factors, in the 1990s as described in the previous bullet, the original cost estimate would be converted to USD 6.9 billion to USD 10.1 billion. Those numbers are very consistent with the total cost of THORP and UP3. The original estimate should factor out the effects of cost overruns and delays, thus being close to the “engineering” cost of the facility. This assumption is supported by the consistency of the Rokkasho original estimates with the

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cost of THORP and UP3, combined with the fact that the Rokkasho facility is largely based on THORP and UP3 technology.

- While the details of the cost overruns are unknown for the Rokkasho facility, the framework developed in [13] can provide some useful information. It is known that regulatory intervention during construction can cause large cost overruns and delays for nuclear and other complex facilities. While the extension of the alterations and changes during the construction phase are unknown, the reason given by JNFL for the most recent delays are that “extra time is needed for the safety screenings by the Japan Nuclear Regulation Authority (NRA), including the stricter safety standards imposed by NRA after the Fukushima Daiichi accident”. [14]. Moreover, it is a general phenomenon that lengthy constructions tend to lead to large cost overruns and can become a significant drag on funding.

- The general (above inflation) escalation for nuclear construction projects, described for LWRs in [15] may help reconciling the estimates for Rokkasho and THORP/UP3. It was found in [13] that regulatory stringency tend to happen during periods of construction, with the total size of the nuclear sector being strongly correlated to the level of regulatory stringency. Additionally, it was found in [15] that for “best experience” LWR constructions in the US, the annual cost increase was about 3% during the observation period of 1978-1987; by applying that same cost increase only during the period of LWR construction in the US (i.e. extrapolating from 1987 to 1996 at 3% annual increase), the best experience overnight cost in constant dollars in 1987 would be very consistent with the best experience overnight cost observed today (of about 100 USD/kWe). If that same rate of 3% is applied to the THORP construction cost between 1994 (year of the completion of construction at THORP) and 2016, year of the expected completion of Rokkasho, the escalation in constant dollars (i.e. above the rate of inflation) of the THORP cost would have been almost a factor of 2 (exactly 1.91), thus being very close to the construction cost estimates provided by JNFL.

At a 0% discount rate, the capital charges for a total final cost of USD 17.5 billion (as reported by JNFL), assuming 40 years of operations and the nominal annual throughput of 800 MT/year, would be 550 USD/kgHM (or 940 USD 2015/kgHM with 3% used for IDC and a 3% discount rate). However, by using the PPP conversion as described above, and the original estimates (which should factor out the cost overruns), the capital unit costs at 0% discount rates would be between 216 USD/kgHM and 317 USD/kgHM, and at 3% discount rate it would be between 370 USD/kgHM and 542 USD/kgHM with 3% used for IDC. The total operational cost of Rokkasho for 40 years is expected to be JPY 6.8 trillion [5], which would amount to USD 2 200/kgHM in 2015 dollars using just the conversion rate, or between 1 150 and 1 700 USD/kgHM using the PPP factor between Japan and the US It is not clear if this large cost includes all the facilities present at Rokkasho or just the reprocessing plant. Assuming that this cost would only be for the reprocessing part of the plant, the total reprocessing cost of Rokkasho when using the conversion rate between Japan Yen and US dollars, would be USD 550+USD 2 200=USD 2 750 per kg of heavy metal, largely driven (i.e. about 80%) by the O&M costs at 0% discount rate, and USD 940+USD 2 200=USD 3 140 per kg of heavy metal at 3% discount rate with 3% used for IDC. It is not clear if this total include the cost of startup and decommissioning, and at 0% discount rate it excludes a return on the invested capital, since the facility was constructed under arrangement that provided 0% financing from the Japanese utilities [5]. In ref. [5] it is calculated that when adding all those extra costs, including realistic financing assumptions for a commercial facility, the unit cost of reprocessing at Rokkasho could be as high as 4 100 USD/kgHM, even though “it is not entirely clear which components of the reprocessing-recycling-waste disposal programme are included in that huge total” [5].
However, if using the PPP factor for a proper conversion, and using the original cost estimates as proxies for the “engineering” cost of such facility, thus attempting to exclude factors that drove delays and cost overruns, the unit costs of reprocessing at Rokkasho would be between approximately 1 350 USD/kgHM and 2 000 USD/kgHM at 0% discount rates and between approximately 1 520 USD/kgHM and 2 240 USD/kgHM at 3% discount rates with 3% used for IDC, still largely driven by the O&M costs. More work is needed in the future to understand the reason for the expected large O&M costs of Rokkasho as compared to those of La Hague and THORP.

Conclusions

In this paper the historical construction and operational costs of major reprocessing facilities are examined, with special focus on THORP, La Hague and Rokkasho, including comparisons to identify consistencies and discrepancies in reported costs, and with proposed considerations on the cost discrepancies between the European and Japanese facilities. Levelised cost calculations were performed to convert capital costs to unit costs.

Only two large commercial facilities are in operation today in the western world, one (La Hague) owned by AREVA in France and the other (THORP) owned by British Nuclear Fuels Limited (BNFL) in the United Kingdom: both plants started operations in the 1990s, and both facilities have similar reported construction and operational costs. When normalising for the throughput of the two facilities, the total construction and operation cost was estimated at about 1 000 USD\textsubscript{2015}/kgHM with a zero percent discount rate and 40 years operational life. With different assumptions on the operational life and discount rates, the unit cost would be different, as discussed in the paper. Construction of the Rokkasho reprocessing facility started in 1993, but, actual reprocessing operations at Rokkasho are forecasted to start not earlier than September 2018. The cost of the Rokkasho facility as provided by JNFL recently [10] are about twice as high as those of THORP and La Hague/UP3, for the same annual nominal reprocessing capacity, and for essentially the same technology. According to other estimates [11, 12], the capital costs are even higher. Considerations on how those high cost could be interpreted were proposed in the paper, also in light of the considerations in [13, 16].

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References


A flexible advanced dry head-end for the processing of used nuclear fuel that can be directly interfaced to solvent extraction or fluoride volatility separative processes

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Abstract

Nuclear power is the largest source of carbon-free energy in the USA (63%), producing large amounts of energy with very little fuel consumption by mass. One kilogram of uranium produces about 25,000 times more energy that one kilogram of a fossil fuel (coal or gas). In addition, more than 90% of the components in the used nuclear fuel (UNF) can be recovered and reused to produce even more energy, leaving behind a very small amount of waste. The recycling cost is relatively small (3 to 5 mills/kWh), and it may be lowered by advances in the recycling processes.

To recycle the usable components from the fuel requires the fuel to be separated from the Zircaloy® cladding and put into a form conducive to chemical separations. These processing steps that enable the chemical separations are usually termed head-end. The infrastructure processes, and wastes from the head-end, including the off-gas system, are some of the larger and more costly components of the entire plant. In particular, the behaviour of key volatile species—such as tritium, krypton, and iodine; processing steps such as leaching/dissolution; and some waste streams such as the high-acidity raffinate—have a substantial impact on the overall process and the plant design, operation, and cost.

A novel dry head-end treatment process for UNF using nitrogen dioxide to oxidise the UO$_2$-based fuel is being developed to facilitate head-end operations and reduce the overall cost and complexity of the processing plant. The up-front process removes tritium, iodine, and other volatile fission products and converts the monolithic fuel material to a fine powder that enables either direct extraction into an organic solvent (such as 30% tri-n-butyl phosphate in dodecane) or flame fluorination. The process is well suited for oxide-based fuels used in light water reactors (including both pressurised water reactors and boiling water reactors) and heavy water reactors fuel (i.e. Canada Deuterium Uranium Reactor systems), and it also may have application to sodium-bonded metallic fast reactor fuel and high-temperature gas-cooled fuel.

The process has been demonstrated successfully as a proof-of-principle, and many aspects have been corroborated multiple times at laboratory scale. Current efforts aim at demonstrating the process with high-burn-up commercial used fuel at kilogram scale and developing the process to a technology readiness level sufficient to evaluate and estimate the cost of an engineering-scale implementation.
Advanced TALSPEAK for separating trivalent actinides and lanthanides

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Abstract

An Advanced TALSPEAK (trivalent actinide-lanthanide separations by phosphorus-reagent extraction from aqueous complexes) concept is being developed that uses 2-ethylhexylphosphonic acid mono-2-ethylhexyl ester (HEH[EHP]) to selectively extract trivalent lanthanide (Ln) ions in preference to trivalent actinide (An) ions. The An ions are held in the aqueous phase by complexing with N-(2-hydroxyethyl)-ethylenediamine-N,N',N'-triacetic acid (HEDTA). The primary advantage of the Advanced TALSPEAK system compared to the traditional TALSPEAK system is that it is much less sensitive to the aqueous solution pH. For this reason, it is viewed as being much more robust for industrial application. In this paper, we describe the basic principles of the Advanced TALSPEAK system, as well as experiments conducted to explore the sensitivity of the process to minor changes in the process conditions (for example, changes in the pH, the HEDTA concentration, etc.). The latter experiments showed that the Advanced TALSPEAK system can be expected to be robust in a plant setting. A flowsheet has been developed and a counter-current flow test using a surrogate feed is discussed.
Research and development on pyrochemical treatment of spent nitride fuels for MA transmutation in JAEA

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Abstract

Uranium-free nitride fuel has been chosen as the first candidate for MA transmutation fuel for sub-critical accelerator-driven systems (ADS) and studied in the Japan Atomic Energy Agency (JAEA). Advantages of nitride fuel include high metal density, good thermal properties and large mutual solubility among actinide elements. By using ZrN as an inert matrix, various compositions of MA and Pu in the solid solution type nitride fuel can be fabricated. On the other hand, reprocessing of spent MA transmutation fuels and reusing MA recovered from the spent MA transmutation fuels is necessary to improve the transmutation ratio.

A pyrochemical process has been proposed as the first candidate for reprocessing of the spent nitride fuel for MA transmutation. This paper provides an overview of the current status of technology development, and introduces our recent study. This recent study includes developments of the anode suitable for electrorefining of nitride fuels and the apparatus for renitridation of the metals recovered in Cd cathode for 100g-Cd scale cold tests. Evaluation of the batch sizes of each process with consideration to the decay heat of MA and FP is also introduced.

Introduction

Transmutation of long-lived radioactive nuclides including minor actinides (MA: Np, Am, Cm) has been studied by the Japan Atomic Energy Agency (JAEA). Pb-Bi cooled sub-critical accelerator-driven system (ADS) is regarded as one of the powerful tools for transmutation of MA under the double strata fuel cycle concept. In the concept, the MA contained in high-level waste (HLW) from the commercial fuel cycle is partitioned and fed into the dedicated MA transmutation fuel cycle with ADS. Uranium-free nitride fuel has been chosen as the first candidate for MA transmutation. The advantages of nitride fuel compared with oxide fuel include high metal density, good thermal properties and large mutual solubility among actinide elements. Addition of Pu to the fuel of the first cycle is necessary to obtain high criticality during the early cycles. An inert matrix is mixed to adjust the power density of the core. By using ZrN as an inert matrix, various compositions of MA and Pu in the solid solution type nitride fuel can be fabricated [1-3].

Previous feasibility research for ADS in JAEA shows that MA transmutation ratio per 1 burning cycle is limited to 20%. Reprocessing of spent ADS fuel and reusing MA recovered from the spent MA transmutation fuels is necessary to improve the transmutation ratio. The recovered MA and Pu from the spent MA transmutation fuels mixed with the MA fed from the commercial fuel cycle are used in the second and later dedicated MA transmutation fuel cycle. Our target is to transmute 99% of MA contained in HLW, in order decrease the radiotoxicity of HLW until it drops below that of natural uranium (i.e. from about 5 000 years to about 300 years). Our previous study indicated that
transmutation of 99% MA can be achieved by suppressing the loss of MA on each reprocessing or fuel (re-)fabrication process below 0.1%, if a transmutation ratio per cycle over 20% was achieved. The development of a suitable method for fabrication and reprocessing of MA nitride fuel is needed. A pyrochemical process has been proposed as the first candidate for reprocessing spent nitride fuel for MA transmutation, because this technique has some advantages over the aqueous process, such as the resistance to radiation damage, which is an important issue for fuels containing large amounts of highly-radioactive MA.

This paper provides an overview of the current status of technology development, and introduces our recent study. The development of the anode suitable for electro-refining of nitride fuels and the development of the apparatus for renitridation of the metals recovered in Cd cathode for 100g-Cd scale cold tests are the main topics of our recent study. An evaluation of the batch sizes of each process, which is necessary for estimating the scale of the engineering-apparatus, with consideration to the decay heat of MA and FP, is also introduced.

**Pyrochemical treatment of spent nitride fuels for MA transmutation**

A pyrochemical process has been proposed as the first candidate for reprocessing the spent nitride fuel, because this technique has some advantages over the aqueous process, such as the resistance to radiation damage, which is an important issue for the fuels containing large amounts of highly-radioactive MA. In addition, the compactness of the facility, margin of criticality, and recycling feasibility of $^{15}$N are also advantages of the pyrochemical process [2,4]. For the pyrochemical process, which was originally developed for metal fuel treatment, the electrolysis of nitride fuels and refabrication of nitrides from the recovered actinides have been our main concerns; other main processes have been developed primarily in metal fuel treatment technology. A process flow outline of pyrochemical treatment of the spent nitride fuel is shown in Figure 2 [2]. Our studies are not only on the electorefining, but also the nitridation-distillation combined method for the fabrication of TRU nitride from TRU recovered in Cd cathode and chemical dissolution of anode residues [2,4].

**Figure 1:** Outline of the proposed MA transmutation fuel cycle
Figure 2: A process flow outline of pyrochemical treatment of the spent nitride fuel

Roadmap of development

Figure 3 shows the roadmap of the development of pyrochemical treatment of MA transmutation nitride fuel, proposed by JAEA [5]. On electrorefining and MA recovery technology, basic data measurements and flowsheet design were achieved. Currently, our first tasks are to advance the technology by conducting bench-scale tests using MA samples including those recovered from irradiated fuels, as well as engineering-scale tests with surrogate materials [2]. After completing these tasks, bench-scale tests of the pyrochemical process using irradiated nitride fuels will be conducted in order to demonstrate the engineering feasibility. The tests will require the construction of new hot facilities in which an appropriate amount of irradiated nitride fuels can be handled under an inert gas atmosphere. The development is targeted around 2050 to process the actual spent MA fuel used in ADS.

Figure 3: Roadmap of the development for pyroprocessing of MA transmutation fuel

Note: Time axis does not represent exact periods of development
Development of the apparatus aiming at engineering-scale tests with surrogates

Our latest tasks on the roadmap are conducting bench-scale tests using MA samples including those recovered from irradiated fuels, and engineering-scale tests with surrogate materials. On the pyrochemical treatment of nitride fuels, electrorefining is applicable because of high electric conductivity and relatively low stability of nitrides, though some of the details are different from metal fuel treatment. We are focusing on two subjects which differ from metal fuel treatment; one is related to the generation of nitrogen gas at the anode during the electrolyses, and the other is the necessity of nitride fuel fabrication from actinides recovered in Cd cathode. To develop the apparatus aiming at engineering-scale tests with surrogates, the anode suitable for electrorefining of nitride fuels and the apparatus for renitridation of the metals recovered in Cd cathode are important.

Development of the anode suitable for electrorefining of nitride fuels

Our previous studies on electrorefining of \((\text{U,Pu})\text{N}\) pellet samples indicate the necessary of improving the design of the experimental apparatus especially concerning on the anode [6]. In the previous studies, we used the nitride pellet set in the basket with small holes on the side wall as the anode and a liquid Cd cathode as the cathode. During the electrolyses, continuous stirring of the molten salt bath was necessary to keep the current, to remove \(\text{N}_2\) gas bubbles evolved at the anode. After the electrolyses, no sample remained in the anode basket, but a powder nitride sample was found at the bottom of the molten salt bath. The recovery ratio of U+Pu in the Cd cathode was lower than 50% because the nitride powder was generated by the electrolyses and it dropped out from the anode basket [6].

We decided to improve the design of the apparatus to avoid deposition of the nitride powder from the basket and to remove \(\text{N}_2\) gas bubbles from the surface of the nitride efficiently. Figure 4 shows the apparatus developed at JAEA [7], in which a large basket having no small holes, and a molten salt stirrer set inside of the anode basket are used. Using DyN pellets as surrogate material, the electrolysis of the pellets was completed in 6.6 hours according to the observed total passed charge [7]. The results indicate that the design of the anode developed in this study can be suitable for electrorefining of nitride fuels [7]. Further studies using the developed apparatus are in progress.

Figure 4: Schematic diagram and photos of the electrorefining apparatus [7]
Development of the apparatus for renitridation of the metals recovered in Cd cathode

For renitridation of the metals recovered at the Cd cathode, a nitridation-distillation combined method was developed, in which the Cd alloys containing actinides are heated under a nitrogen gas flow [4, 8]. The studies however have been limited to small scale experiments using 10 mg to 10 g of Cd alloys [6, 8, 9]. Aiming at scale-up tests, development of the apparatus for renitridation of the metals recovered in the Cd cathode for 100g-Cd scale cold tests have been carried out.

Figure 5 shows the larger-scale apparatus which has been developed and installed in an Ar atmosphere glove box. Cd alloy sample is heated under nitrogen gas flow or in a vacuum and distilled Cd is recovered at Cd reservoir located at the bottom of the apparatus. Preliminary experiments using surrogate materials such as Cd-Dy shows that nitride sample can be obtained by the nitridation distillation combined method using this apparatus [10]. Further studies on the renitridation reaction using this apparatus under various conditions are in progress.

Evaluation of the batch sizes of each process

To develop the engineering-scale apparatus for pyroreprocessing, an evaluation of the batch sizes of each process is necessary. The batch sizes of the main processes are being evaluated, such as electrorefining and renitridation. We considered that the batch sizes of treatments are limited by decay heat. Using a typical composition of the spent MA transmutation fuel and conditions of operation, we calculated specific decay heats of spent fuel, liquid Cd cathode with recovered TRU in electrorefining process and the nitride powder obtained by the renitidation of TRU recovered in the Cd cathode. The temperatures of these materials were calculated versus their amounts under the condition of radiation cooling in Ar gas atmosphere. The maximum batch size is defined as the amount of each material reaching the defined limit temperature. Our preliminary calculation indicates that the amounts of materials are in the order of kg-TRU for electrorefining and smaller than kg-TRU for the renitridation process [11].
Conclusion

The status of research and development on the pyrochemical treatment of spent nitride fuels for MA transmutation at JAEA was introduced. Our latest tasks on the roadmap were conducting bench-scale tests using MA samples including those recovered from irradiated fuels, and engineering-scale tests with surrogates. To develop the apparatus aiming at engineering-scale tests with surrogates, we are focusing on the experimental studies on an anode suitable for electrorefining of nitride fuels and the larger-scale apparatus for renitridation of the metals recovered in Cd cathode.

References


Numerical analysis of the zone refining process for fission products separation from the waste salt by using non-equilibrium segregation coefficient

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Abstract
The refining process of the waste salt is necessary for the disposal of the fission products in waste salt as well as the reuse of purified salts during pyroprocessing. In this research, the zone refining process is used for the purification of waste salt which is distinguished from the existing fission product separation methods. We conducted three-dimensional simulation of the process considering heat transfer, fluid flow, mass transfer, and solute trapping in the solid. The distributions and decontamination factors of the fission products are calculated as functions of the heater speed with the non-equilibrium segregation coefficient, and enrichment of the fission products at the interface. For multi-pass cases, analytical solutions are used to calculate the distributions and decontamination factors of the fission products.
Flowsheet development and process safety of GANEX and i-SANEX flowsheets

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Abstract

A range of flowsheet options is being developed for either homogeneous or heterogeneous recycling of minor actinides in advanced fuel cycles (AFCs). At the National Nuclear Laboratory (NNL) Central Laboratory, work has been undertaken to support the development of both scenarios through single stage and multi-stage centrifugal contactor trials and development of models to predict the extraction behaviour in these processes. A “EURO-GANEX” (Grouped ActiNide EXtraction) flowsheet was developed through collaboration with EU partners under the ACSEPT (Actinide reCycling by SEparation and Transmutation) project for the homogeneous recycling of minor actinides. A variation of this “EURO-GANEX” flowsheet has subsequently been tested to obtain separate plutonium/neptunium and americium/curium products to provide a heterogeneous route for recovery of the actinides within a single solvent extraction cycle.

Design and optimisation of these flowsheets has been primarily focused upon achieving the desired performance, essentially high recovery of the minor actinides to minimise losses to waste streams, and good decontamination factors from fission products to meet the required product specification for fuel manufacture. However, the safety of the proposed process is also an important consideration in the design of advanced reprocessing flowsheets. Consequently, it is essential to underpin the normal operating envelope of the flowsheet and assess the impact of potential maloperations upon the process performance. In collaboration with EU partners under the SACSESS project, research at NNL has been directed towards assessment of process safety in both homogeneous (GANEX) and heterogeneous (i-SANEX) flowsheets. This has employed both experimental (rig trials) and modelling approaches to investigate the impact of maloperations upon the process performance. An initial safety review of the EURO-GANEX process was completed under the EU SACSESS project to highlight potential hazards and maloperations in the process that would need to be considered as part of future R&D requirements. Following this initial assessment a trial was carried out using miniature centrifugal contactors to assess the potential impact of plutonium and americium recycle in the extract-scrub contactors of the process. A dynamic model has also been developed for the extract-scrub section of the i-SANEX flowsheet to predict the behaviour of the lanthanides and americium under a range of maloperation conditions. Single stage centrifugal contactor studies were also undertaken to assess the effect of various process parameters upon the extraction of lanthanides and americium in the i-SANEX flowsheet.
Alpha-ray degradation of CMPO adsorbents for the extraction chromatography

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Abstract

The extraction chromatography technology is a candidate technique for minor actinides (MA; Am and Cm) recovery from high-level liquid waste. The Japan Atomic Energy Agency (JAEA) has been conducting research and development on extraction chromatography technology for MA separation. For the safety management of this technology, the focus was put on the degradation of the adsorbent caused by irradiation from adsorbed MA. In this study, characterisation of the degradation products due to irradiation was carried out to gain better understanding of the mechanism of the degradation.

Common chemical structures were found in the alpha-ray and gamma-ray degradation products. Specific positions in the CMPO molecule must be weak and easy to be dissociated by the irradiations. For some degradation products, two oxygen atoms in the CMPO molecule, which contribute to extraction of MA, still remained even after the irradiations. Our previous study showed that elution of Am from CMPO (Octyl(phenyl)-N,N-diisobutylcarbonylmethylphosphine oxide) adsorbent is suppressed by alpha-ray irradiation. Therefore, back-extraction performance from the degradation products with two oxygen atoms might be poorer than that of CMPO. In order to prevent the accumulation of MA inside the packed column, MA elution behaviour from the products should be evaluated and an appropriate wash solution for MA should be proposed.

Introduction

Extraction chromatography technology is one of the candidate techniques for the recovery of minor actinides (MA; Am and Cm) from high-level liquid waste (HLLW), and it is expected to reduce radiotoxicity as well as the amount of final level vitrified waste [1]. In this technology, the adsorbent of porous silica particles, on which styrene-divinyl benzene copolymer is coated (referred to as SiO₂-P) and an extractant for the MA recovery is impregnated into the copolymer [2], are packed into a column, then adsorption/elution reactions enable selective recovery of MA from the HLLW. The Japan Atomic Energy Agency (JAEA) has employed this technology in the advanced aqueous reprocessing process [3] as shown in Figure 1 and conducted various kinds of researches on this technology such as MA recovery trials from genuine HLLW [4, 5], optimisations in the adsorbent for better MA recovery performance [6], etc.

In our previous study, MA recovery from genuine HLLW was achieved through a 2-step column operation [7]. The first column recovered MA and lanthanides (Ln) from the HLLW, and the second column separated MA from Ln. For the adsorbents of the first and second columns, octyl(phenyl)-N,N-diisobutylcarbonylmethylphosphine oxide (CMPO) [8] and bis(2-ethylhexyl) hydrogen phosphate (HDEHP) [9] extractants are typically used, respectively.
Since an engineering-scale apparatus [10] was already developed and its performance demonstrated through 10 times repeated separation operations under inactive environment [11], MA recovery demonstration using the large-scale device will be required for the implementation of the technology as the next step. However, ahead of the engineering-scale hot experiment, a safety management for this technology needs to be established to prevent severe accidents such as fire or explosions. Several accidents on ion exchange process employed in separation of radioactive elements have been reported [12], and most of those were considered to be caused by irradiation degradation of organic compounds consisting of resins.

As the first step of research of the safety management, the behaviour of H₂ gas generated by irradiation inside the packed column was evaluated [13]. In the next stage, investigations are currently being initiated to study degradation of the adsorbents. Changes in the adsorption/elution performance caused by alpha-ray and gamma-ray irradiations and on degradation products of TODGA/SiO₂-P adsorbent produced by the irradiations were previously reported [14, 15]. In this study, the irradiation degradation products of CMPO/SiO₂-P adsorbent were identified, and an attempt to understand the mechanism of the degradation was made.

**Figure 1:** Advanced aqueous reprocessing process

---

**Experimental**

A plate of styrene-divinyl benzene copolymer with 20 mm ×30 mm × 2 mm was prepared, and CMPO as shown in Figure 2 was impregnated into the plate as described in the article by Wei et al [2]. CMPO/SiO₂-P adsorbents were also prepared according to the same article.
He\(^{2+}\) ion beam with 5 MeV, which simulates alpha-ray from \(^{241}\)Am, was irradiated on the plate set inside a vacuum chamber. A slurry of CMPO/SiO\(_2\)-P adsorbent mixed with the same volume of water was also irradiated by He\(^{2+}\) ion beam, using the same irradiation conditions described in [16]. The He\(^{2+}\) ion beam irradiation experiments on the plate and the slurry were carried out at tandem accelerators in Wakasawan Energy Research Centre, Japan and Takasaki Ion accelerators for Advanced Radiation Application (TIARA) of National Institute for Quantum and Radiological Science and Technology (QST), Japan, respectively. For both experiments, the current of the beam was 200 nA and the integrated irradiation dose was 2 MGy.

Gamma-ray from \(^{60}\)Co source was irradiated on the slurry with 3 kGy/h up to 2 MGy. The dose rate was calculated from the measured value obtained by the alanine-ESR method [17, 18]. The irradiation experiment was performed in the irradiation room No 1 of Co-60 Gamma-ray Irradiation Facility No 1 in Takasaki Advanced Radiation Research Institute, QST, Japan.

The irradiated plate or the adsorbents separated from the water was washed with 1,2-dichloroethane, then the degradation products remaining on the adsorbent were transferred to the organic phase. The chemical species extracted in the 1,2-dichloroethane from the plate and the adsorbent were analysed by GC/MS, NMR and FT-IR measurements.

**Results and discussion**

**Alpha-ray degradation products**

The alpha-ray degradation products observed in the irradiated plate are shown in Table 1. The degradation products No 1 and 2 are considered to be produced by dissociation of P-C bond in the CMPO molecule and of the bond between C neighbouring P and C in the carbonyl base, respectively. Dissociations of those two positions must be a dominant interaction between alpha particle from \(^{241}\)Am and the CMPO molecule.
Table 1: Alpha-ray degradation products of CMPO observed in the irradiated plate

<table>
<thead>
<tr>
<th>ID</th>
<th>Structure</th>
</tr>
</thead>
<tbody>
<tr>
<td>Degradation product No 1</td>
<td><img src="image1" alt="Structure" /></td>
</tr>
<tr>
<td>Degradation product No 2</td>
<td><img src="image2" alt="Structure" /></td>
</tr>
</tbody>
</table>

The degradation products found in the irradiated slurry are shown in Table 2. The products are different from those observed in the irradiated plate, and dissociations of C-N bonds in the CMPO molecule seem to be a main influence of the irradiation on this system. Since bond dissociation energy of C-N in organic compounds is smaller than those of C-O, P-O and C-P [19-21], shielding against He$^{2+}$ ion beam by water in the slurry might lead to the difference in the influence. Dioctylphosphinic acid might be produced by dissociations of P-phenyl and P-C bonds

Table 2: Alpha-ray degradation products of CMPO observed in the irradiated plate

<table>
<thead>
<tr>
<th>ID</th>
<th>Structure</th>
</tr>
</thead>
<tbody>
<tr>
<td>Degradation product No 3</td>
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</tr>
<tr>
<td>Degradation product No 4</td>
<td><img src="image4" alt="Structure" /></td>
</tr>
<tr>
<td>Degradation product No 5</td>
<td><img src="image5" alt="Structure" /></td>
</tr>
<tr>
<td>Dioctylphosphinic acid</td>
<td><img src="image6" alt="Structure" /></td>
</tr>
</tbody>
</table>

Gamma-ray degradation products

Table 3 shows the degradation products of CMPO/SiO$_2$-P adsorbent produced by gamma-ray irradiation, where a product dissolved in the nitric acid solution is also shown. The degradation product No 1 and 2 were also produced by He$^{2+}$ irradiation on the plate, thus the dissociated bonds in the CMPO molecule must be easily influenced by the effect of irradiation. The product No 6 was
observed only after the gamma-ray irradiation, and it must be produced by oxidation of dissociated CMPO molecule due to oxidising water-derived radicals such as H and OH.

Most of the degradation product No 2 remained on the adsorbent, and a small fraction of the product was also found in the co-existing solution and only this product was observed in the solution. Therefore contamination of the product solution by degraded organic compounds might not be significant.

Table 3: Gamma-ray degradation products of CMPO

<table>
<thead>
<tr>
<th>ID</th>
<th>Structure</th>
<th>ID</th>
<th>Structure</th>
</tr>
</thead>
<tbody>
<tr>
<td>Degradation product No 2</td>
<td>C₆H₁₂O</td>
<td></td>
<td>PCH₃</td>
</tr>
<tr>
<td>Degradation product No 1</td>
<td>C₆H₁₂O</td>
<td></td>
<td>POH</td>
</tr>
<tr>
<td>Degradation product No 6</td>
<td>C₆H₁₂O</td>
<td></td>
<td>PCH₂COOH</td>
</tr>
</tbody>
</table>

Comparison in the degradation products

Figure 3 shows the bonds of the CMPO molecule in which dissociations by the irradiations were observed in this study. Dissociations at several positions were commonly observed after gamma-ray and alpha-ray irradiations as seen in our previous study on TODGA/SiO₂-P adsorbent [15]. Those positions are considered to be easily dissociated by the interaction with the ion beam or radicals due to their relatively small bond energies as discussed above. Dissociation at the position ○ was observed for both irradiations, however the resultant chemical species depended on the type of the irradiation. The difference might be caused by existence of the water and oxidising water-derived radicals. Existence of the co-existing water might work as a shield for He⁺⁺ ion beam irradiation from outside of the adsorbent as discussed above since the irradiation on the slurry seems to degrade the CMPO molecule less than the irradiation on the plate. Those irradiation experiments and analysis employed in this study possibly missed volatile degradation products, therefore TOF mass analysis simultaneously with the irradiation must be required to observe such kind of products.
In our previous study, alpha-ray irradiation on the CMPO/SiO$_2$-P adsorbent by adsorption of $^{241}$Am has been carried out [14]. Some parts of the adsorbed $^{241}$Am was eluted from the adsorbent during the irradiation. In the experiments, elution of the remaining $^{241}$Am was also performed. The ratio of the remaining $^{241}$Am during irradiation and the elution ratio of $^{241}$Am after the irradiation are shown in Figure 4 and 5, respectively. Both the amount of the remaining $^{241}$Am and their elution ratio decreased to 80 % of the initial values by 0.4 MGy irradiation, and those properties decreased with the increase in the irradiation dose. These results imply that degradation products which do not have the extraction ability of Am are produced by the irradiation and that some of the degradation products of CMPO can extract, Am even in a low acidic environment. The extraction of Am by CMPO occurs through co-ordination of 2 oxygen atoms in the carbonyl base of the CMPO molecule around the Am$^{3+}$ ion. Those two oxygen atoms remain in the molecule for the degradation products No 3, 4 and 5, thus they may still have the ability for extracting Am from acidic media. In order to explain the poor elution behaviour from the irradiated adsorbents, the elution performance of Am from those products should be investigated. The degradation product No 1 has a similar structure with that of HDEHP which can extract Am from low acidic media. If the product forms a stable complex with Am under low acidity environment, it may also influence the poor elution property from the irradiated adsorbent. Extraction and back-extraction performances of those products for Am need to be investigated to reveal the mechanism of reduction in the performance of the adsorbent.
Conclusion

The irradiation degradation products of CMPO/SiO$_2$-P adsorbent for MA recovery process were analysed after He$^{2+}$ ion beam (alpha-ray) or gamma-ray irradiations. Dissociations at several bonds in the CMPO molecule were commonly observed for gamma-ray and alpha-ray irradiations, and common degradation products were produced. However, oxidation of dissociated CMPO molecule due to oxidising water-derived radicals such as ·H and ·OH was also confirmed for the gamma-ray irradiation. Some of the degradation products are considered to influence the adsorption/elution performance of the adsorbent, therefore a detailed understanding of the chemical behaviour of those products needs to be investigated for the safety management of the extraction chromatography technology.

References


Iodine adsorption in fixed bed adsorber tests

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Abstract

Compliance to air emissions regulations for future used nuclear fuel reprocessing facilities in the United States depends on the ability to control gaseous emissions of radionuclides, including iodine-129. $^{129}$I evolves in gaseous iodine-bearing species during several phases of used nuclear fuel aqueous reprocessing, including head-end processes, fuel dissolution, and waste processing.

Control of gaseous $^{129}$I emissions has been studied worldwide for at least the past 40 years. Both wet and dry scrubbing processes have been designed, tested, and used in certain applications. Chemisorption processes using Ag as the active ingredient on a relatively inert support have been identified and used with the most success. In these processes, gas streams flow through a fixed bed of support media which contains Ag. As the gaseous iodine species adsorb onto the media, the iodine reacts with the Ag to produce relatively non-volatile AgI.

In the US, $^{129}$I emissions control has been studied specifically under the Off-gas Sigma Team of the Material Recovery and Waste Form Development Campaign (and its predecessors) since 2009. This research has included bench-scale deep-bed tests with different gas mixtures (to emulate primarily dissolver off-gas at this time) and different sorbents (mainly Ag mordenite, also known as Ag zeolite [AgZ], and Ag aerogel [AgAero]). The deep-bed adsorption testing has focused on determining (a) iodine adsorption efficiencies, (b) sorbent capacities and iodine utilisation efficiencies, and (c) mass transfer zone (MTZ) parameters.

Test results so far have indicated:

- High iodine adsorption efficiencies are possible, with decontamination factors (DFs, the flowrate of gaseous iodine into the adsorber divided by the flowrate gaseous iodine exiting the adsorber) up to about 10,000 for I$_2$, about 3,000 for CH$_3$I for the test conditions.

- MTZ depths ranged between 1.2 cm (0.5 in.) to 5 cm (2 in.) for I$_2$ and 10-20 cm (4-8 in.) for CH$_3$I, but increased over time, reducing iodine sorption capacities at time of bed break-through; deeper sorbent beds may be needed for efficient CH$_3$I capture.

- Ag utilisation ranged between about 30% and 70% for most test durations; most were about 50%.

- Maximum I$_2$ sorption capacities averaged ~10 g per 100 AgZ and ~20 g per 100/g AgAero.

- Nearly all adsorbed iodine was strongly-held physisorbed iodine for both I$_2$ and CH$_3$I.

Additional tests are needed to provide adsorption results for a wider range of off-gas conditions and additional iodine species, to provide additional sorbent performance and selectivity information, and to advance the technology readiness level for Ag sorbents for use in the US.
Synthesis and characterisation of actinide fluorides for studies on molten salt reactor fuel cycle

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Abstract

One of the objectives of the advanced nuclear reactor concepts developed within the Generation IV International Forum (GIF) is to reduce the amount and radiotoxicity of the spent fuel and also effective utilisation of the resources. Minimisation of the generated nuclear waste is likewise the goal of P&T scenarios, which aim at reducing the long-term radiotoxicity of spent fuel by transmuting Pu, minor actinides and long-lived fission products in dedicated reactors.

Molten salt reactor (MSR) is one of the systems studied within GIF. A wide variety of MSR concepts were proposed, e.g. a breeder reactor operated under \(^{233}\)U-\(^{232}\)Th fuel cycle or a transmuter burning Pu and other transuranic elements; different systems were based on fast, non-moderated or thermal neutrons; the fuel can be dissolved in fluoride or chloride molten salt media, etc. Studies on properties of actinides dissolved in the MSR fuel salt are crucial for designing and establishing safe reactor long-term operation and for achieving satisfactory fuel salt clean-up with recovery and recycling of actinides.

This work deals with the synthesis and characterisation of selected actinide fluorides for studies on their thermodynamic and electrochemical properties. A unique, specially designed and manufactured experimental set-up is described, enabling fluorination of actinides by pure gaseous hydrogen fluoride. It consists of a glove box operated under pure argon atmosphere, a high-temperature furnace with a horizontal fluorination reactor and a HF gas. Procedures for syntheses of selected actinide fluorides are mentioned as is the established analytical procedure for controlling the purity of the obtained fluorides. The starting materials for fluorination were oxides, obtained from oxalate decomposition at moderate temperature to form fine powders with high-surface area. After fluorination at temperatures of 450-600°C, using static or flow-through procedure, the products were characterised by X-ray diffraction (XRD) and differential scanning calorimetry (DSC) methods, enabling precise detection of possibly remaining oxide or other impurities. The obtained fluorides were pure as in each case the XRD showed a composition of one phase corresponding to the expected product and at the same time, the DSC also revealed the presence of a single compound with correctly corresponding melting points.

The work was carried out within the European EC/EURATOM Horizon 2020 research project SAMOFAR, which deals with the safety assessment of the molten salt fast reactor (MSFR) concept for both the reactor standard and transient operations and for the chemical plant proposed for the fuel salt cleaning and recycling.
Aspects of off-gas treatment for advanced head-end and back-end processes in UNF recycle plants

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Oak Ridge National Laboratory, United States

Abstract

The chemical processing of used nuclear fuel (UNF) will result in the release of significant fractions of the volatile and semivolatile fission product radionuclides during the head-end treatment steps. The headend processing steps traditionally include the mechanical shear and the dissolution process. Advancements in the head-end processing systems can change the step at which specific volatiles and semivolatiles are separated from the fuel, the fractions of the volatile and semivolatile compounds that are released from the fuel at each step, the concentrations of the volatiles and semivolatiles in the effluent gas stream, and the chemical composition of the carrier gas stream (caused by the use of alternative process reagents).

Tritium pre-treatment has been proposed as an additional head-end step to simplify tritium management. Standard tritium pretreatment is a dry process wherein the fuel is oxidised in the presence of air or oxygen, and it is expected to result in near-complete release of tritium from the oxide fuel matrix. The process also releases radiologically significant quantities of other volatile radionuclides contained in the fuel, including carbon (\(^{14}\)C), iodine (\(^{129}\)I), and krypton (\(^{85}\)Kr). The fractions released have been reported at ~50% of the carbon (as CO\(_2\)), ~1% of the iodine, and ~10–50% of the krypton.

Advanced tritium pre-treatment using alternate reagents has the potential of releasing larger fractions of the volatile radionuclides and a number of the semivolatile components, all of which must be addressed in the off-gas treatment systems. Chemical de-cladding adds reagents that volatilise the zirconium in the alloy and may increase the release of trapped gases. There is also the potential to volatilise additional species contained in the fuel matrix. Dissolution of the fuel in nitric acid releases the remainder of the volatile fission gases and may produce semivolatile species that will also be found in the dissolver off-gas.

The dissolver off-gas stream usually contains large concentrations of acid vapour, by-product acid gases (NOX), and water vapour. In the absence of tritium pre-treatment, other tritium management techniques may be needed to address the accumulation of tritium in both the aqueous streams and off-gas streams within the facility. Processes are also being explored to recycle zirconium from the cladding. Tritium management may also be required to address the tritium contained within the cladding. The addition of pre-treatment processes or chemical de-cladding to the head-end processes could result in off-gas systems significantly different from those currently found in commercial reprocessing facilities.

The systems may be required to handle both dry and humid gas streams, streams laden with additional reagent gases, and streams that are potentially more concentrated in the volatile radionuclides. Off-gas treatment systems may include the potential for the recycle of reagent gases.
as a waste reduction measure. This paper will provide descriptions of off-gases from several advanced head-end and backend operations and the associated off-gas treatment systems. Impacts of processing parameters on the relative quantities of volatile components released and recovery efficiencies will be discussed.
Investigation of the radical oxidation chemistry of fuel cycle aqueous buffers and holdback reagents

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²Idaho National Laboratory, Aqueous Separations & Radiochemistry Division, United States

Abstract

The performance and efficiency of actinide (An) separation from the lanthanides (Ln) in processes such as TALSPEAK, Advanced TALSPEAK, and ALSEP is highly dependent on the aqueous solvent conditions; particularly the aqueous solution pH and the presence of radiation that will degrade both the diluent and added solutes. While much previous work has demonstrated that the presence of differing concentrations of buffers such as citric or lactic acid protects the radiolytic degradation of the holdback reagents used in these extraction processes, little is known about the kinetics involved in this chemistry.

Since the utilisation of aqueous actinide holdback reagents is essential for these An/Ln separations, understanding the role played by the buffers in preventing reagent radiolytic degradation is critical for continuing development of these advanced aqueous separations processes. Therefore, we have initiated an investigation of the radiation-induced chemistry of the various components of these solvent systems. Here we report upon our kinetic measurements for the hydroxyl (\(^{\cdot}\)OH) radical induced degradation of DTPA and citric acid.

These measurements were performed using the linear accelerator system at the Radiation Research Laboratory, University of Notre Dame to quantitatively isolate this radical in aqueous solution. As no direct transient absorbance change could be used, these data were measured using competition kinetics, with 100 ppm KSCN as a standard. This standard was chosen as its hydroxyl radical reaction rate constant has been well established:

\[^{\cdot}\text{OH} + \text{SCN}^- \rightarrow (\text{SCN})_2^+ + \text{OH}^- \quad k_3 = 1.15 \times 10^{10} \text{ M}^{-1} \text{ s}^{-1} (1)\]

These data are shown in Figure 1. We have also used this approach to perform kinetic measurements at multiple pH’s, which, when combined with literature temperature-dependent pKa values, have also allowed us to deconvolute specific rate constants for all individual ionised species for these chemicals.
Electrochemical study on LiCl-KCl-LuCl₃ using W and Bi cathode

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¹Seoul National University, Korea, ²Korea Institute of Nuclear Safety, Korea

Abstract

Spent nuclear fuel reprocessing is one of the most important issues in the nuclear industry, with particular regard to the safety and security of high-level radioactive materials as well as the recycling of useful resources. Pyroprocessing technology has been studied to co-recover electrochemically transuranic elements from lanthanide series elements with high proliferation resistance and accident tolerance. In this study we focused on electrochemical studies of LuCl₃ in LiCl-KCl eutectic salt. Lutetium has low concentrations of spent nuclear fuel composition but there are only a few previous studies on Lutetium. Therefore we investigated cyclic voltammetry and electrolysis in LiCl-KCl-LuCl₃ on W cathode and Bi cathode to provide information of Lu³⁺ in molten salt.
Characterisation of material non-uniformity to quantify uncertainty in material accountancy in pyroprocessing

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Abstract

In order to explore how the non-uniform nuclide composition in spent fuel affects uncertainties in material accountancy in pyroprocess, high-fidelity depletion simulations by the SERPENT code, with the necessary resolution in the radial and axial directions, have been conducted to determine location-dependent nuclide compositions in the representative spent fuel assembly. To mitigate the computational limitation for a large number of radial and axial segments in the assembly-scale simulation, we have decoupled radial non-uniformity from axial non-uniformity. The radial non-uniformity for each axial location has been predetermined by additional assembly-scale simulations in which the fuel rods are modelled with multiple segments in the radial direction. Then, the radial distributions have been applied proportionally to the results of the assembly-scale simulation by matching the results with the axial height of the assembly.

The probability distribution for nuclide composition after the chopping process, which is the first process in the head-end process, is plotted. Based on that, in order to quantitatively evaluate the uncertainty associated with material accountancy, we have applied a random-sampling approach. The normality for distributions obtained by the random-sampling method is tested by a Q-Q plot in a graphical manner and the Jarque-Bera test in a statistical manner. The results of tests approximately show that the data sets for the sample size 5, 10, 20, 30, and 40 cases are normally distributed. In addition, the uncertainty in determination of the Pu-244 Cm-ratio after the chopping process would not be appropriate, because that of 244 Cm concentration in samples would be a significant level due to the non-uniformity of 244 Cm in spent fuel. This approach will be applied to evaluate uncertainties for material accountancy after the voloxidation and pre-treatment processes in the future.

Introduction

One of methods to evaluate safeguards for a nuclear facility is material accountability [1]; therefore, material accountability for the nuclear facility, for instance, the pyroprocessing system, should be evaluated. In order to measure the Pu amount in pyroprocess, the Pu to 244 Cm-ratio method, which is based on Equation 1, has been suggested [2].

\[
\text{(Pu mass)} = \left( \frac{\text{Pu}}{\text{244 Cm}} \right) \times \left( \text{244 Cm mass} \right)
\]

The two basic principles in this method are i) the dominant neutron source in spent fuel is 244 Cm; therefore, the mass of that can be measured by a neutron counting rate [2][3], ii) it is claimed that the Pu to 244 Cm-ratio has not varied during the key-pyroprocess such as electrolytic reduction,
electro refining, and electro winning [4]. Based on the two concepts, once we set the ratio before the key-pyroprocess, the Pu mass can be indirectly evaluated by measuring the $^{244}\text{Cm}$ mass.

The uncertainty of this method can be from the evaluation of the ratio and the neutron detecting to measure the $^{244}\text{Cm}$ mass. Technologies to enhance the neutron detecting have been developed [5][6]. The ratio can be determined either by instruments [2] or numerical methods [7]. The ratio in spent fuel rods or assemblies can be varied with respect to the axial and radial locations, because of the non-uniformity of burnup [8] that can lead the non-uniform nuclide compositions in discharged spent fuels. However, when the Pu to-$^{244}\text{Cm}$-ratio method is being applied in pyroprocess, how the non-uniform distributions of ratio and nuclide composition effect on the Pu material accountability has not been discussed.

In order to evaluate the uncertainty of the Pu to-$^{244}\text{Cm}$-ratio method by the non-uniformity of nuclide composition, the following works are planned in this study; i) the determination of representative spent fuel assemblies for which the non-uniformity is fully characterised, ii) the development of a process-flow model for the head-end process in which the ratio is evaluated, and iii) the evaluation of uncertainty in the ratio determination resulting from the material non-uniformity in spent fuel.

Characterisation for the representative spent fuel assembly

The representative spent fuel assembly

In order to select the representative spent fuel assembly, the types and number of nuclear power plants in South Korea has been investigated. The most frequent type of nuclear power plants is optimised power reactor (OPR)-1000 as 12 among 21 pressurised water reactors (PWR) [9]. According to Yu’s report [10], the 3 types of assemblies (Type-0, -1, and -2) are loaded in the core. The difference among the 3 types of fuel assembly is a number of gadolinia-bearing fuel (GBF) and its loading location in an assembly as shown in Table 1. This study is only focusing on the Type-0 fuel assembly. The detail design parameter for the assembly, which is PLUS7, is shown in Table 2. The average power of one assembly (15.90MW/assembly) is evaluated by the total thermal power of the OPR-1 000 core (2815MW [10]) divided by the number of assembly in the core (177FAs/core). The chemical composition of cladding, ZIRLO, is shown in the reference [11].

<table>
<thead>
<tr>
<th>Type</th>
<th>Num. of FAs/core</th>
<th>Initial Enrichment [w%]</th>
<th>Num. of fuel rods/FA</th>
<th>Num. of GBFs/FA</th>
</tr>
</thead>
<tbody>
<tr>
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<td>4.5/4.0</td>
<td>184/52</td>
<td>0</td>
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<tr>
<td>Type-1 FA</td>
<td>72</td>
<td>4.5/4.0</td>
<td>184/52</td>
<td>8</td>
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<tr>
<td>Type-2 FA</td>
<td>57</td>
<td>4.5/4.0</td>
<td>184/52</td>
<td>12</td>
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Table 2: Design parameters for PLUS7 fuel assembly [10]

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Value</th>
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</thead>
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<tr>
<td>Pellet density [g/cm³]</td>
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</tr>
<tr>
<td>Pellet diameter [cm]</td>
<td>0.819</td>
</tr>
<tr>
<td>Clad inner diameter [cm]</td>
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<tr>
<td>Clad outer diameter [cm]</td>
<td>0.952</td>
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<tr>
<td>Cladding</td>
<td>ZIRLO [11]</td>
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<tr>
<td>Active length [cm]</td>
<td>381</td>
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<tr>
<td>Fuel pitch [cm]</td>
<td>1.288</td>
</tr>
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<td>Fuel assembly</td>
<td>16X16</td>
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<tr>
<td>GBF</td>
<td>6w% Gd2O3</td>
</tr>
<tr>
<td>Power [MW/Assembly]</td>
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</tbody>
</table>

The depletion simulations by SERPENT

In order to achieve the nuclide composition in spent fuel, the depletion simulations have been conducted by SERPENT, which is the continuous-energy Monte Carlo burnup code [12]. For depletion conditions, the numerical recipes developed in the previous study have been applied. The summary of the numerical recipes are following; 1) the xenon equilibrium calculation in SERPENT is applied, 2) the burnup step is less than 25 days, 3) the active fuel rod region for each fuel pin cell in the assembly is divided by 30 meshes. For the middle mesh, the larger size of mesh is given, while the smaller sizes of meshes are given for meshes at the top and bottom of the fuel rod. And 4) the axial moderator temperature distribution evaluated by the energy conservation method is applied for the entire burnup step. Because the assembly is symmetric, the quarter of assembly is modelled to save the running time. Because the depletion for fuel assemblies are differently scheduled depending on the loading location in the core, the different depletion periods and cycles (1-, 2-, and 3-cycle depletion period) for Type-0 fuel assembly has been considered. In addition, assumptions are applied due to lack of information; 1) the length of 1 cycle is 18 months with the 90% capacity factor, 2) there is no boron in the moderator, 3) the power of assembly is constant during the entire depletion period, and 4) the cooling time of each spent fuel assembly after discharging is 5 years. Based on the above information, the quarter of Type-0 fuel assembly and its fuel pin cell generated by SERPENT are shown in Figure 1. The numbers in the top-view for the assembly indicate the lattice number for modelling.

The depletion simulation results are shown in Figure 2. The figures in the left and right are the axial distributions for Pu and $^{244}$Cm mass densities of each fuel rod in the Type-0 spent fuel assembly, respectively. The 3 different colour lines in the figures are indicating the different depletion periods as 1- (blue), 2- (black), and 3- (red) cycle depletion periods with the 5 year cooling time. The dot lines are indicating the average mass density for the entire assembly.
Figure 1: Modelling of the fuel pin cell in the assembly simulation

Figure 2: Axial mass density distributions for Pu (left) and $^{244}$Cm (right) of each fuel rods in the Type-0 FA with respect to 1 (blue lines), 2 (black lines), and 3 (red lines) cycle depletion periods

Because the particle size of powder after the voloxidation process in the head-end process of pyroprocess is approximately ten micro-metre, the radial non-uniformity of nuclide composition of fuel pellets could also have a strong impact on the material accounting. Thus, the radial non-uniformity in fuel rods should be taken into account for a more realistic simulation of sampling. However, the previous simulations do not take into account the radial distribution of nuclide composition, because simulating fuel rod burnup with sufficient meshes in both the axial and radial direction requires excessive computation time and computer memory. Therefore, the radial nuclide composition distribution for each fuel rod is predetermined by analysing that of the representative fuel rod. The representative fuel rod consists of 10 meshes in the radial direction and 30 meshes in the axial direction, which is equivalent with the axial mesh configuration applying for the assembly-scale simulation, as shown in Figure 3. In the previous simulations, each fuel pin cell in the assembly is named with the different lattice number for modelling as shown in Figure 1, while the identical lattice number is given for the identical fuel rod type as shown in Figure 3. By giving this equal lattice number, the representative radial distribution as a function of the axial location for each fuel rod type (4.0w%, and 4.5w% fuel rod) in the assembly can be achieved. The achieved representative radial distribution of nuclide composition is applied proportionally to the results of the axial distribution simulations by matching the results with the axial height of the fuel rods. In this way, the approximate non-uniformity of nuclide composition in the radial and axial directions of each fuel rod in the assembly can be achieved.
**Figure 3**: Modelling for the fuel pin cell with the radial and axial meshes for the active fuel rod region

In order to decouple two independent simulations, the normalised mass densities for each axial location in the fuel rod (axial and radial) are evaluated as the mass density of each radial mesh divided by the average mass density of that axial layer of meshes, as shown in Equation 2 and Figure 4. Then, the mass density of each axial mesh for the assembly-scale simulation is substituted into the Equation 3 in order to produce the radial distribution of nuclide composition of each axial mesh.

\[
\text{Normalized } MD_{ij} = \frac{\text{MD of } i^{th} \text{ mesh on } j^{th} \text{ layer}}{\text{Avg. MD of } j^{th} \text{ layer meshes}}
\]  

(2)

where, MD: Mass density  
i: ith mesh in the radial direction, j: jth (layer) mesh in the axial direction

\[
\text{MD}_{ij} = \text{Normalized } MD_{ij}^{AxRad-sim} \times MD_{ij}^{Ax-sim}
\]

(3)

- i: ith mesh in the radial direction, MD: Mass density  
j: jth (layer) mesh in the axial direction  
- AxRad - sim: results simulated for a fuel rod designed with axial radial meshes  
- Ax - sim: results simulated for a fuel rod designed with axial meshes

The total number of discrete meshes is 30 in the axial direction and 10 in the radial direction, for a total of 236 fuel rods. Different radial and axial mesh sizes are applied for the depletion simulations; therefore, the volumes of each mesh are different. To account for the fact that some meshes are larger than others, the SERPENT simulation result is rearranged into 381 and 20 equally sized segments for the axial and radial directions, respectively, for each fuel rod. The larger meshes are thus broken up into smaller meshes that all have the uniform nuclide composition. Finally, the nuclide composition data for the assembly is rearranged in a 381-by-20-by-236 matrix form.
The probability distributions for nuclide composition in Head-end process

**Chopping process**

A spent fuel assembly, which is generally around 4 m in length, could not be an appropriate form as a feed material for pyroprocessing. It should be disassembled and pre-treated to enhance efficiencies in pyroprocess. To begin with, the disassembling and chopping is assigned. The length of chopping has not been decided, however, literatures have mentioned a potential and conceptual length. The one to two inches in length of chopped pieces is introduced in Williamson’s paper [13]. The IAEA report mentions that the sheared fuel sections are generally 3 to 5cm in length [14]. In order to calculate the necessary reactor size for decladding and voloxidation process, the length of piece considered is 3, 5, 7, and 10cm [15]. The length of UO₂ pellet as a feed material for voloxidation experiments are 1.5cm [16] and 1.01 cm [17]. In this study, the 1cm in length of chopped piece, which is generally closed to the length of one pellet, is assumed, because not only the latest publication considers 1.01cm in length [17], but also the smaller length of piece would increase the non-uniformity of nuclide composition distribution in the products. The total number of pieces from one assembly is 889 916 (=381pieces/rod*236rods/assembly). Based on this, the histograms for Pu mass density after the chopping process (1cm) for the Type-0 spent fuel assemblies depleted during 1- (top), 2- (middle), and 3- (bottom) cycles are plotted as shown in Figure 5 and Table 3.

**Random sampling after the chopping process**

In this section, uncertainties for samples associated with Pu and ²⁴⁴Cm mass densities have been statistically evaluated. For that, the mean and standard deviation of sample mean is evaluated by the random-sampling approach. The various sample size has been considered for that such as 5, 10, 20, 30, and 40 pieces. In order to be consistent the total number of sampled pieces for the entire sampling repetition, the number of trials for 5, 10, 20, 30, and 40 sample size cases are 240 000, 120 000, 60 000, 40 000, 30 000 times, respectively. The histograms for the mean of sample mean in each sample size case for the Type-0 assembly with 3 different depletion periods are plotted in Figure 6. The different colour bars are utilised to distinguish different sample size cases (sample size: 5 (blue), 10 (red), 20 (green), 30 (black), and 40 (yellow)). The statistical summary for these is shown in Table 4.
Figure 5: Histograms for Pu mass density in the chopped pieces (Type-0 FA; 1-cycle depletion results [top], 2-cycle depletion results [middle], and 3-cycle depletion results [bottom])

Table 3: Means and standard deviations for Pu mass density in chopped pieces

<table>
<thead>
<tr>
<th>Pu</th>
<th>1 cycle</th>
<th></th>
<th>2 cycles</th>
<th></th>
<th>3 cycles</th>
<th></th>
</tr>
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<tbody>
<tr>
<td></td>
<td>Mean</td>
<td>Std</td>
<td>Mean</td>
<td>Std</td>
<td>Mean</td>
<td>Std</td>
</tr>
<tr>
<td>Mass density [g/cm³]</td>
<td>5.45E-02</td>
<td>7.98E-03</td>
<td>8.49E-02</td>
<td>8.22E-03</td>
<td>1.04E-01</td>
<td>8.42E-03</td>
</tr>
</tbody>
</table>
Figure 6: Histograms for Pu mass density by random sampling of chopped pieces (Type-0 FA; 1-cycle (top), 2-cycle (middle), and 3-cycle (bottom) depletion results)
Table 4: Statistical results for the random-sampling simulations

<table>
<thead>
<tr>
<th>Sample Size (# of pellet)</th>
<th>Depletion periods</th>
<th></th>
<th></th>
<th></th>
<th></th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>1 cycle depletion</td>
<td>2 cycles depletion</td>
<td>3 cycles depletion</td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>Mean</td>
<td>Std</td>
<td>Mean</td>
<td>Std</td>
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<tr>
<td>5</td>
<td>5.45E-02</td>
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<td>8.49E-02</td>
<td>3.68E-03</td>
<td>1.04E-01</td>
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<tr>
<td>10</td>
<td>5.45E-02</td>
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<td>8.49E-02</td>
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<td>1.04E-01</td>
</tr>
<tr>
<td>20</td>
<td>5.45E-02</td>
<td>1.78E-03</td>
<td>8.49E-02</td>
<td>1.84E-03</td>
<td>1.04E-01</td>
</tr>
<tr>
<td>30</td>
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<td>1.46E-03</td>
<td>8.49E-02</td>
<td>1.50E-03</td>
<td>1.04E-01</td>
</tr>
<tr>
<td>40</td>
<td>5.45E-02</td>
<td>1.26E-03</td>
<td>8.49E-02</td>
<td>1.31E-03</td>
<td>1.04E-01</td>
</tr>
</tbody>
</table>

**Normality test**

The normality for obtained distributions in Figure 6 is evaluated in this section. There are several methods to evaluate the normality of a distribution. In this study, two methodologies, the Quantile-Quantile (Q-Q) plot as a graphical method and the Jarque-Bera (JB) test as a statistical method [18], have been applied. For Q-Q plots, the y- and x-axis are the quantiles of obtained data and standard normal quantiles, respectively. The blue lines lie close to the red linear lines as the obtained data is close to the normal distribution. As shown in Figure 7, by increasing the sample sizes, the obtained distribution by random sampling is getting close to the normal distribution.

**Figure 7:** Q-Q plot for Pu concentration distributions in Type-0 spent fuel assembly depleted during 3 cycles depending on different samples sizes (5, 10, 20, 30, and 40)

In order to apply the JB test method, two parameters, skewness and kurtosis, for the obtained data should be evaluated. Based on two, the JB value, which is approximately following the chi-square distribution, has been evaluated by Equation 4. Then, the chi-square test with the two degree
of freedom can be conducted to evaluate the goodness of fit between the obtained distribution and the normal distribution. The significant values for sample sizes are shown in Table 5 [18]. If the evaluated JB value is less than the significant value, the null hypothesis, which is the observed distribution follows the normal distribution, is accepted. If not, the null hypothesis is rejected.

\[
JB = \frac{N}{6} \left[ S^2 + \frac{(K-3)^2}{4} \right] \sim \chi^2
\]

\[
S = \frac{1}{n} \sum (x_i - \bar{x})^3, K = \frac{1}{n} \sum (x_i - \bar{x})^4
\]

where, \( S \) : skewness, and \( K \) : kurtosis, \( N \) : sample size

Table 5: Significant values and its level for different sample sizes

<table>
<thead>
<tr>
<th>N</th>
<th>( \alpha=0.10 )</th>
<th>( \alpha=0.05 )</th>
</tr>
</thead>
<tbody>
<tr>
<td>20</td>
<td>2.13</td>
<td>3.26</td>
</tr>
<tr>
<td>30</td>
<td>2.49</td>
<td>3.71</td>
</tr>
<tr>
<td>40</td>
<td>2.70</td>
<td>3.99</td>
</tr>
</tbody>
</table>

The JB values with skewness and kurtosis for the Pu mass density distributions generated by the random-sampling method for each sample size and depletion period case are shown in Table 6. The all of skewness is negative values which mean that the distributions show the right tail is longer than the left tail. The skewness value is closed to zero by increasing the sample size that means the histogram for data sets is getting more symmetric. The kurtosis for normal distribution is three as a reference. That for all cases is closed to three. By increasing the sample size, we would say that the distribution is close to being the normal distribution, because the skewness and kurtosis are closed to zero and three, respectively. Even though Table 5 is not showing the significant value for 5 and 10 sample size cases, it would be concluded that the obtained data sets are normally distributed at the 10% significant level.

Table 6: Skewness, kurtosis, and JB for the obtained Pu data sets for each sample size case
Discussion

The magnitude of non-uniformity for nuclide composition in spent fuel has been varied depending on burnup. As shown in Figure 2, the standard deviations for Pu and $^{244}$Cm mass densities have been decreasing as increasing burnup. According to the burnup credit study [8], the non-uniformity of normalised axial burnup is decreased as increasing the entire discharged burnup in spent fuel. This phenomenon proportionally influences on the axial distributions for Pu and $^{244}$Cm concentration. Straightforwardly, the decreasing axial non-uniformity could lead the smaller variation of Pu mass density for pieces generated after the chopping process as shown in Table 3 and Figure 5.

Based on these distributions, the random sampling has been conducted to quantify the uncertainty associated Pu and $^{244}$Cm mass density by the non-uniform nuclide composition in spent fuel. The relative standard deviations for Pu mass density in chopped pieces for 3 different burnup cycle cases are less than 3%, if the sample size is greater than 30. In contrast, that for $^{244}$Cm is still around 10%. Therefore, the uncertainty in determination of ratio could be negatively and greatly affected by the significant non-uniformity of $^{244}$Cm. In other words, it would be claimed that the determination of ratio would not be appropriate using the products after the chopping process in the head-end process.

Conclusion

The high-fidelity depletion simulations by SERPENT have been conducted to achieve the detail and reliable non-uniformity of nuclide composition in the representative spent fuel. For that, the decoupling of results in between the axial and radial high-resolution simulations has been developed. The main concept of this coupling is the obtained representative radial distributions for each fuel rod type are applied proportionally to the results of the axial distribution simulations by matching the results with the axial height of the fuel rods. In this study, the characterisation for the Type-0 fuel assembly has been conducted. Other representative fuel assembly types (Type-1 and -2) will be characterised by the method shown this study in the future.

The head-end process consists of chopping process, voloxidation process, and pre-treatment process. In this study, the probability distributions and uncertainty of nuclide composition have been evaluated after chopping process. Those after the voloxidation and pre-treatment processes will be conducted in the future. Finally, the evaluated uncertainty in determination of the ratio in the head-end process will be propagated through the entire process to discuss the material accountability of Pu, when the Pu-$^{244}$Cm-ratio method is applied.

References


The effect of nitric acid and americium concentrations on Am(VI) reduction kinetics

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Abstract

Numerous solvent extraction processes have been proposed in an attempt to fully close the nuclear fuel cycle and incorporate recycling of the reusable materials. One common component among the industrially relevant separations systems is the oxidation state manipulation of Pu and Np. Recent efforts to streamline the processes necessary to recycle used nuclear fuel have included oxidising americium, one of the minor actinides found in the fuel after irradiation. Oxidising Am(III) to Am(VI) can simplify used fuel recycling by incorporating a group actinide decontamination where U, Pu, Np, and Am are co-extracted in one step and then selectively back extracted into appropriate product streams. Americium(III) oxidation to Am(VI) is easily achieved in high nitric acid medium using the powerful oxidant sodium bismuthate. This process is represented in the following equation:

$$2\text{Am}^{3+} + 3\text{BiO}_3^- + 10\text{H}^+ \rightarrow 2\text{AmO}_2^{2+} + 3\text{Bi}^{3+} + 5\text{H}_2\text{O}$$ (1)

This type of process would require a complete understanding of not only Am(III) oxidation to Am(VI) but also subsequent reduction of the Am(VI) back to Am(III). For this system, the auto reduction rates for Am(VI) to Am(V) and Am(III) are complicated by secondary reactions with radiolysis products produced by americium alpha decay. Another complicating factor which could affect the Am(VI) reduction kinetics includes disproportionation reactions in higher acid media. The principle secondary oxidation/reduction reactions (Equations 2-5) and the Am(V) disproportionation reaction (Equation 6) are represented using the following expressions:

$$\text{AmO}_2^{2+} + \cdot \text{OH} \rightarrow \text{AmO}_2^{2+} + \text{OH}^-$$ (2)

$$2\text{AmO}_2^{2+} + \text{HNO}_2 + \text{H}_2\text{O} \rightarrow 2\text{AmO}_2^{2+} + 3\text{H}^+ + \text{NO}_3^-$$ (3)

$$2\text{AmO}_2^{2+} + \text{H}_2\text{O}_2 \rightarrow 2\text{AmO}_2^{2+} + 2\text{H}^+ + \text{O}_2$$ (4)

$$\text{AmO}_2^{2+} + 2\text{HO}_2^+ + 2\text{H}^+ \rightarrow \text{Am}^{3+} + 2\text{O}_2 + 2\text{H}_2\text{O}$$ (5)

$$3\text{Am(V)} \rightarrow 2\text{Am(VI)} + \text{Am(III)}$$ (6)

The goal of this project is to monitor the reduction of Am(VI) to Am(V) and Am(III) and determine the kinetic reaction rates, the kinetic reaction orders, and the simultaneous secondary reaction mechanisms. It is envisioned that the layers of complexity associated with these types of kinetics can be peeled back and a deeper fundamental understanding of Am(VI) reduction in the presence of higher concentrations of americium metal ions and nitric acid can be obtained.
ALSEP chemistry: From fundamental to applied testing

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Pacific Northwest National Laboratory, United States

Abstract

There has been a significant interest in the development of an extraction scheme to separate the trivalent lanthanides from minor actinides to decrease the long-term radiotoxicity of used nuclear fuel. One process, referred to as the Actinide-Lanthanide Separation Process – ALSEP – combines the neutral diglycolamide N,N,N',N'-tetra-2-ethylhexyl diglycolamide (T2EHDGA) with the acidic extractant 2-ethylhexylphosphonic acid mono-(2-ethylhexyl) ester (HEH[EHP]) in an alkane diluent. The ALSEP solvent extracts trivalent actinides and lanthanides from this acidic solution, which can then be separated in a stripping step using buffered polyaminocarboxylic acid solution. Research conducted at Pacific Northwest National Laboratory (PNNL) has investigated fundamental and applied aspects of the ALSEP process chemistry. This paper describes our current results on the speciation and transport behaviour of individual and combined T2EHDGA and HEH[EHP] extractants under conditions relevant to the ALSEP system. The extraction speciation analysis is supported by the obtained information on the structure of the organic phase complexes by various spectroscopic techniques. To evaluate ALSEP performance under complexity of the actual used fuel, it was tested using dissolved high burn-up ATM-109. In this experiment, ATM-109 fuel dissolved in nitric acid was first subjected to the tributyl phosphate extraction to remove uranium, plutonium, and neptunium. The obtained nitric acid-based high-level raffinate served as a feed to the ALSEP extraction, scrub, and strip steps. The results of this testing are presented.
Effects of gamma irradiation on the adsorption characteristics of xerogel microcapsules

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¹Japan Atomic Energy Agency, Japan
²Tohoku University, Japan

Abstract

Innovative nuclide separation systems with alginate gel polymers (ALG) enclosing various extractants (xerogel microcapsules) have been developed. Among xerogel microcapsules, ALG containing no extractants, ammonium tungstophosphate encapsulated into ALG (AWP-ALG) and potassium copper hexacyanoferrate encapsulated into ALG (KCuFC-ALG) were found to be suitable for the separation of Zr, Cs and Pd, respectively. In this study, these xerogel microcapsules were irradiated with gamma rays and the dependence of their adsorption characteristics on dose was investigated. The adsorption characteristics of AWP-ALG and KCuFC-ALG were nearly independent of dose due to the high irradiation resistance of the inorganic exchangers (AWP and KCuFC). While, the uptake amounts of Zr onto ALG decreased as the dose of gamma rays increased. Zirconium adsorbed onto the ALG itself. Thus, the change of adsorption characteristics of ALG on Zr is caused by the degradation of ALG by gamma rays. These results will contribute to the design of a separation process with xerogel microcapsules.

Introduction

Innovative nuclide separation systems with alginate gel polymers (ALG) enclosing various extractants (xerogel microcapsules) have been developed for the reduction of radioactive wastes as well as the effective utilisation of resources in high-level radioactive liquid wastes (HLLW) [1-4]. Xerogel microcapsules are composed of a carrier matrix (ALG, silica gel, etc.) and specific extractants or inorganic exchangers (ammonium tungstophosphate ((NH₄)₃PW₁₂O₄₀) (AWP), potassium copper hexacyanoferrates (K₂Cu₉/2[Fe(CN)₆]₃) (KCuFC), etc.) included inside the matrix. For the adsorption of Cs and Pd on xerogel microcapsules, AWP and KCuFC were identified as specific extractants or inorganic exchanger, respectively. ALG itself, which is one of the xerogel microcapsules, is suitable for the adsorption of Zr. An example of the flow with xerogel microcapsules is shown in Figure 1. Zirconium is removed by ALG to prevent the formation of precipitates comprising hydrated zirconium molybdate. Cesium is removed by AWP-ALG to utilise Cs-137 as a heat and radiation source. Palladium is removed by KCuFC-ALG to utilise as catalysts. Although it was confirmed that unirradiated xerogel microcapsules (ALG, AWP-ALG and KCuFC-ALG) show relatively higher uptake to Zr, Cs and Pd, respectively, the adsorption characteristics of ALG, AWP-ALG and KCuFC-ALG after gamma irradiation over 17.6 kGy were not reported. In this study, these xerogel microcapsules were irradiated with gamma rays up to 3 898 kGy and the dependence of their adsorption characteristics on dose was investigated.
Experimental

Preparation of xerogel microcapsules

ALG was prepared according to the following procedure. Sodium alginate solution (50 cm$^3$, 3.0 wt%) was added dropwise to 0.5 M Ca(NO$_3$)$_2$ solution while stirring at room temperature to form spherical gel particles. After standing overnight, the gel particles had settled and become rigid. The particles were separated from the calcium solution, washed with deionised water and air-dried at 30 °C for 2 days. The gel particles were then treated with 0.1 M HNO$_3$ solution to replace Ca$^{2+}$ with H$^+$. Then the gel particles were washed with distilled water and finally air-dried at 30 °C for 1 day.

AWP-ALG was prepared in the following way. Fine AWP powders (0.5 g) were kneaded with NaALG solution (50 cm$^3$, 1.5 wt%) and fully dispersed using a kneader equipped with rotation and revolution functions. The well-kneaded sol was then dropped into 0.5 M Ca(NO$_3$)$_2$ solution via a medical needle under constant stirring at room temperature to form xerogel microcapsules. These were then stirred gently for one night to enhance ageing, separated from the solution, washed with distilled water and finally air-dried at 30 °C for 2 days. KCuFC-ALG was prepared by the gelation of kneaded sol of NaALG (sodium alginate) and ferrocyanide ions with 0.5 M Cu(NO$_3$)$_2$ solution. The subsequent procedure was the same as that for AWP-ALG.

Gamma-ray irradiation

The prepared xerogel microcapsules were irradiated with gamma rays. ALG, AWP-ALG and KCuFC-ALG, in a dry condition, were irradiated under a gamma dose rate of 2.8 kGy/h by Co-60 gamma-ray source for 5 hours, 5 days and 58 days. Each irradiation time corresponds to a dose of 14 kGy, 470 kGy and 3 898 kGy, respectively.

IR spectra measurements

The infrared (IR) spectra of xerogel microcapsules before and after gamma-ray irradiation were obtained by KBr method. The specimens were prepared in the following way. Two-hundred mg of KBr powder and 1 mg of xerogel microcapsules were mixed and pressed and, disc-shaped samples were prepared. IR spectra measurements were conducted by Fourier transform infrared spectroscopy (FT-IR, HORIBA, FT-730).
Determination of relative uptake

Metal ion solutions (Zr, Cs or Pd) were obtained by diluting standard solutions (1000 ppm). The uptakes of metal ions for xerogel microcapsules were determined by batch method. Aqueous solutions (5 cm$^3$) containing 10 ppm of the metal ions were shaken in contact with 50 mg of xerogel microcapsules at 25±1 °C. The experimental conditions are shown in Table 1. The concentrations of Cs$^+$ and other metal ions in aqueous phase were measured by atomic absorption spectrophotometry (AAS, Thermo Fisher Scientific K.K., AA-890) and inductively coupled plasma-atomic emission spectrometry (ICP-AES, SII, SPS7800). The uptake (R, %) of metal ion removed from the solution and relative uptake (RR) are defined as:

$$R_i = \left( \frac{C_0 - C_f}{C_0} \right) \times 100 \quad (1)$$

$$RR = \frac{R_{\text{after}}}{R_{\text{before}}} \quad (2)$$

where $C_0$ and $C_f$ (ppm) are the concentrations of metal ions initially and at equilibrium, respectively; $R_{\text{after}}$ and $R_{\text{before}}$ (%) correspond to the uptake of metal ion before and after irradiation.

<table>
<thead>
<tr>
<th>Adsorbent</th>
<th>ALG</th>
<th>AWP-ALG</th>
<th>KCuFC-ALG</th>
</tr>
</thead>
<tbody>
<tr>
<td>Metal ion</td>
<td>Zr</td>
<td>Cs</td>
<td>Pd</td>
</tr>
<tr>
<td>Aqueous phase</td>
<td>Volume (cm$^3$)</td>
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<td>5</td>
</tr>
<tr>
<td></td>
<td>Nitric acid concentration (M)</td>
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<td>2</td>
</tr>
<tr>
<td></td>
<td>Metal ion concentration</td>
<td>Zr 10 ppm</td>
<td>Cs 10 ppm</td>
</tr>
<tr>
<td>Solid phase</td>
<td>Weight (g)</td>
<td>0.05</td>
<td>0.05</td>
</tr>
<tr>
<td>Shaking time (hours)</td>
<td>20</td>
<td>24</td>
<td>12</td>
</tr>
</tbody>
</table>

Results and discussion

Appearance and structure after gamma irradiation

The appearances of xerogel microcapsules before and after gamma irradiation are shown in Figure 2. The colour of ALG and AWP-ALG became brown with increased dose. The brown coloration of xerogel microcapsules showed the generation of chromophore-like ethylene groups and carbonyl groups, which was expected to be generated by radiolytic degradation of the ALG.

The IR spectra of xerogel microcapsules before and after irradiation are shown in Figures 3-5. The increase of carboxyl groups (COOH and/or COO$^-$) was observed for ALG and AWP-ALG with an increased dose. A degradation mechanism of alginate by gamma irradiation was proposed by Luan et al. [5]. They showed the generation of carbonyl groups and carbonyl groups by link cleavage of glycoside bonding in ALG caused by gamma rays. Since ALG composed of xerogel microcapsules were expected to be degraded by gamma rays, ALG and AWP-ALG turned into brown.
The degradation of KCuFC-ALG was expected to be inhibited by Cu$^{2+}$ inside the KCuFC-ALG. Several forms of cation-exchange resins with different cross-linkages were subjected to gamma irradiation by Ichikawa et al. [6]. The results showed that the reduction behaviour of metal ions adsorbed onto cation-exchange resin during irradiation inhibited the degradation to a significant extent, since H radicals produced by radiolysis of resin itself were converted to H$^+$ by redox reaction with reducing agents like Fe$^{3+}$, Cu$^{2+}$ and UO$_2$$^{2+}$. In this study, ALG, AWP-ALG and KCuFC-ALG were composed of alginitic acids, calcium alginites and copper alginites, respectively. Among these cations (H$^+$, Ca$^{2+}$ and Cu$^{2+}$) in ALG, copper ions are most likely to reduce. Thus, there is possibility that the degradation of ALG in KCuFC-ALG was probably inhibited by reduction of Cu$^{2+}$.

The absorption peak around 800 cm$^{-1}$ in Figure 4 was ascribed to W-O bonding of AWP [7]. Since an intensity of the peak is almost constant during gamma irradiation, AWP inside the AWP-ALG was found to be not degraded by gamma rays up to 3 898 kGy.

**Figure 2: Appearance of xerogel microcapsules**

<table>
<thead>
<tr>
<th>Unirradiated</th>
<th>14 kGy</th>
<th>470 kGy</th>
<th>3898 kGy</th>
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<tbody>
<tr>
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<td><img src="image2" alt="Image" /></td>
<td><img src="image3" alt="Image" /></td>
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<tr>
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<td><img src="image7" alt="Image" /></td>
<td><img src="image8" alt="Image" /></td>
<td><img src="image9" alt="Image" /></td>
</tr>
</tbody>
</table>

**Figure 3: IR spectra of ALG**

![Graph showing IR spectra of ALG with peaks at different wavenumbers](image10)
The relative uptakes of xerogel microcapsules after irradiation are shown in Table 2. The adsorption characteristics of AWP-ALG and KCuFC-ALG were nearly independent of dose in this range. The relative uptakes of Cs onto AWP-ALG and Pd onto KCuFC-ALG were almost constant up to a dose of 3 898 kGy. Cesium and Pd adsorbed onto inorganic exchangers (AWP or KCuFC) inside the ALG. Due to the high irradiation resistance of the inorganic exchangers [8-9], the adsorption characteristics of AWP-ALG and KCuFC-ALG were maintained during gamma-ray irradiation. While, the relative uptakes of Zr onto ALG decreased as the gamma-ray dose increased. Zirconium adsorbed onto ALG itself. ALG is known to be degraded into lower-molecular-weight polymers by gamma-ray irradiation [10]. The change in adsorption characteristics of ALG to Zr was caused by the degradation of ALG.
Table 2: Relative uptakes of xerogel microcapsules

<table>
<thead>
<tr>
<th>Adsorbent</th>
<th>ALG</th>
<th>AWP-ALG</th>
<th>KCuFC-ALG</th>
</tr>
</thead>
<tbody>
<tr>
<td>Metal ion of interests</td>
<td>Zr</td>
<td>Cs</td>
<td>Pd</td>
</tr>
<tr>
<td>Uniradiated</td>
<td>1.00</td>
<td>1.00</td>
<td>1.00</td>
</tr>
<tr>
<td>Irradiated</td>
<td>14 kGy</td>
<td>1.00</td>
<td>1.00</td>
</tr>
<tr>
<td></td>
<td>470 kGy</td>
<td>0.28</td>
<td>0.98</td>
</tr>
<tr>
<td></td>
<td>3 898 kGy</td>
<td>0.08</td>
<td>0.98</td>
</tr>
</tbody>
</table>

Conclusion

Three kinds of xerogel microcapsules, ALG, AWP-ALG and KCuFC-ALG, for adsorption of Zr, Cs and Pd were irradiated with gamma rays and the dependence of their adsorption characteristics on dose was investigated by Co-60 gamma-ray source. AWP-ALG and KCuFC-ALG showed high stabilities of the adsorption characteristics to the gamma irradiation up to 3 898 kGy. The degradation of ALG by gamma rays was confirmed based on changes of IR spectra and appearance before and after gamma irradiation. The relative uptakes of Zr onto ALG decreased with increased dose. These results will contribute to the design of separation process with xerogel microcapsules.

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References


Study on cesium removal using heteropolyacid salts in HCl system

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Abstract

A novel reprocessing process with nuclide separation was proposed, based on chromatographic techniques in HCl solution system. In this reprocessing system, the pyridine resin was used for the main process. The highly-radioactive nuclides should be removed before sending the spent fuel solution to the main process, although the pyridine resin shows high radiation resistance. Radioactive cesium is one of the main radiation generators in the spent fuel. It is well known that the heteropolyacid salts have a high cesium selectivity and a high cesium adsorption capacity. Ammonium molybdophosphate (AMP) and ammonium tungstophosphate (AWP) were tested for the investigation on the feasibility of cesium removal from a radioactive solution in HCl system. The distribution coefficients in HCl solution were evaluated, and the tests confirmed that both AMP and AWP have a high cesium selectivity. The stabilities of AMP and AWP in HCl solution were also investigated. The slight dissolution of molybdenum and tungsten were confirmed. The dissolution of both the AMP and AWP are suppressed in the higher concentration of HCl. Using these results, the appropriate process for the removal of cesium was discussed for our proposed reprocessing system.

Introduction

Many elements co-exist in spent nuclear fuel. Uranium and plutonium can be recycled by reprocessing. The remaining elements (i.e. minor actinides, lanthanides, platinum group elements (PGM), etc.) are considered as high-level waste (HLW), if they are not extracted from the spent nuclear fuel. However, if extracted from the spent fuel, these elements will become valuable resources. Thus, the recovery of these valuable elements was proposed as an addition to the reprocessing process. The proposed reprocessing system with nuclide separation processes is based on the chromatographic technique in hydrochloric acid solution and consists of a dissolution process, a reprocessing process, a MA separation process, and a nuclide separation process [1]. The schematic diagram of the process is shown in Figure 1. A pyridine resin was chosen as the main ion exchange resin because of its higher radiation resistance in comparison with quaternary ammonium resin (especially in HCl system) [2]. Another important advantage of the pyridine resin is that it has two functions; one is the anion exchanger, and another is the soft-donor ligand. Thus, many nuclides can be separated or removed. For example, group separation between the trivalent actinides and lanthanides was achieved in HCl solution [3]. We demonstrated this process of STEP II and III (except STEP I) in continuity using an irradiated MOX solution (20%Pu MOX irradiated in “Joyo” with a burnup of 148GWd/t) [4] and simulant spent fuel solution [5]. Step II confirmed that PGM (platinum group metal) and Tc can be recovered and removed from spent fuel by pyridine resin filter. In Step-III, the recovery of U and Pu and the separation of MA(III) from lanthanides was confirmed. Another experiment [6] showed that Np can be recovered with U and Pu. In the proposed process, the cesium
removal was yet untouched. The high radiation nuclides tend to be removed before sending the spent fuel solution to main process, although the pyridine resin has high radiation resistance. Radioactive cesium is one of the main radiation generators in the spent fuel. It is well known that the heteropolyacid salts have a high cesium selectivity and a high cesium adsorption capacity [7-9]. In the present work, the ammonium molybdophosphate (AMP) and the ammonium tungstophosphate (AWP) were tested to investigate the feasibility of cesium removal from a radioactive solution in hydrochloric acid system.

**Experiment**

Commercially available type AMP and AWP (WAKO, Co. JPN) were used, these were powder type n-hydrates. The adsorption experiments were carried out by batch-wise experiment. Test solutions were prepared by dissolution of alkali chlorides or alkaline earth chlorides in 0.1, 0.5, 1, 2, or 5 mol/L of HCl solution. The concentration of each ion of alkali metal ions or alkaline earth ions was adjusted to 10 mmol/L. 1g of AMP or AWP was inputted into 10cm$^3$ of each test solutions. These solutions with AMP or AWP were shaken 24 hours while keeping the temperature at 25°C. After shaking, the sample solutions were prepared by the removal of AMP or AWP by filtration (pore size: 0.45 µm). Concentrations of alkali metal ions and alkaline earth ions in the sample solutions were measured by ICP-MS (Agilent 7 700). From the data obtained, the distribution coefficients were calculated using the following equation,

$$K_d \ [cm^3/g] = \frac{(C_b - C_a)V}{m}$$

$C_a$, $C_b$, $V$, and $m$ are the concentrations in the sample solution after the adsorption experiment, before the adsorption experiment, volume of solution [cm$^3$], and mass of AMP or AWP [g], respectively. The stabilities of AMP and AWP in HCl solution were also investigated. The concentration of molybdenum and tungsten dissolved in sample solutions were measured by ICP-MS, and the percentage of dissolved AMP or AWP was calculated.

**Figure 1**: Schematic diagram of new reprocessing process with separation system for valuable elements in spent nuclear fuel.
**Results and discussion**

The obtained distribution coefficients of alkali metal ions are shown in Figure 2. It was confirmed that both AMP and AWP have a high cesium selectivity in alkali metal ions for all studied concentrations of HCl. Both distribution coefficients of cesium on AMP and AWP decrease with HCl concentration, however, the distribution coefficients is large enough for the removal of cesium. It was also confirmed that alkaline earth ions are not strongly adsorbed on AMP and AWP. The obtained distribution coefficients of alkaline earth ions are shown Figure 3. To conclude about the selective removal of cesium from spent fuel, it appears that other elements included in spent fuel could be separated.

The stabilities of AMP and AWP in HCl solution are shown in Figure 4. The dissolution of molybdenum and tungsten are confirmed. Results confirmed that AWP is more stable than AMP in HCl solution. However, the percentage of dissolved AMP is low. It was also confirmed that the dissolution of both the AMP and AWP are suppressed in the higher concentration of HCl. This information helped determine operation conditions. The stability of AMP and AWP in HCl solution with alkali metal ions is lower than with alkali earth ions. This is probably due to the chemical transformation by the ion exchange: both the stabilities and the distribution coefficients decrease with HCl concentration, i.e. the stabilities decrease with the increase in the ion exchange ratio.

**Figure 2:** Distribution coefficients of alkali metal ions on (a) AMP and (b) AWP in HCl solution
Figure 3: Distribution coefficients of alkaline earth ions on (a) AMP and (b) AWP in HCl solution

To determine if AMP or AWP should be used, further data should be obtained and discussed using additional information such as a cost for example. If AMP is adopted, the molybdenum recovered from spent fuel has a potential as a source material. It is one of the important points. Powder-type AMP or AWP were used in the present work, but the granular-type AMP or AWP is needed for the column use. The granulation of AMP and AWP is one of the important issues.

Figure 4: Stability of AMP and AWP in HCl solution

Conclusion

The cesium selectivity of AMP and AWP in HCl solution was confirmed. The stabilities of AMP and AWP in HCl solution were also confirmed. For the actual use, more data should be collected, and further research and development is necessary.
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References


Nuclear separations process control and accountability

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Abstract

The Department of Energy (DOE) Office of Nuclear Energy has recently initiated a two-phase, multi-year experimental study at the Pacific Northwest National Laboratory (PNNL) to evaluate the current technological capability to control the preparation of MOX (U/Pu) mixtures using aqueous co-decontamination separations. During Phase I, cold, simulation tests will be performed to establish flowsheet conditions and test advanced control systems. During Phase II, the flowsheet and control system will be tested with irradiated fuel at a kilogram scale. The two-phase study is called the CoDCon project. The control system capability will be measured by the uncertainty or predictability bands while targeting a specific U/Pu ratio. Only chemical control methods will be used, eliminating the use of physical blending up or down to reach the target mixture.

The major tools to be used for process control and error-band measurement are the separations process model and near real-time on-line instrumentation. The process model, as a mathematical description of the process, will be capable of predicting the distribution of key process components throughout the entire system during start-up, steady-state operations and shutdown. The online instrumentation will measure the actual distribution of those components and the current operating conditions. Since both model and measure will have uncertainty bands, a quantitative methodology recently published by Benjamin Cipiti, et al,[1] will be used to provide a numerical description of the combined uncertainties.

Since the same technologies for process control can be used for inventory control, the project will also establish error bands for near real-time material balances and accountability for key elements, quantified by the Cipiti method. Following the 1981 lead of the commercially-funded Barnwell, South Carolina separations project, measurement will be feasible during both Phase I and Phase II to detect simulated diversion scenarios. Further, through sensitivity analyses, information will be collected during cold and hot testing that will allow the identification of those process measurements needing improvements to significantly enhance process control and reduce errors in safeguards and accountability, thus helping identify priority R&D needs.

Although the IAEA has established diversion detection goals for separations plants devoted exclusively to peaceful applications, future requirements may change, depending on unknown and unknowable circumstances. In any case, the CoDCon project will provide information on current detections capabilities for aqueous co-decontamination separations at a kilogram scale. Although those results cannot be extrapolated directly to full-scale plants, the measured uncertainties during Phase II will be a good measure of current capabilities. Because of the close linkage of process control and accountability, the Office of Nuclear Energy intends close CoDCon collaboration with NNSA to maximise the project’s safeguards value. The NRC will also...
be kept informed since the quantitative measure of safeguards values may be useful in licensing future domestic separation plants. When the project results warrant such consideration, they may ultimately represent significant steps towards process and safeguards monitoring in support of world-wide nuclear disarmament.

Introduction

A good material balance is a fundamental requirement of an effective chemical processing operation. If the material balance or accountability is near real-time, it can provide process control information that, in the limit, may lead to feedback control of the important process variables such as flow rate, temperature and acidity. Ultimately, it could result in process automation.

Unaccounted-for material may indicate unexpected retention, leakage or, in the case of nuclear separations of fissile materials, the possibility of diversion. Thus material balances, accountability, and process control of nuclear separations are closely associated with the generic term “safeguards”, that is, the protection against misuse of chemical separations plants designed for the peaceful use of nuclear energy.

The mission of the US Department of Energy Fuel Cycle Research and Development Program sponsored by the Office of Nuclear Energy is to provide options for future commercial or governmental applications through the development of advanced technologies. One facet of the programme is the preparation of process models and measurements that allow near real-time material balances. A process model is a virtual separation system in the form of a mathematical description of a technical process, capable of predicting within an uncertainty range the location and concentration of all elements of interest throughout the system in space and time. Its real-world counterpart is the system of measurements of the concentrations of those elements at a series of sampling locations as well as process conditions such as flow rates, temperature, pressure and acidity.

When the concentration measurements can be made “on-line”, that is, by using continuous sampling methods such as spectroscopy, the combination of modelling and measurement has the potential for providing near real-time material balances of the selected elements. If applied to nuclear systems such as used fuel partitioning in preparation for transmutation, the combined system could have potential safeguards value by providing near real-time actinide accountability data.

The CoDCon Project: in September 2015, the Material Recovery and Waste Form Development campaign in co-operation with the Materials Protection, Accounting and Control Technology campaign of the DOE Office of Nuclear Energy’s Office of Fuel Cycle Technologies initiated a co-decontamination flowsheet study (the “CoDCon Project”) to determine the present technical capability for near real-time process control and material accountability. Using a sensitivity analysis applied to the process model, the error bands associated with the project measurements will be examined to identify those offering the greatest potential for improving the overall material balance and accountability.

The co-decontamination process under experimental study at the Pacific Northwest National Laboratory is a simple PUREX process coupled with the use of hydrazine-stabilised U(IV) as the plutonium reductant. Providing an excess of the stoichiometric requirement to reduce the plutonium to Pu(III) will produce a U/Pu nitrate solution with a potentially wide range of element ratios. The final ratio will be determined by the flow rate of a solvent back extractant which will remove “excess” uranium and produce the desired product mixture.
The target of the CoDCon project nitrate solution will be 70% uranium/30% plutonium, a convenient ratio for the preparation of fast reactor fuel. The object of the project is to determine how closely that ratio can be obtained, both as an aqueous solution and in final mixed-oxide form (MOX). The control of the flow rate of the back extractant used to produce the selected U/Pu ratio will be based on near real-time spectroscopic measurements of the concentration and valence of the uranium and plutonium in the various process streams, both before and after back extraction. The distribution of such instrumentation is shown in the flowsheet diagram in Figure 1. Figure 2 illustrates the application of on-line spectroscopy in the detection of a test solvent extraction diversion, using a rare earth simulant.

**Figure 1:** Simplified flow sheet indicating measurement collection points for the various instruments

**Figure 2:** Online spectroscopic detection of simulated diversion
The CoDCon experimental study will be divided into two phases. Phase I will involve cold-testing in glove boxes using pure uranium and plutonium feed streams followed by later experiments using U/Pu plus simulants. In Phase II, the dissolved content of irradiated LWR fuel rods will be used as feed to separations equipment within hot cells to more nearly simulate conditions in a commercial separations facility, with the feed per batch containing a total quantity of dissolved fuel approximately equal to that in a single PWR fuel rod (between one and two kg). A decision to transition from Phase I to Phase II will necessarily await the new federal administration and will be influenced by Phase I results.

The final research results will be the value of the error bars associated with the product concentration variations as well as the identification of those process variables requiring improved controls to minimise the error bars. Thus the ultimate value of the project will be the prioritisation of follow-on fuel cycle research and development activities for future process control and accountability enhancement.

A legitimate question that can be assigned to most R&D programmes is that of the ultimate goal. What reduction in process control error bars will be “good enough”? Asked to a separation process designer, the answer would probably be similar to that used in environmental protection goals: As low as reasonably attainable (ALARA). The open-endedness of such a goal can support a very long-range research and development programme. However, if applied to nuclear safeguards, a quantitative value can be tentatively identified.

In the International Atomic Energy Agency’s Safeguards Glossary of the international Nuclear Verification Series, the detection goal is of the possible diversion of a significant quantity of fissile material (for plutonium containing less than 80% of Pu-238, a significant quantity is 8 kilograms) of direct use material within one to three months. For a low throughput facility, the “required” IAEA accountability target would be relatively easily attained. For a large commercial separations plant with a design throughput of 800 tonnes of irradiated fuel per year, the amount of unaccountable materials (combined errors of model and measurement or real diversion) would be plus or minus 1-3%, a challenging goal. For the experimental studies of the CoDCon Project, a goal of plus or minus 1% has been rather arbitrarily established. The actual errors will be a measure of current capabilities.

**Significance of results**

The use of a dynamic model of a separation system coupled with advanced instrumentation should provide a measure of current capabilities for process control, near real-time material balances, accountability and attainable safeguards. The latter two measurements can also be quantitatively evaluated by following the precedent of the commercial Barnwell separations plant sited at the edge of the DOE Savannah River Site. After the project’s federal support was cancelled by executive order in 1977, DOE-sponsored tests of simulated diversion using depleted uranium were conducted to measure the then capability of detection. Both the cold tests in Phase I and the Phase II hot tests will be designed to allow similar tests using the advanced on-line instrumentation and dynamic models currently available. The CoDCon Project will provide a good measure of current capabilities and allow future R&D needs to be prioritised.

Because of low uranium prices and a slow-down in nuclear reactor construction, it is unlikely that a commercial separation plant will be built in the U.S. for many decades, so full-scale domestic application of the advanced safeguards afforded by the model/measurement system being studies by the CoDCon project may also be delayed. In any case, both the DOE National Nuclear Security Administration (NNSA) and the U.S. Nuclear Regulatory Commission (NRC) will be kept informed of
the project’s progress: the former, because of parallel interests between the CoDCon project and the NNSA’s Next Generation Safeguards Initiative and the latter, because a quantitative measure of accountability uncertainty using the Cipiti methodology may have value as a safeguards measure as part of future NRC regulatory requirements for commercial separation plants.

Separations plants are under consideration elsewhere in the world, so cooperative studies involving a foreign country considering such a facility and the NE CoDCon project could provide mutual benefits. The foreign partner could benefit from the opportunity to test a co-decontamination flowsheet producing a desired U/Pu nitrate and MOX mixture directly rather than indirectly through separate product mixtures, using advanced online instrumentation and controls. DOE could benefit by the application to a realistic near-term process design, rather than a hypothetical mid-century plant of unknown design and requirements. The IAEA might also benefit from information about the contribution advanced process control instrumentation can make towards near real-time accountability measurements. The IAEA oversight of the Japanese Rokkasho separations plant is expensive and may require around-the-clock IAEA presence when the plant restarts within the next several years. If similar circumstances were to be encountered in the future, the model/measurement approach under study by the CoDCon project may afford an opportunity for remote surveillance at a cost reduction and a potential increase in accuracy compared with the current Rokkasho oversight system.

Because a simple separation process will be tested during the CoDCon project using instrumentation well described in the open literature, it is expected that the project results will be publicly available. However, if there are restrictions on the release of project results, it is the Office of Nuclear Energy’s intent to share the results on a quid-pro-quo basis with those international partners with common interests and, with who appropriate 123 Agreements are in place.

Looking very far into the future, widespread use of the model/measurement approach for quantifying near real-time material balances and accountability in commercial separations plants could provide a future international oversight organisation similar to the IAEA with the tools to help enforce the safeguards needed for nuclear disarmament. The diversion detection requirements of such an international agreement are of course unknown, but the pathway towards such a capability will probably need to be similar to that of the CoDCon project: measure current capabilities quantitatively, identify the needed improvements to reduce the error bars, conduct the needed R&D, re-measure and re-evaluate. Even an open declaration by the U.S. that the country is following such a path with such an end goal may help stimulate progress in other disarmament areas and offer encouragement that such a goal, espoused by all signatories of the Nuclear Non-Proliferation Treaty, is truly attainable.

References

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