NUCLEAR ENERGY AGENCY
COMMITTEE ON THE SAFETY OF NUCLEAR INSTALLATIONS

IN-VESSEL AND EX-VESSEL HYDROGEN SOURCES

Report by NEA Groups of Experts

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The Working Group on the Analysis and Management of Accidents (GAMA) is mainly composed of technical specialists in the areas of coolant system thermal-hydraulics, in-vessel protection, containment protection, and fission product retention. Its general functions include the exchange of information on national and international activities in these areas, the exchange of detailed technical information, and the discussion of progress achieved in respect of specific technical issues. Severe accident management is one of the important tasks of the group.
FOREWORD

The following document is composed of two parts:

- Part I, “GAMA Perspective Statement on In-vessel Hydrogen Sources”, has been prepared by B. Clément (IPSN), K. Trambauer (GRS) and W. Scholtyssek (FZK), on the basis of information collected from GAMA members and the previous Principal Working Group on Coolant System Behaviour (PWG2). It was endorsed by GAMA in April 2001 and approved for publication by CSNI in June 2001.

- Part II, “GAMA Perspective Statement on Ex-vessel Hydrogen Sources” has been prepared by M. Petit (IPSN), H. Alsmeyer (FZK) and V. Gustavsson (SwedPower), on the basis of information collected from members of the previous Principal Working Group on the Confinement of Accidental Radioactive Releases (PWG4). It was published earlier under the reference NEA/CSNI/R(2000)19.
PART I

PERSPECTIVE ON IN-VESSEL HYDROGEN SOURCES

Background

The CSNI had initiated an activity related to hydrogen sources during a severe accident in nuclear power plants. Concerning ex-vessel sources, a “Perspective Statement” was prepared by the PWG4 Task Group on Severe Accident Phenomena in the Containment (SAC) [1]. PWG2 took the action to provide a similar paper on in-vessel sources. A first draft was presented to the newly formed Working Group on the Analysis and Management of Accidents (GAMA) in September 2000. GAMA recommended a re-draft more consistent with the paper on ex-vessel sources. The revised paper was discussed and endorsed during the April 2001 meeting of the group.

Introduction

Various risk studies have shown that hydrogen combustion is one of the major risk contributors to early containment failure in the case of a severe accident in a nuclear power plant. In the past two decades, the different aspects of this issue, namely hydrogen sources, distribution in the containment, combustion behaviour and loads, have been investigated in many research programmes, including single effect tests and integral experiments, model and code development and nuclear plant analysis. Hydrogen mitigating and controlling systems like recombiners, igniters or inertisation measures have been proposed and developed and, in some cases, their practical implementation is underway. To assess the related risk for hydrogen specific accident scenarios, and to design and optimise mitigating systems, quantitative hydrogen analysis has to be carried out. Necessary input for such an analysis is the knowledge of the hydrogen release conditions, especially source rates, total mass and time sequence.

The objective of this perspective paper is to identify potential in-vessel hydrogen sources and to review the state of knowledge on underlying physical and chemical processes. The availability of models and codes for accident analysis is to be addressed, including the areas of remaining uncertainties. Finally, the needs for further experimental investigation and model development should be identified.

Perspective on in-vessel hydrogen sources

Core degradation is usually categorised into two phases: The “early phase” covers the period starting with core uncovering and heating, possibly with melting and relocation of reactor materials with relatively low melting point (cladding, absorber materials), but with core geometry remaining essentially intact. The “late phase” covers the period of melting of ceramic materials, formation of a molten pool in the core area, relocation of the pool to the lower plenum of the pressure vessel, up to failure of the lower head. The degradation process consists of situations with defined geometry – the core, the pool with crusts in the core area or in the lower plenum – and of transient processes like melting and relocation in between. Further transients may occur, for instance during the early phase, if availability of coolant water is regained.
and the possibly overheated core is flooded and quenched as an accident management measure, or, during the late phase, if a fuel coolant interaction should occur. Reactor materials, for instance Zircaloy, steel and B\textsubscript{4}C, are oxidised and hence hydrogen is produced at different stages of the melt progression. The amount produced and the production rate depend heavily on boundary conditions including participating masses, surface areas, availability of water and steam, temperature ranges, or degree of pre-oxidation of materials.

A thorough synthesis on the “Status of Degraded Core Issues” has been given by Bandini [2], which includes an overview on the existing databases and on the availability of models and codes. Some conclusions with relevance to hydrogen specific aspects may be shortly repeated:

− The early phase of core melt progression is reasonably understood.
− The uncertainty of the oxide layer failure of the cladding has the largest impact in the evaluation of timing and amount of hydrogen generation.
− Major uncertainties exist concerning the oxidation behaviour of material mixtures and B\textsubscript{4}C, which may significantly contribute to the hydrogen formation.
− No detailed mechanistic models are available in system codes to describe core quenching. The need for further experiments and model development is recognised.
− The possibility of conducting experiments on debris bed quenching should be considered.
− The melt relocation process (in-core pool to lower head) is an almost completely unresolved issue in view of timing, pouring rate and amount of involved materials.

In the following, a more detailed discussion of hydrogen source aspects is presented. It is partly based on results, summarised in Table 1, from a Specialist Meeting on this topic that was held in spring 1999 with the participation of CEA and FZK experts. Where applicable and available, other recent information is included [3], [4], [5].

**In-vessel hydrogen sources**

**Zircaloy oxidation**

During core heat up, hydrogen is produced by oxidation of the Zircaloy cladding with steam. The underlying phenomena are well understood. Oxidation kinetics is described by largely consistent diffusion models and parabolic correlations, based on experimental investigations. Some uncertainties exist due to the autocatalytic nature of the process. Another source of uncertainty are processes like ballooning and deformation of the cladding during the early core degradation phase. They can lead to reduction or blockage of cooling channels and reduced steam flow. This reduces heat removal from the affected region. On the other hand, oxidation processes and hence hydrogen production could be slowed down since less steam is available. Nevertheless, it is commonly agreed that prediction of the hydrogen source rate, typically about 0.2 kg/s for a 1000 MWe PWR, is sufficiently accurate as long as the core geometry remains intact.

**Steel oxidation**

Steel oxidation may contribute by about 10 to 15 % to the total hydrogen production. Similar to Zircaloy, sufficiently accurate correlations are available.
**B,C oxidation**

B,C is used as absorber material in BWR, VVER and some western type PWR. The oxidation process with steam is understood for the temperature range below ~1400 K, and correlations are available. Above 1400 K, oxidation kinetics is widely unknown. B,C can add significantly to the hydrogen source term because its oxidation in steam is more exothermic and produces more hydrogen (and other gases, for instance CH₄, which is important for the iodine source term) per gram material than Zircaloy does. The effect has been observed in CORA tests [3], but quantitative predictions for the high temperature range are not possible at the moment. Further tests which can contribute to the database on B,C oxidation behaviour are intended to be carried out within an ISTC project and in the QUENCH facility.

Conclusions:

- Experimental programmes on this topic are underway.
- The issue should be reconsidered in due time.

**Hydrogen production during reflood and quenching**

Reflooding and quenching of the uncovered core is the most important accident management measure to terminate a severe accident transient. If the core is overheated, this measure can lead to increased oxidation of the Zircaloy cladding which in turn can trigger a temperature escalation. Relatively short flooding and quenching times can thereby lead to high hydrogen source rates which must be taken into account in risk analysis and in the design of hydrogen mitigation systems.

Until recently, the experimental database on quenching phenomena was rather scarce. The available Zircaloy/steam oxidation correlations were not suitable to determine the increased hydrogen production in the few available tests (CORA, LOFT LP-FP-2).

At FZK the QUENCH programme was started, using electrically heated test bundles to investigate relevant phenomena like cracking and fragmentation of the oxygen-embrittled cladding due to thermal shocks, which leads to the generation of new metallic surfaces and to enhanced oxidation and hydrogen generation. Another process to be studied is the possible uptake of hydrogen in Zircaloy and additional release during oxidation which influences release kinetics. Further tests are proposed, e.g., within the ISTC-1648 project, which includes aspects of irradiated fuel, or in PHEBUS-2k in-pile tests with nuclear heated bundles.

An analysis of hydrogen production during reflood in a large PWR (EPR), using SCDAP/RELAP5 and MAAP4, has been presented in [6]. The main conclusions are:

- Strong hydrogen production can be expected only in the case of power recovery at the time where the core is strongly heated but still predominantly intact. However, this scenario is rather unlikely because of the narrow time window (about 10 – 20 min).
- Hydrogen production rates during reflood were predicted rather similarly with the two codes. They are in reasonable agreement with rates scaled from QUENCH experiments.
- Very different total hydrogen masses were calculated with the two codes. This was attributed mainly to the different treatment of relocated cladding and core material.

Conclusions:
− It has been recognised that reflooding phenomena are important in view of hydrogen production.

− The available experimental database is limited and there is a lack of mechanistic models; this introduces uncertainties in hydrogen analysis.

− Presently performed and proposed experimental activities are suited to significantly improve the situation.

− The issue should be re-assessed in due time.

**Hydrogen production during core melt-down**

The late-phase core degradation is characterised by high temperature regimes leading to various processes that influence hydrogen production. These include:

− Melt formation and relocation. The effective surface of reactive materials is changed, and melt is relocated to areas with lower temperatures and freezes, with the potential of blockage or rubble bed formation and reduced steam flow in the regions above. Since unoxidised materials are relocated first, it is to be expected that hydrogen production is reduced.

− Physico-chemical interactions of reactor materials. These affect physical properties, for instance in reducing the melting temperature of material mixtures, which may accelerate the core degradation process. The oxidation behaviour of material mixtures is not well known and this contributes to the uncertainties in hydrogen production.

A major uncertainty in determining the hydrogen production during a severe accident is the timing of the cladding failure and loss of core geometry. Code comparisons [6] have resulted in significant differences for the hydrogen mass produced, which can be mainly attributed to the different models used for cladding failure and related U-O-Zr melt relocation and oxidation.

Studies of different accident scenarios for a PWR [7] predicted a degree of Zircaloy oxidation of about 30% in a fast sequence (LBLOCA, about two hours to RPV failure), and of about 50% if core geometry failure occurs late (SBLOCA, SBO, about five hours to RPV failure). However, estimates performed for a typical PWR show that the degree of cladding oxidation is in the range of 25-70 % in fast sequences, and may increases to 90% if core geometry failure occurs late. The main parameters, which can influence the results, were identified in sensitivity studies. These are critical minimum thickness of unoxidised Zircaloy, ZrO₂ dislocation criteria and the liquefaction temperature of U-Zr-O mixtures. Further, the choice of the equivalent diameter of particulate debris has an important influence on the prediction of the source rate and the total hydrogen mass.
Conclusions:

- There is a need for experimental activities, which could increase the database for better characterisation of cladding and fuel behaviour and respective failure criteria, and of debris beds and their oxidation behaviour to reduce related uncertainties.

- This should be accompanied by development of suitable models and their implementation in severe accident codes. The PHEBUS programme is well suited to contribute to this issue.

**Hydrogen production during fuel-coolant interaction**

In the late core degradation phase, hot melt from the in-core area is relocated to the lower plenum, which may be filled with water. Injection of the melt into water, for instance in form of a jet, and fragmentation of the melt would lead to an increase of the reaction surface and strong oxidation of not yet oxidised metals. Experiments with Zr/ZrO₂ and Zr/stainless steel, with oxidation degrees of up to 40%, e.g. ZREX [8], have indicated that typically 5 to 25% of the metals are oxidised if no steam explosion occurs, and between 70 to 100% in the case of a triggered steam explosion. Depending on the amount of participating masses and the degree of pre-oxidation of the melt, significant hydrogen masses could be produced during a short period.

FARO tests have demonstrated that injection of pure oxidic melts, even without steam explosion, may produce hydrogen in significant quantities [9], with an estimated 2 kg hydrogen per Mg UO₂. Changes of the stoichiometry of Urania have been considered as an explanation but were not found to be consistent with FARO results [1]. The conclusion in [1] that associated phenomena are not understood and require further investigations is supported.

**Influence of irradiated and MOX fuel**

Very few experimental data are available concerning the impact of burn-up and MOX characteristics on core degradation and on the oxidation behaviour of mixed materials. Adequate models are generally not available in severe accident codes. The direct influence on the oxidation behaviour of metallic materials is probably not very important. However, burn-up and MOX use are assumed to accelerate the liquefaction of the fuel and therefore they have an influence on the timing of the accident sequence. As mentioned above, earlier loss of core geometry generally leads to less hydrogen generation, which means that, in view of the hydrogen issue, no negative effects are to be expected.

**Conclusions**

During cladding oxidation of intact fuel rods by steam, the hydrogen source term can be predicted within the measurement errors of the experiments. Steel oxidation is modelled, whereas B₄C oxidation is widely unknown for temperatures above 1400 K. Major uncertainties still exist for oxidation during reflood processes. This is under study in ongoing experimental programmes. A very limited data base is available for the late phase of severe accident scenarios, and where the rod-like geometry has degraded to a debris bed or a molten pool. In these configurations, main uncertainties in prediction of the hydrogen source term are related to oxidation behaviour of Zr-rich mixtures including both the kinetics of oxidation and the steam-debris interaction surface. Better knowledge in this area would enable more reliable hydrogen specific containment analysis and support design and assessment of control and mitigation systems. For in-vessel molten core coolant interaction, only few data sets exist which provide limited information on the hydrogen source term.
REFERENCES


<table>
<thead>
<tr>
<th>In-Vessel Hydrogen Generation Phases</th>
<th>Cladding Oxidation</th>
<th>Steel Oxidation</th>
<th>Boron Carbide (B4C) Oxidation</th>
<th>Reflood</th>
<th>Molten Pool Formation</th>
<th>Interaction in the Lower Head (MFCI)</th>
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<tbody>
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<td>Are the relevant H2 generation processes understood?</td>
<td>yes</td>
<td>open question</td>
<td>yes</td>
<td>partly yes</td>
<td>partly yes</td>
<td>large uncertainties (steam explosion: unknown)</td>
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<td>Which models are available?</td>
<td>diffusion models; parabolic correlations</td>
<td>Correlation</td>
<td>correlation</td>
<td>&lt; 1100 °C; correlation</td>
<td>extended DBA models, SVECHA1</td>
<td>a few specific models on debris bed oxidation; parametric model for core slump; user specified fragmentation models</td>
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<td>Are modifications necessary?</td>
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<td>Yes</td>
<td>no</td>
<td>yes</td>
<td>yes</td>
<td>yes</td>
</tr>
<tr>
<td>Which models are implemented into computer codes?</td>
<td>parabolic correlations</td>
<td>Correlation</td>
<td>correlation</td>
<td>extended DBA models</td>
<td>shattering models</td>
<td>scaled down (by Zr in melt content) models</td>
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<td>How accurate are the models?</td>
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<td>sufficient</td>
<td>insufficient</td>
<td>under improvement</td>
<td>insufficient</td>
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<td>Are experimental data available?</td>
<td>yes</td>
<td>yes, within DBA2</td>
<td>&lt; 1100 °C, only powder, no pellets</td>
<td>yes, under DBA</td>
<td>yes (CORA, LOFT)</td>
<td>very few</td>
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<td>Which experimental programs are currently performed?</td>
<td>DAMRAGUE, QUENCH, PHEBUS-FP, AEKI</td>
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<td>DBA</td>
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<td>all, except of ASTEC V0</td>
<td>no code, except partially MELCOR</td>
<td>ASTEC within DBA, MAAP extended DBA models; ICARE within DBA; SCDAP: ext. DBA models</td>
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<td>0.5 kg/s</td>
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<td>10-15% of total H2 prod. of TMI-2</td>
<td>unknown</td>
<td>5 - 10 kg/s</td>
<td>&lt; 0.2 kg/s (SCDAP from debris oxidation)</td>
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</table>

1 not for reactor applications till now
2 Design Basis Accident (Temperatures < 1500 K)
3 Codes under consideration: ASTEC V0, ICARE/CATHARE V1, MAAP-4; MELCOR 1.8.4, SCDAP/RELAP5
4 For Zr-ZrO2 melts
PART II

PERSPECTIVE ON EX-VEssel HYDROGEN SOURCES

Ex-vessel hydrogen sources

The purpose of this perspective document is to identify the potential ex-vessel hydrogen sources and to address the question whether, considered the uncertainties associated to these sources, further investigations are required. The statement is established with reference to the needs for safety evaluation of nuclear reactors under severe accident conditions. It is recognised that the views could be different if one looks at these issues from another standpoint.

Since the TMI-2 accident in 1979, there had been a large interest in the nuclear reactor safety community for studying the behaviour of hydrogen in case of a severe accident. As a result, different ‘state of the art’ reports were produced. Examples of these documents are NUREG/CR-1561 and EUR 14307. In particular, they identified potential hydrogen sources during accidents, including ex-vessel sources.

Various ex-vessel hydrogen sources, covering a variety of physical and chemical processes, were identified. Although their precise quantification and relative importance is to be established on a case by case basis with respect to the specific reactor design of interest, general trends can be formulated. The sources to be considered are the followings:

- radiolysis of water;
- corrosion reactions,
- reaction of urania with steam and water;
- core-concrete interaction;
- debris-atmosphere interaction.

These sources are discussed hereafter.

Radiolysis of water

Radiolysis of water occurs both during normal operation and under accident conditions. It may take place in the core and in the sump. It involves the decomposition of water molecules by radiation producing various radicals. The net result is the production of hydrogen and oxygen molecules in essentially a stoichiometric ratio. The phenomena are reasonably understood for pure water at room temperature. More uncertainties exist for elevated temperatures and the presence of solutes or impurities.
However, evaluations showed that under accident conditions the rate of hydrogen production is low. Typical figures are in the order of some hundreds of kilograms after three months. This low production rate can easily be accommodated with existing mitigation means such as hydrogen recombiners. Provisions are usually already taken with respect to water radiolysis for post design basis accidents (LOCA). PWG4 therefore considers that no specific work is required on this issue.

**Corrosion reactions**

In a reactor containment, the only significant sources of hydrogen from corrosion are reactions of zinc and aluminum. Zinc is present in some types of paint and in galvanised steel. These reactions are of importance for high and low pH values. Evaluations show that the amount of hydrogen that could be produced by corrosion reactions is in the order of 100 kg in some hours, far less than from other sources. As for radiolysis, this production can easily be accommodated by existing mitigation means. PWG4 considers that no specific work is required on this issue.

**Reaction of urania with steam and water**

Urania may come in contact with steam, for example in the case of the release of corium from RPV to a flooded cavity. Uranium dioxide reacts with steam to produce UO$_{2+x}$ and hydrogen. The extent of the reaction is dependent on partial pressures of steam and hydrogen. It was reported in earlier works that the fraction of uranium dioxide that reacts is limited in presence of a large excess of steam over hydrogen and that it becomes very small when hydrogen is present.

However, recent experimental results, produced during the course of the FARO program, showed that hydrogen can be produced in significant amounts during the quenching of corium by water, even though the melt is already oxidised. The interpretation of measurements gave figures of about 0.2 kg of hydrogen produced for 100 kg of melt.

It seems unlikely that the mechanism involved be an oxidation from stoichiometric to hyperstoichiometric urania (see above). Other processes such as oxidation from hypostoichiometric to stoichiometric urania or from hypostoichiometric to hyperstoichiometric urania can be postulated. Up to now, none of them was identified as being consistent with FARO experimental results.

A simple extrapolation from the FARO test to the large-scale reactor situation would give a H$_2$ source of 200 kg. Because of limitations of the mixing process of melt and water, it is recognised that the large melt masses would certainly not undergo the same degree of chemical reaction as in a small experiment. Therefore, realistic H$_2$ rates are expected to be significantly smaller.

In conclusion, it must be stated that the associated phenomena are not understood to date and require further investigations. PWG4 considers that this issue is not specific to ex-vessel situation and that it is also of great interest for FCI studies, which have been conducted in the frame of PWG2. PWG4 therefore recommends that the issue be investigated by the new Working Group on the Analysis and Management of Accidents.

**Core-concrete interaction**

Dry core-concrete interaction has been investigated in a number of experimental and theoretical programs. The process of hydrogen and carbon monoxide production during MCCI is well understood and characterised. It is essentially due to the oxidation of metals by gases (H$_2$O and CO) which are released during the interaction. These metals are oxidised in the following order: Zr, Si, Cr and Fe. Zr, Cr and Fe
come from the molten core, molten parts of the RPV and the concrete re-bar. Si (and some SiO) is produced by initial reduction of SiO$_2$ present in the concrete by Zr. The main effect of SiO$_2$ is thus to delay the hydrogen production. A significant part of hydrogen is produced during the early phase of the core-concrete interaction while Zr is being oxidised. The uncertainty here is essentially due to the initial amount of Zr to be taken into account, which differs for different accident sequences. However, this quantity is clearly dependent on the level of oxidation during the in-vessel phase of the accident. Overall, it can be assumed that the remaining mass of Zr, which is the major source of early hydrogen production, will be oxidised during the early phase of the MCCI. The corresponding amount of H$_2$ released into the containment by complete oxidation of all Zr is in the order of 1000 kg for a typical PWR. However, for those plants equipped with mitigation means such as passive autocatalytic recombiners, hydrogen production during MCCI would occur at a time at which the hydrogen concentration in the containment would have already been depleted to a certain extent, thus limiting the associated threat to the containment. After depletion of Zr and its follow-on products, long term H$_2$ release during MCCI is governed by Fe oxidation with typical release rates of 2 mol/s (4 g/s) which would continue over several days. This release is, however, accompanied by a larger rate of steam flow, which reduces the flammability. Based on these elements, PWG4 considers that the issue of hydrogen production by zirconium oxidation has to be dealt with as a whole (in-vessel and ex-vessel phases), and that the main area of uncertainty, which requires further analysis, is the oxidation during the in-vessel accident progression.

It might be recalled, however, that uncertainties still exist in long-term heat transfers during MCCI, which may result in uncertainties in basemat penetration time estimates. It is also pointed out that, depending on the particular plant design, long-term overpressurisation may remain a safety issue.

**Debris-atmosphere interaction**

In case of RPV failure at high pressure, the corium melt can be entrained out of the cavity in the form of droplets. It has been shown experimentally that these droplets, because of their very large area, can interact efficiently with the atmosphere to form hydrogen. The primary process is the oxidation of the metals. As for core-concrete interaction, the primary source of uncertainty in the analyses will be the initial amount of metals in the corium, which is in turn dominated by the extent of Zr oxidation during the in-vessel phase of the accident. PWG4 then considers that no specific work is required on this issue.

**Summary**

To recapitulate, the PWG4 perspective on Ex-vessel Hydrogen Sources can be summarised in the following statements:

1. The issue of hydrogen sources must be considered as a whole and cannot be separated into in-vessel and ex-vessel issues. For significant sources that may not be accommodated by mitigation means associated to DBA, the uncertainty is largely dominated by the unknown extent of Zr oxidation during the in-vessel phase.

2. PWG4 notes that hydrogen production during corium quenching by water is still not adequately understood and requires some additional work to be performed. This issue is of interest not only for MCCI in an initially flooded cavity but also for Fuel Coolant Interaction studies. PWG4 recommends that the issue be investigated by the new Working Group on the Analysis and Management of Accidents.
REFERENCES

