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**NUCLEAR ENERGY AGENCY
COMMITTEE ON THE SAFETY OF NUCLEAR INSTALLATIONS**

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PWG4 Perspective on Ex-vessel Hydrogen Sources

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ORGANISATION FOR ECONOMIC CO-OPERATION AND DEVELOPMENT

Pursuant to Article 1 of the Convention signed in Paris on 14th December 1960, and which came into force on 30th September 1961, the Organisation for Economic Co-operation and Development (OECD) shall promote policies designed:

- to achieve the highest sustainable economic growth and employment and a rising standard of living in Member countries, while maintaining financial stability, and thus to contribute to the development of the world economy;
- to contribute to sound economic expansion in Member as well as non-member countries in the process of economic development; and
- to contribute to the expansion of world trade on a multilateral, non-discriminatory basis in accordance with international obligations.

The original Member countries of the OECD are Austria, Belgium, Canada, Denmark, France, Germany, Greece, Iceland, Ireland, Italy, Luxembourg, the Netherlands, Norway, Portugal, Spain, Sweden, Switzerland, Turkey, the United Kingdom and the United States. The following countries became Members subsequently through accession at the dates indicated hereafter: Japan (28th April 1964), Finland (28th January 1969), Australia (7th June 1971), New Zealand (29th May 1973), Mexico (18th May 1994), the Czech Republic (21st December 1995), Hungary (7th May 1996), Poland (22nd November 1996) and the Republic of Korea (12th December 1996). The Commission of the European Communities takes part in the work of the OECD (Article 13 of the OECD Convention).

NUCLEAR ENERGY AGENCY

The OECD Nuclear Energy Agency (NEA) was established on 1st February 1958 under the name of the OEEC European Nuclear Energy Agency. It received its present designation on 20th April 1972, when Japan became its first non-European full Member. NEA membership today consists of 27 OECD Member countries: Australia, Austria, Belgium, Canada, Czech Republic, Denmark, Finland, France, Germany, Greece, Hungary, Iceland, Ireland, Italy, Japan, Luxembourg, Mexico, the Netherlands, Norway, Portugal, Republic of Korea, Spain, Sweden, Switzerland, Turkey, the United Kingdom and the United States. The Commission of the European Communities also takes part in the work of the Agency.

The mission of the NEA is:

- to assist its Member countries in maintaining and further developing, through international co-operation, the scientific, technological and legal bases required for a safe, environmentally friendly and economical use of nuclear energy for peaceful purposes, as well as
- to provide authoritative assessments and to forge common understandings on key issues, as input to government decisions on nuclear energy policy and to broader OECD policy analyses in areas such as energy and sustainable development.

Specific areas of competence of the NEA include safety and regulation of nuclear activities, radioactive waste management, radiological protection, nuclear science, economic and technical analyses of the nuclear fuel cycle, nuclear law and liability, and public information. The NEA Data Bank provides nuclear data and computer program services for participating countries.

In these and related tasks, the NEA works in close collaboration with the International Atomic Energy Agency in Vienna, with which it has a Co-operation Agreement, as well as with other international organisations in the nuclear field.

CSNI

The NEA Committee on the Safety of Nuclear Installations (CSNI) is an international committee made up of senior scientists and engineers, with broad responsibilities for safety technology and research programmes, and representatives from regulatory authorities. It was set up in 1973 to develop and co-ordinate the activities of the NEA concerning the technical aspects of the design, construction and operation of nuclear installations insofar as they affect the safety of such installations. The Committee's purpose is to foster international co-operation in nuclear safety amongst the OECD Member countries. CSNI's main tasks are to exchange technical information and to promote collaboration between research, development, engineering and regulation organisations; to review the state of knowledge on selected topics of nuclear safety technology and safety assessments, including operating experience; to initiate and conduct programmes to overcome discrepancies, develop improvements and reach consensus on technical issues; to promote co-ordination of work, including the establishment of joint undertakings.

PWG4*

CSNI's Principal Working Group on the Confinement of Accidental Radioactive Releases (PWG4) has been given two tasks: containment protection, and fission product retention. Its role is to exchange information on national and international activities in the areas of severe accident phenomena in the containment, fission product phenomena in the primary circuit and the containment, and containment aspects of severe accident management. PWG4 discusses technical issues/reports and their implications, and the results of International Standard Problem (ISP) exercises and specialist meetings, and submits conclusions to the CSNI. It prepares Technical Opinion Papers on major issues. It reviews the main orientations, future trends, emerging issues, co-ordination and interface with other groups in the field of confinement of accidental radioactive releases, identifies necessary activities, and proposes a programme of work to the CSNI.

SAC*

The Task Group on Severe Accident Phenomena in the Containment (SAC) is a specialised extension of PWG4. Its main tasks are to exchange information, discuss results and programmes, write state-of-the-art reports, organise specialist workshops, and perform ISP exercises in the field of severe accident phenomenology.

*. Since the preparation of this report; the Working Groups of the CSNI have been restructured. The functions of the PWG4 and the SAC have now been taken over by a new Working Group on the Analysis and Management of Accidents.

FOREWORD

The attached “PWG4 Perspective Statement on Ex-vessel Hydrogen Sources” has been prepared by Mr. M. Petit (IPSN Task Leader), Dr. H. Alsmeyer (FZK) and Dr. V. Gustavsson (SwedPower). It was endorsed by PWG4 during its meeting of 28-29 February 2000.

This note is devoted to hydrogen sources outside the reactor pressure vessel. A complementary document on in-vessel hydrogen sources has been prepared by CSNI’s PWG2 (Principal Working Group on Coolant System Behaviour). The draft of the PWG2 will note will be submitted to the Working Group on the Analysis and Management of Accidents in September 2000.

PWG4 PERSPECTIVE ON EX-VESSEL HYDROGEN SOURCES

Perspective on 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 uranium 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_2O and CO_2) 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 SiO₂ present in the concrete by Zr. The main effect of SiO₂ 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 H₂ 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 H₂ 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

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