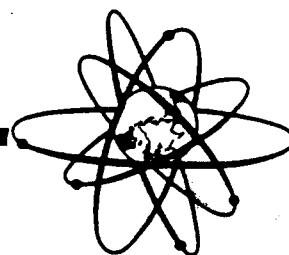OECD

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SOURCE TERM MITIGATION ASPECTS OF ACCIDENT MANAGEMENT

Report by an OECD/NEA Group of Experts
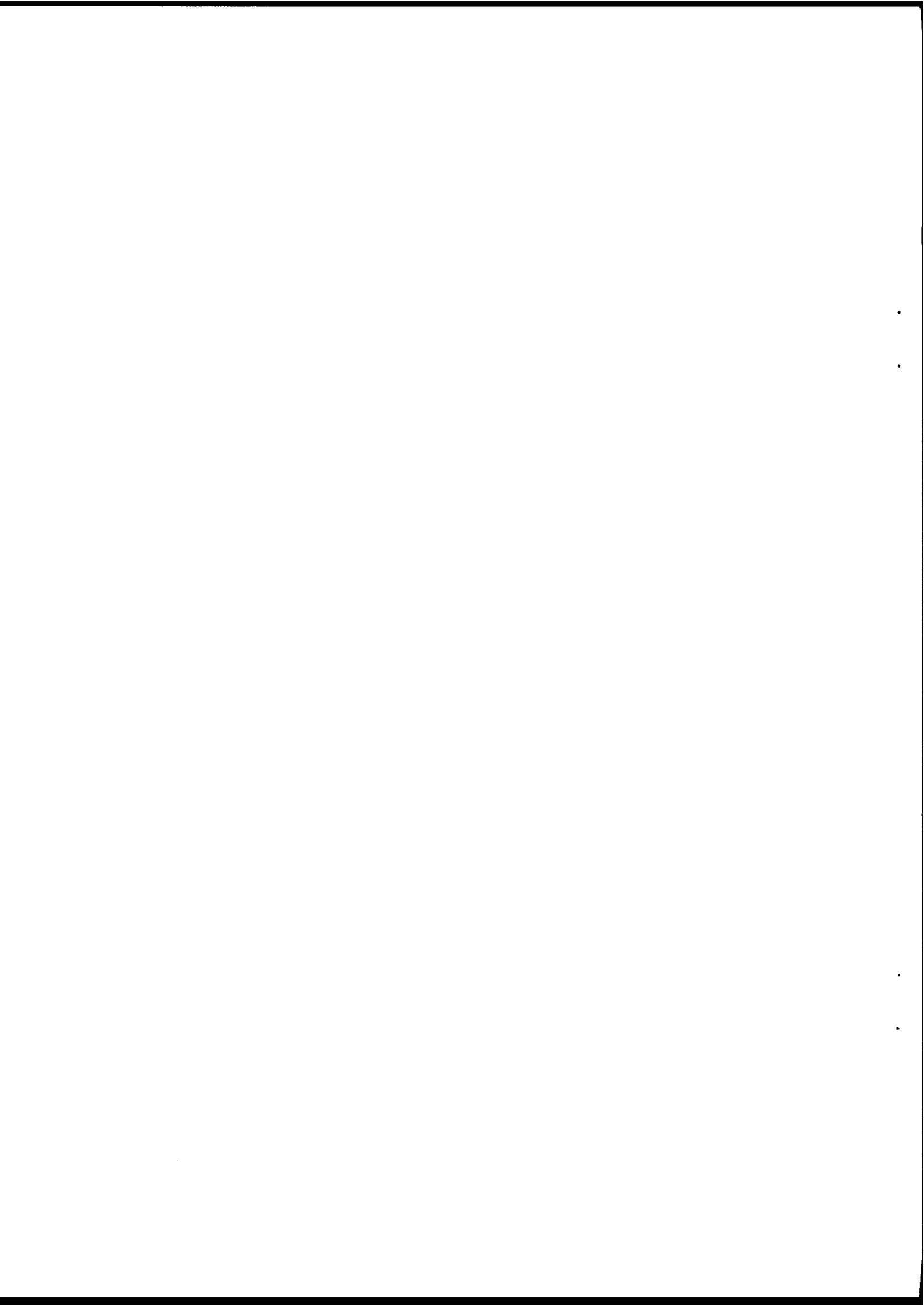
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SOURCE TERM MITIGATION ASPECTS

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December 1989

Committee on the Safety of Nuclear Installations
Nuclear Energy Agency
Organisation for Economic Co-operation and Development

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O E C D

In 1948, the United States offered Marshall Plan aid to Europe, provided the war-torn European countries worked together for their own recovery. This they did in the Organisation for European Economic Co-operation (OEEC).

In 1960, Europe's fortunes had been restored; her standard of living was higher than ever before. On both sides of the Atlantic the interdependence of the industralised countries of the Western World was now widely recognised. Canada and the United States joined the European countries of the OEEC to create a new organisation, the Organisation for Economic Co-operation and Development. The Convention establishing the OECD was signed in Paris on 14th December 1960.

Pursuant to article 1 of the Convention, which came into force on 30th September 1961, the Organisation for Economic Co-operation and development 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 this 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 Signatories of the Convention were Austria, Belgium, Canada, Denmark, France, the Federal Republic of Germany, Greece, Iceland, Ireland, Italy, Luxembourg, the Netherlands, Norway, Portugal, Spain, Sweden, Switzerland, Turkey, the Unided Kingdom and the United States. The following countries acceded subsequently to the Convention (the dates are those on which the instruments of accession were deposited): Japan (28th April 1964), Finland (28th January 1969), Australia (7th June 1971) and New Zealand (29th May 1973).

The Socialist Federal Republic of Yugoslavia takes part in certain work of the OECD (agreement of 28th October 1961).

N E A

The OECD Nuclear Energy Agency (NEA) was established on 20th April 1972, replacing OECD's European Nuclear Energy Agency (ENEA, established on 20th December 1957) on the adhesion of Japan as a full member.

NEA now groups all the European Member countries of OECD and Australia, Canada, Japan and the United States. The Commission of the European Communities takes part in the work of the Agency.

The primary objectives of NEA are to promote co-operation between its Member governments on the safety and regulatory aspects of nuclear development, and on assessing the future role of nuclear energy as a contributor to economic progress.

This is achieved by:

-- encouraging harmonisation of governments' regulatory policies and practices in the nuclear field, with particular reference to the safety of nuclear installations, protection of man against ionising radiation and preservation of the environment, radioactive waste management, and nuclear third party liability and insurance;

-- keeping under review the technical and economic characteristics of nuclear power growth and of the nuclear fuel cycle, and assessing demand and supply for the different phases of the nuclear power to overall energy demand;

-- developing exchanges of scientific and technical information on nuclear energy, particulary through participation in common services;

-- setting up international research and development programmes and undertakings jointly organised and operated by OECD countries.

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

C S N I

The NEA Committee on the Safety of Nuclear Installations (CSNI) is an international committee made up of scientists and engineers. It was set up in 1973 to develop and coordinate the activities of the Nuclear Energy Agency 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 constitutes a forum for the exchange of technical information and for collaboration between organisations which can contribute, from their respective backgrounds in research, development, engineering or regulation, to these activities and to the definition of its programme of work. It also reviews the state of knowledge on selected topics of nuclear safety technology and safety assessment, including operating experience. It initiates and conducts programmes identified by these reviews and assessments in order to overcome discrepancies, develop improvements and reach international consensus on technical issues of common interest. It promotes the coordination of work in different Member countries including the establishment of co-operative research projects and international standard problems, and assists in the feedback of the results to participating organisations. Full use is also made of traditional methods of co-operation, such as information exchanges, establishment of working groups, and organisation of conferences and specialist meeting.

The greater part of CSNI's current programme of work is concerned with safety technology of water reactors. The principal areas covered are operating experience and the human factor, reactor coolant system behaviour, various aspects of reactor component integrity, the phenomenology of radioactive releases in reactor accidents and their confinement, containment performance, risk assessment, and severe accidents. The Committee also studies the safety of the fuel cycle, conducts periodic surveys of reactor safety research programmes and operates an international mechanism for exchanging reports on nuclear power plant incidents.

In implementing its programme CSNI establishes co-operative mechanisms with NEA's Committee on Nuclear Regulatory Activities (CNRA), responsible for the activities of the Agency concerning the regulation, licensing and inspection of nuclear installations with regard to safety. It also co-operates with NEA's Committee on Radiation Protection and Public Health and NEA's Radioactive Waste Management Committee on matters of common interest.

FOREWORD

In 1987, CSNI's Principal Working Group No. 4 on Confinement of Accidental Radioactive Releases asked one of its Task Groups to prepare a note on Source Term Mitigation Aspects of Accident Management. The Soviet response to the Chernobyl accident had indeed demonstrated that even when a severe accident is well under-way, actions can be taken to reduce the radioactivity release. The Soviet actions at Chernobyl of depositing dry shielding and filtering materials on the core debris were initially effective, but appear to have had an adverse delayed effect in insulating the core and promoting temperature rise. One might well ask whether alternative actions, for example involving wet rather than dry deposition of materials, might have been even more effective.

Some of the issues to be addressed by the Task Group were the following:

(i) The accident scenarios of concern pre-suppose core melt and vessel failure. These stages of the accident are usually short in timescale, and involve substantial releases of radionuclides to the containment. The first question to be addressed was, given sufficient warning of the imminent development of vessel failure, are there any irregular uses of engineered safety systems which may still be functioning which might effectively reduce airborne activity concentrations in the containment. In broader terms, are there any short-term mitigative actions which can be taken?

(ii) The state of containment failure or by-pass is a key factor in determining the magnitude of the source term, and also in controlling what actions may be feasible to mitigate longer term release. In the case of by-pass into an auxiliary or other building, it may be possible in some instances to cause deliberate flooding to promote activity hold up but the associated clean up problems need to be assessed. For a failed containment, the size of any breach, and whether it will permit the introduction of materials into the containment shell, is an important question. If the containment is failed by multiple fissuring, the options to be considered may be either "plastering over the cracks", or deliberately making a larger hole in order to get materials into the building. The advisability of the latter course of action is strongly dependent on the confidence that the mitigative effect of the materials will be substantial.

(iii) Specific source term aspects to be considered were:

> (a) Long term iodine release. The analysis in NUREG-1150 suggests that this can be a dominant effect in certain sequences. Retention of iodine will be promoted by wet conditions, high pH, and the presence of silver or other getters. The most effective course of action and the available time need to be considered.

> (b) Resuspension of other materials. Dry out of sump pools and heat up of residues may lead to the resuspension of volatile constituents in the longer term. Is the most effective course

of action to try to keep the containment flooded, or might the introduction of dry materials be more effective, and assist clean up? For example, it is established that the presence of boric acid will greatly reduce the potential for resuspension of caesium. This question may not only be relevant to the containment, but to resuspension from the primary circuit if the timescales permit mitigative action.

(c) Core/concrete interaction may be a source of long term release. Again, could this best be controlled by keeping the containment flooded? Or would addition of dry filtering materials, or of large quantities of fusible material which would become incorporated in the melt and reduce the temperature, be a better response? Would addition of oxidants to the melt be beneficial?

These specific aspects were considered in three chapters of the report, the lead authors of which are mentioned on the following page. The full report was discussed and endorsed by PWG4's Task Group on Fission Product Phenomena in the Containment (FPC), PWG4 itself and CSNI. A list of the members of the Task Group is given in an Appendix to the report.

CONTENTS

Summary

Although the occurrence of a core melt accident accompanied by containment
failure in an LWR is extremely improbable, some prior consideration of the
measures which could be taken to mitigate the continuing release of
radioactivity is needed as part of overall accident management. This review
discusses possible mitigation measures for minimising the release of iodine
from water pools, the revaporisation of fission products from debris beds,
and the release of radionuclides in molten core-concrete interaction, from a
failed reactor containment. It is clear that the most effective mitigation
measure for all of these processes is to maintain water flooding of the
containment, with suitable chemistry control to suppress the formation of
volatile iodine chemical species. Some alternative dry mitigation methods
are considered, and problems associated with these are discussed.

Introduction

The severe accident at the Chernobyl Unit 4 Nuclear Power Plant in the USSR, which occurred in April, 1986, has stimulated a number of developments in reactor safety.

Careful analyses of the Chernobyl accident have shown that it has hardly any relevance for the safety of water-cooled reactors of western design (1, 2). However   , a determination to maximise the benefit of any lessons which could be learned from Chernobyl has led, amongst other developments, to an increased emphasis on severe accident management as part of safety strategy. Most of this activity concentrates on those measures which can be taken to arrest an accident, and prevent core melt. In the unlikely event that an accident does progress to the most severe state of a molten core and a failed reactor containment, one of the clear lessons of the Chernobyl accident is that emergency measures can still be taken to mitigate the release of activity to the environment. These measures were ultimately successful at Chernobyl, but a very large radiological release occurred in the meantime. A further lesson to be learned is that, with some pre-planning, an even more successful mitigation strategy might have been implemented.

This review is concerned with source term mitigation in the later stages of the severest types of accidents which have been hypothesised for LWRs of western design. It presupposes that a core melt and reactor vessel failure have occurred. If the reactor containment building also fails early, within the first half-day or so of accident initiation, current analyses suggest that in certain accident sequences, there could be an immediate release of a significant fraction of the core inventory of radioactivity. Even in these cases, the majority of the activity would remain in the containment, and every step needs to be considered to keep it there. If the failure of containment is delayed by a day or more, present analyses suggest that the immediate release upon failure will amount to only a small fraction of the core inventory. In this situation, the long-term release processes considered here, and their mitigation, assume even greater relative importance.

Three important phenomena which could contribute to the delayed source term from a failed containment are discussed in the chapters of this review. Chapter 1 considers measures to mitigate the release of volatile iodine from water pools, where most of this fission product is predicted to collect. The second chapter reviews the situation after these pools boil dry, when residual beds of debris could heat up by decay heat to temperatures where volatile fission products might vaporise. Chapter 3 discussed what might be done to prevent, or mitigate, molten core-concrete interaction releases from the reactor cavity.

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CHAPTER 1. IODINE SOURCE TERM MITIGATION

1.1 Introduction

The potential for long-term releases of fission products following the initial phases of an accident stimulates the need to consider potential actions that could be taken to mitigate releases. In this chapter, potential actions to minimise the long-term release of radioiodines are discussed. The importance of iodine derives from its radiotoxicity, abundance and ready transport in air. The ability to form volatile iodines long after an accident is "terminated" makes mitigation strategies particularly relevant for radioiodine control.

This paper briefly discusses short-term and long-term inorganic iodine releases, and then focuses on the mitigation of organic iodine release, since, in the long term, organic iodides are expected to form the dominant fraction of iodine releases.

Releases and mitigation strategies are discussed here only with respect to the behaviour of iodine in containment. A comprehensive source term analysis must consider the fact that radioiodine is the daughter of fission product tellurium. One particular iodine isotope (I-132, half-life 2.3 h) will be created as a result of decay of its parent Te-132 (half life 78 h) in significant quantities during an accident. This iodine isotope could, under special circumstances, constitute a large fraction of the radioiodine source term to the environment. The long-term behaviour of tellurium should be considered in developing a complete strategy for mitigation of radioiodine release.

1.2 Short-Term Iodine Release

The iodine releases during the early phase of a severe accident will be dominated by the physical processes occurring then. The most important of these will be the rate of mass transfer from the containment to the environment and the formation, transport and deposition of aerosol-borne radioiodine. The mass transfer rate will be influenced by the temperature and pressure history within containment. The bulk of the iodine is expected to be associated with aerosol particles (dry) or water droplets (wet). Aerosol releases will be strongly influenced by the total mass of aerosol material and the timing of containment failure. Computer code predictions of the behaviour of aerosols during an accident show that the bulk of the aerosol material generated will be deposited within containment during the initial period of the accident [1].

1.3 Long-Term Iodine Release

In the long-term, the release of radioiodine will be strongly influenced by chemical processes. There may be continued generation of aerosol material because of core-concrete interactions, or the gas sparging, boiling or rapid depressurisation of sump pools. The rates of aerosol generation from these sources will decrease with time as the decay heat decreases or additional cooling is provided. At some point, releases of iodine will be dominated by the formation of volatile iodides in solution or on surfaces. For practical purposes, the only relevant iodine species will

be I_2 or organic iodine. The only other possible iodine species, HI, has long been shown to have a negligible vapour pressure in equilibrium with a solution phase [2].

An exception to the above statement is the creation of iodine airborne in water droplets as a result of emergency cooling recirculation that might be started at a late stage of an accident following a severe core damage event. During such a recovery phase, water may be recirculated through the core and released to containment through a break in the reactor coolant system. If the water is released under a moderate pressure, the resulting jet may break up and generate water droplets containing iodine. In general, such jets form relatively large droplets upon break up [3]. Large droplets will rapidly settle onto surfaces under the influence of gravity and will not be available for release from containment. The jet break up will, however, lead to the generation of a small number of aerosol-sized particles, some of which may remain suspended long enough to be transported through a leak in containment.

The fraction of airborne iodine available for release via jet fragmentation of recirculating water will normally be very small because of the concentration of iodine in the water and the small fraction of the recirculation water that forms an aerosol. However, the recirculation water will pass through the primary coolant circuit, where it may dissolve substantial quantities of previously deposited iodine, or leach iodine from failed fuel elements. This may lead to a temporary high concentration of iodine in the recirculation water and may enhance the importance of recirculation in releasing airborne iodine.

It is not clear what steps could be taken to mitigate iodine releases enhanced by this recirculating cooling. Substantial water flows are required to cool a damaged core for more than one month after an accident. It might be desirable to decrease the flow rate to minimise aerosol spray formation at a break point. In the likely absence of detailed information regarding spray behaviour, such considerations will clearly be secondary to the need to provide rapid cooling to the core.

1.4 Mitigation of Organic Iodide Release

While it is possible to create I_2 in aqueous solution and release it to the containment atmosphere, the conditions that favour its formation are unlikely to be achieved. As Lemire et al. [4] have shown, the formation of I_2 requires an oxidising solution with a low pH. In contact with air, the aqueous phase within containment will be buffered by CO_2. Pressurised water reactors normally operate at a near-neutral pH buffered by the presence of boric acid, and there is a tenfold excess of cesium (to form CsOH) over iodine in the core, which, after an accident will tend to make the sump water mildly basic. Atmospheric CO_2 will tend to lower the pH, but the sump water will remain basic (pH > 7). Boiling water reactors normally operate with pure water (pH 7 and no buffer capacity). For these reactors, the sump water pH will be more affected by atmospheric chemistry, although fission products such as CsOH will tend to make the sump water after an accident neutral or mildly basic (pH > 7) as well. Under these conditions, the equilibrium I_2 concentration in solution will be low. There will still be some I_2 released to the air due to the low partition coefficient of I_2, but

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release of large quantities of I_2 to the environment will be unlikely unless there are very high mass transfer rates of air through the containment building and turbulence in the sump water.

The dominant form of released iodine will be organic iodine. Post-accident analyses of the radioactive iodine released from the containment building during the TMI-2 reactor accident showed that the bulk of this iodine was in the form of an organic iodide [5]. Laboratory experiments have demonstrated that substantial quantities of organic iodides can be generated in solution [6]. The generation of organic iodides from this dissolved iodine is also favoured by oxidising conditions and a low pH, but the requirements are not as rigorous as those for the formation of I_2 [7]. In addition, experiments have shown that the generation of organic iodides is enhanced by a radiation field [8-11] (discussed below).

The primary tasks in mitigating the long-term release of radioiodines after an accident, therefore, involve the prevention of organic iodide formation and its release from containment. There are at least three basic strategies that can be pursued to attain this objective:

1.　control the solution chemistry to reduce the rate of organic iodide formation,

2.　control the solution chemistry to reduce the stability of organic iodides, and

3.　provide engineered systems to reduce the release of organic iodides.

The first two of the above strategies assume that the bulk of the iodine that may be released is located in the pools of sump water in the basement of the reactor buildings. This neglects the potential importance of iodine deposited on vertical or horizontal surfaces above the sump level during the early phase of the accident. On many of these surfaces, iodine may be physically or chemically absorbed and may not have been washed into the sump.

The most attractive strategy is the first option. If the formation of organic iodides can be prevented, then less iodine will be available for release. Research is under way in several laboratories around the world to develop a full understanding of the mechanisms for formation of volatile organic iodides in solution. An established route for organic iodide formation involves the reaction of I_2/HOI as an intermediate. Both I_2 and HOI are oxidation products of the I- anion initially introduced into the sump solution as CsI. Prevention of the oxidation of I- will reduce the rate of organic iodide formation.

There are again several potential means of reducing the rate of I- oxidation:

a　lowering the pH of the sump water,

b　addition of chemical reducing agents,

c   elimination of oxidising agents from solution, and

d   eliminating chemical agents from solution that catalyse the oxidation of I^- by dissolved O_2.

By far the easiest and most effective option is addition of a chemical to raise the pH of the water in containment. In addition to oxidising conditions, conversion of I^- to I_2/HOI requires acidic conditions [7]. In basic solutions, I_2 is not stable and HOI dissociates to form IO^-. thus maintaining a basic sump water chemistry will very effectively prevent the formation and release of organic iodides. In some reactors, the primary coolant water is already at a high pH, for corrosion control, and additives to provide a high pH after an accident are common and cheap (eg., NaOH, used at TMI-2 immediately following the accident [12]). The sump pH will be affected by the fission products (eg, CsOH), coolant system additives (eg, boric acid, LiOH) and the buffering capacity of the CO_2 from the containment atmosphere. Nevertheless, it should be a relatively simple procedure to ensure that the sump water pH is maintained at a high value (pH > 9) long after an accident. The great disadvantage of this strategy is that it requires the use of very caustic, corrosive solutions, the misuse of which (eg, accidental spray system operation) could lead to high plant clean-up costs.

It is clearly very difficult to prevent the presence of any oxidising agent in the sump water following an accident, particularly since dissolved oxygen alone will tend to make the solution oxidising. Likewise it would be very difficult to guarantee that there are no catalysts in solution, such as transition metal ions, that would tend to accelerate I^- oxidation.

The introduction of reducing chemical agents is the next most feasible for the above options. Such agents are routinely used for corrosion control in water systems. Hydrazine is a popular choice for a reducing agent. Experiments and calculations have shown that it can be very effective in reducing the rate of organic iodide formation in solution [13]. Hydrazine has many attractive features for oxidation control, including the fact that it reacts rapidly with I_2. It has a major disadvantage, however it tends to decompose rapidly and thus needs to be continuously added to a system to maintain reducing conditions. Therefore, it is most attractive for short-term use, immediately after an accident. Long-term hydrazine control will be particularly awkward for water pools having very open access to the atmosphere, where there is rapid mass transport of oxygen to the water.

The second strategy focuses on the potential for the destruction of organic iodides in solution before they can be released to the containment atmosphere. In solution, organic iodides can react with other chemical reagents, particularly nucleophilic radical anions. Hydrolysis by OH^- is particularly effective [14]. This is naturally enhanced by high OH^- ion concentrations (high pH). Hence, maintaining a high pH in the sump water has the double-edged effect of limiting oxidation of I^- and destroying organic iodides that are formed.

The third strategy for controlling long-term release is the use of some engineered system for retention of organic iodides, within the containment building, at lease long enough to allow the radioactive iodine to decay.

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The basic engineered system for any reactor is the intact containment building. Mitigation of the release of all fission products is enhanced by the degree of leak tightness the containment maintains following an accident. However, all containment buildings are subject to a small amount of leakage that may be increased by the accident.

A standard technique for mitigating fission product release is the addition of a filtration system to the containment (the filtered vent concept). Deliberate venting through a filter assembly is a strategy that permits the relief of very high pressure during the early phase of an accident to maintain containment building integrity. The organic iodides could be retained in such filter systems by deep-bed charcoal filters, but the systems currently under consideration or already installed do not include such filters. The CANDU multi-unit stations, with a vacuum building, do possess such filters for long-term pressure relief and iodine retention. Charcoal filters are also available as part of the normal air filtration systems on the containments of other reactors [15].

Charcoal filters are very effective in removing all forms of volatile iodides from the air, but they do not perform equally well under all conditions; high humidity and high temperatures degrade their performance [16]. In addition, a controlled filtered release of the containment atmosphere may not be available following all reactor accidents.

There exist other potential engineered systems, which have not been as well explored. One possibility is the introduction of an organic fluid sink into the containment. Organic iodides are much more soluble in organic solvents than in water. In equilibrium within containment, there would be an effectively reduced vapour pressure (hence airborne concentration) of organic iodides and a gradual mass transfer of iodine from the water in the sump to the organic fluid. Paquette et al. [17] have shown that an oil film is effective in preventing the release of organic iodides to the air because of the preferential solubility of the organic iodides in the organic film. This technique has the advantage of being a potential sink for organic iodides originating not only in the sump water, but also from the other surfaces in the containment where the iodine was originally deposited.

The location of the organic fluid, its method of introduction and potential problems with cleanup have never been addressed, and the practicality of this option is open to debate. The introduction of large quantities of an organic fluid could lead to an unacceptable combustion hazard unless properly engineered.

For those reactors containing silver/cadmium control rods, the silver released into the sump will provide an excellent chemical sink for iodine [18]. The formation of AgI will compete with the formation of organic iodides in the sump and effectively suppress the release of organic iodides. There are other additives that could be added to containment to reduce the rate of organic iodide production or its volatility but this option has not been well explored.

1.5 Radiolysis Reactions

In the brief review of iodine release mitigation, there has been limited discussion regarding the formation of volatile iodides following an accident. While normal redox chemistry will undoubtedly be important, it should be remembered that there will be intense radiation fields within containment following a severe accident. Radiolysis of aqueous solutions containing iodine is potentially the dominant mechanism for generating organic iodides in solution. There are at least two significant processes that can be enhanced by radiolysis, oxidation of I^- to I_2/HOI [19] and the production of reactive organic radicals (eg, $CH_4 \rightarrow CH_3 + H$). Beahm has demonstrated that radiolysis of methane-saturated iodine solutions can lead to substantial release of organic iodides [8].

In previous reviews of iodine chemistry, it was thought that radiolysis could also have a beneficial effect in further oxidising I_2 to IO_3^-. Under most pH and redox conditions of interest, IO_3^- is the most stable form of iodine in solution. It is also very unreactive with respect to formation of organic iodides under normal conditions. Recent experimental evidence, however, shows that radiolysis of iodate solutions saturated with methane can also lead to the release of organic iodides [20]. The mechanism of this unexpected process is not yet well established. It does appear clear that radiolysis is undesirable under all conditions in terms of its potential to enhance the long-term release of organic iodides.

If radiolytically active impurities (eg, ions subject to redox reactions) are present in concentrations greater than that of iodine, the primary radiolysis radicals will be scavenged and the radiolytic impact on iodine will be attenuated. Also, radiolysis can destroy organic iodides. Since destruction is a primary process (ie, $R-I + OH, H, e, O_2^-$), the yield will be higher than the yield of organic iodide formation, which is a secondary process. hence in the long term, the steady state should favour organic iodide destruction.

The implications of the effects of radiolysis in terms of mitigation of long-term iodine release are not clear. It would be desirable to minimise the radiolysis of the iodine-containing solutions in the containment. This would require separating the radiation source and the solution. Clearly this is not a trivial task during post-accident recovery. It is unlikely that this could be possible for all accidents and the cost of engineered safety systems to achieve this objective would be very uneconomical in terms of the cost/benefit relationship.

The point of the above discussion is to emphasise of the importance of radiation in influencing the long-term behaviour of fission product iodine following an accident. The goal of current work in the intermediate-scale Radioiodine Test Facility is to determine more quantitatively the relationship between a radiation field and the generation and release of organic iodides [21]. One aspect of this work is to determine more precisely the influence of all the complex chemical system parameters, including radiation, on the release of organic iodides. The results may confirm the efficacy of maintaining a sufficiently high solution pH in mitigating long-term iodine release and may lead to the development of other strategies to mitigate release.

1.6 Summary

The long-term release of radioiodine following a reactor accident will be dominated by the generation and release to the atmosphere of volatile organic iodides. There exist a number of mitigation strategies capable of reducing the rate of organic iodide formation and release. Some of these are very simple in concept and execution (eg, addition of a base to raise the sump water pH), but all have some disadvantages.

Experimental programs are now in progress to fully identify the important factors controlling organic iodide generation and release. Through these programs, we expect to obtain an improved understanding of the most important chemical and physical processes. This should lead to an optimisation of mitigation strategies.

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CHAPTER 2:   CONTROL OF REVAPORISATION FROM DEBRIS BEDS
IN A FAILED CONTAINMENT

2.1 Introduction

     We discuss here the possible measures which might be taken to
mitigate the releases of radionuclides from debris beds remaining in the
reactor containment building after this has failed. Initially, the
containment will be wet, but decay heat will eventually result in
boil-off of all of the water, and the residual dry debris will then heat
up, possibly to temperatures at which significant revaporisation of the
more volatile species occurs. The emphasis here is on PWR accidents,
since it is assumed that in BWR accidents where the suppression pool
becomes the main repository of activity released from the core, means
will be found of preventing pool dry-out. Core-concrete interaction in
the reactor cavity is considered in another section of this report, and
we consider here only the other areas of the containment floor,
including the sump pools. Much of the material is based upon a paper on
long-term post-accident chemistry in source term analysis presented at
the ACS Severe Accident Chemistry Symposium, Toronto, 7-10 June 1988
(1). Although the situation following accidents which fail the
reactor containment is given prime attention, some discussion of
containment by-pass accidents is included.

2.2 Conditions in a Failed Containment

     Even for a specific core melt accident sequence resulting in
reactor vessel melt-through, there are large uncertainties concerning
the likely distribution of fission products and core materials in the
containment before and after failure. Rather than attempting a detailed
assessment of individual sequences, an approach of scoping a range of
possible scenarios is adopted. In all cases, a major failure of the
containment building is assumed. The chief variables to consider are:

a.   Containment failure time. Both early failure, within an hour or
two of accident initiation, and delayed overpressure failure taken to
occur at an arbitrary but reasonable time of $10(5)s$ (27.8 hr), are
considered as scoping situations. In the case of early failure, the
puff release source term upon containment failure is likely to be
substantial, and the fractions of inventory of some fission products,
particularly iodine and caesium, remaining in the containment may be
significantly less than unity. Nonetheless, measures to minimise the
continuing release of activity will certainly be required, as in the
accident at Chernobyl Unit 4 in 1986 (2). The decay heat in the
residual material at these early times remains high. For delayed
containment failure situations, most of the activity in the form of
aerosols will have settled out of the containment atmosphere by the time
of failure, and the puff release source term will be low, typically < 1%

for radionuclides other than the noble gases. In this case, minimisation of the revaporisation contribution is relatively more important, since it could dominate the overall release. Decay heat, which acts as the driving force both for dry-out and for debris heating, will be somewhat reduced at these later times. An additional source of heat important in driving up core temperatures during the early, in-vessel stages of severe accidents is the reaction of steam with zircaloy fuel cladding. Subsequent oxidation of zircaloy in the containment may also contribute. However, these chemical heat sources represent only a small perturbation on the overall decay heat and water balance, and are omitted in the following calculations.

b.   Initial water inventory. Clearly, the more water present in the containment during the early stages of the accident, the longer it will take for this to boil off after containment failure, and the better the chances of being able to take countermeasures. The minimum amount of water which could be present for a PWR is the reactor coolant system (RCS) plus accumulator (ACC) inventory. For a large, dry-containment 1100 MWe PWR, this amounts to about 350 m3. The largest amount likely to be present is this plus the full inventory of safety injection (SI) water, taken here to total 2055 m3. The dry-out times calculated below are based on these limiting water inventories.

c.   Radionuclide composition. There are large uncertainties regarding the fractions of core inventory of individual radionuclides released from the core at various stages of an accident, and their distributions in the containment at the time of failure. Reference 1 defines three scoping mixtures, intended to span the range from a minimum in-vessel core melt aerosol to a high pressure melt ejection scenario involving dispersal of 25% of the core. The scoping cases are reproduced here in Table 1, and employed in the assessments which follow.

d.   Debris composition and dispersal. In addition to the radionuclides and core materials, the debris beds will generally contain inactive materials. For a PWR, boric acid from the coolant and safety injection water will be a major contributor. Additionally, there may be inactive concrete constituents derived from core-concrete aerosols. These materials will add to the thermal capacity of the debris, affecting the timescales for heat-up, and thus for countermeasures. Moreover, the distribution of the debris over the containment floor is very uncertain. In general, it may be supposed that it will be far from uniform. There is a possibility that the soluble fission products iodine and caesium will get washed from walls and other surfaces by condensing steam, and collect entirely in pools on the containment floor. As these pools dry out following containment failure, the iodine and caesium may tend to become concentrated in the containment

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concentrated in the containment sumps. Thus, in one extreme considered in Reference 1, concentration of active and inactive materials in a sump of nominal area 68 m2 is considered. At the other extreme, uniform distribution of the debris over the full geometric area of the containment, 1650 m2, is taken. The matrix of debris bed compositions and geometries analysed in Reference 1, which forms the basis for the assessments below, is reproduced in Table 2.

2.3 Timescales for debris dry-out

While conditions remain wet, the most important resuspension processss is likely to be the revolatilisation of iodine from water pools. This is discussed in another section of this report, where mitigating actions are reviewed. It is important to establish the timescales likely to be available both for these actions, and for any further actions which might be needed to control releases from the dried-out debris. In reality, there may be non-uniform distribution of water in the containment, which will be very design-specific. In some containment designs, the reactor cavity will flood, and boil-off from the cavity may represent the main fate of the water. Other designs are intended to preserve a dry cavity in all foreseeable circumstances. The latter situation is addressed here for scoping purposes. It may be assumed that in the case of a flooded reactor cavity, heat transfer rates from the whole core debris to the water will be higher than are considered here, and dry-out times will be correspondingly shorter.

Table 3 shows the calculated dry-out times from initiation of an 1100 MWe PWR core melt accident with early containment failure. Results for the three scoping mixtures of radionuclides, representing different decay heats, are shown for the minimum and maximum intial water inventories. Complete transfer of the decay heat from the debris to the total water is assumed. The scoping mixtures include no allowance for loss of iodine and caesium in the puff release associated with early containment failure. As a sensitivity study, the final row in Table 3 shows the dry-out times calculated if only 50% of the decay heat from the full iodine and caesium inventories is available i.e. assuming 50% puff release fractions for these fission products. The extension of the dry-out time is considerable, but the inclusion of only these two radionuclides is rather unrealistic.

Table 4, reproduced from Reference 1, shows the dry-out times calculated in a set of scoping situations in the event of delayed containment failure, from the time of failure at 10(5)s. If these times are referred back to accident intiation, they turn out in all cases to be fairly similar to the times for the coresponding cases in Table 3. This is expected, since if the water is assumed to be the main sink for the decay heat, the steam generated prior to containment failure goes into pressurising the containment, and is abruptly released on failure. The second column of Table 4 highlights the fact thar a substantial amount of the total water inventory is present as steam in the containment atmosphere at the time of failure, taken to occur at 160 C

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and 8 bar pressure. A further fraction flashes to steam in the depressurisation upon failure.

The main conclusions to be drawn from these scoping results are that if only the minimum amount of water is initially present, there may be quite limited time available to take measures to prevent dry-out. In one of the scoping situations examined for a delayed failure scenario, which is not unduly pessimistic, a period of only 8 hours is calculated before dry-out. However, if the amount of water initially present is much larger, of the order of the full safety injection volume, then dry-out times even in the worst case are suffiently long that some means of maintaining wet conditions can surely be organised, always assuming that this is the preferred source term management option.

2.4 Debris bed heating

Accident management measures to mitigate releases from dry debris beds will only be required if there is actually a threat that such beds could heat up to temperatures where revaporisation of volatile species is likely. This problem has been addressed in Reference 1, where application of the code HOTBED to model the temperature histories of beds of the compositions and configurations summarised in Table 2 is described. The results of those scoping studies are reproduced in Figure 1. From this, it is apparent that rapid heat-up of the debris following dry-out is predicted in all cases, but that the asymptotic temperatures reached are sensitive particularly to the spread of the debris. In the situation where debris becomes concentrated into a relatively small area such as a sump, temperatures in excess of 1000K are predicted. In these circumstances, as discussed in Reference 1, substantial revaporisation of iodine is possible, and revaporisation of tellurium and ruthenium may become of concern. However, if the debris is widely and uniformly dispersed over the whole containment floor, much more modest temperatures are expected at which revaporisation may be a minor effect.

The chief conclusion to be drawn from this study is that, particularly since a non-uniform distribution of material is likely to result in hot-spots, there is a strong possibility of revaporisation of certain volatile fission products from decay-heated debris beds, and that mitigating actions need to be considered.

2.5 Mitigating actions

Approaches to mitigating the revaporisation source term which readily suggest themselves are prevention of dry-out by continued supply of water, or deposition of a dry bed of filtering material on the debris. A combination of these two, in sequence or simultaneously, might also be considered. We now discuss these approaches in turn.

2.5.1 Water flooding.

This appears to be the simplest and most attractive option. Whatever the damage state of the containment, it is likely that some means of access for getting water into the containment can be identified.

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However, several factors need to be considered in deciding whether in particular circumstances this is a viable approach:

a) Energetic interactions. If the debris beds are already completely dry and have reached elevated temperatures, there is a danger that reflooding could result in energetic interaction with water, resulting in significant physical resuspension of radioactive material. The likely presence of hot-spots means that this could be a serious problem. Considerable care in deciding on the initial delivery rate of water is needed.

b) Is an adequate water supply available? Table 5 gives the water supply rates required to balance the boil-off rate due to the full core decay heat of an 1100 MWe PWR at various times after shut-down. From this, it is seen that the required rates of supply are well within the capabilities of mobile units such a fire tenders which might be quickly brought in. More problematic, though, might be the integral quantities of water required to maintain wet conditions. For the plant in question, the integral decay energy between $10(5)$ and $10(6)$ s is sufficient to boil off just over 4000 tonnes of water. Roughly 5 times this amount would be needed by $10(7)$ s (116 days). A substantial reservoir of suitable water, such as a lake or river, would be needed to supply such quantities if other means of cooling could not be instituted.

c) Would resuspension of fission products from the water be a problem? The question of iodine revolatilisation from water pools has been addressed elsewhere in this report, and it is concluded that some means of water chemistry control would be required to minimise this problem. Very recently, it has been shown that organic tellurides might be formed radiolytically in pools under severe accident conditions (3), and this is clearly a topic in need of further investigation. The physical entrainment of activity in water droplets appears from recent results (4) to be a very minor process.

d) Would there be problems with water leakage from the containment and long term clean-up of the water? The TMI-2 experience offers some guidance here. Clearly, water-borne activity is potentially mobile, and it might be difficult to prevent migration of water from the containment to other parts of the reactor complex. Such migration is a design-specific matter to which attention should be given. In particular, it is important to be sure that routes will not exist outside the reactor complex which could lead to the contamination of surrounding soil and groundwater. Thus, control of the total quantity of water is essential. The TMI-2 clean-up has shown that decontamination of large volumes of water is possible, albeit with considerable difficulty and expense.

2.5.2 Deposition of filtering materials.

The most obvious problem to this approach (although notably absent in the Chernobyl accident!) is the obstruction which the residual shell of the containment might pose to giving access to the direct deposition of filter materials on the debris. Even if sufficiently large apertures

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did exist in the structure to give access, for example for air dropping, the uncertainties about the distribution of active materials in the containment discussed above would mean that a total blanketing strategy would have to be adopted. Roughly 2000 tonnes of sand would be required to provided a filter bed of depth 1 m over the entire floor area of the PWR containment used for illustration purposes here. Judging by the response at Chernobyl, it would be possible to deliver this or even larger amounts of sand or other more specific filter materials by air drop in a reasonably short time. However, deliberate enlargement of the apertures in the containment would probably be needed to ensure good access and a wide distribution over the floor area.

In addition to the problems of delivery, the other drawback to a filter-bed approach made apparent by the Chernobyl experience is the thermal insulating effect which can drive up the core debris temperature in the long term, and may lead to resuspension of previously-trapped material. A scoping study of this effect has been performed using the HOTBED code (5). This is based on the model debris bed denoted as case 4 in Table 2, that is, the full core inventory of I, Cs and Te, half of the Ba and Sr, and a quarter of the residual core inventory, together with maximum amounts of boric acid and core-concrete aerosols, spread uniformly over the full containment floor area. The change in heat up has been investigated when an overlying uniform bed of sand 0.1m deep covers the debris. The thermal properties of the sand bed required as input to the HOTBED code are generally well-defined, with the exception of the thermal conductivity. There is considerable uncertainty over the thermal conductivity of porous sand beds since the properties of air and quartz are widely different in this respect (0.1 and 2.87 Wm-1K-1 respectively). The thermal conductivity of a sand bed is dominated by the air component, and for present purposes a value of 0.29 Wm-1K-1 has been used, one tenth of the value for fuzed quartz.

The predicted temperature-time histories for the debris on its own and with the overlying sand are shown in Figure 2 (curves 1 and 2a respectively). The presence of the sand has a dramatic effect in pushing up the initial rate of temperature rise, and a continuos rise is predicted rather than an asymptotic value being approached. In fact, the quantitative predictions of HOTBED are not valid for such temperatures. The effects of concrete ablation, and the mel of the sand itself, will become important once temperatures reach about 1800 K. Also, the calculation assumes no convective he losses to the air through the sand bed, and so represents an limit on the real situation. Nonetheless, the qualitative conclusion that the sand bed will have a major adverse effect insulating the debris and driving temperatures up, thus promo revaporisation of the volatile fission products, is clear-cut Curve 2b of Figure 2 shows the results of a sensitivity study which the thermal conductivity of the sand bed was given an u limiting value, namely that appropriate to fused quartz. The asymptotic temperature is reduced considerably, but is still region where significant revaporisation of volatile fission products is likely.

Given this overheating problem, it is clear that a simple san bed filter can only be used as a temporary holding measure in source term
- mitigation. The action of the sand bed is almost entirely physical,

providing a large area of cool surface for condensation of volatile
fission product vapours, and for the trapping of aerosol particles by a
variety of mechanisms. So the problems of thermal insulation could be
reduced by using carefully graded gravel and sand bed, such that
convective he losses to the air were promoted whilst filtration
efficiency little impaired. However, the possibility of being able to
construct such a bed in an ad hoc manner in post-accident circumstances
is remote. A more promising approach would be addition of chemically
specific trapping materials in the upp cooler regions of a sand or
gravel filter bed, which would re effective in resisting the temperature
challenge. This conce seems to have been the reason for the
incorporation of clay minerals in the filter bed constructed by the
Soviets at Cher (2). Some candidate materials are now considered in
relation trapping of the key volatile fission products I, Cs, Te and R
Silver. This is a well-known and effective getter for gaseous forms of
iodine. AgI is much less volatile than CsI. It is possible that there
will already be large amounts of control silver in the core debris
beds, in which case addition of sil to the filter bed may be pointless.
Also, elemental silver m be too expensive. However, silver impregnation
of other trap agents (silica, alumina, zeolites) can greatly increase
their effectiveness for retention of iodine, and probably also of
tellurium.

Zeolites. These can be highly effective molecular sieves, but they need
to be selected to trap molecules in a specific size range. Sources of
abundant (and preferably cheap) zeolites which would have high retention
efficiencies for the volatile fission products would need to be
identified in advance. The temperature resilience of zeolites is good
for cationic species such as Cs, but less satisfactory for iodine.
Silver impregnation might remedy the latter problem.

Clay minerals. A variety of clay minerals act as effective cation
exchange media, and could be employed in trapping Cs, Ba, Sr, Ru and
possibly the lanthanides and actinides. In general, so long as the
initial trapping process occurred at fairly low temperatures whilst the
minerals maintained their structures, retention would be maintained
during subsequent heating to elevated temperatures.

Silica Gel. Much the same comments apply in this case as for the clay
minerals. Silica gel will only be effective if the initial adsorption
takes place at temperatures not much above 100 C.

Charcoal. Although activated charcoal is widely applied as a filtering
medium for iodine and other volatile species (eg RuO4) at ambient
temperatures, its poor temperature resilience and flammability mean that
it can be ruled out as a trapping medium in this context.

2.5.3 Wet filter bed.

Some of the benefits of the presence of water may be achieved with
smaller quantities than are required to keep the containment flooded if
a combined approach of deposition of dry filter material and addition of
water is followed. For example. In some situations dry filter
materials may be more promptly available than water, which can only be

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introduced later. The presence of water can have benefits such as increasing the efficiency of the filter bed for fine aerosol particles, and trapping some volatile forms of iodine. Since the water is not in direct contact with the debris, water losses will be lower, and quite small quantities could have a large effect in mitigating releases, at least in the short term. A further advantage of this approach would be the elimination of the possibility of energetic interactions which might occur on direct reflooding of hot debris beds. A preliminary covering with quite a shallow layer of sand or gravel prior to reflood could be of benefit here.

2.6 Containment By-pass

Containment by-pass accidents can potentially give rise to large source terms without any structural failure of the containment building. Classically, much attention has focussed in the interfacing LOCA or V-sequence of WASH-1400, in which a direct path opens from the reactor coolant system to the auxiliary building via a rupture in the low pressure injection system pipework. More recently, by-pass accidents involving multiple steam generator tube failure have received increasing attention. In the latter, if the accident progresses to the stage of vessel melt-through, most of the residual core activity will be retained within the containment, and the obvious management strategy is to reflood the debris and maintain cooling. Simultaneously, some means of filtering activity which might escape along the by-pass route is needed. The possibilities will depend strongly on the reactor design, and some ad hoc arrangement for pool scrubbing or dry sand bed scrubbing may be feasible.

In interfacing LOCAs, large quantities of water will be discharged into the auxiliary building during the early stages of the accident. In general, the EC/SI water will be present in addition to the RCS water, giving typically in excess of 2000 m3 total volume. Again, the fate of this water and its influence on the source terms is highly dependent on the detailed design of the interfacing system in question and the auxiliary building. In some designs, the break point in the low pressure pipework will be submerged, and the fission products released from the core during core melting, and subsequently by core-concrete interaction, will be largely trapped in the pools in the auxiliary building (6). In this situation, the early release of radionuclides may be quite low (7,8), and it is essential that dry-out and revaporisation from debris beds is prevented. The results presented in Section 3 above on dry-out times then become important. If the EC/SI and RCS water is present, there will be plenty of time to arrange for an emergency supply of water, but if only the RCS water is present, rapid action will be needed. In all circumstances, one or more of the measures to control iodine volatility discussed in the previous chapter should be implemented .

2.7 References, Chapter 2

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Table 1

Scoping Cases - pool dry-out/revolatilisation

Heat Sources

a   I   Cs

b   I   Cs   Te   0.5Ba   0.5Sr

c   I   Cs   Te   0.5Ba   0.5Sr   0.25 core

Water

RCS & Accumulators - 350 m3

RCS & Accumulators & Emergency Coolant or Spray Injection

-2055 m^3

Table 2

Debris bed studies

	Case				
	1	2	3	4	5
Radionuclides	a	b	b	c	c
B_2O_3 1590 kg	*	*	*		*
12240 kg				*	
MCCI aerosol, 4900 kg			*	*	
Area Sump 68m^2	*	*			
Floor 1650 m^2			*	*	*

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Table 3

Containment Pool Dry-out Times for Early Failure

Radionuclide composition	Initial water inventory, m3	
	350	2055
a	53hr	>116d
b	40hr	64d
c	15.6hr	9d
0.5 a	9d	>116d

Table 4

Pool Dry-Out

Water Inventory, m3			Dry-out time		
Initial	Before failure	After failure	Radionuclide case		
			a	b	c
350	71	64	25hr	19hr	8.4hr
2055	1760	1600	95d	43d	8.7d

Table 5. Water supply rates to balance decay heat

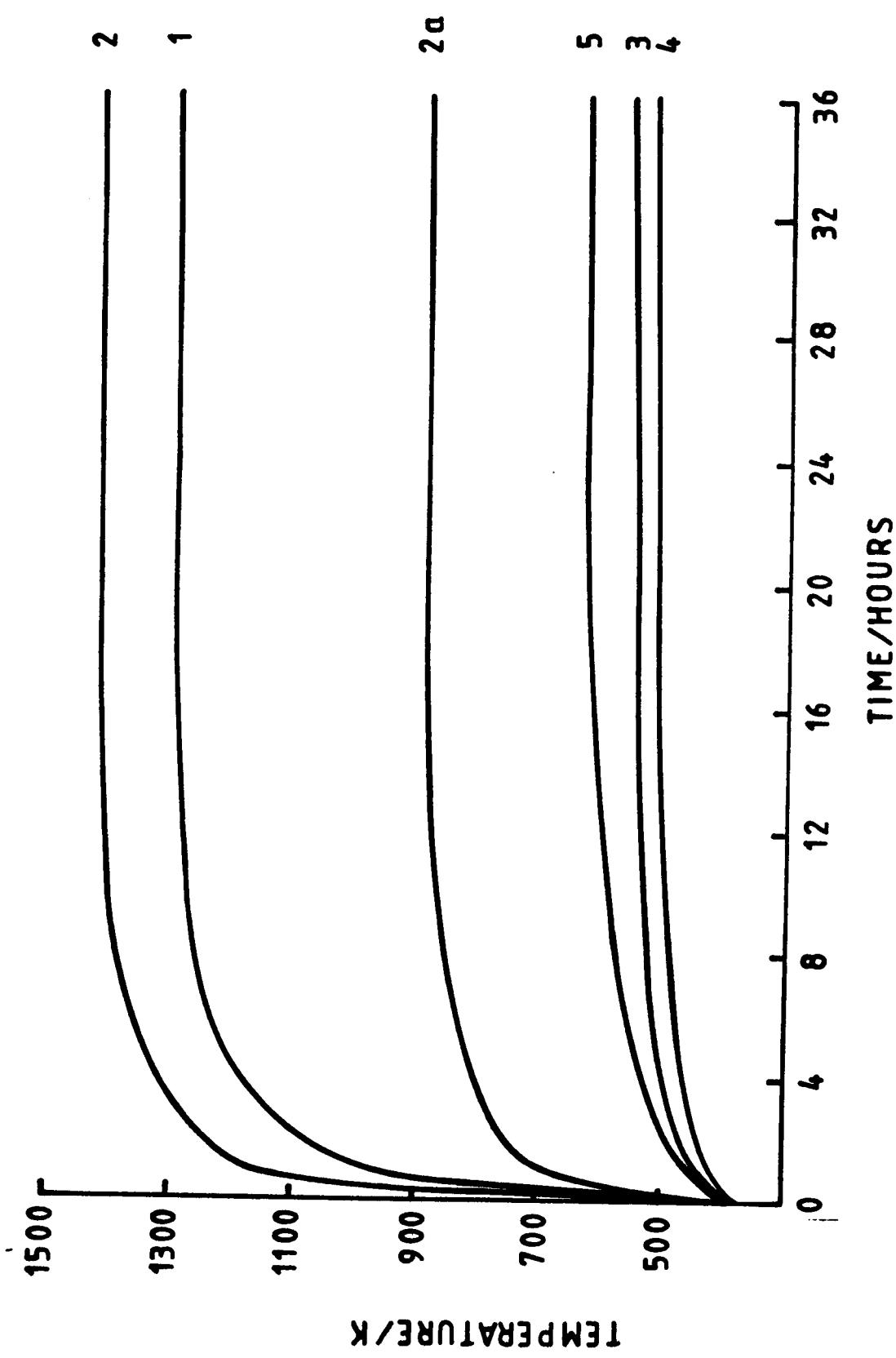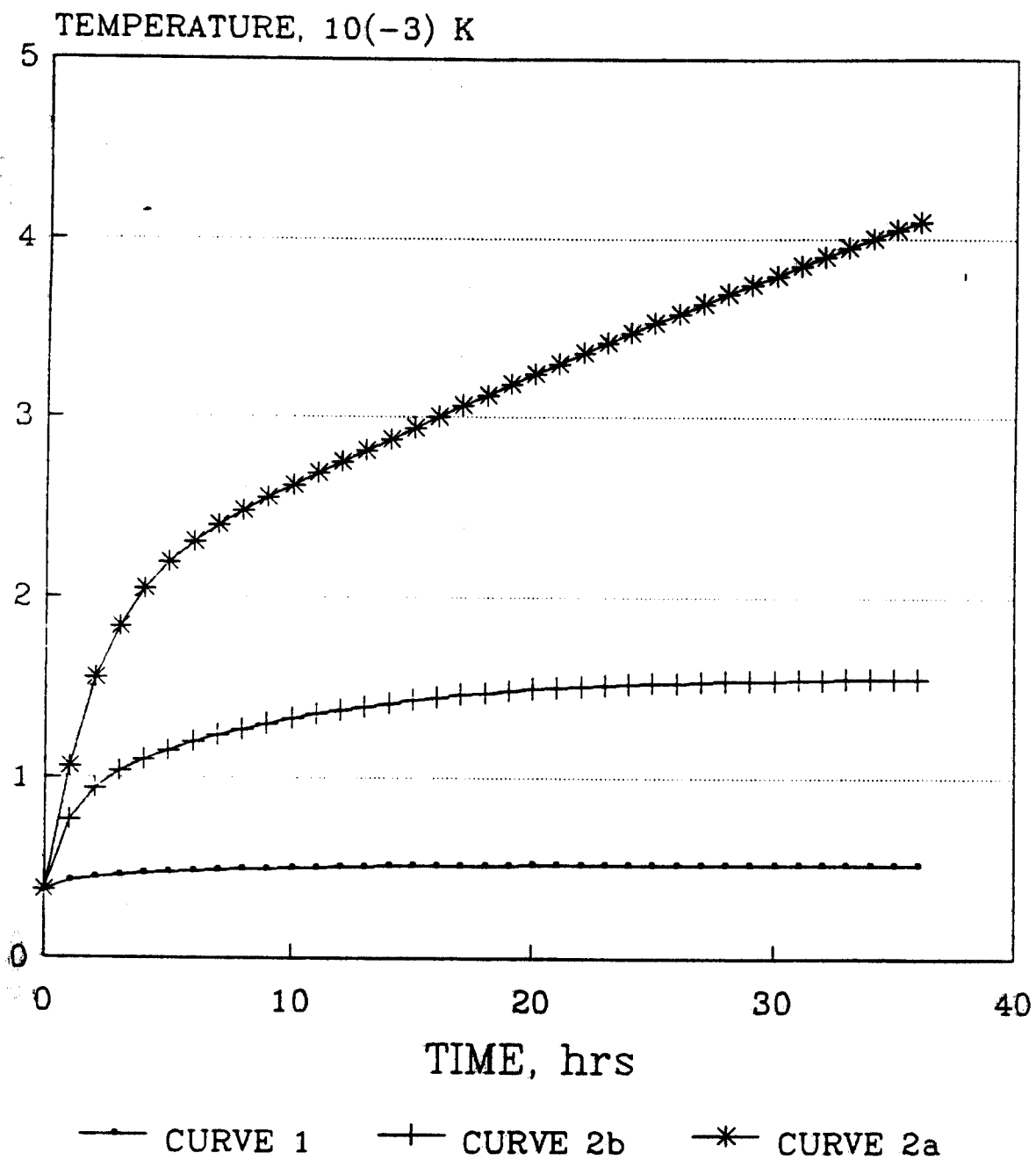| Time, s	10(4)	10(5)	10(6)	10(7)
Rate, kg/s	17.3	8.0	3.2	0.89

FIGURE 1   DEBRIS BED TEMPERATURES AS A FUNCTION OF TIME

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FIGURE 2

INFLUENCE OF OVERLYING SAND ON PWR DEBRIS BED TEMPERATURES

TEMPERATURE, 10(-3) K

TIME, hrs

——— CURVE 1   —+— CURVE 2b   —*— CURVE 2a

CHAPTER 3. MITIGATION OF MOLTEN CORE CONCRETE
INTERACTION RELEASE OF RADIONUCLIDES

3.1 Introduction

The accident scenarios under consideration presuppose core melt and vessel failure in LWRs. In the absence of mitigating actions the decay power of the molten core material will supply sufficient heat over a considerable period of time to affect a penetration of the pressure vessel lower head region. This results in a physico-chemical interaction at high temperatures between the molten core material and the structural concrete of the containment building. The process, molten core concrete interaction (MCCI) affects an ablation of the concrete and is accompanied by the formation of considerable amounts of gases and aerosols, carrying the radiotoxic fission products. Therefore MCCI is a potential threat to the integrity of the containment building due to containment overpressurisation caused by the explosive generation of steam and the formation of noncondensible combustible gases (hydrogen and carbonmonoxide). In addition melt-through of the reactor cavity solid structure (consisting of concrete and steel) can result in a below-ground release of radioactivity to the soil beneath the reactor.

The objective of the report is to describe potential short-term (t < 24h) mitigating actions to be considered for application in the prevailing (MCCI-scenario) conditions in order to reduce the release of radioactivity to the environment (source term).
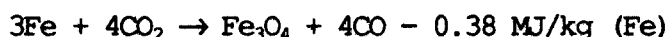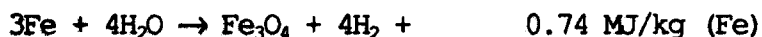
3.2 General Description of MCCS

The interaction between core debris and concrete in a severe (LWR) reactor accident is a well-recognised aspect of severe reactor accident analysis (1). The complex physical and chemical phenomena, associated with MCCI, are extensively described in the literature (2-7). Computer models of the thermal hydraulics of core debris/concrete interactions, the aerosol production and radionuclide release associated with these interactions have been developed (6). However, the level of understanding of the phenomenology of MCCI is still incomplete. The magnitude of core-concrete release is regarded as a key source of uncertainty for all NUREG-1150 reference plants (8). Moreover, the experimental data base for code validation is largely restricted to molten steel/core debris interactions. However, extensive experimental research projects (eg ACE) are planned at various scales to improve the level of understanding of MCCI including aspects of coolability of the molten core debris (9).

Temperature levels, typical for MCCI are in a temperature range between 2000 - 2500°C. The time and duration of MCCI for a given reactor is dependent on the specific accident scenario. Generally the time scale is in the order of hours.

During the course of the MCCI water and carbon dioxide are produced as thermal decomposition products of concrete. MCCI itself is an endothermic - process. However, with respect to the MCCI-thermal hydraulics a number of exothermic chemical reactions between core metals, carbon dioxide and steam

has to be taken into account. The reaction products hydrogen and carbonmonoxide are produced in significant amounts. Both compounds are non condensible and combustible gases. The important chemical reactions involved, including the reaction heat, are presented in table 1:

Metallic reaction:

$$Zr + 2H_2O \rightarrow ZrO_2 \quad + 2H_2 + 6.53 \text{ MJ/kg (Zr)}$$

$$Zr + 2CO_2 \rightarrow ZrO_2 \quad + 2CO + 5.83 \text{ MJ/kg (Zr)}$$

$$3Fe + 4H_2O \rightarrow Fe_3O_4 + 4H_2 + \quad 0.74 \text{ MJ/kg (Fe)}$$

$$3Fe + 4CO_2 \rightarrow Fe_3O_4 + 4CO - 0.38 \text{ MJ/kg (Fe)}$$

3.3 Mitigating Strategies

In general the ideal mitigating strategy is to prevent MCCI to the largest possible extent by the exclusion of a physical contact and heat transfer between the molten core material and the concrete walls and floors of the reactor cavity. If this can be achieved, the carbondioxide and steam production (caused by the thermal decomposition of concrete) are suppressed. Consequently, a potential heat source (reactions of zirconium with carbondioxide and steam) is removed. At the same time the formation of hydrogen and carbonmonoxide is prevented.

3.3.1 Ad hoc Catchers in the Cavity

One of the few available ad hoc measures is to arrange for core catching material to be deposited as a barrier layer between the reactor vessel and cavity during the course of the accident. A variety of passive systems can be used to dump the material, for instance from an external reservoir under gravity forces, preferably as free-flowing granular powder or as larger spherical particles. Access either through man hatches or through a separate penetration in the reactor chamber is feasible. The main problem, however, is the absence of an efficient heat sink to cool the core debris over a long period of time, if fusable materials are used as a temporary heat sink. If infusable barrier material is used, it is difficult to predict whether or not molten core material will penetrate the porous bed and reach the basemat or adversely will attack the drywell shell. The introduction of a layer of fusible material (glass, silica) on top of the infusable layer is a possibility both to dilute the melt and to trap fission products within the glass matrix. Because of the large uncertainties in the actual knowledge of the physical-chemical interactions between molten core material and barrier layers extensive experimental research with regard to modelling and selection of materials will have to be conducted. At the moment the method of dry deposition should be regarded as highly questionable.

3.3.2 Alternative Measures

If the possibilities to install an efficient barrier layer between molten core and concrete are absent, the mitigation of the consequences of MCCI is the important issue. In other words, what measures can be taken to

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increase the retention of aerosols, carrying the fission products and to prevent excessive pressure build-up in the containment. According to Powers (6) "the current accident management strategy of choice for dealing with core debris/concrete interactions is to attempt to quench the melt. To date, experimental studies of combined core debris/concrete/coolant interactions have shown water to have little effect on the attack on concrete. Neither has the attack been arrested nor has there been enhanced hydrogen production. Water pools overlying core debris with concrete are found to go into film boiling enhanced by the barbotoge of gases evolved from the concrete. Overlying water pools have been shown, however, to dramatically attenuate the aerosol production by core debris/concrete interactions. There remains, however, the need for additional experimental data to resolve questions on how existing results would change at increasing scale and whether steam explosions can be triggered when water pools overlie molten core debris". Conclusively, pool scrubbing, effected by an overlying water pool has to be addressed as mitigating strategy, specifically the short-term use of chemical additives to increase the retention of fission products, eg of radioiodine, which is promoted at p_H-values > 9 and reducing conditions eg by the supply of hydrazine (10, 11). Alternatively, the cavity can be provided with a dilute (alkaline) aqueous solution of sodium silicate. Evaporated to dryness $Na_2O.2SiO_2$ forms a water soluble glass with a melting point of 792 °C to immobilise fission products.

3.3.3 Future Concepts - Core Catchers/Coolers in the Cavity

For new plants, a large number of detailed pre-engineered designs to exclude MCCI have been extensively described in the literature, including a broad variety of ingenious patent applications. *) These so-called core catchers are installed in the pedestal region (BWR) or the containment cavity (PWR). Generally the concepts are based on high temperature (>2000 °C) resistant device, claimed to be mechanically strong enough to accommodate the molten core debris and provided with an active heat removal system (12-14). It is noted that a variety of compounds has been suggested as potential construction materials resistant to molten core material.

In the absence of oxygen, both tungsten (mp 3400 °C), molybdenum (mp 2617 °C) and graphite (sublimation > 3550 °C) can withstand very high temperatures. The feasibility of metals as core catching material is questionable, however, because the presence of large amounts of zirconium in the core debris will possibly promote corrosive alloy formation with metallic compounds. Since the Chernobyl accident the choice of carbon is highly controversial. Moreover, carbide (and carbonitride in the presence of nitrogen) formation with UO_2 is probable (15), as well as carbide formation with zirconium (16). Thorium oxide (mp 3220 °C), uraniumdioxide (mp 2878 °C) and magnesiumoxide (mp 2852 °C) have been proposed as core catching materials (12).

Mg, Cr, Zr, Cr(III) oxide based refractories can withstand very high temperatures. The melting points of MgO and Cr_2O_3 are 2800 and 2435 °C respectively. MgO and Cr_2O_3 form binary oxides with UO_2; the melting points

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*It is emphasised that the Task Cell has addressed these references as sources of information without taking any specific position on the issue of core-catchers.

of UO_2/MgO (50 mole %) and UO_2/Cr_2O_3 (50 mole %) are 1750 and 2050 °C respectively (17). Under reducing conditions of high temperature (maximal 1450 °C) coal gasification the errosion-corrosion resistance of these compounds towards molten coal ashes has been demonstrated (18-21). The experiments were carried out with commercially available non-porous bricks. Generally the penetration of the slag components into the bulk material was low due to the small porosity. The observed average wear rates were smaller than 40 μm/hour. Application of this material as protective liner for the cavity wall and floors is an interesting possibility.

In existing reactor plants the installation of a core-catcher/cooler in the reactor cavity is more complicated or largely impossible, depending on specific local conditions, particularly high radiation dose levels which limit or exclude the access of personnel. Also, the geometry and available space of the cavity and the presence of instrumentation and equipment, such as control rod drive systems in a BWR, are important parameters. Probably there is a limited number of possibilities to install core catchers and/or protective ceramic liners during maintenance and repair periods, for instance a coolable structure of refractory blocks, provided with cooling pipes and supported by a metal frame. To maintain mechanical stability at the prevailing temperatures the melting point of the core-catching material has to be high. In addition, physico-chemical and metallurgical interactions with the metallic and oxide phase of the melt should be absent. Also the formation of (non condensable) gaseous reaction products should be minimal. Finally, costs and commercial availability are important parameters. In support of NUREG-1150 the feasibility and costs of installing various mitigating systems in LWRs were investigated recently (22). The study includes a number of designs of Core Concrete Interaction Control Subsystems (CCICS) for PWRs and BWRs. As high temperature resistant materials the application of siliconcarbide (SiC), aluminiumoxide (Al_2O_3) and mageniumoxide (MgO) were proposed. The structure is cooled by flowing cooling media. In NUREG-1150, MCCI-mitigative options M2 and M4 are described for the Peach Bottom reference plant. Both include the construction of a concrete barrier in the drywell to prevent core debris contacting the drywell shell. The total costs of the M4 safety option is in the order of 10^6 USD.

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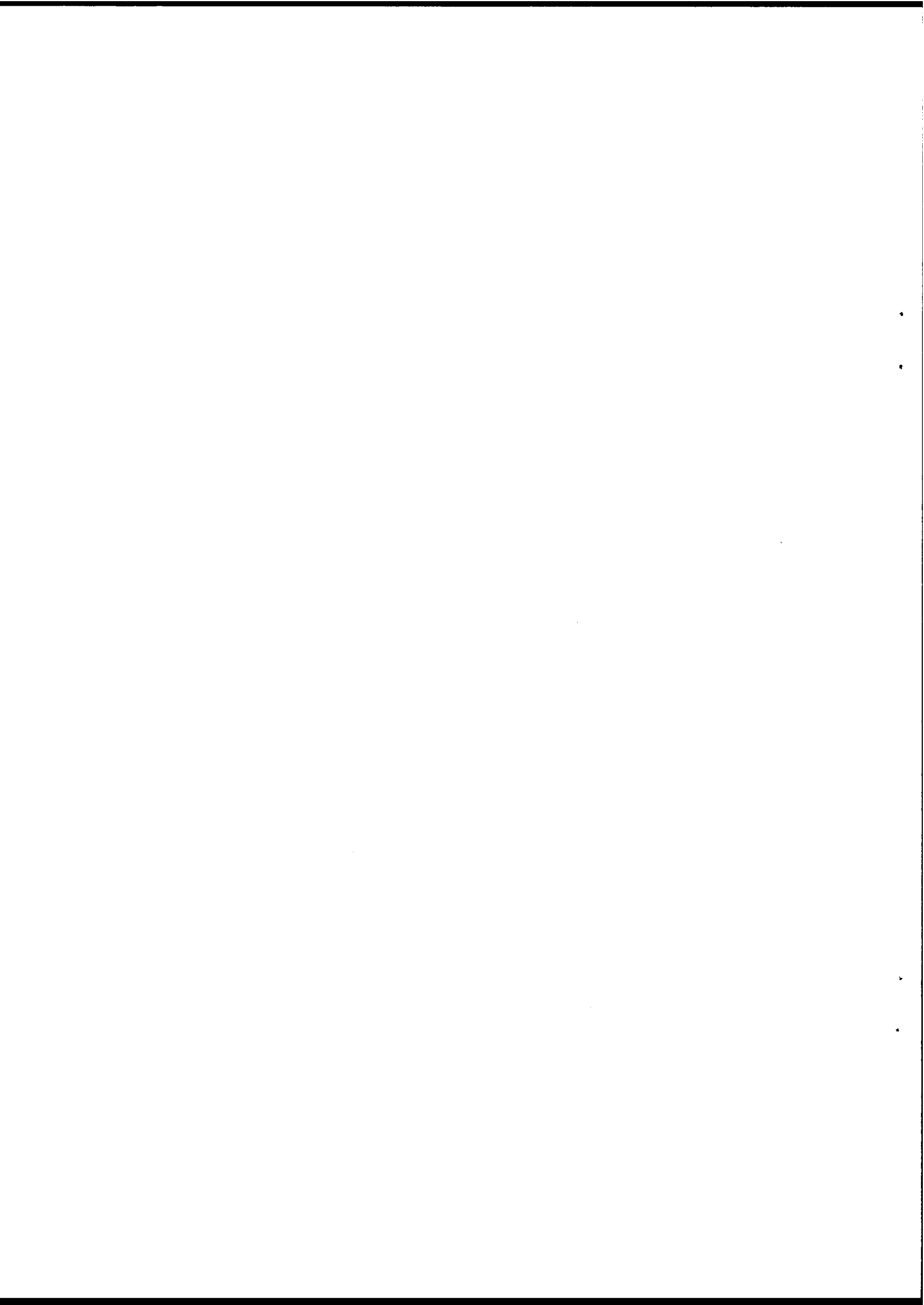
CHAPTER 4. CONCLUSIONS

There are many uncertainties concerning the state of the containment, and
the distribution of fission products and core debris in it, in the later
stages of an LWR core melt accident which results in containment failure.
This review has been oriented towards PWR severe accidents, although much of
the material is also pertinent to BWR accidents involving by-pass of the
suppression pool. One fairly certain feature is that there will be large
amounts of water present, and that much of the iodine fission product
inventory released from the fuel will be dissolved in water pools. The
analyses of the three main delayed source term phenomena in this review show
that maintainance of wet conditions offers the overall best approach to
control of activity release. The complex redox chemistry of iodine
determines the balance between volatile and involatile species in solution,
but the fraction in volatile form can be minimised by maintaining high pH.
This can be easily achieved by addition of bulk alkaline reagents such as
sodium hydroxide to the water. This approach will suppress the formation of
both inorganic and organic volatile iodine species. Other approaches, such
as addition of a reductant, may have some short-term benefit, but radiolytic
destruction of reducing reagents may limit the effectiveness.

The analysis in Chapter 2 shows that, unless emergency coolant water is
present already, fairly quick action is needed to arrange for a water supply
into the failed containment. Dry-out of the containment, or parts of it,
could occur in as little as 8 hours. The maintainance of an adequate water
supply to prevent dry-out is not onerous, although the integral demands over
a long period are substantial. Fortunately, almost all LWRs lie close to
large bodies of water which could act as adequate emergency reservoirs.
Water quality may not be an important consideration in such a situation,
although the use of sea-water, for example, could give considerable problems
at later stages of clean-up if ion-exchange methods are to be used. Careful
control of the supply rate, to eliminate escape of contaminated water to the
surroundings, is also needed.

The main potential problem in the use of water as a control measure is that
reflooding of large masses of hot debris might result in energetic
interactions which resuspend active material. This is particularly the case
if molten or very hot solid core debris in the reactor cavity is reflooded.
Much work is still in progress to assess the effectiveness of overlying
water pools in quenching core debris during MCCI, although there is ample
evidence that such pools will greatly attenuate fission product release.

If the approach of maintainance of flooding fails, then a second approach is
to add dry filtering materials to blanket the sources of resuspended
activity in the containment. The Chernobyl example provides a clear
illustration of the advantages and disadvantages of this method. The
modelling studies reported in Chapter 2 confirm the observation at
Chernobyl, that the long-term insulating effect of a filter blanket tends to
negate the benefits, by promoting much higher temperatures in the
decay-heated debris underneath. Dry blanketing is only a viable option if
some means of long-term heat removal can also be organised. A combination
of dry blanketing and subsequent flooding may have advantages in some
circumstances.

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In the absence of a pre-engineered core catcher, the options available for control of long-term MCCI are limited, largely due to problems of access to the reactor cavity region. The flooding approach is discussed above. Addition of fusible material, which might cool the melt, or form an inactive molten pool over the core debris to trap fission products, might be feasible in some circumstances.

Appendix

CSNI TASK GROUP ON FISSION PRODUCT PHENOMENA
IN THE CONTAINMENT
(FPC)

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