

Unclassified

NEA/CSNI/R(2000)12



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

OLIS : 07-Mar-2000
Dist. : 08-Mar-2000

PARIS

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

English text only

NEA/CSNI/R(2000)12
Unclassified

WORKSHOP ON IODINE ASPECTS OF SEVERE ACCIDENT MANAGEMENT

Summary and Conclusions

88381

Document complet disponible sur OLIS dans son format d'origine
Complete document available on OLIS in its original format

English text only

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.

CAM

The Task Group on Containment Aspects of Severe Accident Management (CAM) 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 on containment accident management and on techniques to protect the containment and their implementation.

FPC

The Task Group on Fission Product Phenomena in the Primary Circuit and the Containment (FPC) 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, perform ISPs in the field of fission product phenomenology.

**OECD Workshop on
Iodine Aspects of Severe Accident Management
Vantaa, Finland, 18-20 May 1999**

SUMMARY AND CONCLUSIONS

INTRODUCTION AND OBJECTIVES

Following a recommendation of the OECD Workshop on the Chemistry of Iodine in Reactor Safety held in Würenlingen (Switzerland) in June 1996 [Summary and Conclusions of the Workshop, Report NEA/CSNI/R(96)7], the CSNI decided to sponsor a Workshop on Iodine Aspects of Severe Accident Management, and their planned or effective implementation. The starting point for this conclusion was the realization that the consolidation of the accumulated iodine chemistry knowledge into accident management guidelines and procedures remained, to a large extent, to be done. The purpose of the meeting was therefore to help build a bridge between iodine research and the application of its results in nuclear power plants, with particular emphasis on severe accident management. Specifically, the Workshop was expected to answer the following questions:

- what is the role of iodine in severe accident management ?
- what are the needs of the utilities ?
- how can research fulfil these needs ?

The Workshop was organized in Vantaa (Helsinki), Finland, from 18 to 20 May 1999, in collaboration with Fortum Engineering Ltd. It was attended by forty-six specialists representing fifteen Member countries and the European Commission. Twenty-eight papers were presented. These included four utility papers, representing the views of Electricité de France (EDF), Teollisuuden Voima Oy and Fortum Engineering Ltd (Finland), the Nuclear Energy Institute (USA), and Japanese utilities.

The papers were presented in five sessions:

- iodine speciation
- organic compound control
- iodine control
- modeling
- iodine management

A sixth session was devoted to a general discussion on iodine management under severe accident conditions.

The meeting was concluded, for interested participants, by technical tours of the VICTORIA and COPO research facilities and of the Loviisa Nuclear Power Plant.

CONCLUSIONS AND RECOMMENDATIONS

Role of Iodine in Severe Accident Management

The role of iodine in Severe Accident Management can be considered on three separate levels:

- the level of the expected operator actions that are governed by the SAM guidelines and procedures,
- the level of the emergency response organisation actions, and
- the level of the plant modifications for the mitigative measures.

A possible role of iodine management is the influence of gaseous iodine on public protection needs when initiating the filtered containment venting. The means for reducing the gaseous iodine would be to increase the sump pH to clearly basic values provided that it is still compatible for long-term management operation.

The optimized filtered venting operation with respect to iodine reflects on all above levels. Some utilities might consider it beneficial to provide robust means to increase the sump pH and scrubber efficiency. Such measures might be passive or might require well-defined operator action to actuate.

Evaluation of possible delaying of manual filtered venting would be, in most cases, a task for the emergency response organization. The decision to delay filtered venting actuation would be based on estimated volatile iodine releases and consequent need for evacuation or sheltering. Such task is a typical interface between SAM and the emergency response organization.

In many countries, however, the decision regarding containment filtered venting actuation may incorporate other criteria. For example, in France, the decision to actuate containment filtered venting is made essentially on actual containment pressure criteria in order to avoid any risk of a gross containment failure. Such decision can be made more than a day after the onset of the accident, once the offsite emergency measures to protect the public have been completed.

Needs of utilities

The safety significance of iodine, in case of accidents, has always been recognized by the industry, and more particularly by the utilities. A prudent engineering approach, based on conservative assumptions, has led to addressing all iodine relevant problems at the design level, and robust systems and components have been provided.

All such components and systems are used for plant safety assessment, and would thus be called upon, if available, in case of accident.

Design and acceptance criteria for such components and systems can vary from one country to another. However, as some countries have designed such systems to address in vessel core-melt scenarios, there is no need, on the utility side, for further development for such systems and components. Concerning accident management, as was seen in the discussions which took place in the SESAM group (CSNI Senior Group of Experts on Severe Accident Management), critical decisions are made without factoring in iodine considerations, as the

major concerns are to stop accident progression and prevent uncontrolled loss of containment integrity. Guidance is generally well established, and here again, no urgent need has been identified at the utility level.

Two issues, however, could be considered, for which data consolidation could be welcome:

- iodine behavior in the secondary side of SGs
- possibility for on-line measurement of iodine concentration inside containment.

This, however, should be understood as a need to summarize existing data for the former, evaluate the interest and feasibility for the latter, rather than a need for further extended development.

It should also be emphasized that the current trend towards electricity market deregulation will put pressure on utilities to decrease their generation costs. Although there is no evidence that utilities will all take the same actions to achieve this objective, there will probably be a trend to reassess the need for some regulatory constraints. Examples of such initiatives were given in the meeting for the U.S. regulatory context, utilities contemplating applications for charcoal removal in some filtration systems or for eliminating the need for post-accident sampling.

Finally, considering the current lack of plans for future development of nuclear power, maintaining and transmitting knowledge could well become a critical issue in the near future. Although no precise need was identified during the meeting, as most presentations were clearly on progress in R&D, it could be wise to evaluate whether additional knowledge on iodine behavior is deemed necessary for the safety of currently operating plants in the years to come, and, if the answer is positive, which level of knowledge would be adequate and how to maintain it.

Related issues such as activities having the potential for attracting young graduates should be discussed in this perspective.

Needs of regulators

Regulators need information, knowledge, an adequate data base and competence in order to perform their duties. This is true in particular for assessing the iodine aspects of severe accident management.

Another reason for regulators to need knowledge on iodine behavior is that they have either to perform or to review level 2 PSAs. Assessing the source term and the impact of accident management actions on the releases requires adequate knowledge of the main fission product chemistry and phenomena in the containment. The needed accuracy depends on the uses of the PSA.

Deregulation puts pressure on utilities to reduce costs and to relax regulatory constraints. The regulators should be able to judge the acceptability of relaxation requests and their justification, and for that they will need up-to-date information.

Finally, during a real crisis situation, regulators may be called upon to take or recommend prompt decisions to protect the public, such as (1) is sheltering better than evacuation, (2) should the public be instructed to take potassium iodide tablets. Careful preparation may not

be enough to take such decisions. A team of people knowledgeable in plant behavior in severe accident situations, and in iodine behavior in particular, may be needed.

Improvement of predictive capabilities

Presentations and discussion during the Workshop brought up the need to finish tasks that are currently underway and would improve the predictions of iodine behaviour. There is need for validated, fast-running models, integrated to plant codes, to predict iodine behavior, for design assist, and safety analysis and accident response decision-making. Implicit is the need to complete and consolidate the current generation of experiments (i.e., PHEBUS) and the need for distillation of the knowledge into practical tools (i.e., IMOD) which is just entering fruition.

Specifically, there is also a need for reliable predictions of time-dependent airborne iodine species concentrations to establish qualification requirements for hydrogen recombiners.

Progress made on major iodine chemistry safety issues since the 1996 Iodine Workshop

Homogeneous Phase Iodine Chemistry

The sump pH is the most important parameter in determining the formation of volatile iodine in the aqueous phase, and the maintenance of a high pH could be an important accident management tool in many sequences. It is therefore important that the factors leading to the acidification of the sump are well understood and quantified.

Recent results on nitric acid formation confirm old calculations and experiments performed at laboratory scale. The effect of surface material on nitric acid formation is recognized to be significant, but no experimental results were presented to quantify the extent of the surface effect.

Pyrolysis of cable insulation as a result of core melt can be extensive and produce HCl and Cl₂. This could have an impact on pH, and filtering and scrubbing in severe accident management, but plant-specific evaluations should be considered. Significant progress has been made on understanding and modeling the effect of organic materials on pH decrease. The radiolysis of organic solvent dissolved from painted surfaces could lead to acid production in the containment sump.

The sump temperature is confirmed to be of secondary importance in determining iodine volatility as the result of further data obtained on hydrolysis and partition coefficient of various organic iodides.

In the gas phase, organic iodine could be higher than originally expected, in particular at low oxygen content, in the presence of chlorine and B₄C control rod material in the BWR case.

The rate of conversion of I₂ to IO₃⁻ by ozone is lower than previously thought, and is expected to be even lower if surface effects and the presence of H₂ are considered. However, further confirmation is needed.

Surface reactions

Reactions of iodine with structural surfaces are better understood in the area of organic iodide and acid production.

New data has been provided by PHEBUS FPT 1 tests on iodine/painted surfaces interactions.

Reactions with surfaces in the containment could affect the iodine volatility either by permanent trapping on the surface, or by changing the chemical form of iodine.

Significant progress has been made on Ag-I interaction. Good data and proper modeling have been obtained on the major mechanisms of $\text{Ag} + \text{I}_2$ reactions. Nevertheless, in addition to the importance of the $\text{Ag}_2\text{O} + \text{I}^-$ reaction, there is still some debate on the stability of AgI and also Ag_2O in the presence of radiation. NO_x and Cl^- may reduce AgI stability; this is to be confirmed by ongoing experiments.

Mass transfer

Overall gas-aqueous interfacial mass transfer uncertainties remain but they are generally considered to be of low impact on iodine source term evaluation.

Modeling of iodine chemistry

Significant progress has been achieved on simulation of intermediate scale tests using various models.

The experts expressed clear needs for further ISP activities focussing on iodine behavior under accident conditions.

The most significant development on modeling of iodine chemistry is the development of simple models that are flexible and readily understandable.

Availability of improved fast running integral codes will help better understand the interactions between severe accident phenomena and the effect of severe accident management measures on the iodine source term and will also aid decision making.

Iodine management

The progress made since the last Iodine Workshop includes the following:

- the effects of paints and cable insulation material are now widely acknowledged and there are calculation tools available so that those effects on pH evolution can be quantified,
- plant modifications to enhance pH control in plants that are most vulnerable to iodine volatilization have been initiated,
- the effects of chlorine on iodine behavior in scrubber filters have been identified and some plant modifications to mitigate these effects have already been made,
- the possibility of poisoning effect of gaseous iodine on catalysts has been restated.

Concluding remarks on the status

Iodine Speciation and Chemical Processes

Iodine behavior in containment is a complex interplay of sump radiolysis and surface chemistry reactions, strongly influenced by key parameters such as temperature, pH and composition of surface and solute materials.

Effects of pH on iodine volatility and the related effects of paints and cable insulation are now widely acknowledged and calculation tools predict iodine behavior with reasonable accuracy.

Modeling

ISP 41 was the first comparison exercise on iodine code. Mechanistic and semiempirical codes were capable of producing satisfactory results, but they showed sensitivity to user experience and selection of input data.

Simple iodine models, suitable for use in fast-running system codes, have shown ability to reproduce integral experimental results and are verified by mechanistic code calculations.

Iodine Control

Significant reduction in iodine releases can be obtained by controlling containment water pH and by filtering containment leakage.

Silver is an effective trap for molecular iodine. Experiments also indicate the probable high trapping efficiency of iodide by extensively oxidised silver aerosols, but uncertainties exist regarding knowledge of the surface state. Stability of AgI to radiation, especially in the presence of Cl⁻ and NO₂, remains to be established, according to recent investigations. An experimental programme (PHEBUS Project - PSI) is addressing this issue.

HCl, arising from pyrolysis of cable insulation, in the absence of strong buffering can cause lowering of pH, increase of volatile iodide formation, and possible impairment of filtered venting scrubber systems by Cl₂.

In ice condenser designs, buffer chemicals (borax) in the ice provide efficient pH control, utilizing favorable mass transfer characteristics within containment.

Charcoal filters are shown to provide an effective and well-characterized barrier to iodine release from the vacuum building in CANDU plants.

New results

PHEBUS experiments have shown that significant amounts of gaseous iodine were transported in the primary circuit during certain phases of the test. The gaseous iodine injected at the break from the circuit was the main cause of the observed gaseous iodine fraction in the containment during the short term (several hours). In the middle term (a few days), the organic iodides are the dominant species. They result from the partial conversion of the iodine that reacted with the atmospheric paints.

General/Accident Management

Iodine aspects enter SAM issues at different levels, primarily at the interface with emergency response planning, safety assessment and possible plant modification design analysis. Should a severe accident happen, iodine issues would not drive accident management decisions. No additional research need was identified with respect to present regulatory requirements.

The Programme Committee points out, however, that further research may still be needed for other applications, such as risk assessment, emergency response planning, and to maintain capability and competence in the field of iodine chemistry and its application to nuclear reactor safety.

Annex I:

PROGRAMME COMMITTEE

Dr; Harri Tuomisto (Fortum Engineering Ltd, Finland) - Chairman *

Mr. Benoît De Boeck (AVN, Belgium)

Mr. Jacques Duco (IPSN, France)

Mr. Salih Guntay (PSI, Switzerland) *

Mr. Grant W. Koroll (AECL, Canada)

Prof. Jan-Olov Liljenzin (Chalmers University of Technology, Sweden) *

Mr. Michel Vidard (EDF/SEPTEN, France)

* Session Chairmen

Additional Session Chairmen:

Dr. Didier Jacquemain (IPSN, France)

Mr. Timo Karjunen (STUK, Finland)

Dr. J. Clara Wren (AECL, Canada)

Programme Committee Secretary:

Mr. Jacques Royen (OECD/NEA)

Annex II:

PROGRAMME

Tuesday 18 May 1999

- 08.00 - 09.00 **REGISTRATION**
- 09.00 - 09.20 **WELCOME :**
- H. Väyrynen, Vice President Nuclear Power, Fortum Engineering Ltd
H. Tuomisto, Fortum Engineering Ltd, Chairman of the Workshop
J. Royen, OECD/NEA, Secretary of PWG4, FPC and CAM
- 09.20 - 09.50 **INTRODUCTION TO THE WORKSHOP**
- 09.20 - 09.30 *Background of the Workshop*
by J. Duco, Chairman of CAM
- 09.30 - 09.50 *The Needs of the Nuclear Industry with Respect to Iodine Aspects of Severe Accident Management : A Utility Perspective*
by M. Vidard
- 09.50 - 13.00 **SESSION I - IODINE SPECIATION**
- Chairman : J.-O. Liljenzin (Chalmers Technical University, Sweden)**
- 09.50 - 10.20 *Iodine Chemical Kinetic Study within the PHEBUS Primary Circuit*
by L. Cantrel and L. van Wijk
- 10.20 - 10.40 **BREAK**
- 10.40 - 11.10 *Iodine Studies at the University of Toronto : Piecing Together a Portrait of the 53rd Element*
by G.J. Evans, F. Taghipour, T. Nugraha, J.R. Ling, E.P.D.B. Reynolds and A. Narayanan
- 11.10 - 11.40 *An Overview of the Iodine Behaviour in the Two First PHEBUS Tests FPT-0 and FPT-1*
by D. Jacquemain, N. Hanniet, C. Poletiko, S. Dickinson, C. Wren, D.A. Powers, E. Krausmann, F. Funke, R. Cripps and B. Herrero
- 11.40 - 12.10 *Radiolytic Oxidation of Molecular Iodine in the Containment Atmosphere*
by F. Funke, P. Zeh and S. Hellmann
- 12.10 - 12.40 *Chlorine Release by Pyrolysis from Hypalon Cable Insulation Material and its Effect on Iodine Speciation in the Containment*
by A. Auvinen, J.K. Jokiniemi and R. Zilliacus

12.40 - 13.00	<i>General Discussion on Session I</i>
13.00 - 14.30	LUNCH
14.30 - 18.10	SESSION II - ORGANIC COMPOUND CONTROL
	Chairwoman : J. C. Wren (AECL, Canada)
14.30 - 15.00	<i>Iodine Volatile Species Production from Painted Surfaces of the Reactor Containment, During a Severe Accident</i> by E. Belval-Haltier and P. Taylor
15.00 - 15.30	<i>Experimental Studies of Organic Iodide Production</i> by C.B. Ashmore, G. Baston, T. Dean, S. Dickinson, A.J. Matcham and H.E. Sims
15.30 - 16.00	<i>Small Scale Experiments on Organic Iodide Production from Iodine-Painted Surface Interaction</i> by C. Marchand and M. Petit
16.00 - 16.20	BREAK
16.20 - 16.50	<i>Data Analysis and Modelling of Organic Iodide Production at Painted Surfaces</i> by F. Funke
16.50 - 17.20	<i>Organic Iodide Formation in BWR Accidents</i> by T. Karjunen and R. Zilliacus
17.20 - 17.50	<i>Studies on the Effects of Organic-Painted Surfaces on pH and Organic Iodide Formation</i> by J.C. Wren, J.M. Ball and G.A. Glowka
17.50 - 18.10	<i>General Discussion on Session II</i>
18.30	RECEPTION

Wednesday 19 May 1999

- 08.30 - 12.40 **SESSION III : IODINE CONTROL**
- Chairman : D. Jacquemain (IPSN, France)**
- 08.30 - 09.00 *Calculations on Iodine Speciation in Gas and Liquid Phases*
by A. Emrén, C. Ekberg and J.O. Liljenzin
- 09.00 - 09.30 *Kinetics of the Uptake of Aqueous Iodine on Silver Surfaces*
by S. Dickinson, H.E. Sims, E. Belval-Haltier, D. Jacquemain, C. Poletiko,
F. Funke, Y. Drossinos, E. Krausmann, B. Herrero, T. Routamo, E. Grindon and
B.J. Handy
- 09.30 - 10.00 *A Model of Heterogeneous Silver-Iodine Reactions in the Liquid Phase*
by E. Krausmann and Y. Drossinos
- 10.00 - 10.20 **BREAK**
- 10.20 - 10.50 *Stability of AgI Colloids in the Presence of a Dissolved High Dose-Rate β -Emitting
Radionuclide*
by R. Cripps, H. Bruchertseifer, B. Jäckel and S. Guntay
- 10.50 - 11.20 *Modeling of Iodine and Nitrogen Oxides Trapping by Acidic Hydroxylamine in Nuclear
Spent Fuel Reprocessing - Consideration of Applicability to Reactor Severe Accident
Management*
by C. Cau dit Coumes, S. Courtois, J. Chopin-Dumas and F. Devisme
- 11.20 - 11.50 *A Model to Simulate the Removal of CH_3I from Airstreams Under Post-Accident
Conditions by Charcoal Filters*
by J.C. Wren, C.J. Moore, M.T. Rasmussen and K.R. Weaver
- 11.50 - 12.10 *General Discussion on Session III*
- 12.10 - 13.30 **LUNCH**
- 13.30 - 17.10 **SESSION IV : MODELLING**
- Chairman : S. Guntay (PSI, Switzerland)**
- 13.30 - 14.00 *A Study of the Effects of Radiation and Impurities on the Gaseous Iodine Production in
a Containment During a Severe Accident*
by M. Takahashi, S. Nukatsuka, T. Hashimoto and Y. Tanaka
- 14.00 - 14.30 *International Standard Problem (ISP) No. 41 : Computer Code Comparison Exercises
Based on a Radioiodine Test Facility (RTF) Experiment on Iodine Behaviour in
Containment under Severe Accident Conditions*
by J. Ball, G.A. Glowa, J.C. Wren, A. Rydl, C. Poletiko, Y. Billarand, F. Ewig,
F. Funke, A. Hidaka, R. Gauntt, R. Cripps, B. Herrero and J. Royen
- 14.30 - 15.00 *A Simplified Model for Containment Iodine Chemistry and Transport : Model
Description and Validation Using Stainless Steel RTF Test Results*
by J.C. Wren, G.A. Glowa and J.M. Ball
- 15.00 - 15.20 **BREAK**

- 15.20 - 15.50 *Simplified Modelling of Iodine Behaviour for Source Term Assessment of French Reactors*
by M. Petit, C. Marchand, Y. Billarand and C. Rouillon
- 15.50 - 16.20 *Iodine Behaviour Calculations for a TMLB' Sequence*
by B. Herrero and E. Verduras
- 16.20 - 16.50 *Iodine Source Term Calculations for the Dukovany VVER-440/213 NPP*
by A. Rydl, P. Vokac and J. Dienstbier
- 16.50 - 17.10 *General Discussion on Session IV*

Thursday 20 May 1999

09.00 - 10.30	Session V - Iodine Management Chairman : T. Karjunen (STUK, Finland)
09.00 - 09.30	<i>Iodine Management in the Finnish NPPs</i> by H. Sjövall, T. Routamo and H. Tuomisto
09.30 - 10.00	<i>Making Practical Use of Iodine Knowledge in Severe Accident Management Strategies: A U.S. Industry Perspective</i> by D.J. Modeen, D. Leaver, R.J. Lutz, Jr. and R. Schneider
10.00 - 10.30	<i>Iodine Behavior Under BWR Severe Accident Conditions</i> by R. Hamazaki, M. Akinaga, Y. Haruguchi and H. Chiba
10.30 - 10.50	BREAK
10.50 - 13.00	Session VI - General Discussion on Iodine Management under Severe Accident Conditions Chairman : H. Tuomisto (Fortum Engineering Ltd)
10.50 - 11.30	<i>Session Chairmen Summaries</i>
11.30 - 13.00	<i>General Discussion</i> , in particular : * What is the role and importance of Iodine Management in the overall Severe Accident Management strategy? * What are the needs of the utilities? How can research fulfil these needs?
13.00	CLOSE OF THE WORKSHOP
13.00 - 14.00	LUNCH
14.00	Visit to the VICTORIA and COPO II research facilities of Fortum Engineering Ltd

Friday 21 May 1999

09.00	Technical Tour of the Loviisa Nuclear Power Plant (limited to 20 participants)
-------	---

Annex III:

SESSION SUMMARIES

Introductory Session

There were two invited lectures to introduce the background and the objectives of the Workshop. The Chairman of the CAM, Mr. Jacques Duco explained the background of the current Workshop and highlighted the conclusions from the 4th Workshop on the Chemistry of Iodine in Reactor Safety held on June 10-12, 1996 at Würenlingen, Switzerland. He stressed that it would be beneficial to review the progress made since that meeting and how the progress can be integrated into accident management.

Mr. Michel Vidard of EdF discussed the needs of the nuclear industry with respect to the iodine aspects of SAM. He concluded that the iodine aspects have been addressed in the design of existing plants using quite conservative assumptions and that no major modifications were expected for Accident Management. Remaining uncertainties are deemed manageable by utilities.

Session I: Iodine Speciation (Prepared by Professor J.O. Liljenzin, Chalmers University of Technology, Gothenburg, Sweden)

Five papers were presented in Session I. These can be divided into three groups according to the areas treated, see Table below.

The first two papers in the Table treated various aspects of the results from the PHEBUS FP tests FPT-0 and FPT-1 of importance for the iodine source term and initial iodine speciation. The current paradigm of release from the overheated core as CsI is questioned as measurements indicate that a fraction of the iodine is transported in a more volatile form during certain phases. InI formation has been postulated, based on kinetic calculations, but no experimental evidence is available. Furthermore, elementary iodine is probably also emitted in non-negligible amounts from the primary system in these tests. Analysis of data indicates that kinetics might play a role in iodine speciation in the primary circuit. These observations may require future changes of the initial chemical form of iodine in many of the currently used severe accident analysis codes. This will then probably have an indirect effect on accident management as such codes are often used to predict the progression of severe accidents and the possible effects of planned management actions.

The second group of two papers described measurements of iodine reactions of importance in air filled containments, but also important for inerted P/S-containments during the start of reactor operation, i.e., before reduction in oxygen concentration is complete. Furthermore, these data are also of interest for both types of containments in case of severe accidents due to loss of cooling during refuelling and maintenance. An interesting observation by Evans et al. is the formation of organic iodides with higher molecular weight than CH₃I, and correspondingly lower volatility. Ozone induced oxidation of elementary iodine is effective in reducing the concentration of gaseous I₂, not only in irradiated dry air but also in the presence of steam.

Paper by	Area	Accident Management Relation	PWR	BWR
Cantrel et al.	Iodine speciation in PHEBUS FPT-0 & -1 tests	- I and Cs released in chemical forms not in accordance with the common assumption of CsI and CsOH - Primary system modeling	YES	Some
Jacquemain et al.				
Evans et al.	Formation of organic iodides and iodine oxides	- Effects of organic impurities in containment water - pH control to minimize RI formation - I volatility reduction by ozone	YES	Some
Funke et al.				
Auvinen et al.	HCl from cable pyrolysis	- pH control - RI trapping in venting filters	?	YES

The third group only comprises one paper, which describes measurements of HCl release from a typical cable insulation, HYPALON, during pyrolysis in the absence of oxygen. The observed amounts of HCl per weight of insulation corroborates earlier scouting measurements made in the Swedish RAMA project in the 1980's. The effect of HCl on iodine behavior and management is threefold. Firstly the resulting very low pH of unbuffered water volumes will increase the fraction of iodine converted to organic iodides (RI). Secondly, radiolytic production of Cl_2 will lead to an increase of the fraction of iodine present as I_2 . Thirdly, HCl will consume some of the amount of iodine

removal reagents present in filtered venting scrubber systems, requiring higher initial amounts. From an accident management point of view this will require earliest possible injection of the proper amount of strongly alkaline liquids or buffer solutions into containments with initially unbuffered water.

Session II: Organic Compound Control (Prepared by Dr. J Clara Wren, AECL, Canada)

The session on organic compound control dealt primarily with organic iodide formation, which is of interest, because of the difficulty of filtering or scrubbing these species. Since the last iodine workshop, significant progress has been made in understanding the role of painted surfaces in formation of organic iodides, and a large data base has been accumulated on the formation of these species.

It is now well established that organic iodides may be formed as a result of the interaction of organic solvents "leached" from painted surfaces and I_2 in the aqueous phase. The general features of the process are well understood and a model has been developed.

The mechanism for organic iodide formation from interaction of I_2 on painted surfaces is not as well understood, however a significant effort has been made in performing experiments, compiling the data and developing an empirical model based on this mechanism.

Because the relative importance of the two processes (homogenous aqueous phase and heterogeneous surface) has not been established, quantifying organic iodide inventory with any certainty in containment under accident conditions, *for those situations where surface reactions are expected to be important*, is still somewhat "problematic".

A suggested approach to address the issue of the quantification of organic iodide is to compare the predictions by each of these models, for a given series of reactor accident conditions. This will establish whether or not it is important to firmly identify a proper mechanism in quantifying the organic iodide production rate. If it is, additional work will be required to establish the mechanism for the heterogeneous formation route.

An additional influence of painted surfaces is their effect on pH of the sump when in contact with irradiated solutions. The radiolysis of organic solvents dissolved from painted surfaces could lead to significant quantities of acid being produced in the sump. The mechanism is now well understood and easily modeled. The impact of this study on SAM is in pH control during severe accidents.

Experiments have also been initiated on the homogeneous gas phase production of organic iodides from CH_4 and I_2 , which could be an important route for formation in BWR reactor accident sequences. The new results indicate that the organic iodide formation under BWR conditions may be higher than originally expected. The effect of chlorine appears to be significant. Large amounts of B_4C in the control rods of some PWRs (EDF 1,300 MW units) could also impact the organic iodide production during a severe accident.

In summary an extensive database now exists for the effect of organic components on iodine volatility, and modeling is reasonably well advanced. Additional effort is required to establish the organic iodide inventory in containment for situations in which the generation of organic iodides on surfaces is expected to be significant.

Session III: Iodine Control (Prepared by Dr. Didier Jacquemain, CEA, France)

Six papers were presented in Session III. These can be divided into two groups according to the areas treated.

The first four papers concerned research linked to the study of limitation of gaseous iodine formation due either to the effect of maintaining high pH's or the efficiency and stability of iodine trap (namely reaction with Ag to form insoluble Ag I). Progress was made to quantify factors possibly leading to the acidification of the sump: release of HCl from cable insulation by pyrolysis in the BWR situation, determination of nitric acid formation under radiation in glass vials.

There is a consensus in saying that high pH should be maintained in the sump to reduce volatile iodine formation from the aqueous phase. However, uncertainties remain to quantify acid production under representative conditions: effect of surfaces on HNO₃ production, effect of released material from fuel degradation, production of CO₂ in case of core-concrete interactions.

Two papers described the substantial progress made to gain a firm understanding of Ag /I interactions in the sump. Modeling of AgI formation in the sump has reached a sufficient level of accuracy to model small scale (SIEMENS and AEAT) and intermediate scale (PHEBUS, RTF, AECL) experiments. The use of Ag/I modeling for the reactor case requires the knowledge of the Ag surface state (oxidation, aerosol size, morphology). There is still no firm understanding of the effect of dissolution of Ag oxides on the Ag/I reaction. There is a consensus in saying that Ag may be a very efficient iodine trap in plants where Ag-In-Cd control rod material are used, provided AgI is stable to radiation. Stability of AgI to radiation, especially in the presence of Cl⁻ and NO₂, remains to be established according to recent investigations.

The last two papers described a technical proposal and a technical system to mitigate volatile iodine. A proposal was made to substitute NaOH by hydroxylamine which offers the potential advantage of reducing I₂ volatilization from acidic sump down to pH = 1.0 and could help to get rid of NO_x and H₂O₂ to some extent. Substantial research work is still required to determine the stability and mitigation efficiency under accidental conditions. TEDA impregnated charcoal were shown to be adequate organic iodides filtering devices when used to temperature up to 80°C and relative humidities up to 70 %. Kinetics of uptake of organic iodides as well as desorption rates were established for those conditions. Since other work showed that organic iodides may contribute significantly and even be in some cases the major contributors to the volatile iodine fractions (PHEBUS situation), making adequate technical systems for the filtration of those compounds appears to be of importance. Additional research may be necessary to show that the results in this presentation could be applied to other reactor systems and different conditions.

Session IV: Modelling (Prepared by Mr. Salih Güntay, PSI, Switzerland)

Six papers that can be grouped in the following 2 categories were presented in this session:

1. Modelling and verification
2. Application of iodine chemistry for plant assessment in conjunction with an integral severe accident system code

Four papers were presented in the first category. The first presented NUPEC's further modelling efforts to improve the IMPAIR3 code in the areas of hydrogen peroxide production and its reaction with iodine, production and reaction of dissolved oxygen, the effect of boric acid, and nitric acid production. Assessment performed using RTF Test RTF3b re-demonstrated the predictive capability of IMPAIR3.

The second paper presented the outcome of the 'International Standard Problem ISP41', which was recommended by the 1996 Workshop. Nine organisations participated in this exercise which was organised for the first time by the OECD in this field. The comparisons indicated that the mechanistic codes (LIRIC and MELCOR-I) reproduced the RTF test data successfully. The exercise also indicated the importance of the user experience in selecting the model parameters (rate constants) used by the empirical codes (IODE and IMPAIR3). It demonstrated the need to extend the validity of the models incorporated in such codes to cover a broader range of conditions affecting the iodine chemistry. The experts expressed clear needs for further ISP activities focussing at other significant aspects of the iodine chemistry.

The third and fourth papers in the first category provided new approaches to treat the iodine chemistry with a very small number of species or to treat species categorised in 6 groups. The simplification of the chemistry has been attempted to obtain fast running computer models but still to predict the gaseous iodine concentration with reasonable accuracy. The approaches taken by both Canadian and French organisations have used rate constants developed based on the extensive sensitivity analysis of the comprehensive mechanistic models. Although further work is needed to complete the development, the demonstrated predictive capabilities have indicated possibilities for future roles of such models if they are incorporated in integral system codes. Availability of such improved fast running integral tools will help to better understand the interactions of the several severe accident phenomena and the effect of several accident management measures on the iodine source term and will also aid the decision making.

The fifth and the sixth papers of this session in the second category presented applications of detailed iodine chemistry using the IODE code with the initial and boundary conditions as predicted by the system codes, MAAP3b and MELCOR for a large PWR and a VVER-440, respectively. The importance of key parameters, like sump pH, spray operation, etc., on the iodine source term into the environment was shown. The papers highlighted the further needs to determine the sump pH based on the conditions evolving during the accident progression and for the pH control. Highlighted also is the need for a coupled treatment of iodine chemistry with the rest of severe accident phenomenology. This argument supported the goals of the further simplified iodine model development introduced above.

Session V: Iodine Management (Prepared by Mr. Timo Karjunen, STUK, Finland)

The original approach to iodine management in a number of countries (TID 14844) was to design nuclear power plants assuming that during an accident a large fraction (50 %) of core inventory is released into containment, where one half is rapidly deposited while the other half stays initially airborne. From the airborne iodine, most (91 %) in elemental form, 5 % appears as aerosols and 4 % in organic compounds. These assumptions lead to containment designs with a relatively stringent requirement for leak tightness and to control of containment leakage by ventilation and filtering.

As knowledge concerning iodine behavior has expanded substantially over the years, some of the early assumptions are now viewed as overly conservative, as stated in the second paper presenting U.S. industry perspectives on iodine management by Mr. D. Modeen. He listed the following mitigating phenomena:

- iodine is released mainly in the form of aerosols,
- the aerosol form of iodine is generally retained in containment more readily than the gaseous form,
- reevolving of iodine from the containment sump is prevented by maintaining neutral or alkaline pH,
- significant fraction of iodine released from the core is retained in the primary circuit.

The current plant designs and operating practices are considered to provide adequate protection of the public from severe accidents. Consequently, the industry is now pursuing regulatory relief from systems such as post-accident sampling system, which are judged to be costly and unnecessary. No uncertainties in accident management have been identified by the U.S. industry that would warrant further studies.

During the discussion following the presentation, Mr. J. Lee from NRC pointed out that the term "severe accident" is used in the U.S.A. to refer to accidents involving both core damage and pressure vessel rupture, which are considered to be beyond the current design basis. This may explain why further iodine studies are seen as unnecessary by the industry, while in practice the industry is pursuing many relaxations in the current operating practices on the basis that they do not significantly increase the risk related to design basis accidents, which thus include also core melt accidents without pressure vessel rupture. Consequently, further studies supporting the evaluation of these and similar applications are needed.

An example of such studies was given by Mr. R. Hamazaki, who presented results from both experimental and analytical studies concerning volatile iodine formation during a core melt accident in a Japanese BWR.

In the calculation presented by Mr. Hamazaki the containment water pH was assumed to be affected by nitric acid formation alone, and therefore the reduction in water pH during the sequence was only moderate from 7 to 6. As suppression pool water in BWRs is not buffered, any additional release of acids can lead to a more sudden and deep reduction. Such a reduction may result from cable insulation pyrolysis releasing large quantities of hydrochloric acid.

The release of hydrochloric acid with its potential consequences to iodine retention in containment and scrubber filter system was described in detail in presentation by Mr. H. Sjövall. Should the suppression pool water become acidic, not only iodine but also chlorine is vaporized. As chlorine may then be carried out to the scrubber filter, where it competes with

iodine in the reactions involving also sodiumthiosulfate, the release of chlorine can lead to degradation of filter iodine retention capability. In order to prevent this the thiosulfate concentration in the filter has been increased to compensate all possible chlorine releases. Modifications are foreseen also in the containment water pH control system, of which capacity will be increased to compensate hydrochloric acid releases. Also possibilities to enhance organic iodide retention in the scrubber filter by suitable additives will be studied. In the discussion that followed it was pointed out by Prof. J. Liljenzin that water pH affects also the formation of precipitates due to containment material oxidation, which may reduce the efficiency of decay heat removal systems in the long run.

The effect of hydrochloric acid on containment sump water pH was also studied by Mr. T. Routamo. However, this effect was shown to be of minor importance at Loviisa VVER due to borax stored in the ice in the ice condensers, which provide efficient buffering capacity during an accident.

The general consensus among the Workshop participants was that a significant reduction in iodine releases can be obtained by controlling containment water pH and by reducing, collecting and filtering the containment leakage. In designs where the containment can be depressurized without venting, these means appear to be efficient enough so that no major modifications in plant systems and current operation practices are foreseen. However, further studies may be warranted, should the current release limits or operation practices be modified. For the designs that apply filtered venting as a part of their severe accident management strategy the situation is less clear, since there the containment water pH has been found to be sensitive to chemicals, such as chlorine, that can be released during an accident. The same chemicals may also affect directly the iodine reactions either in the containment or in a scrubber filter enhancing iodine volatilization. Some modifications have already been made to accommodate these effects, and possibilities to enhance iodine management will be sought also in the future.

Session VI: Session Chairmen Summaries and General Discussion (Prepared by Dr. Harri Tuomisto, Fortum Engineering Ltd, Finland)

The Session Chairmen reported the conclusions of their sessions as presented in the previous sections. There was an extensive discussion on these reports.

The Chairman of the Workshop emphasized the need for broad considerations of various aspects concerning the role of iodine in the SAM. Prevention and mitigation of fission product releases and related environmental and health consequences is the final goal of SAM once the core degradation has started. The objective of the Workshop is to discuss, whether additional measures with respect to the iodine should be taken. All such considerations should take into consideration the overall plant-specific approach to SAM and iodine aspects should not be treated in isolation from the other SAM measures. When national criteria exist for the allowable Severe Accident releases of fission products, the iodine releases are mainly relevant for evaluation of acute health effects and evacuation, sheltering and stable iodine distribution needs. It is obvious that there are large differences among various plant designs and the considerations have to be very plant specific.

For discussion it is useful to separate the role of iodine to the operator level (SAM guidelines and procedures), the emergency organization level (avoiding or timing of early evacuations) and plant modification level.

Allocation of resources on the iodine R&D should be compared with respect to the risk reduction potential of other relevant SAM research. Finally, remaining uncertainties are to be managed by applying sufficiently robust decisions and measures.

Dr. Jay Lee of NRC preferred to focus on iodine research for design basis accidents instead of looking at measures for Management of Severe Accidents. Furthermore, it is to be expected that the regulatory bodies will need capabilities to evaluate safety cases that the utilities may present already in the near future. Such safety cases would aim at relaxation of such overly strict requirements that may cause an unnecessary cost burden in the deregulated electricity market. He added that the plant life management may bring respective cases e.g. for allowing higher than existing leakage rates and increased timing for containment isolation.