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SHORT OVERVIEW ON THE DEFINITIONS AND SIGNIFICANCE OF THE LATE PHASE FISSION PRODUCT AEROSOL/VAPOUR SOURCE

by an NEA Group of Experts

September 1994

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O E C D

The Convention establishing the Organisation for Economic Co-operation and Development (OECD) was signed on 14th December 1960.

Pursuant to Article 1 of the Convention, the 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 current Signatories of the Convention are Australia, Austria, Belgium, Canada, Denmark, Finland, France, the Federal Republic of Germany, Greece, Iceland, Ireland, Italy, Japan, Luxembourg, Mexico, the Netherlands, New Zealand, Norway, Portugal, Spain, Sweden, Switzerland, Turkey, the United Kingdom and the United States. The Commission of the European Communities takes part in the work of the OECD (Article 13 of the OECD Convention).

N E A

The OECD Nuclear Energy Agency (NEA) was established on 1st February 1958 under the name of the OEEC European Nuclear Energy Agency. NEA membership today consists of all European Member countries of OECD as well as Australia, Canada, Japan, Republic of Korea, Mexico 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 among the governments of its participating countries in furthering the development of nuclear power as a safe, environmentally acceptable and economic energy source.

NEA works in close collaboration with the International Atomic Energy Agency (IAEA), 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 OECD 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.

FOREWORD

The attached paper has been prepared by J. Sugimoto, M. Kajimoto, K. Hashimoto and K. Soda on the basis of information made available and discussed by members of the Task Group on Fission Product Phenomena in the Containment (FPC) of CSNI's Principal Working Group on the Confinement of Accidental Radioactive Releases (PWG4). It has been endorsed by the FPC and PWG4, at meetings held in March and September 1994 respectively. The report has been approved by CSNI for publication as a CSNI report in November 1994.

A list of FPC members is given in an Appendix.

Short Overview on the Definition and Significance of the Late Phase Fission Product Aerosol/Vapor Source

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1. INTRODUCTION

1.1 Background

Fission product once deposited on the surface structure or dissolved in the water in a containment may be resuspended and/or revaporized and/or reentrained during the course of a severe accident. These phenomena are supposed to take place in several situations such as rapid pressure decrease due to the containment failure or containment venting, or hydrogen combustion in containment. The mechanism of resuspension, revaporization or reentrainment is either hydraulic, thermal, chemical or both. In some of the early work by the nuclear industry revaporization issues were identified. Those are work by IDCOR on the Mark I which showed revaporization continuing for some 50 hours after accident initiation[1], work by Stone and Webster employees on the Mark II[2], and work sponsored by the New York Power Authority which involved modification of the STCP to include revaporization[3].

These phenomena, generally referred as "resuspension" would pose some safety concern on a long-term release of fission products to the environment.[4] This late phase fission product aerosol/vapor release was placed as one of unresolved issues in the analysis of severe accident phenomena, since this additional fission product source at or near the time of containment failure might reduce the benefits gained by delayed failure of the containment.[5] The present paper gives an overview on the definition and significance of the late phase aerosol/vapor fission product source in severe accident condition.

1.2 Definition

In describing these phenomena in which the fission product is reentrained by flashing of sump water due to depressurization, the term "resuspension" is frequently

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used, but "reentrainment" is also used by some authors. The revaporization and revolatilization are similar phenomena in which FP aerosols are suspended by different mechanism. Resuspension sometimes represents all of four phenomena discussed in the present paper. Therefore in order to avoid confusion among experts, the terms "resuspension", "revaporization", "reentrainment", and "revolatilization" should be clearly distinguished with each other.

In the present paper, the followings are the proposed definition of these terms as illustrated in Figure 1:

(1) Resuspension

Deposited aerosol or condensed materials on the structure surface is suspended as fine fragments due to drag force by fluid flow. (It may be noted that the resuspension can be initiated by the steam spikes from the water injection on relocated melt or the flow due to hydrogen burns.)

(2) Revaporization

Chemical compound in the deposited aerosol or condensed materials on the structure surface is vaporized when the vapor pressure at the surface is larger than its partial pressure in the gas due to, for example, FP decay heating or the heat by chemical reactions. (It may be noted that revaporization can be initiated by a change of chemical conditions such as occurs when air from the containment enters the reactor coolant system[6].)

(3) Reentrainment

Chemical compound once dissolved in the liquid or deposited on walls is entrained as droplets by the boiling of liquid or steam flashing due to, for example, depressurization.

(4) Revolatilization

Dissolved materials in the pool water is evaporated due to the conversion to volatile form by chemical reactions in particular under the radiation field.

In the present paper, "resuspension", "revaporization", "reentrainment", and "revolatilization" issues are discussed and the need for further research is addressed from the viewpoint of reactor safety.

2. EXPERIMENTAL AND ANALYTICAL EFFORTS

2.1 Experiments

(1) Resuspension

The fundamental work on resuspension of deposited aerosols was performed by Corn in 1960s.[7] The more empirical work on resuspension of plutonium was performed using cascade impacters in a field experiments.[8] Aerosol resuspension tests have been conducted at ORNL in the efforts to develop an analytical model in TRAP-MELT code for the reactor coolant system or reactor secondary containment for resuspension of deposited aerosols.[9] The tests were performed with a test section of a 1.83 m-long, 0.076 m-diameter Pyrex pipe and a 1.83 m-long, 0.051 m-wide deposition surface. The test results showed that the resuspension rates were influenced not only by the flow velocities, but also by particle material, size, density, and by the amount of material deposited on the surface. It was noted that aging of the deposited materials also affected resuspension.

Resuspension of the dried aerosol particles from surfaces was investigated in an experimental program, PARESS, and a model was developed for the phenomena at the Program Light Water Reactor Safety in Paul Scherrer Institute.[10] The investigation indicated that the amount of resuspension and the area affected are strong functions of the flow velocities on the surfaces and through the containment break, and break area. For small openings (up to 10 cm diameter) the amount of resuspended mass is believed to be insignificant. For large openings (up to 3 m diameter) occurrence and the amount depend on the flow velocities in the containment as well as through the opening area. It was shown that there could be continuing resuspension of surface deposits.

The deposition and translocation of deposited wet CsOH aerosol on the tube surface were investigated in Finland.[11] Two horizontal straight tubes of 0.56 cm and 1.80 cm in diameter and a 0.56 cm diameter tube with 90° bend were used in the experiments. In some experiments, a novel XRF-method was applied to avoid interfering the deposition process of CsOH in pipes. They found that the deposition in the bend tube increased the deposition rate after the bend about twofold as compared with a straight tube. They also compared the experimental results with the theoretical predictions.

Experiments have also been conducted at AEA Technology in England[12] to follow the resuspension of the control rod Ag-In-Cd alloy aerosol from stainless steel under both laminar and turbulent flow. In this study, significant resuspension of aerosols were observed in the turbulent conditions. In addition, the results showed that processes of aerosol resuspension involved the removal of surface layer and the removal of a more adherent layer.

Recently the STORM project has been initiated to investigate the mechanical deposition and resuspension of soluble and insoluble aerosols in turbulent flow.[13] A large-scale experimental facility is being built at JRC Ispra. Currently the STORM project is dimensioned to allow the study of resuspension in straight pipes under prototypical flow conditions, with a maximum velocity of 50 m/s in the widest pipe (203 mm in diameter) and more than 200 m/s in the narrowest pipe (63 mm in diameter). The experiments will be eventually extended to a variety of geometries (bends, obstacles, steam generators).

(2) Revaporization

There is very few work on revaporization due to FP decay heating or chemical reactions. The down stream behavior of fission products has been investigated by injecting mixtures of CsOH, CsI and Te into a flowing steam/hydrogen stream at ANL.[14] It was found that the reactions within the gas phase produced compounds which controlled the vapor deposition, aerosol formation, and revaporization. Some experimental works have been performed at AEA Technology for control rod materials such as Ag, In and Cd from stainless steel coupons using thermal gradient technology to determine the effective vapor pressure that control the revaporization.[12] Cesium revaporization from stainless steel coupons exposed to cesium hydroxide vapor has been investigated with Knudsen cell and mass spectrometry.[15] Revaporization of mixtures of simulant fission products and structural materials generated from Falcon experiments has been investigated with a combination of Knudsen cell mass spectrometry and thermal gradient techniques.[16,17]

These phenomena are also being addressed as part of the PHEBUS-FP programme and are seen as an important part of the CEC's Reinforced Concerted Action on Source Term involving both experiments and modeling studies on revaporization.

Recently a large-scale experimental program has been initiated to investigate the mechanical deposition and revaporization in the WIND project at Japan Atomic Energy Research Institute (JAERI). Currently the analytical study has started on the FP aerosol behaviors in primary pipings and on the thermal effect on piping wall due to decay heating of the deposited FP aerosols. Test facility is under design stage and the revaporization behavior will be investigated at up to 1,400° C. It is noted that the particle size of resuspended or revaporized material is to be precisely measured.

(3) Reentrainment

Rapid boiling of water in the sump is regarded as a mechanism for reentrainment. There is very limited work on reentrainment by the depressurization. The pool boiling reentrainment from the failed containment was simulated in the LACE program using 850 m^3 test vessel and a 3-m deep water tank by a rapid depressurization of the test vessel. A mixed aerosol of CsOH and MnO was introduced to the containment test vessel, and the reentrainment of aerosol material from the flashing pool was determined by measuring the aerosol concentration of Li_2SO_4 added to the pool. The experimental results showed that an amount of reentrainment aerosol was insignificant.[18] The depressurization rate in the LACE experiment may be too small compared with the situation of the catastrophic containment failure.[5] On the contrary, however, it is still far large compared with the situation of slow containment failure due to leakage.[19] The depressurization rate strongly depends on the containment failure mode.

Experimental works in the REST project were conducted at KfK to examine the reentrainment of particles from the boiling sump as a potential long-term source of radioactivity with 3.7m^3 vessel.[20] The experimental results showed that the reentrainment rates were in the expected range but a better understanding of their dependence on the influence parameters would be required.

As a part of ALPHA program, reentrainment of aerosol from flashing pool due to containment failure has been investigated at JAERI.[21] The model containment is 50 m^3 in volume and the maximum operable pressure is 2 MPa. Currently the first scoping experiment has been conducted. The effect of pressure decreasing rate on the reentrainment rate is mainly focused and the analytical model is to be assessed.

(4) Revolatilization

The study on the revolatilization of iodine in the containment has been conducted at ORNL.[22] In this study, iodine behavior in containment was evaluated during early stage of an accident (up to 1200 min.). According to this results, the revolatilization of iodine is strongly dependent on pH of water solution. If the pH drops below 7, a large fraction of I^- will be converted to I_2 and be released into the containment atmosphere. The overall iodine behavior is also being addressed in PHEBUS-FP programme.

Experimental works have been also conducted at UK, CEA, AECL and JAERI to examine the effects of radiolysis and hydrolysis of iodine in liquid. From these works it was concluded that much of the homogeneous solution chemistry of iodine was understood, but the key uncertainty area is the role of organic solutes on the pH behavior, organic iodine production and inorganic iodine behavior in a radiation field.[23]

2.2 Analysis

Analytical efforts have been devoted to describe the resuspension and revaporization behaviors.[24,25,26,27,28,29] Most analytical results including NUREG-1150 indicated that the revaporization from sump water during flashing at the time of containment failure would not significantly increase fission product release from the failed containment.[30] Modeling effort was also made by SRD of UK on a long term release of fission products from a failed containment. A theoretical work for the aerosol resuspension have developed at CEGB, Berkeley Nuclear Labs. based on consideration of particle binding energy at structure surfaces.[31] The effect of chemical form on cesium revaporization behavior has been analytically investigated at SNL.[32] The report by CSNI FPC describes the current status on this issue.[30]

The THALES-2 code, which has been developed at JAERI to analyze the thermal-hydraulics and radioactive nuclide behavior under the severe accident conditions, incorporates a revaporization model. Analysis of source terms for BWRs with THALES-2 indicated that the retention of FPs in the reactor coolant system would be strongly affected by the revaporization.[33] Most of all CsI deposited in the RCS was revaporized and transported to the containment while the effect of revaporization of CsOH was negligible. It also showed that the effect of revaporization on the source terms depend on the timing of the containment failure and the temperature of reactor coolant system at the time of containment failure. Analysis of source term for LWR severe accident sequences (station blackout for BWR and PWR and small LOCA for PWR) have been performed by ENEL and JRC [29] with ECART code [34] which incorporates a resuspension model. The results show that after the core slump into the vessel lower head and the following water flashing, high steam and gas velocities can cause the resuspension of a significant part of aerosols previously deposited in the primary cooling system. The amount of aerosol released to the containment can be largely underestimated by the codes if the resuspension is ignored. In the new version of VICTORIA code, resuspension and revaporization of aerosol species are modeled along with detailed chemical model including interaction with structure wall materials.[35]

In a comparative study of source terms of a BWR Mark-I with THALES-2, STCP [36] and MELCOR [37] codes, it was found that models of revaporization have a significant effect on the source term evaluation.[38] Figure 2 shows a typical example of the calculated results of mass distribution for CsI by three codes at 1,000 minutes after the accident initiation. The release fraction of CsI to the environment was 8 %, 0.006 % and 1 % of core inventory in THALES-2, STCP and MELCOR calculations, respectively. In THALES-2 and MELCOR calculations, the amount of CsI released to the environment was increased due to the revaporization of deposited CsI in the RCS at the containment failure. On the other hand, the STCP predicted little amount of CsI source term since the revaporization in the RCS after the vessel failure is not modeled in the STCP. It is also noted that the database of saturation vapor pressures of chemical compounds with high accuracy at high temperature (over 1,000 K) is not enough along with the problem of the true speciation when FP reacts with the substrate to describe the revaporization behavior in analytical codes.

In the sensitivity study on source terms at JAERI, the reentrainment of droplets in the containment during the suppression pool flashing following a containment failure at the BWR plant was examined with THALES/ART [39,40] and REMOVAL codes [41]. The results indicate that a large amount of entrainment occurs at the near surface region.[42] The liquid mass released from the containment as droplet was 0.069, 0.0025 and 0.00043 of suppression pool inventory for the break area of 10, 2, and 0.4 m^2, respectively. Thus it was found that the break area has a large effect on resuspension rate.

In the analysis of revolatilization, there is further need for the development/validation of existing models against realistic integral tests. Dealing with surface reactions was identified as one of the most significant uncertainties in the modeling.[23]

It is noted that the existing experiments and analyses indicate resuspension can occur, but they do not precisely tell us the particle size of resuspended material.[9,35,43] It may be that resuspended materials are so large that they promptly redeposit without adding to the consequences of an accident. This seems especially likely if the deposits have had time to sinter or react.

3. CONCLUSIONS AND RECOMMENDATIONS

The issue on the late phase fission product aerosol/vapor source has been reviewed based on the current understandings of the phenomena and the developed analytical models. The following conclusions and recommendations are obtained from the present overview as issues to be investigated and resolved for the future research as summarized in Table 1:

(1) Resuspension

Resuspension is referred to the fragmentation of once deposited fission products on the structure surface due to the drag force by fluid, possibly from steam spikes when injected water contacts with relocated melt, or from hydrogen burns. Experimental information on resuspension behavior in the pipings should now be concentrated on the behavior of representative multi-layered deposits under flow conditions expected during severe accidents such at bends, at positions with high thermal gradients, and at places of discontinuities in the gas flow. Most investigations of mechanical resuspension have considered gradual increases in steady flow rather than abrupt, transient, highly turbulent flows associated with sudden depressurization. Shock waves and vibrations of deposition surfaces occurring along with changes in flow velocity have been almost neglected in research to date. Future models should incorporate the major physico-chemical parameters involved for such mechanism.

(2) Revaporization

Revaporization is referred to the vaporization of once deposited aerosols on the RCS or containment wall due to FP decay heating, chemical heat, or change of chemical condition. Experimental information on revaporization behaviors is not enough and the reasonable analytical modeling have not been developed and assessed. It is also noted that the database of saturation pressure at high temperature is not enough along with the problem of the true speciation when FP reacts with the substrate. Therefore more experimental and analytical study on revaporization is needed.

(3) Reentrainment

Reentrainment is referred to the resuspension of aerosols once dissolved in the sump water by rapid boiling of water or dump deposits by steam flashing due to depressurization at the time of containment failure or containment venting. Currently the experimental information on reentrainment is limited. Also the proposed analytical models on this reentrainment has not been satisfactorily assessed for the typical condition of severe accident. Therefore more experimental and analytical study on reentrainment is still to be considered.

(4) Revolatilization

Revolatilization is referred to the evaporation of dissolved iodine in the pool of water in containment due to the conversion to volatile iodine more rapidly under the radiation field. Experimental and analytical study on revolatilization behavior has mostly been completed but still some uncertainties remain such as the role of organic solutes on the pH dependence or the surface reactions. The effects of revolatilization on source terms should be examined along with the possible severe accident sequences.

It is noted that the effectiveness of mitigative measures to reduce the effect of resuspension/ revaporization/reentrainment/ revolatilization on source terms should be investigated from a viewpoint of accident management. The experimental and analytical findings obtained from the study on these issues will be utilized for this purpose.

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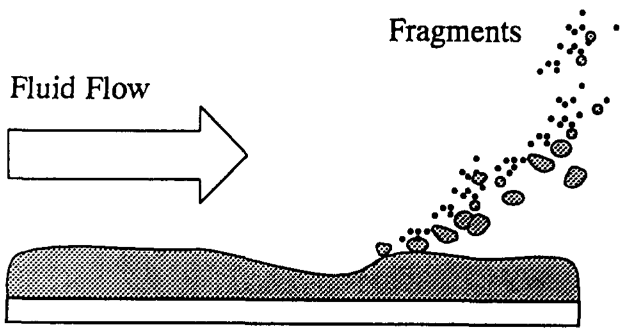
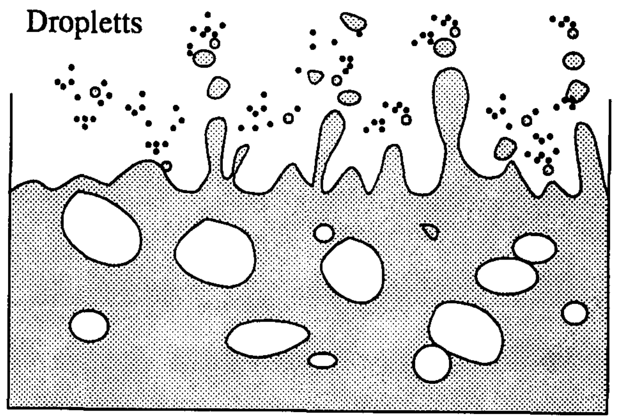
Table 1 Summary of Late Phase Fission Product Aerosol/Vapor Source

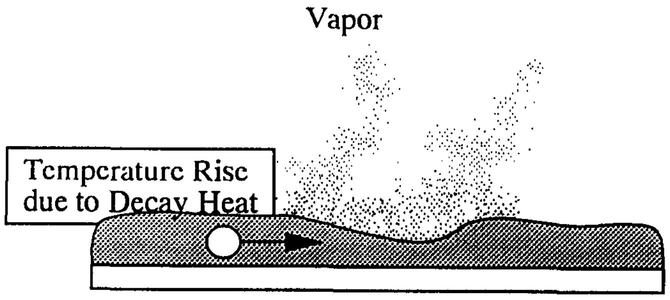
Mechanism	Major findings obtained	Future works needed
Resuspension	• Fundamental mechanism and parametric effects (flow rate , particle material size, bread area, flow conditions etc.) • Possible significant effect on calculated source terms	• Experiment with representative multi-layered and complicated deposit conditions • Physco-chemical and sophisticated model development
Revaporization	• Very few work performed on this issue • Possible significant effect on calculated source terms	• More detailed experiment on the phenomena • Reasonable analytical model development • Database of saturation vapor pressures of chemical compounds at high temperature (over 1,000K)
Reentrainment	• Reentrainment strongly affected by depressurization rate • Possible significant effect on calculated source terms	• More detailed experiment on the phenomena • Reasonable analytical model development
Revolatilization	• Strong dependence on pH of water solution • Homogeneous solution chemistry of iodine well understood	• Reduce uncertainties in experiments and analytical models(organic solutes on pH dependence, surface reactions)



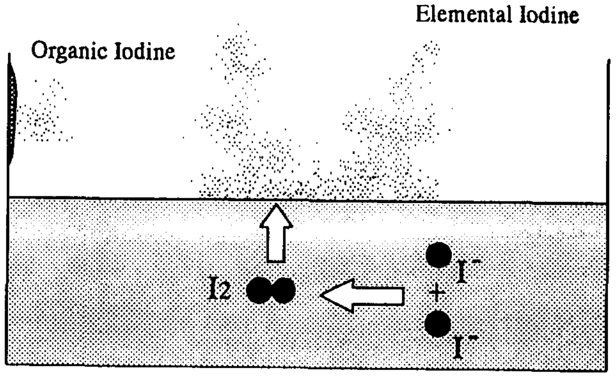
(a) Resuspention



(c) Reentrainment



(b) Revaporization



(d) Revolatilization

Figure 1. Transport Behavior by Resuspention, Revaporization, Reentrainment and Revolatilization

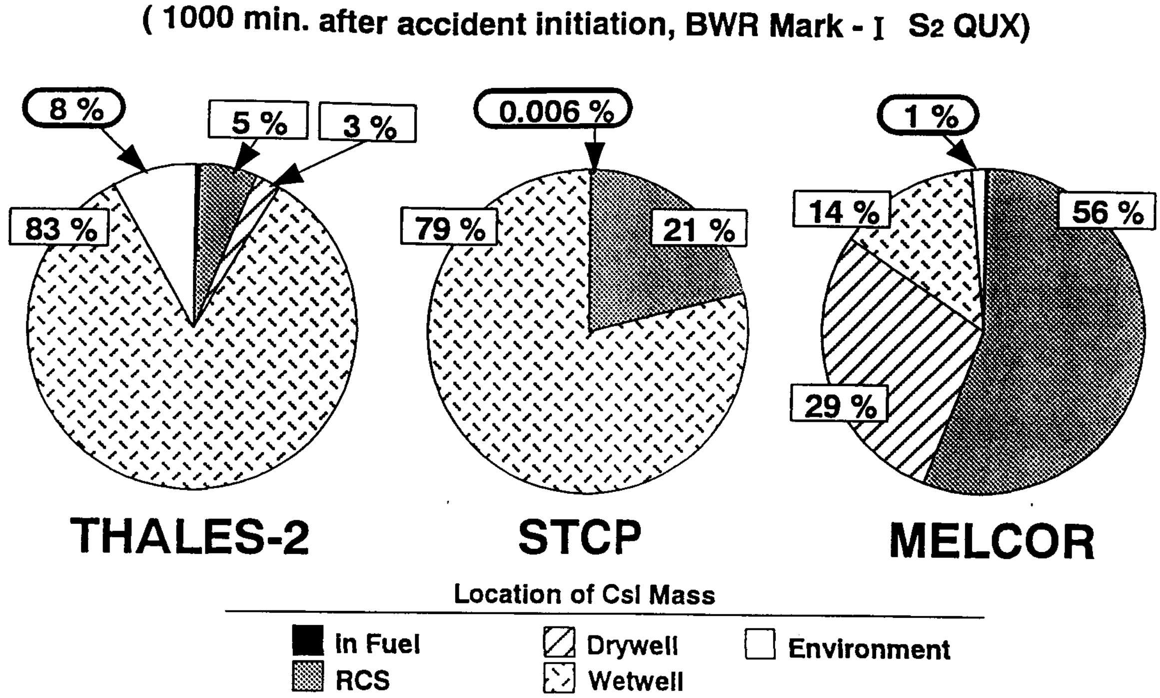


Figure 2. Comparison of the calculated CsI Distribution by the THALES-2, STCP and MELCOR *

* Based on a calculation performed at JAERI[38] with the MELCOR version available at that time, just to show the significance of revaporization model as an example. Recent MELCOR version may predict about 4 % rather than 1 %.

APPENDIX

List of Members of PWG4's Task Group on Fission Product Phenomena in the Containment (FPC)

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