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Journal of Nuclear Materials 248 (1997) 226–232

journal of nuclear materials

Experimental analyses of iodine behavior under severe accident conditions with ART

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Abstract

The fission products (FP) behavior analysis code ART developed at JAERI simulates a FP transport and deposition in a reactor coolant system and containment during severe accidents. As part of the code development and verification, several experimental analyses have been conducted. In the JAERI's WAVE experiment, the effect of nitrogen or steam carrier gases on the cesium iodide (CsI) behavior in piping was recently investigated. The ART analysis for nitrogen agreed with the experimental results by reflecting the detailed thermo-fluiddynamic calculation on the CsI aerosol behavior analysis. On the contrary, the analysis for steam did not agree well with the experimental results because observed enhancement of aerosol growth cannot be explained by existing models. Moreover, the newly developed empirical models on iodine chemistry in water were examined for the ACE/RTF 3B experiment. The analysis showed that those models have a fundamental analytical capability. © 1997 Elsevier Science B.V.

1. Introduction

It is recognized in the source term analyses of light water reactors (LWRs) that large uncertainties still remain in the fission products (FP) aerosol behavior in reactor coolant systems during severe accidents [1]. In order to reduce the uncertainties in the analyses, the complete integration of thermohydraulics and FP behavior was pointed out as one of the solutions [2]. In the present study, the complete integration method was evaluated and the other parameters which affect the source term calculation were also investigated from the viewpoint of the iodine chemistry.

The WAVE (Wide range Aerosol model VErification) experiment has been performed at the Japan Atomic Energy Research Institute (JAERI) to investigate the FP behavior in piping under severe accident conditions [2]. In the experiments, the effect of carrier gas, nitrogen and steam on the CsI behavior was investigated. Analyses of the experiments have been conducted with the three-dimensional fluiddynamic code SPRAC [3] and the FP behavior analysis code ART (Analysis of Radionuclide Transport) [4,5].

The empirical models on iodine chemistry in sump water which was recently incorporated into ART were examined against the Radionuclide Test Facility (RTF) 3B experiment [6] in the ACE (Advanced Containment Experiment) Phase-B effort.

2. Outlines of computer codes

The SPRAC code has been developed by JAERI and the Kawasaki Heavy Industries, to analyze the air ingress accident of the High Temperature engineering Test Reactor (HTTR) at JAERI [3]. The code can treat incompressible flows with complex geometries using a boundary-fitted

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PII S0022-3115(97)00207-9

coordinate (BFC) system. The SPRAC code used in the present study is suitable for analyzing the present WAVE thermohydraulic conditions.

The ART [4,5] code has been developed at JAERI for the analysis of FP behavior in reactor coolant systems and in containment under severe accident conditions. The radionuclide behavior modeled in ART is shown in Fig. 1.

The ART code can model the systems by an arbitrary number of volumes and solves the governing equations for multi-component aerosol and gaseous radionuclides. The 'sectional method' developed by Gelbard [7] is used to describe the aerosol size distribution. The phase change of chemical species is considered in the ART code while the chemical reactions among FP elements in the gas phase is not modeled. The ART code is a module of the JAERI's source term analysis THALES-2 [8] code and has a fast running capability for probabilistic safety assessment (PSA).

3. Wave experiments

The test section shown in Fig. 2 mainly consists of a dish containing CsI powder, electrical heaters for CsI evaporation, a straight pipe and three stainless steel (SUS304) coupons located at the floor, wall and ceiling of the pipe. The conditions for thermohydraulics and CsI aerosol generation were defined based on those of typical severe accidents for the PWR hot-leg inlet (see Table 1). Two experiments were performed using the carrier gases of nitrogen and steam.

A dish containing CsI powder was set in the test pipe before the experiment initiation. In the post-test measurement, the iodine and cesium deposited onto coupons were measured by the organic solvent with a spectro photometer and the liquid ion chromatography method, respectively.

3.1. Nitrogen carrier gas

The measured CsI deposition mass onto the floor, wall and ceiling coupons are shown in Fig. 3. A relatively large amount of deposition was found at the upstream floor of the pipe. The appearances of deposition onto the coupons at downstream is shown in Plate 1. A larger amount of deposition was found at the ceiling than downstream at the floor. The electron probe microanalyzer (EPMA) analysis for the deposited coupons showed that the cesium distribution was almost the same as that of iodine. This supports that most of the cesium and iodine might have been transported and deposited in the chemical form of CsI after the vaporization.

The aerosol size distribution measured by a cascade impactor agrees most with the lognormal distribution (see Fig. 4). The mass median aerodynamic diameter (MMAD) and mass median diameter (MMD) were 1.2 and 0.5 μm, respectively.

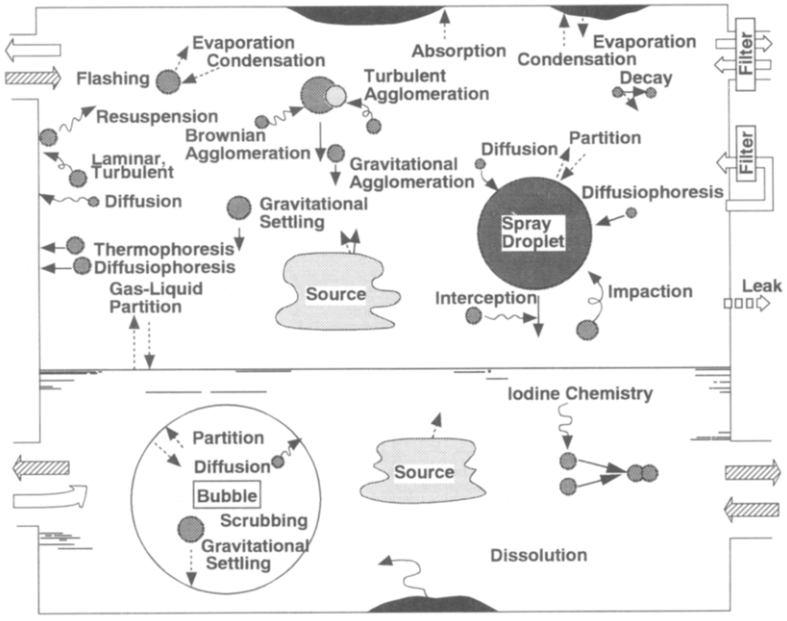

Fig. 1. FP transport and deposition models in ART.

Table 1
Conditions for thermohydraulics and CsI aerosol generation

	Nitrogen	Steam
Injection rate	3.3×10^{-4} m^3/s at 293 K	2.3×10^{-4} m^3/s at 293 K
Pipe wall temperature	1073–400 K	1073–400 K
Inlet gas temperature	960 K	914 K
Flow velocity at inlet	1.05 m/s	0.61 m/s
Test duration	60 min	60 min
Mass of vaporized CsI	2.59 g	1.66 g

3.2. Steam carrier gas

In the experiment with steam carrier gas, the carrier gas was changed to nitrogen gas after the experiment to avoid the condensation of steam onto the coupons. The tendency of deposition distribution in steam was mostly the same as that in nitrogen. However, there is a difference in deposition at the pipe downstream. In steam, the deposition mass onto the floor became larger than that onto the ceiling. The appearances of deposited CsI onto downstream coupons is shown in Plate 2. The diameter of the deposited aerosol in steam is clearly larger than that in nitrogen (see Plate 1) [9].

The aerosol size distribution in steam did not follow the lognormal distribution (see Fig. 4). The obtained mass median aerodynamic and mass median diameters were 6.0 and 2.5 μm, respectively. The observed aerosol size distribution showed two peaks (at 0.7 and 15 μm) in steam while one peak at 1.5 μm in nitrogen. This suggests that steam (water) plays an important role in the aerosol growth. The EPMA analysis showed that the element ratios of oxygen is different between large and small aerosol sizes (see Table 2). This indicates that the atomic ratio of H_2O in aerosol could affect the aerosol growth.

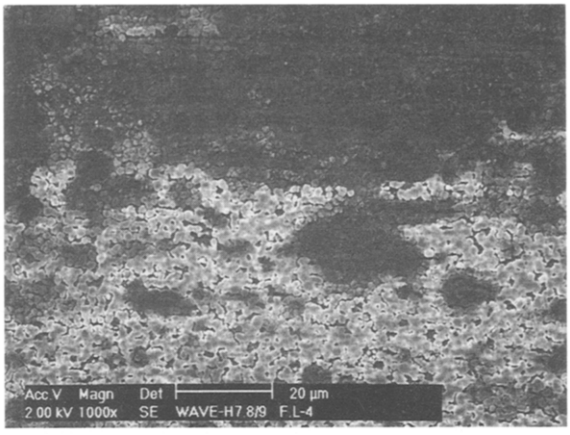

Plate 1. Deposited CsI onto SUS304 coupons at pipe upstream (nitrogen).

4. Analytical results

4.1. Nitrogen carrier gas

The thermohydraulic analysis with SPRAC was first performed for preparing a more realistic boundary condition for the ART analysis to evaluate a specific ART model. In the ART analysis [2], the pipe cross-section was subdivided into five sections based on the SPRAC noding.

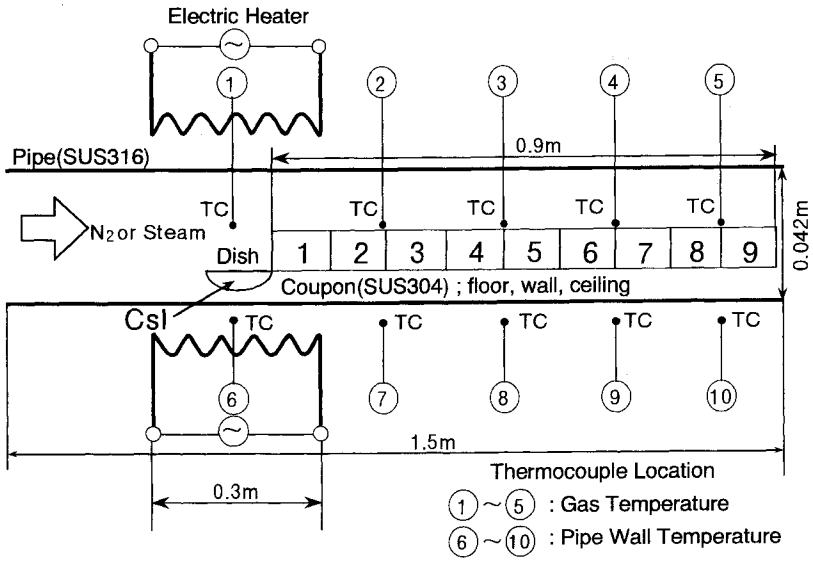

Fig. 2. Schematic of WAVE test section.

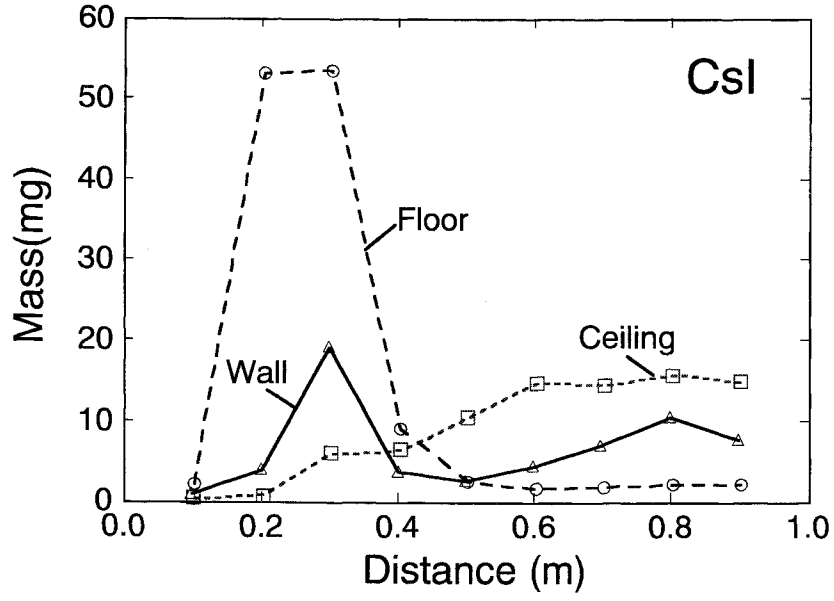

Fig. 3. Deposited CsI mass onto coupons (nitrogen).

The detailed gas flow and temperature distribution calculated by SPRAC were averaged in each section and used in the ART analysis as the input so that the SPRAC results could be well reflected.

The ART results with five sections are in reasonable agreement with the experiment within a factor of two and are a better prediction than those of the previous method with one node pipe cross-section. The present analysis showed that the major deposition mechanism for a CsI aerosol in nitrogen is thermophoresis which depends on the thermal gradient in gas. Therefore, the coupling of FP behavior and the detailed fluiddynamic analyses was found

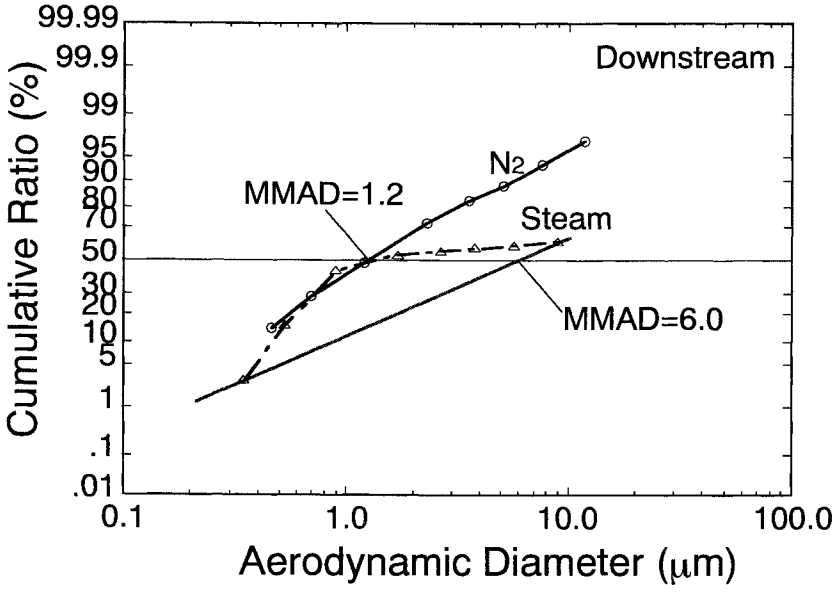

Fig. 4. Measured aerosol size distribution.

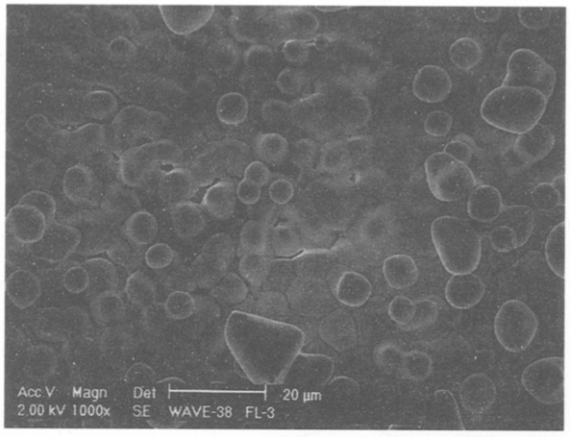

Plate 2. Deposited CsI onto SUS304 coupons at pipe upstream (steam).

to be essential to accurately predict the CsI deposition in piping, to which little attention has been paid in the previous studies [2].

4.2. Steam carrier gas

In the case of steam, the thermohydraulics in piping calculated by SPRAC was almost the same as that in nitrogen. However, the present ART analysis did not reproduce the CsI deposition distribution in the pipe because the analysis could not predict the observed enhancement of aerosol growth in steam. That is, the existing aerosol growth models such as agglomeration or condensation at the aerosol surface cannot explain this phenomenon.

Table 2
EPMA analysis for different aerosol size in steam

	Element ratio in aerosol	
	iodine/Cs	$O(H_2O)/Cs$
Small aerosol (0.53–0.9 μm)	1	~0.5
Large aerosol (>15 μm)	1	~1

In the ART modeling, a part of the FP gas which exceeds the saturation vapor pressure is assumed to become an aerosol due to homogeneous nucleation and the generated aerosol is assigned to the minimum aerosol mass class of which the default value is 10^{-18} kg. In order to simulate the observed aerosol size distribution with existing models in ART, a sensitivity analysis was performed by changing the minimum aerosol mass class from 10^{-18} to 10^{-12} kg. The analytical results shown in Fig. 5 indicated that the minimum aerosol mass class of 10^{-13} kg well reproduced the observed tendency, that is, larger deposition mass at the floor than that at the ceiling. The reason for the better prediction is that the deposition due to the gravitational settling of which the deposition velocity increases in proportion to the aerosol mass became as large as due to thermophoresis.

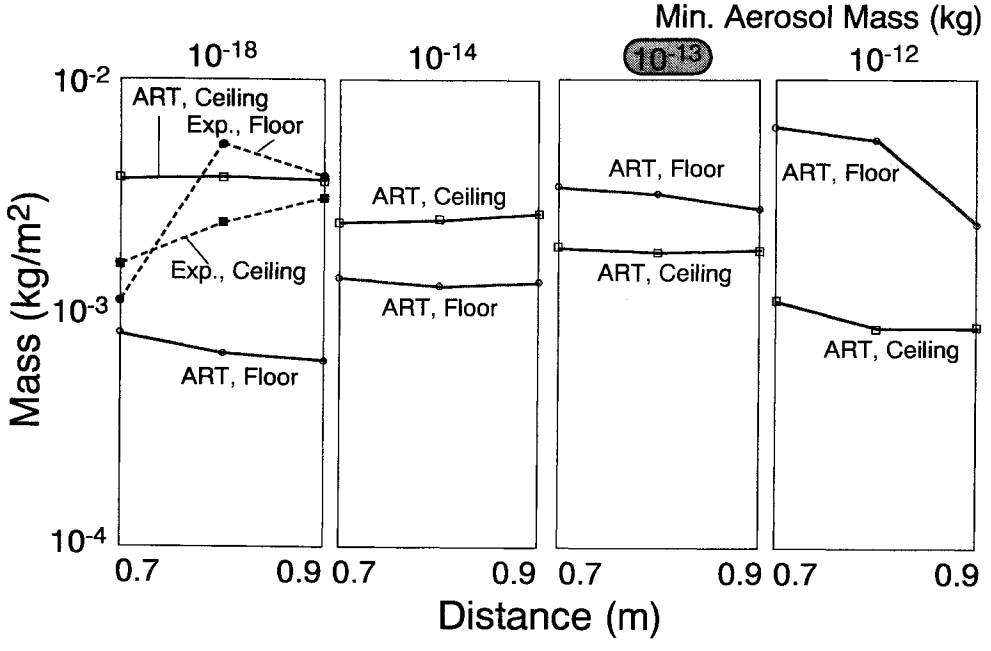

Fig. 5. Effect of minimum aerosol mass on deposition calculation (steam).

4.3. Discussions

The possible mechanisms for the observed enhancement of aerosol growth in steam are as follows. Although the Brownian, turbulent or gravitational agglomeration is one of the possible reasons, the enhancement cannot be explained only by one mechanism. There is a possibility of condensation of other chemical compounds, e.g., CsOH, at the aerosol surface, but this may be ignored because the dominant chemical form in the test section is considered to be CsI as discussed in Section 3. The hygroscopicity is also unlikely because the relative humidity was small enough due to the injection of superheated steam in the experiment.

There is a possibility that water molecules might work as an attractive force between the CsI aerosol particles and enhance the aerosol growth in superheated steam.

5. ACE/RTF 3B experiment

The RTF facility [6] consists of a cylindrical main vessel painted inside by epoxy painting. At the initiation of the experiment, CsI was injected into the water pool of 3.5×10^{-2} m^3 as a test solution. The average dose rate was approximately 2.0 kGy/h. The temperature of the test facility was set at 333 K and the pool pH was increased from 5.5 to 9.9 at 73 h after initiation of the experiment by injection of an LiOH solution into the pool.

The empirical iodine chemistry models were developed based on the IMPAIR-2 models [10]. The models incorporated into ART are shown in Table 3. For saving computation time, the differential equation on the concentration of each chemical form was integrated by the Euler method after a variable conversion. The interaction of iodine with the epoxy painting was ignored for simplification. In order to take into account the decrease in I_2 concentration due to that interaction, a slightly lower rate constant for radiolysis of I^- than the original IMPAIR-2 was assumed.

Table 3
Iodine chemistry models incorporated into ART

Radiolysis	$2I^- + h\nu \rightarrow I_2 + 2e^-$ [a]
	$IO_3^- + h\nu \rightarrow I^- + 1.5O_2$
Hydrolysis	$I_2 + H_2O \Leftrightarrow HOI + I^- + H^+$
Oxidation/reduction	$2I^- + 0.5O_2 + 2H^+ \Leftrightarrow I_2 + H_2O$
Disproportionation	$3HOI \Leftrightarrow IO_3^- + 2I^- + 3H^+$

[a] h: Planck constant; ν: frequency of γ-ray.

The measured and calculated I_2 concentrations in gas and liquid are shown in Fig. 6. The calculation predicted well the decrease of I_2 concentration in liquid and gas when the pH was changed. In the present model, the HOI concentration in liquid mainly increased due to hydrolysis in compensation for a decrease of the I_2 concentration when the pH was changed. However, the I_2 concentrations after the pH change were underpredicted by approximately one order of magnitude due to no modeling for organic iodine. The present analysis showed that the incorporated

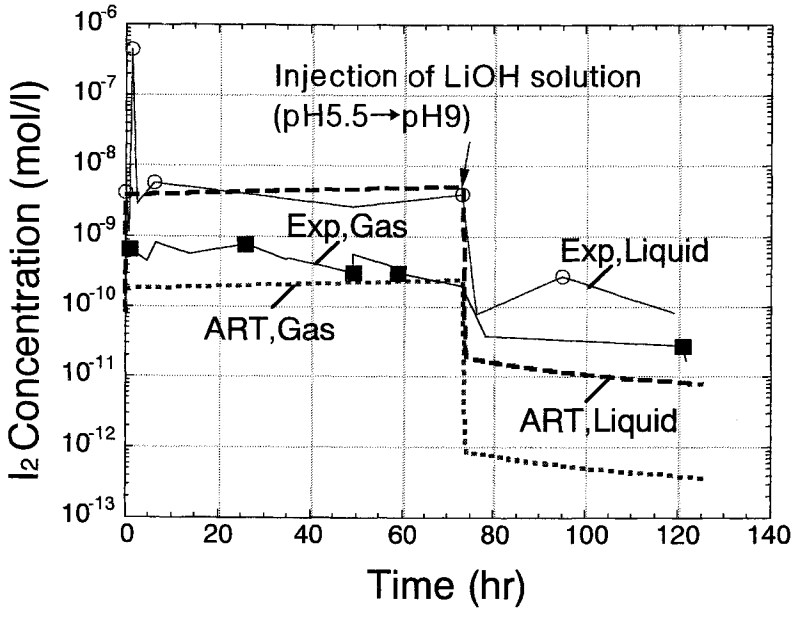

Fig. 6. I_2 concentration in liquid and gas in the ACE/RTF 3B experiment.

models have a fundamental analysis capability for the iodine chemistry.

6. Conclusions

The analytical capability of the ART code for CsI behavior in piping was assessed against a WAVE experiment. A clear difference was found in the aerosol size between nitrogen and steam carrier gas. For nitrogen, the analyses predicted well the measured deposition by reflecting the detailed thermo-fluiddynamic calculation on the CsI aerosol behavior analysis. For superheated steam, on the contrary, the experiment could not be well reproduced without the use of a larger aerosol size than the predicted. Since the observed enhancement of aerosol growth in superheated steam could not be explained by existing models, further investigation on the mechanism is needed.

The empirical models on iodine chemistry were examined against the ACE/RTF 3B experiment. The calculation well reproduced the decrease of the iodine concentration in liquid due mainly to hydrolysis at the time of pH change. It was confirmed that the present models have a fundamental analytical capability for the iodine chemistry.

The present studies showed that the close coupling between thermo-fluiddynamics and FP behavior analyses, the precise prediction of aerosol size distribution and the appropriate treatment of the chemical reaction are of great importance to accurately evaluate the source term.

Acknowledgements

Authors would like to express their sincere gratitude to Messrs. H. Ito, K. Komori and T. Aizawa of JAERI for their technical assistance and comments for the WAVE experiment. Special thanks are due to Dr M. Fumizawa of JAERI and Dr M. Akamatsu of Kawasaki Heavy Industries, Ltd. for their valuable advices on the application of the SPRAC code. The authors also thank Mr M. Naritomi of Nuclear Engineering Corporation and Mr N. Yamano of JAERI for their useful comments on CsI aerosol behavior.

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