Development and Verification of the Coupled AnCheBi&I-COSTA Framework for Iodine Chemistry and Aerosol Dynamics

Miran Park[a,b], Yoonhee Lee[a,*]
[a]*Dept. of Energy Engineering, Korea Institute of Energy Technology (KENTECH), 21 Kentech-gil, Naju-si, Jeonnam-Gwangu, 58330, Republic of Korea*
[b]*Dept. of Applied Plasma and Quantum Beam Engineering, Jeonbuk National University, 567 Baekje-daero, Deokjin-gu, Jeonju-si, Jeonbuk-do 54896, Republic of Korea*
**Corresponding author: yooney@kentech.ac.kr*

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1. Introduction

During a severe accident of a nuclear power plant, fission products undergo various physical and chemical behaviors which affect their transport and release to the environment. Recently, the interactions between aerosol and chemical behaviors have been highlighted through the OECD/NEA international research projects. In the STEM-2 project [1], it was observed that deposited iodine oxide (IOx) aerosols can be chemically decomposed and revaporized into the gas phase. In the ESTER project [2], more complex interactions were investigated, including the chemical decomposition of deposited cesium iodide (CsI), formation of volatile iodine species, and subsequent formation of IOx aerosols. From these experimental studies, it can be found that the fission products can be transferred between the gas and aerosol phases by chemical reactions. Therefore, it is necessary to consider both chemical behaviors and aerosol dynamics for the mechanistic analysis of fission product behaviors.

In order to establish an integrated analysis framework for the chemical and aerosol behaviors, AnCheBi (Analysis of Chemical Behaviors of Iodine) [3] and I-COSTA (In-Containment Source Term Analysis) [4] are coupled in this study. AnCheBi has been developed to analyze chemical reactions and the evolution of iodine species, while I-COSTA calculates aerosol dynamics based on a sectional approach. For the coupling of the two codes, the formation and growth of aerosols caused by the evolution of chemical species should be consistently transferred to the aerosol dynamics model. Therefore, conservation of iodine inventory on a molar basis between the gas and aerosol phases is important for the proper coupling of AnCheBi and I-COSTA.

In this paper, we verify the numerical implementation of the coupled AnCheBi & I-COSTA framework by examining the conservation of the iodine inventory during nucleation and condensational growth. In addition, sensitivity analyses on the model constants for nucleation and condensational growth are performed to investigate the response of the coupled model.

2. Coupling of AnCheBi & I-COSTA

2.1. Aerosol Formation and Growth Models

In the coupled AnCheBi–I-COSTA framework, nucleation and condensational growth are considered to describe the transfer of iodine oxide from the gas phase to the aerosol phase. The nucleation rate is calculated using an empirical equation based on the Reiss theory as follows:

$$J = k_{nucl} \cdot \left[\mathrm{IO_{x(g)}}\right]^{n}, \tag{1}$$

where J is the nucleation rate [mol/dm^{3}sec], k_{nucl} is nucleation constant [(dm^3)$^{n-1}$/sec/(mol)$^{n-1}$ and n is the exponent for the nucleation rate. The aerosol mass generated by nucleation rate is transferred to the first section in I-COSTA.

For the condensational growth, the growth rate formulated based on the approach used in VICTORIA [5] is adopted as follows :

$$G\left(r,\left[\mathrm{IO_{x(g)}}\right]\right) = 4 \cdot \pi \cdot r \cdot D_{\mathrm{IO_x}} \cdot \left[\mathrm{IO_{x(g)}}\right] \cdot F_r, \tag{2}$$

where r is the radius of an aerosol particle [m], D_{IO_x} is the coefficient used for the condensational growth of gaseous iodine oxide gas [/sec], F_r is the Fuchs-Sutugin correction factor. The concentration of gaseous iodine oxide calculated by AnCheBi in mol/dm^3 is converted to the number concentration [#/m^3] before calculating the condensational growth rate. The resulting G represents the condensational growth rate used in the sectional aerosol equations and has the unit of /sec.

The condensational growth in I-COSTA is calculated using a central difference scheme. Through the condensation, aerosol mass is transferred between adjacent sections, and the mass in each section is also increased by the condensation of iodine oxide.

For the coupling of the two codes, the decrease of gaseous iodine oxide in AnCheBi is calculated by considering both nucleation and condensation as follows:

$$\frac{d\left[\mathrm{IO_{x(g)}}\right]}{dt} = -\frac{1}{1000 \cdot N_A} \cdot \sum_{l=1}^{n_{bin}} \left\{ {}^{1}G_{l,k}\left(\left[\mathrm{IO_{x(g)}}\right]\right) \cdot \frac{\bar{Q}_l}{m_l} \right\} - J_{nucl}\left(\left[\mathrm{IO_{x(g)}}\right]\right), \tag{3}$$

where N_A is Avogadro's number, n_{bin} is the number of aerosol sections, $\bar{Q}_l$ is the total aerosol mass concentrations in section l and ml is the mass of a single aerosol particle represented by section l. Therefore,

$\bar{Q}/m_l$ represents the number concentration of aerosol particle in each section.

The corresponding increase of aerosol mass is calculated in I-COSTA. For the first section, the aerosol governing equation associated with nucleation and condensational growth is expressed as

$$\frac{dQ_{1,k}}{dt} = {}^{2}G_{l,i} \cdot Q_{l,k} + {}^{1}G_{l,k} \cdot \bar{Q}_l + J_{nucl}\left(\left[\mathrm{IO_{x(g)}}\right]\right) \cdot MW, \quad (4)$$

where (MW) is the molecular weight of the chemical species. For the other sections, the aerosol mass is transferred between adjacent sections by condensational growth. Therefore, the decrease of gaseous iodine oxide in AnCheBi is directly linked with the increase of aerosol mass in I-COSTA. This formulation is used to conserve the iodine mass during the coupled calculation.

2.2. Numerical Coupling of AnCheBi &I-COSTA

For the numerical coupling of AnCheBi and I-COSTA, the governing equations of the two codes are formulated in a monolithic form. The concentrations of chemical species in AnCheBi and the sectional aerosol mass concentrations in I-COSTA are treated as unknown variables in a single system of nonlinear equations. The governing equations are discretized using the implicit Euler method, and a single Jacobian matrix is constructed for the coupled system.

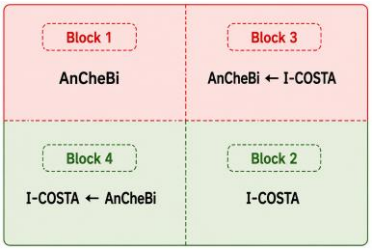


Fig. 1. Structure of the Jacobian matrix for the monolithic coupling of AnCheBi and I-COSTA.

Fig. 1 shows the structure of the Jacobian matrix for the coupled AnCheBi–I-COSTA framework. Blocks 1 and 2 represent the Jacobian matrices originally constructed for AnCheBi and I-COSTA, respectively. Blocks 3 and 4 are additionally constructed to consider the interactions between the two models. Block 3 represents the effects of aerosol mass concentrations in I-COSTA on the chemical species calculated by AnCheBi. In this section, the derivatives associated with the condensational loss of gaseous iodine oxide are considered. Block 4 represents the effects of chemical species in AnCheBi on the aerosol mass concentrations in I-COSTA, where the derivatives associated with nucleation and condensational growth are considered.

Through the additional coupling terms in Blocks 3 and 4, the changes in gaseous iodine species and aerosol mass described in Section 2.1 are simultaneously considered in the numerical solution. Therefore, the chemical behavior calculated by AnCheBi and the aerosol formation and growth calculated by I-COSTA are directly coupled within a single numerical system.

3. Numerical Results

3.1. Verification of the Coupled AnCheBi–I-COSTA Calculation

The coupled calculation is performed to verify the chemical and aerosol behaviors calculated by the integrated AnCheBi–I-COSTA framework. Fig. 1 shows the chemical and aerosol processes considered in the verification calculation. CsI aerosols deposited on the steel surface are given as an initial condition.

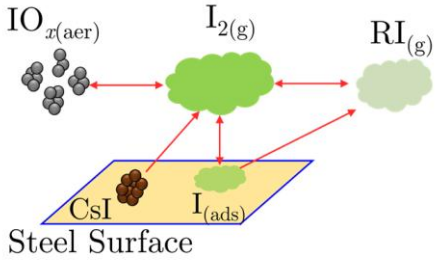


Fig. 2. Scenario for verification of coupled calculation

Under the radiation field, the deposited CsI is decomposed to form gaseous iodine species through the chemical reactions considered in AnCheBi. The gaseous iodine species subsequently form iodine oxide, which is transferred to the aerosol phase through nucleation and condensational growth calculated by I-COSTA.

The major numerical conditions used for the calculation are summarized in Table 1. The time step size is set to 1.0 s, and the aerosol size distribution in I-COSTA is discretized into 20 sections. The other numerical conditions and initial conditions used for the calculation are also given in Tables 1 & 2.

Table 1. Computation and initial conditions for verification

Parameter	Values
Time step [sec]	1.0
Simulation time [sec]	7,200
Range of radius for aerosol particles [μm] (min~max)	0.01~100
Number of sections for aerosol dynamics	20
Initial concentration of CsI (mol/dm^2)	1.86E-04
Surface area of steel [dm^2]	2.4E-05
Volume of the vessel [dm^3]	4.8E+00

Table 2. Coefficients in nucleation and condensational growth

Parameter	Values
k_{nucl} [/sec]	6.0E+07
n	2
D_{IO_x} [/sec]	5.0E-14

Fig. 3 shows the temporal evolution of the iodine inventories during the coupled calculation. The inventories of the iodine-containing chemical species and aerosols are converted to the equivalent mole of iodine atoms for comparison. The variations in the individual inventories show the chemical evolution of iodine from the initially deposited CsI to gaseous iodine species and subsequently to IO_x aerosols. Although the iodine inventory is redistributed among different chemical species and phases, the total iodine inventory remains nearly constant throughout the calculation. This result indicates that the iodine inventory is consistently conserved during the chemical reactions and aerosol formation processes calculated by the coupled AnCheBi&I-COSTA framework. The maximum relative deviation of the total iodine inventory from its initial value is approximately 10^{-4}.

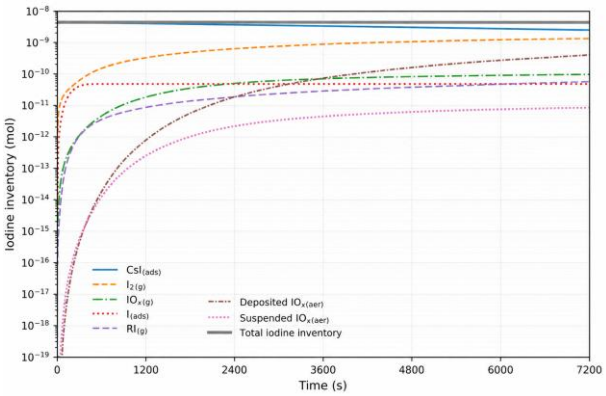


Fig. 3. Temporal evolution of iodine inventories during the coupled calculation

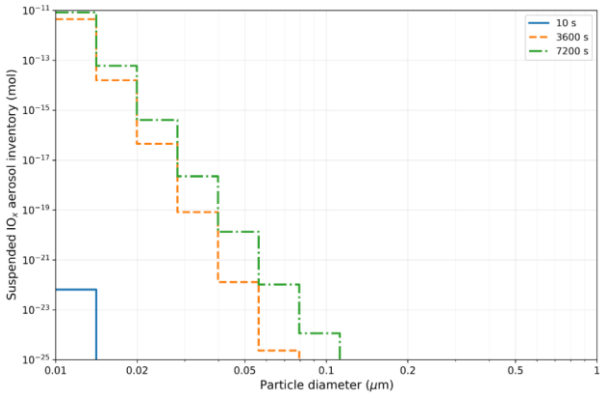


Fig. 4. Evolution of size distribution of aerosol particles during verification

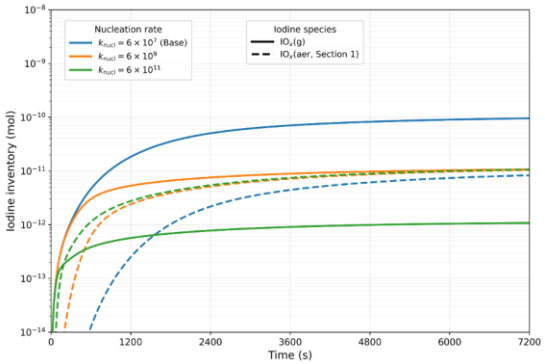
Fig. 4. shows the temporal evolution of the aerosol size distribution during the verification calculation. The aerosols generated by nucleation are initially introduced into the smaller particle-size sections and subsequently transferred toward larger sections through condensational growth. The gradual shift of the aerosol inventory toward larger particle sizes indicates that the aerosol formation and growth processes are consistently calculated in the coupled framework.

3.2. Sensitivity Analysis of Aerosol Formation Models

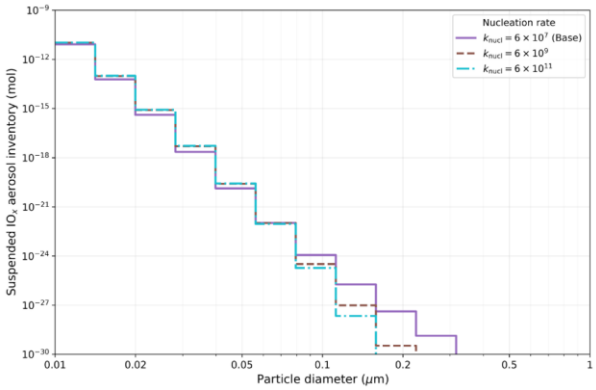
Sensitivity calculations are performed to examine the responses of the implemented nucleation and condensational growth models. The nucleation rate constant k_{nucl}, and the condensational growth parameter, D_{IO_x}, are varied to investigate their effects on aerosol formation and growth, respectively. For the sensitivity analysis of k_{nucl}, the reference conditions used in the verification calculation are maintained except for k_{nucl}. For the sensitivity analysis of condensational growth, k_{nucl} is fixed at 6.0×10^{11} /sec to obtain a sufficient aerosol population and thereby clearly identify the effect of D_{IO_x} on the subsequent aerosol growth. The conditions for the sensitivity calculations are summarized in the following table

Table 3. Calculation conditions for the sensitivity analyses of the aerosol formation and growth models

k_{nucl} [/sec]	D_{IO_x} [/sec]	Values	
Base	Base	6.0E+07	5.0E-14
× 100		6.0E+09	
× 10000		6.0E+11	
Base	Base	6.0E+11	5.0E-14
	× 10		5.0E-13
	× 100		5.0E-12



(a) Change of $IO_{x(g)}$ and $IO_{x(aer)}$ in section 1

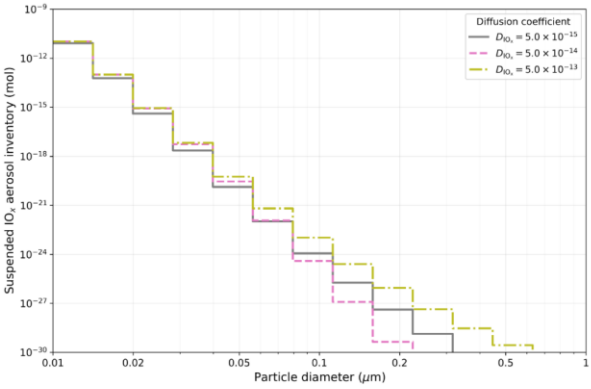


(b) Aerosol size distribution at 7,200 s

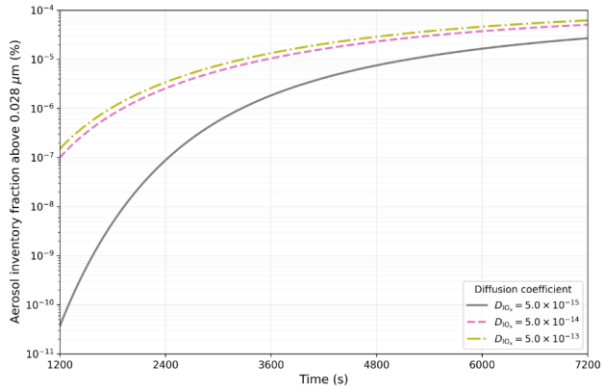
Fig. 5. Effect of the nucleation rate constant on aerosol formation and size distribution

Fig. 5 compares the calculated aerosol behaviors with different nucleation rate constants. As k_{nucl} increases, the

conversion of gaseous iodine oxide to newly formed aerosol particles is enhanced, resulting in an increase in the aerosol inventory. The change in the aerosol formation also affects the subsequent size distribution through condensational growth. These results indicate that the implemented nucleation model responds consistently to the variation in the nucleation rate constant.



(a) Aerosol size distribution at 7,200 sec



(b) Change in fraction of aerosol inventory above 0.028 μm vs. time

Fig. 6. Effect of the diffusion coefficient of iodine oxide on condensational growth.

Fig. 6 shows the effect of the diffusion coefficient of gaseous iodine oxide on condensational growth. As D_{IO_x} increases, the condensational uptake of gaseous iodine oxide by aerosol particles is enhanced. Consequently, the aerosol inventory is transferred more rapidly toward larger particle-size sections, and the fraction of aerosol inventory above 0.028 μm increases more rapidly. These results show the expected response of the implemented condensational growth model to the variation in D_{IO_x}.

4. Conclusions

In this study, AnCheBi and I-COSTA were coupled to establish an integrated framework for the analysis of chemical and aerosol behaviors of iodine during a severe accident. The chemical species calculated by AnCheBi and the sectional aerosol dynamics calculated by I-COSTA were coupled in a monolithic manner by constructing a single Jacobian matrix. Nucleation and condensational growth models were considered for the transfer of iodine oxide from the gas phase to the aerosol phase.

The coupled framework was applied to a verification calculation starting from CsI aerosols deposited on the surface. The calculation described the chemical evolution from deposited CsI to gaseous iodine species and the subsequent formation and growth of IOx aerosols. Although the chemical form and phase of iodine changed during the calculation, the total iodine inventory on a molar basis was conserved. The temporal change in the aerosol size distribution also showed the formation and subsequent growth of IOx aerosols calculated by the coupled framework.

Sensitivity calculations were additionally performed for the nucleation rate constant and the diffusion coefficient of gaseous iodine oxide. An increase in the nucleation rate constant enhanced the formation of IOx aerosols, whereas an increase in the diffusion coefficient enhanced condensational growth and accelerated the transfer of aerosol inventory toward larger particle-size sections. These results showed that the implemented nucleation and condensational growth models responded consistently to the variations in their governing parameters.

From the verification and sensitivity calculations, the numerical implementation and fundamental behavior of the coupled AnCheBi-I-COSTA framework were verified. Further studies will focus on validation of the coupled framework using experimental data involving interactions between iodine chemistry and aerosol dynamics.

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