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Preliminary Analyses on Importance of Iodine Chemistry Models in Aqueous Phase during a Severe Accident

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1. Introduction

Iodine is one of the major contributors to the potential health risk for the public due to its high affinity to thyroid of a human and relatively long half-life nature of the isotopes. In addition, it has multiplicity of the oxidation states under radiation field, high temperature, and humidity in the containment during the severe accident. Under such conditions, radioactive iodine has various chemical forms, e.g., aerosol, organic, inorganic and etc. Depending on the chemical forms, effective dose coefficients, which are essential to evaluate the public risk are various [1]. The aforementioned properties of the iodine during the severe accident, therefore, make the iodine chemistry an area of particular relevance in the source term studies of the nuclear accident.

KINS has investigated the iodine chemistry in the containment as one of the regulatory issues in a relationship with a quantitative safety goal of new and/or operating nuclear power plants and has been developing AnCheBi (Analysis on Chemical Behavior of Iodine Species in the containment during a severe accident) code [2]. Recently, the authors performed analyses on the importance of the models in the gaseous phase and have provided information on the dominant phenomena for estimation of the source term if the iodine is released as gaseous form [3].

In this study, similarly with previous study, we are going to perform numerical analyses on P10T2 experiment [4] for the validation of the iodine chemistry model in aqueous phase. Then we are going to perform the analyses on the importance of the models in aqueous phase.

2. Importance Analyses on the Models in Aqueous Phase via Latin Hypercube Sampling

2.1 Latin Hypercube sampling to sample the reaction coefficients

It is necessary for the reaction coefficients in the iodine chemistry model of aqueous phase to be sampled randomly in order to perform the importance analyses. The samplings in this work are done by in-house code, R-SAPhe which is implemented as an additional function of AnCheBi at this moment [2]. With given probability distribution functions (PDF) and the *n* non-overlapping intervals on the basis of equal probability for each reaction coefficient, R-SAPhe calculates cumulative distribution functions (CDF) and a single value in each interval is selected randomly for each reaction coefficient. Then by picking up randomly to form *n* vectors, the order of *n* values of each reaction coefficient is mixed. In other words, the mixing is done by associating random permutation of the *n* values with reaction coefficients. Then, we have *n* sets of the reaction coefficients to perform sensitivity analyses as output of R-SAPhe.

2.2 Correlation coefficients for the relationship between reaction coefficients and iodine chemical species

CC-SAPhe calculates the correlation coefficients on the reaction coefficients and the concentrations of iodine species with the results of sensitivity analyses via AnCheBi [2]. Values of the correlation coefficients lies between -1 and 1. The values lie between -1 and 1, indicating that if the absolute value of the coefficient is greater than 0.7, the figure of merit has strong linear relationship with the reaction coefficient [5]. In CC-SAPhe, the following four correlation coefficients are calculated.

Pearson coefficient is a covariance of the reaction coefficient and the figure of merit divided by the production of their standard deviations. Spearman coefficient is Pearson coefficient obtained with the rank of reaction coefficient and that of figure of merit. Standardized regression coefficient shows direct measure of the relative importance of the reaction coefficient to the figure of merit since it can remove the problem arising with different reaction coefficients using different units in the numerical analyses. The aforementioned problem is eliminated by standardizing all reaction coefficients, i.e., subtracting mean value of the reaction coefficients from individual reaction coefficient and dividing it by the standard deviation of the reaction coefficients.

Meanwhile, partial correlation coefficient measures degree of association between reaction coefficient and the figure of merit with the effect of other reaction coefficients removed. It can help avoid misleading of the relationship between the reaction coefficient and the figure of merit if there is confounding coefficient in AnCheBi.

We perform the importance analysis as we have done in gas phase [3]. However, in this work, we consider the case when the iodine species are aqueous phase at initial.

The overall procedure of the analyses on the importance is summarized in Fig. 1.

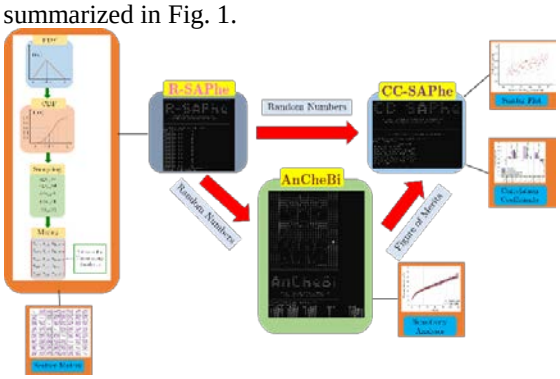

Fig. 1. Framework of importance analyses

3. Numerical Results

We applied the aforementioned procedures to the analyses on P10T2 experiment performed by AECL [4]. The experimental conditions are summarized in Table 1. The computation condition is summarized in Table 2. The range of uncertainty and the number of sampling are derived from the previous works [6]. According to Ref. 6, the uncertainty of experimental data from iodine chemistry experiment is around 25~30% of the measured values. Therefore, it is considered enough to have samplings of 2000 to cover the range of combination of data uncertainty in the samplings.

Table 1. Experimental condition of P10T2

Parameters	Data
Purpose	Iodine behavior in an epoxy painted vessel
Vessel	Epoxy painted
Initial concentrations	$(3.2 \pm 0.2) \times 10^{-4}$
Temperature [K]	298
Dose rate [kGy/h]	0.61
Duration of irradiation	283

Table 2. Computation conditions on the importance analyses

Parameters		Data
Sampling	Number of variables	52
	Type of PDF	Uniform
	Uncertainty range	±30%
Number of Latin Hypercube sampling		2,000
Numerical Method	Time discretization	Implicit Euler method
	Time step control	Step doubling method
	Time step size [sec]	7.0E-03 ~1.0E+00
Figure of merits		$NVI_{(aq)}$, $I_{(aq)}$, $I_{2(g)}$, $HVRI_{(g)}$, $HVRI_{(aq)}$, $LVRI_{(g)}$, $LVRI_{(aq)}$

The results of sensitivity analyses are shown in Figs. 2~6. And the correlation coefficients of the various reaction coefficients on the figure of merits are shown in Figs. 6~9. Note that the numerical results are within the range of the experimental data.

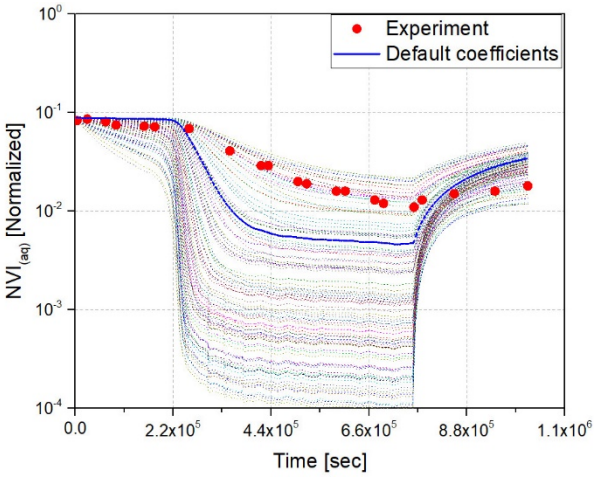

Fig. 2. Change of $NVI_{(aq)}$ in aqueous phase

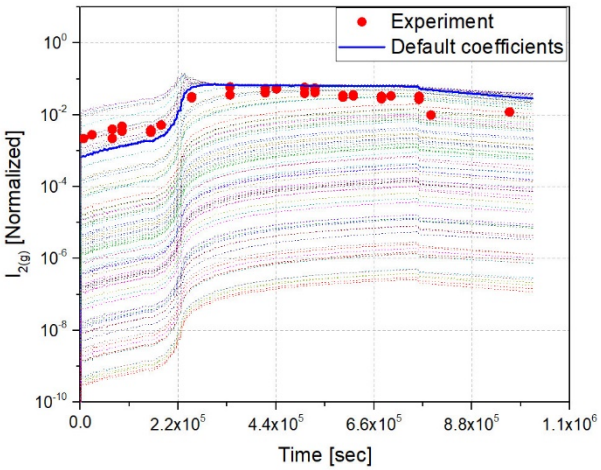

Fig. 3. Change of $I_{2(g)}$ in the gas phase

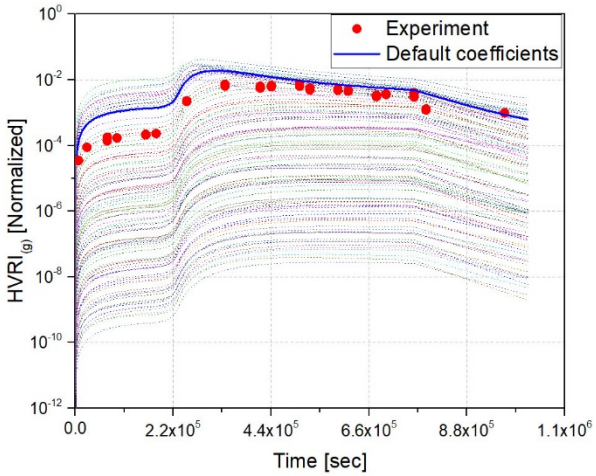

Fig. 4. Change of organic iodide ($HVRI_{(aq)}$+ $HVRI_{(aq)}$) in the aqueous phase

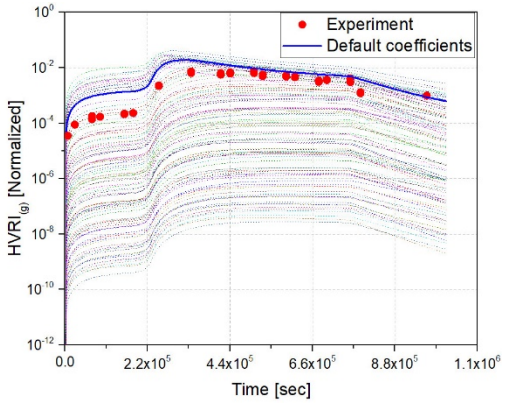

Fig. 5. Change of $HVRI_{(g)}$ in gas phase

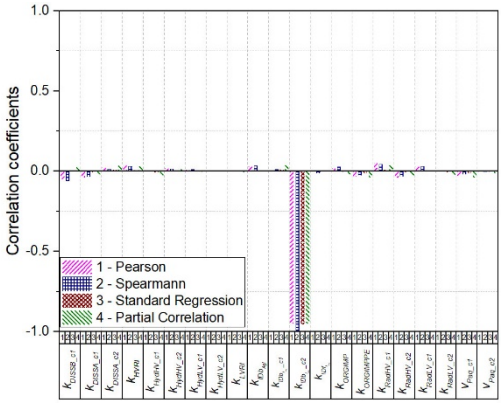

Fig. 6. Correlation coefficients for $NVI_{(aq)}$ and the selected reactions on the aqueous phase

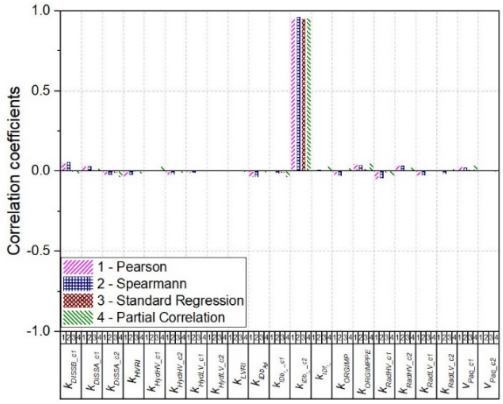

Fig. 7. Correlation coefficients for $I_{2(g)}$ and the selected reactions on the aqueous phase

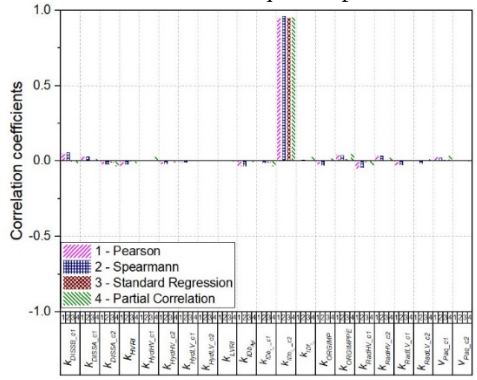

Fig. 8. . Correlation coefficients for $HVRI_{(g)}$ and the selected reactions on the aqueous phase

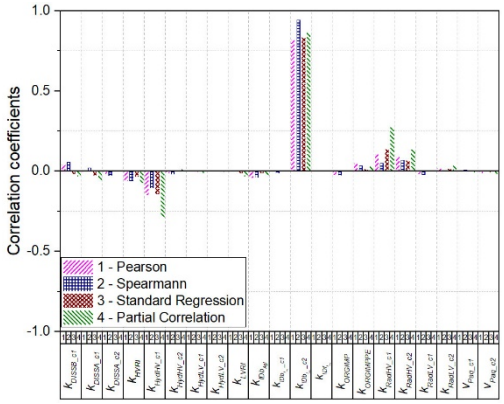

Fig. 9. Correlation coefficients for $HVRI_{(aq)}$ and the selected reactions on the aqueous phase

As shown in Figs. 6~9, the concentrations of the iodine species in the analyses show strong linear relationship with the coefficients on backward reaction between nonvolatile iodine species and volatile iodine species. The results of the analyses show that the evolution of iodine species are mainly affected by the aforementioned backward reaction if the iodine species are in the aqueous phase at initial. The results also imply that the activation energy is the most important factor to determine the backward reaction rate.

Note that it is the case when CsI aerosols are released into the containment and those aerosols are usually deposited onto sump in the containment. The release of CsI aerosols is dominant if the neutron absorption material in the control rod is Ag-In-Cd.

In addition, according to the relationship between activation energy and temperature, represented as Arrhenius equation, the uncertainty range in this study can be converted into the temperature range of 290~430 K. in the sump of vessel. Therefore, it would be possible that the temperature in the sump of the containment also strongly influences the concentration of the iodine species.

4. Conclusions

In this paper, we performed importance analyses on the iodine chemistry model in the gas phase via Latin hypercube sampling by R-SAPhe, in which reaction coefficients are sampled with given range of uncertainty. Sensitivity analyses, then, were performed by iodine chemistry code, AnCheBi, with the sets of reaction coefficients. Correlation coefficients are calculated between concentrations of iodine species, figure of merits, and the reaction coefficients. The followings are main findings from this study :

- The coefficients on the backward reaction between nonvolatile iodine species and volatile iodine species showed strong linear relationship with the concentrations of the iodine species in the aqueous phase.

- The temperature could also influence the evolution of iodine species from those in the aqueous phase as the aforementioned reaction coefficients strongly depend on the temperatures.
- The temperature in the sump would strongly influence the concentrations of the iodine species since uncertainty ranges of the aforementioned coefficients considered in this study are equivalent to the range of temperature in the sump of the containment during severe accident.

From the aforementioned finding, we can expect that the behaviors of the source term for the real scenario if CsI particle is dominant. In that case, the production of iodine species are mainly affected by the backward reaction between $NVI_{(aq)}$ and $I_{(aq)}$. As those aerosols usually deposited onto sump in the containment, the initial condition of the source term in the containment is similar with the case when the iodine species are in the aqueous phase at initial. The aforementioned case occurs if the neutron absorption material in the control rod of the reactor is Ag-In-Cd.

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REFERENCES

[1] ICRP Publication 119 – Compendium of Dose Coefficients based on ICRP Publication 60, ISSN 0146-6453, Elsevier, Oxford, UK.
[2] L. Bosland et al., “Modeling of iodine radiochemistry in the ASTEC severe accident code : Description and application to FPT-2 Phebus test,” *Nucl. Technol.*, **171**, pp. 88-107, 2010.
[2] Y. Lee et al., Analysis on Chemical Behavior of Iodine Species in the Containment during a Severe Accident (AnCheBi) Version V0.00 – Theory and User Manual, KINS/RR-2057, Daejeon, Korea, 2020.
[3] Y. Lee and Y. J. Cho, Preliminary Analyses on the Importance of Iodine Chemistry Reactions in the Gas Phase, Trans. Kor. Nucl. Soc., Spring Meeting, Online, May 13-14, 2021.
[4] G. A. Glowa and C. J. Moore, OECD/NEA Behavior of Iodine Project – Final Summary Report, NEA/CSNI/R(2011)11, Organisation for Economic Co-operation and Development (OECD) Nuclear Energy Agency (NEA), Boulogne-Billancourt, France, 2012.
[5] B. Ratner, The correlation coefficient : Its values range between +1/-1, or do they?, *J. Target., Meas., Anal., Market.*, **17(2)**, pp. 139-142, 2009.
[6] L. Bosland and J. Colombani, “Study of iodine releases from epoxy and polyurethane paints under irradiation and development of a new model of iodine-epoxy paint interactions for PHEBUS and PWR severe accident applications, *J. Raional. Nucl. Chem.*, **314**, pp.1121-1140, 2017.
[7] J. Colombani et al., S2-6-11 EPICUR test report, SERCI-2010-013-DR, Institut de Radioprotection et de Sûreté Nucléaire (IRSN), Fontenay-aux-Roses, France, 2010