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Preliminary Analyses on the Importance of Iodine Chemistry Reactions in the Gas Phase

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1. Introduction

Due to high affinity of a human thyroid and relatively long half-life nature, potential radiological impact of radioactive iodine is significant during a severe accident. Under conditions of the severe accident, such as radiation field with high temperature and humidity in the containment, the radioactive iodine has multiplicity of the oxidation states, which leads to have various chemical forms, e.g., aerosol, organic, inorganic, and etc. Considering that effective dose coefficients are various depending on the chemical form [1], chemical behavior of the iodine is an area of particular relevance in the source term studies of the nuclear accident.

In order to analyze the chemical behaviors of the radioactive iodine, computer codes on the iodine chemistry have been developed based on the combination of separate experimental results on the various phenomena related with iodine [2, 3]. Therefore, it is difficult to find the dominant and important phenomena on the source term of radioactive iodine during severe accidents.

In this paper, based on Latin hypercube sampling on the reaction coefficients in the iodine chemistry code, we will perform importance analyses on the iodine chemistry model in the gas phase. From the analyses we will find dominant and important phenomena for the source term on the iodine, which will provide direction on the evaluation of source term in regulation and the source term research in terms of experiment and computation.

2. Importance Analyses via Random Number Sampling

2.1 Random Sampling of Reaction Coefficients

For the importance analyses, it is necessary for the reaction coefficients in the iodine chemistry model of AnCheBi [4] to be sampled randomly. The samplings in this paper are done by in-house code, R-SAPhe. With given probability distribution function and the *n* non-overlapping intervals on the basis of equal probability for each reaction coefficients, R-SAPhe calculates cumulative distribution functions and a single value in each interval is selected randomly for each reaction coefficient. Then the order of *n* values of each reaction coefficient are mixed by picking up randomly to form *n* vectors. In order words, the mixing is done by associating random permutation of the *n* values with reaction coefficients. As output of R-SAPhe, we have *n* sets of the reaction coefficients to perform sensitivity analyses.

2.2 Correlation Coefficients from the Sensitivity Analyses

With the results of sensitivity analyses via AnCheBi [4], CC-SAPhe calculates the correlation coefficients on the reaction coefficients in order to show the relationship between the reaction coefficients and the figure of merits in the sensitivity analyses, i.e., concentrations of iodine species. The values lie between -1 and 1, indicating that -1 refers to perfect negative linear relationship between the two variables. In contrast, 1 refers to perfect positive linear relationship between the two variables and 0 refers to no relationship. If the absolute value of the coefficient is greater than 0.7, it indicates that the figure of merit has strong linear relationship with the reaction coefficient [5]. In CC-SAPhe, four types of correlation coefficients, i.e., Pearson coefficients, Spearmann coefficients, standard regression coefficients, and partial correlation coefficients are calculated [6].

2.3 Overview of Importance Analyses

In the importance analyses, the first, the reaction coefficients are sampled by in-house code named R-SAPhe, which is based on Latin hypercube sampling (LHS) with the uncertainty ranges of the coefficients used. Then, the sensitivity analyses are performed by iodine chemistry code, AnCheBi [4] with the sets of the aforementioned coefficients to provide the range of concentrations of iodine species. With the results of the sensitivity analyses, correlation coefficients are calculated by an in-house code named CC-SAPhe. We, then, figure out the reaction coefficients which are important for the source term. The procedures are summarized in the following figure.

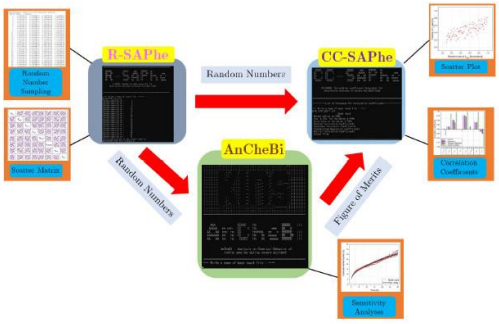

Fig. 1. Overall procedures of the importance analyses

3. Numerical Results

The aforementioned procedure is applied to S2-6-11 experiment performed by IRSN [7]. The experimental conditions are summarized in Table 1. The computation conditions in the analysis are summarized in Table 2.

Table 1. Experimental conditions of S2-6-11 test

Parameters		Data
Purpose		Organic iodide production
Coupon surface		Painted surface
Concentration [mol/m^2]		$\sim 8.9\times10^{-6} M$
Operating temperature [^{o}C]		80
R. H. [%]		60
Dose rate [kGy•h^{-1}]	Coupon	1.54
	Vessel	2.13
Duration [hrs]		8

Table 2. Computation conditions for the importance analyses

Parameters		Data
Variables	Number	20
	Distribution	Uniform
	Uncertainty range	20 % of the reference
# of LHS		300
Numerical methods	Discretization	Implicit Euler
	Time step control	Step doubling method
	Time step size [sec]	7.0E-03~1.0
Figure of merits		$I_{2(g)}$
		$IO_{x(g)}$
	Organic iodides	$HVRI_{(g)}$
		$LVRI_{(g)}$

Figs. 2~4 show results of sensitivity analyses on the figure of merits listed in Table 2. Note that the numerical results show good agreement with experimental data.

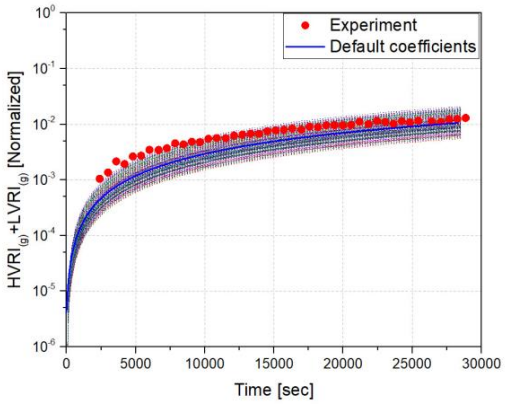

Fig. 2. Change of organic iodides with LHS variables

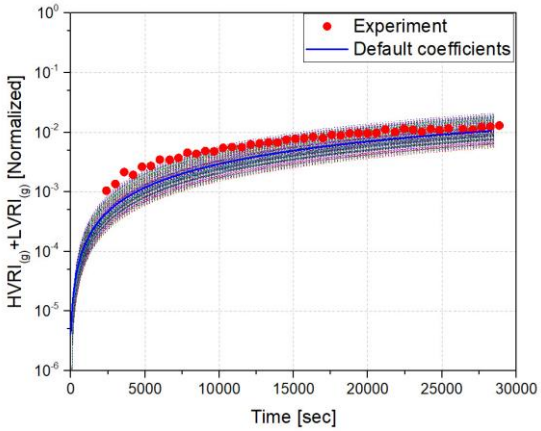

Fig. 3. Change of $IO_{x(g)}$ with LHS variables

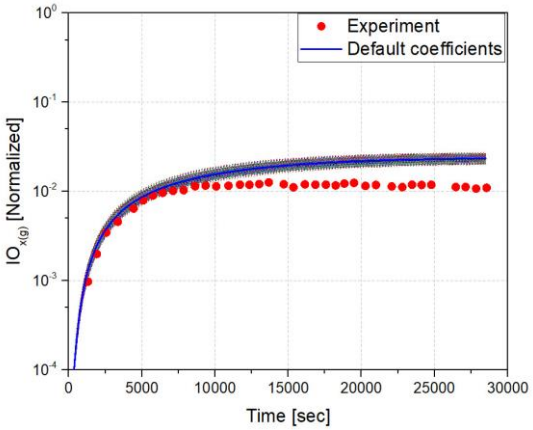

Fig. 4. Change of $I_{2(g)}$ with LHS variables

Correlation coefficients on the organic iodides are shown in Figs. 5 and 6. Note that the activation energy of the reaction related with formation of organic iodides from iodine on the painted surface, $kHVRI_{gRc2}$, and $kLVRI_{gRc2}$, show strong negative linear relationship with concentration of organic iodides. Scattered plots between the concentrations and the reaction coefficients are shown in the following Figs. 7 and 8.

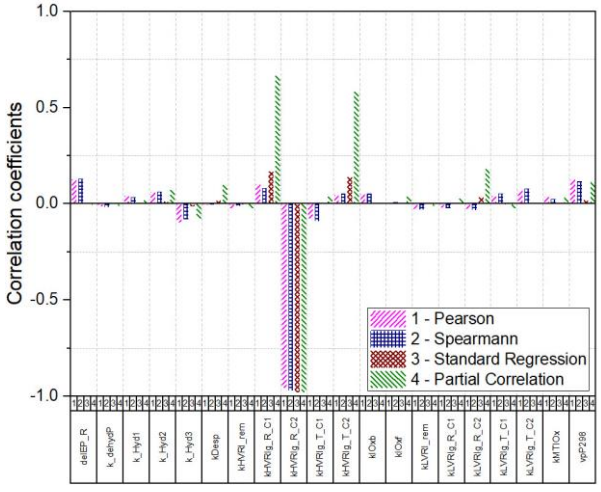

Fig. 5. Correlation coefficients on $HVRI_{(g)}$

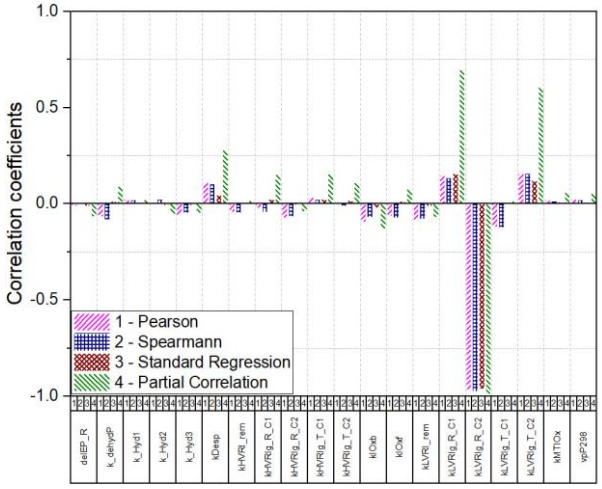

Fig. 6. Correlation coefficients on $LVRI_{(g)}$

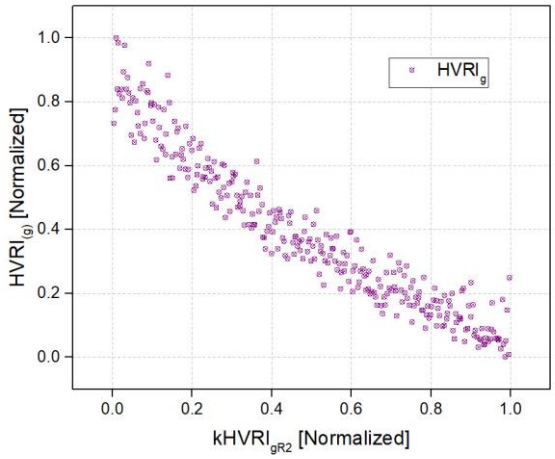

Fig. 7. Scatter plot on HVRI(g) for kHVRI $_{gRc2}$

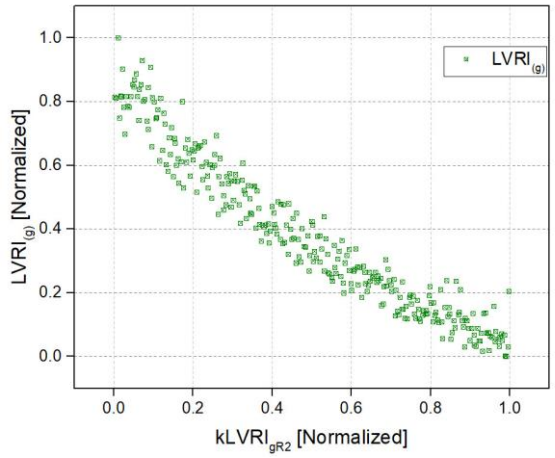

Fig. 8. Scatter plot on LVRI(g) for kLVRI $_{gRc2}$

In the case of molecular iodine and iodine oxide aerosol in the gas phase, reaction coefficients related with behaviors of iodine oxide aerosols, such as kIOxb, (destruction of iodine oxide aerosol to I2 gas), kIOxf (formation of iodine oxide from I2), kMTIOx (deposition of iodine oxide aerosol) and those related with iodine on the painted surface show strong relationship as shown in Figs. 9~12.

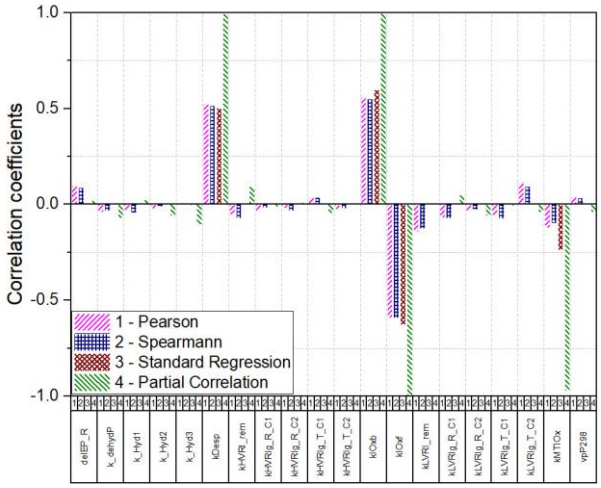

Fig. 9. Correlation coefficients on $I_{2(g)}$

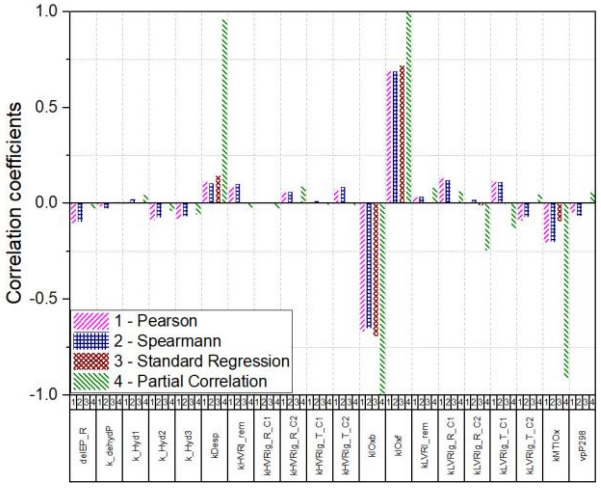

Fig. 10. Correlation coefficients on $IO_{x(g)}$

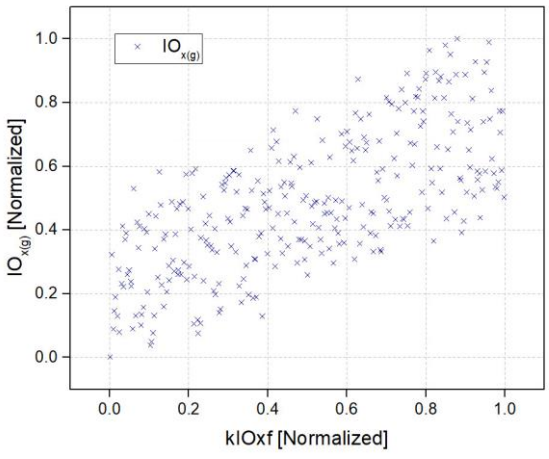

Fig. 11. Scatter plot on $IO_{x(g)}$ for kIOxf

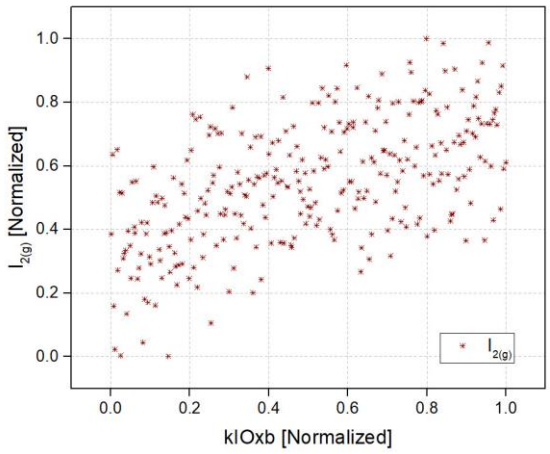

Fig. 12. Scatter plot on $I_{2(g)}$ for kIOxb

From the importance analyses, we can conclude that the reactions of the iodine on the painted surface, those of the iodine oxide aerosols should be reviewed carefully for the regulation. In terms of experimental and computational research, research activity on those reactions should be done. In addition, considering that the activation energy of the reaction is strongly related with the temperature, we also have to examine the temperature of the painted surface carefully.

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.

From the analyses, we found that reactions related with iodine on the painted surface show strong negative linear relationship with the activation energy. In the case of reactions related with iodine oxide aerosols, molecular iodine and iodine oxide aerosol in the gas phase, reaction coefficients related with behaviors of iodine oxide aerosols, such as destruction of iodine oxide aerosol to I2 gas, formation of iodine oxide from I2, deposition of iodine oxide aerosol, and those related with iodine on the painted surface show strong relationship.

From the importance analyses, we can conclude that the reactions of the iodine on the painted surface, those of the iodine oxide aerosols should be reviewed carefully for the regulation. In terms of experimental and computational research, research activity on those reactions should be done. In addition, considering that the activation energy of the reaction is strongly related with the temperature, we also have to examine the temperature of the painted surface carefully.

As future work, we will perform the importance analyses on the aqueous phase and the thermo-hydraulic parameters related with the iodine chemistry. We will also extend the framework of importance analyses to the other phenomena related with source term studies, such as pool scrubbing and aerosol dynamics in the containment.

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