Study of Iodine Behavior in the Gas Phase during a Severe Accident

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1. Introduction

Through extensive research in the area of severe accidents, the radiological consequence for most accident sequences can be predictable. However, it is still necessary to reduce the uncertainties involved in the estimation of the source term, especially regarding the release of iodine and ruthenium from the reactor core and reactor coolant system and transport to the containment [1]. Among the iodine species, the organic iodides produced from the reaction between iodine and organics such as paint, are not easily trapped by the filters during the containment venting following a severe accident.

Korea Institute of Nuclear Safety (KINS) has been studying this issue, joining international research programs such as ISTP-EPICUR, OECDBIP and OECD-STEM. In the course of this study, a simple iodine model, RAIM (Radio-Active Iodine chemistry Model) has been developed (Oh et al., 2011) [2], based on the IMOD methodology [3], and other previous studies [4, 5]. This paper deals with our recent activities on this study, including the development of the model for the iodine reactions in gas phase.

2. Model for Iodine Reactions in the Atmosphere

In gaseous atmospheres, organic iodides can be produced from painted surfaces by radiolysis or thermal reaction between molecular iodine and organics. They also decompose into molecular iodine or iodine oxides by radiolysis, and destruction of iodine oxide contribute to the formation of organic iodides. In these processes, reactions with air radiolysis products (ARPs) such as ozone play an important role. These reactions are modeled for RAIM, referring to the previous research results [4, 5] as follows;

- Formation of ARP (Ozone is the representative)

  Stoichiometry: $3O_2 \overset{\gamma}{\leftrightarrow} 2O_3$

  Kinetics:

  $$\frac{d[O_{3(g)}]}{dt} = D_g \frac{P}{P_0}\left(k_1 - k_2[O_{3(g)}]\right) - \frac{[O_{3(g)}]}{V_g}\left(k_3 S_{pd} + k_4 S_{sd}\right)$$

- Iodine oxide formation

  Stoichiometry: $I_2 + 2O_3 \leftrightarrow \frac{1}{2}(I_4O_9 + \cdots) \leftrightarrow 2IO_{3(g)}^-$

  Kinetics: $-\frac{d[I_{2(g)}]}{dt} = k_1[O_{3(g)}][I_{2(g)}] - \frac{1}{2}k_{-1}D_g[IO_{3(g)}^-]$

- Iodine oxide destruction into RI (under study)

- Formation of organic iodides in the gaseous phase

  1) Formation of CH_3I by radiolysis

     Stoichiometry: $\frac{1}{2}I_{2(absorbed\ onto\ paints)} + R \overset{\gamma}{\rightarrow} CH_3I$

     Kinetics: $\frac{d[CH_3I_{(g)}]}{dt} = k_r D_g [I_{2(p)}] \frac{S_{pd}}{V_g}$

  2) Formation of CH_3I by thermal reaction

     Stoichiometry: $\frac{1}{2}I_{2(absorbed\ onto\ paints)} + R \rightarrow CH_3I$

     Kinetics: $\frac{d[CH_3I_{(g)}]}{dt} = k_T [I_{2(p)}] \frac{S_{pd}}{V_g}$

- Destruction of organic iodides in the gaseous phase (under study)

Fig. 1 shows the diagram for these processes treated by RAIM.

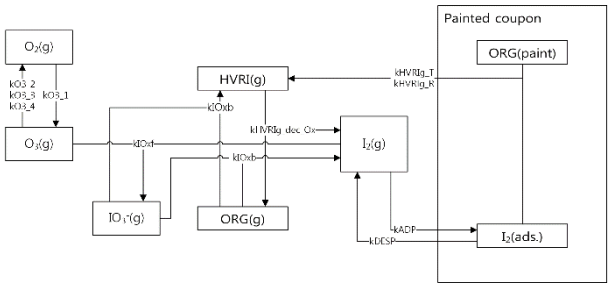

Fig. 1. Iodine behavior in the gas phase treated in the RAIM code

3. Simulation of the EPICUR Experiment

3.1 ISTP-EPICUR S2 Experiment

The EPICUR program, which was operated by the IRSN as part of the ISTP, dealt with the kinetics of organic iodide formation through reactions with paint, reactions in gas phase and formation of volatile iodine in liquid phase. Fig. 2 shows a schematic of the EPICUR loop, in which the irradiation vessel and May-pack device are connected by a stainless steel tube. Thus, the volatile species produced in the irradiation vessel are transferred to the May-pack device via the gas bubble. The May-pack system is composed of several steps with quartz fiber filter, knit-mesh impregnated and activated carbon filter, so that iodine aerosol, iodine molecule, and organic iodide may be captured and quantified. On-line γ measurement is provided by the NaI (Tl) counters placed on top of each stage of the filter of the May-back device.

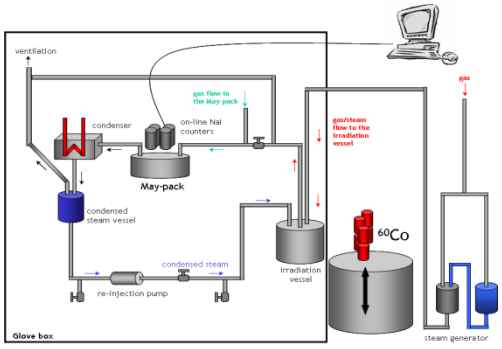

Fig. 2. Simplified view of the experimental EPICUR loop.

The EPICUR program consists of several test series; the paint coupons of the S1 series were put in the iodine solution, whereas the S2 series iodine-loaded coupons were tested in gas phase. The purpose of the S2 series was to generate information of the formation of organic iodide from a painted coupon loaded with molecular iodine and placed in gaseous phase. Among them the S2-6-5-2 test was chosen to be analyzed by RAIM as it was a reproducibility test of the previous one, performed for evaluation of the scattering of the quantities of the iodine trapped on the May-pack filters. The test conditions are shown in Table I.

Table I: Experimental conditions of the S2-6-5-2 test

Kind of condition	Value
Iodine surface concentration on the painted coupon before irradiation ($mol_{I_2} \cdot m^{-2}$)	2.5×10^{-4}
Liquid volume (mL)	0
Gas (mL)	4800
% RH	60
$T_{\text{irradiation vessel}}$ (℃)	80 120
Gas flow in the liquid phase ($L \cdot min^{-1}$)	0.21 0.41
Gas flow in the May-pack ($NL \cdot h^{-1}$)	273
Dose rate at the level of the painted coupon ($kGy \cdot h^{-1}$)	1.66
Dose rate in the gaseous phase ($kGy \cdot h^{-1}$)	2.30

3.2 Analysis of the S2-6-5-2 Test

RAIM was applied to analyze the S2-6-5-2 test. Fig. 3 shows the volatile iodine concentration with the assumption of no desorption from the painted coupon, and its comparison with the experimental data. As shown in Fig. 3, inorganic iodine concentration was underestimated for this case. In Fig. 4, the volatile iodine concentration is compared with the case of the desorption rate constant of about 10^{-6} s^{-1} since this value is used by the recent studies [4, 5]. Therefore, it was decided that this value was also applicable for this analysis. Fig. 5 and Fig. 6 show the concentrations of organic iodides and iodine oxides respectively, with the new assumption. The reason that they were significantly overestimated for the whole test period seems to be due to the inappropriate rate constants for formation of iodine oxides and organic iodide. Thus, further study will be necessary to determine reasonable rate constants for these reactions.

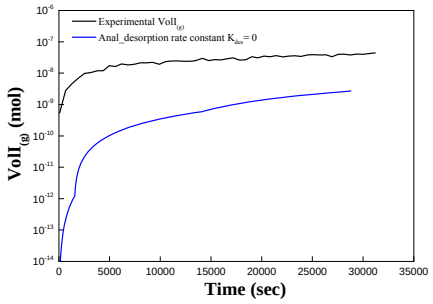

Fig. 3. Volatile inorganic iodine concentration.

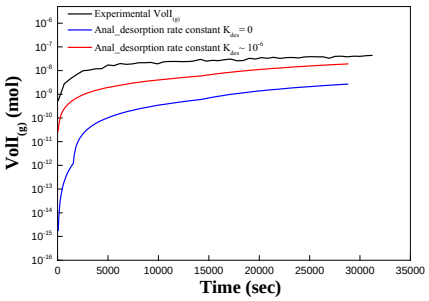

Fig. 4. Volatile inorganic iodine concentrations with different desorption rate constants.

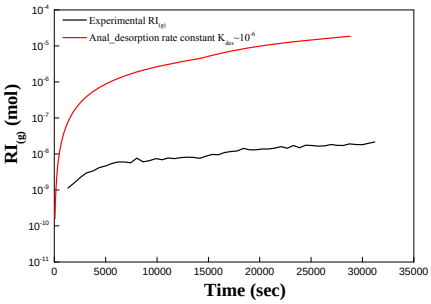

Fig. 5. Organic iodide concentration.

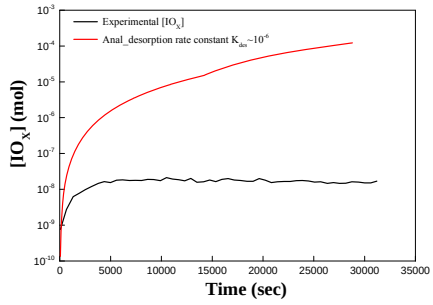

Fig. 6. Iodine oxide concentration.

4. Conclusions

Iodine reactions in gas phase were modeled and added to the RAIM code, taking into account several relevant reactions such as formation of ARP, iodine oxide, and organic iodides in gas phase. RAIM was then applied to analyze the S2-6-5-2 test for which iodine-loaded coupons were tested in gas phase. The analysis results show a reasonable estimation of volatile iodine concentration with the desorption rate constant of about 10^{-6} s^{-1}, while those of the other iodine species overestimated for the whole period of the test. It reveals the need to determine appropriate values for the rate constants for formation of iodine oxides and organic iodides.

Acknowledgments

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