Parametric Study on the Organic Iodide Behavior during a Severe Accident

Myung-Hyun Ryu[a*], Han-Chul Kim[a], Do Sam Kim[a]
[a]Korea Institute of Nuclear Safety, 62 Gwahak-ro, Yuseong-gu, Daejeon 305-338
[*]Corresponding author: s145rmh@kins.re.kr

1. Introduction

Iodine is a major contributor to the potential health risk for the public following a severe accident from a nuclear power plant. Most of metal-iodides, the major form of iodine that enters the containment, can be readily dissolved in the sump water and result in iodide ions. These will be oxidized to form volatile I_2 through a large number of reactions such as radiolysis and hydrolysis. The organic radicals, made from organics such as paint in the sump water, react with iodine to produce organic iodides.

Volatile iodine moves from the sump water to the atmosphere mainly by diffusion and natural convection, and react with surfaces and air radiolysis products (ARPs).Painted surfaces act as a sink for I_2 and as a source for organic iodides through adsorption and desorption. ARPs react with I_2 to form iodine oxides, which leads to the decrease of I_2 and organic iodides [1].

Among the large number of iodine species, organic iodides have been extensively studied recently due to their volatility and very low retention. Qualified tools for modeling these phenomena have been developed and validated by several experiments such as EPICUR, PARIS and OECD-BIP. While mechanistic codes model a large number of reactions and species, semi-empirical codes such as IODE or IMOD treat major ones. KINS developed a simple iodine model, RAIM (**R**adio-**a**ctive **i**odine chemistry **m**odel) [2], based on the IMOD methodology in order to deal with organic iodides conveniently, coupling with an integrated severe-accident analysis code.

There are a number of mechanisms that affect the behavior of organic iodides. In this study, effects of pH of the aqueous phase, temperature, radiation dose rate, surface area of organic paints, initial iodine loads that are known to be important to organic iodide formation were studied analytically with RAIM, and also theoretically**.**

2. Parameters that Affect Organic Iodide Behavior

It has been shown that RAIM could model the behavior of organic iodides and inorganic iodine reasonably, as shown in Fig. 1. Therefore, RAIM was applied to examine the effects of major parameters on the behavior of organic iodides, referring to the previous study [3]. EPICUR S1-7 experimental data [4] were used for this parametric study.

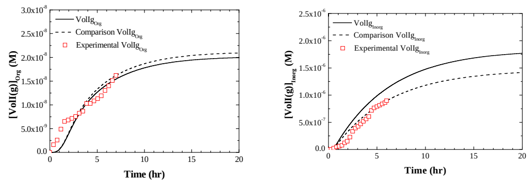

Fig. 1. Comparison of calculated values of volatile organic iodides (S1-7 experiment) and inorganic iodides (S1-5 experiment)

2.1 Effect of temperature

The effect of temperature on the amount of organic iodides formed has been examined while other factors were fixed for RAIM calculation. Fig. 2 shows that when temperature increased by 40℃, the amount of organic iodides decreased by almost two times. Therefore, it can be interpreted that higher temperature could form less organic iodides.

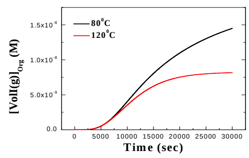

Fig. 2. Calculated values of volatile organic iodides at 120℃ and 80℃

2.2 Effect of the aqueous phase pH

Fig. 3 shows the amounts of organic iodides formed when the pH values of the aqueous phase were5 and 7 respectively. Under the condition of pH of 5, approximately 6 times bigger amount of volatile organic iodides were produced, compared to that of pH 7. Therefore acidic condition could form more volatile organic iodides than the neutral or alkaline condition.

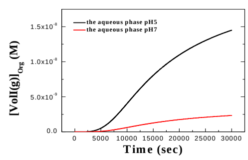

Fig.3. Calculated values of volatile organic iodides at pH 5 and pH 7

2.3 Effect of dose rate

Fig. 4 shows change of the amount of organic iodide formation when radiation dose rate per hour increased. As dose rate doubled, the amount of volatile organic iodide formation increased by 16%.

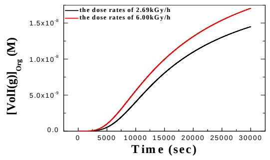

Fig. 4. Calculated values of volatile organic iodides at the dose rates of 2.69 kGy/h and 6.00 kGy/h

2.4 Effect of organic paint area

Fig. 5 shows change of the amount of organic iodide formation as the area of the organic paint increased from 25cm^{2}to 50cm^2. As the paint area was doubled, the amount of volatile organic iodide formation also became doubled.

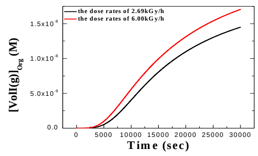

Fig. 5. Calculated values of volatile organic iodide formation with the areas of organic paint of 25 cm^2 and 50 cm^2

2.5 Effect of initial iodine concentration

Fig. 6 shows the amount of organic iodide formation with the initial concentration of iodine is 10^{-5} M and 10^{-4} M. As the amount of initial iodine increased 10 times, volatile organic iodides also increased 10 times.

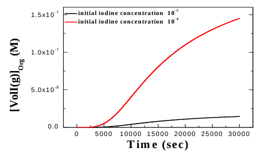

Fig. 6. Calculated values of volatile organic iodide formation with initial iodine concentrations of 10^{-5} M and 10^{-4} M

3. Discussion

Among the five factors listed above, four factors except radiation dose rate affected volatile iodine formation more sensitively. Firstly, the effect of temperature on the amount of volatile iodine can be explained by the following equations [2, 5];

$$\mathrm{NonVolI(aq)} \underset{k_b}{\overset{k_f}{\leftrightarrow}} \mathrm{volI(aq)} \leftrightarrow \mathrm{volI(g)} \quad (1)$$

$$k_b \propto \exp(-1/T_{aq})\cdot[H^+]^{-1/Taq} \quad (2)$$

$$\mathrm{ORG\bullet(aq)+volI(aq)} \rightarrow \mathrm{RI(aq)} \leftrightarrow \mathrm{RI(g)} \quad (3)$$

As temperature increases, the amount of volatile inorganic iodine decreases and so does that of organic iodides by sequence. Secondly, the reason why the amount of organic iodides increases as the amount of the initial iodine concentration increases can be explained similarly. Thirdly, the effect of pH on the amount of organic iodides can be explained by the following equations;

$$\underset{(\mathrm{NonVolI(aq)_{Inorg}})}{\mathrm{HOI + H^+ + I^-}} \overset{K_{eq}}{\leftrightarrow} \underset{(\mathrm{VolI(aq)_{Inorg}})}{\mathrm{H_2O + I_2(aq)}} \leftrightarrow \underset{(\mathrm{VolI(g)_{Inorg}})}{\mathrm{I_2(g)}} \quad (4)$$

$$k_{eq} = \frac{[I_2(aq)]}{[HOI][H^+][I^-]} \quad (5)$$

The amount of volatile organic iodide decreases when pH increases in accordance with equations (3) through (5). Finally, the effect of the area of organic paint on the amount of organic iodides follows equations (3) and (6).

$$\mathrm{RH(aq) + OH\bullet \rightarrow R\bullet(aq) + H_2O} \quad (6)$$

In the above equation RH(aq) is dependent on the paint area. Therefore, this area could increase formation of organic iodides [4].

4. Conclusions

The effects of pH of the aqueous phase, temperature, dose rate, the area of organic paint, initial iodine concentration, that are known to be important for organic iodide behavior were studied using RAIM. These effects were also theoretically discussed. Through this study it was confirmed that applying the accident management strategies such as control of pH, metal-iodide complexation and decrease of the usage of organic paint could reduce release of volatile organic iodides.

Acknowledgments

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