Simulation of ISTP-EPICUR Iodine Chemistry Tests with RAIM

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1. Introduction

Following a severe accident, radioactive iodine is a major contributor to the external dose and the thyroid dose resulting in early fatalities. The amount of iodine release largely depends on its volatility in the containment. Iodine has several chemical forms including aerosols, vapor, and gas. Among them gaseous iodine such as I_2 and organic iodide are dominating due to their high volatility. Therefore, such iodine behavior has been extensively examined [1, 2].

Korea Institute of Nuclear Safety (KINS) has been joining the relevant international programs such as ISTP-EPICUR, OECD-BIP and OECD-STEM. In the course of this study, a simple iodine model, RAIM (Radio-Active Iodine chemistry Model) has been developed [2], based on the IMOD methodology [3] and other previous studies [4, 5]. This model deals with chemical reactions associated with formation and destruction of iodine species in the containment atmosphere and the sump in a simple manner, as shown in Fig. 1. It also treats adsorption and desorption of volatile iodine on the paint surface. The iodine species modeled are inorganic volatile iodine (I_2), organic iodides of high volatility (HVRI) and low volatility (LVRI), non-volatiles, non-aqueous iodine, and iodine oxide aerosols (IO_x). Many other material participating in the iodine reactions, e.g., air radiolysis products (ARP) such as ozone, are also modeled. This paper especially shows the analysis results after addition of gaseous reaction model to RAIM [6], which was further accompanied by adjustments of the existing reaction rate constants even for the aqueous reactions.

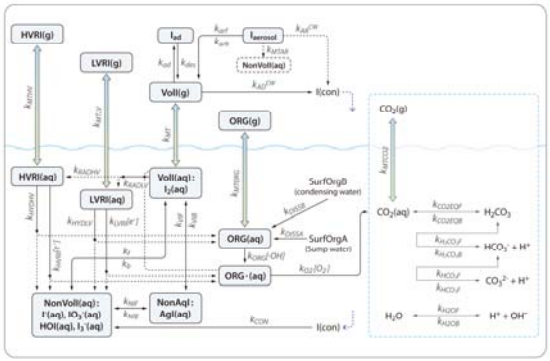


Fig. 1. Iodine behavior treated in the RAIM code

2. Simulation of the ISTP-EPICUR Tests

3.1 EPICUR test facility

The EPICUR program, which was operated by the IRSN (Institut de Radioprotection et de Sûreté Nucléaire) as part of the ISTP (International Source Term Programme), 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 diagram of the EPICUR loop, in which the irradiation vessel and May-pack device are connected by a stainless steel tube. 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. Therefore, iodine aerosol (IO_x), molecular iodine (I_2), and organic iodide (RI) can be captured and quantified serially. 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 [7].

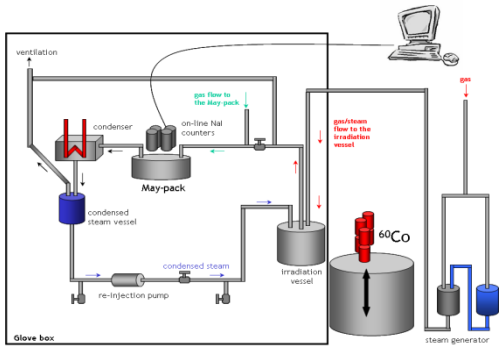


Fig. 2. Simplified view of the experimental EPICUR loop [7].

The EPICUR program consists of several test series; the S1 series tests were conducted to study the organic iodide formation from a painted coupon in the iodine solution, whereas the S2 series was carried out with a painted coupon loaded with molecular iodine and placed in gaseous phase. Among them the S1-9, S1-11, and S2-6-5-2 tests were chosen to be analyzed by RAIM; the first two tests represent the S1 series with the objectives of RI formation and I^- radiolysis or radiolysis only in aqueous phase, while the last stands for the S2 test series with the aim of RI formation in gaseous phase. The test conditions are shown in Table I.

Table I: Experimental conditions of the selected tests

Kind of condition	S1-9	S1-11	S2-6-5-2
Test objective	I^- radiolysis & RI formation	I^- radiolysis	RI formation & I_2 desorption
Iodine surface concentration on the painted coupon before irradiation ($mol_I \cdot m^{-2}$)	No coupon in gas phase	No coupon	2.5×10^{-4}
Iodine solution ($mol_{I^-} \cdot L^{-1}$)	1.03×10^{-5}	1.03×10^{-5}	-
Liquid volume (mL)	2,000	2,009	0
Gas volume (mL)	2,800	2,791	4,800
% RH	-	-	60
$T_{irradiation\ vessel}$ (℃)	66.6→ 71.5	80.5	80→120
pH	7	7	-
Gas flow in the liquid phase ($L \cdot min^{-2}$)	0.25→ 0.50	0.25→ 0.50	0.21→ 0.41
Gas flow in the May-pack ($NL \cdot h^{-2}$)	3.73	3.68	273
Dose rate at the level of the painted coupon ($kGy \cdot h^{-2}$)	1.81	No coupon	1.66
Dose rate in the gaseous phase ($kGy \cdot h^{-2}$)	2.29	2.34	2.30
Average sump dose rate ($kGy \cdot h^{-2}$)	2.84	2.90	-

3.2 Analysis of the Aqueous-phase Tests (S1-9 and S1-11)

Fig. 3 through Fig. 5 show the concentrations of volatile iodine, inorganic iodide, and iodine oxide (IO_x) aerosols for the S1-9 test, respectively. Similarly, Fig. 6 through Fig. 8 show the concentrations for the S1-11 test. Both the results for S1-9 and S1-11 show that the concentrations of inorganic molecular iodine and IO_x were overestimated by RAIM, while that of organic iodide was underestimated at first and then overestimated for S1-9. As the S1-11 test did not use any painted coupon, there were no organic iodide calculated, but in the experiment it was detected with the similar amount of S1-9. In this test, the amount of iodine measured on the Knit-mesh might be slightly underestimated since a fraction of molecular iodine might have been retained on the charcoal stage instead of the Knit-mesh stage due to a low efficiency of the Knit-mesh [8].

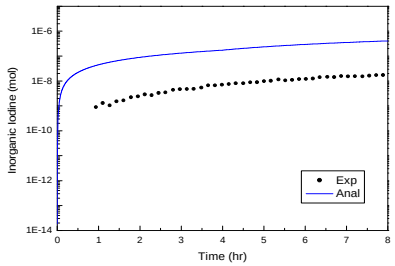


Fig. 3. Volatile inorganic iodine concentration (S1-9).

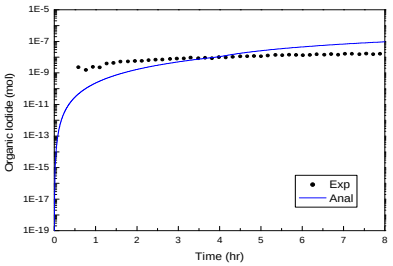


Fig. 4. Volatile organic iodide concentration (S1-9).

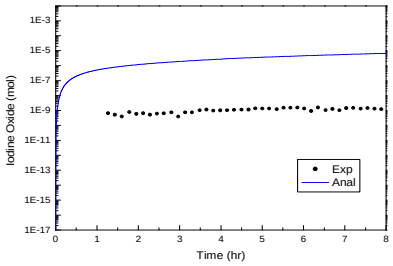


Fig. 5. Iodine oxide concentration (S1-9).

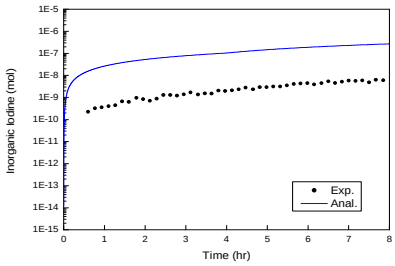


Fig. 6. Volatile inorganic iodine concentration (S1-11).

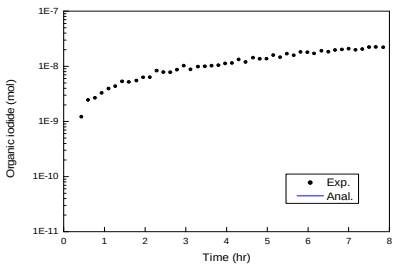


Fig. 7. Volatile organic iodide concentration (S1-11).

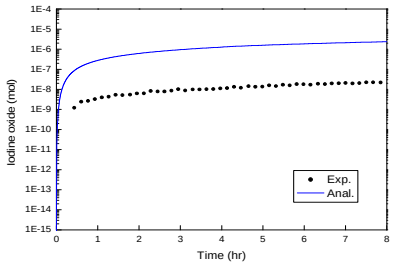
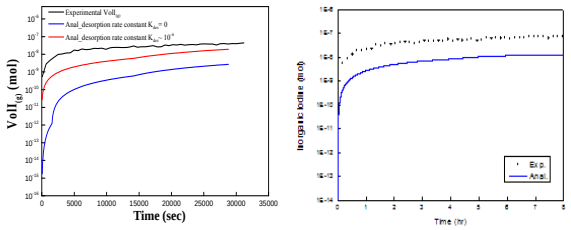


Fig. 8. Iodine oxide concentration (S1-11).

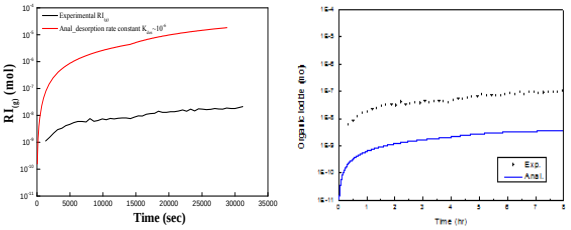
3.3 Re-analysis of the Gaseous-phase Test (S2-6-5-2)

Fig. 9 through Fig. 11 show comparison of the previously calculated concentrations [6] and new ones of volatile iodine, inorganic iodine, and IO_x aerosols for the S2-6-5-2 test, respectively. It should be noted that the previous experimental data were not corrected by comparing the post-test measurement and the on-line data at the end of the irradiation period. As shown in these figures, this revised model predicts better than before. However, the trend of underestimation of molecular and organic iodine is reverse to that of the aqueous tests previously mentioned. Therefore, further study will be necessary to determine more reasonable rate constants for the reactions including mass transfer between aqueous and gaseous phases.



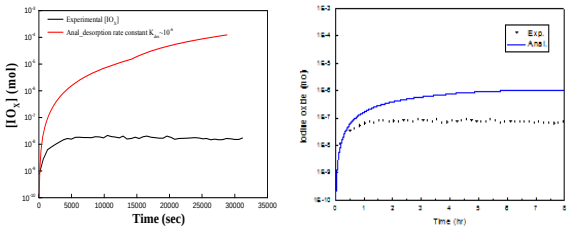
(a) Previous result (b) Present result

Fig. 8. Volatile inorganic iodine concentration (S2-6-5-2).



(a) Previous result (b) Present result

Fig. 9. Volatile organic iodide concentration (S2-6-5-2).



(a) Previous result (b) Present result

Fig. 10. Iodine oxide concentration (S2-6-5-2).

4. Conclusions

After integration of iodine reaction models for gas and aqueous phase, RAIM was applied the S1-9 and S1-11 tests which were carried out in aqueous phase. In addition, re-analysis of the S2-6-5-2 test, for which iodine-loaded coupons were tested in gas phase, was also performed. The analysis results show improvements of estimation, while there were overestimation of molecular and organic iodine for the aqueous tests and a reverse trend for the gaseous test. Therefore, further study is needed to determine more reasonable values for the rate constants including those for mass transfer between aqueous and gaseous phases.

Acknowledgments

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