Development of Assessment Methodology of Chemical Behavior of Volatile Iodide under Severe Accident Conditions Using EPICUR Experiments

Jae Yong Oh[a*], Jong-Il Yun[a], Do Sam Kim[b], Han-Chul Kim[b],
[a]*Dept. of Nuclear and Quantum Eng., KAIST, Daejeon 305-701, Korea*
[b]*Korea Institue of Nuclear Safety, Daejeon 305-338, Korea*
[*]*Corresponding author: onlymywill@kaist.ac.kr*

1. Introduction

Iodine is one of the most important fission products produced in nuclear power plants. Under severe accident condition, iodine exists as a variety of species in the containment such as aqueous iodide, gaseous iodide, iodide aerosol, etc. [1]. Following release of iodine from the reactor, mostly in the form of CsI aerosol, volatile iodine can be generated from the containment sump and release to the environment. Especially, volatile organic iodide can be produced from interaction between nonvolatile iodine and organic substances present in the containment. Volatile iodide could significantly influence the alienated residents surrounding the nuclear power plant. In particular, thyroid is vulnerable to radioiodine due to its high accumulation.

Therefore, it is necessary for the Korea Institute of Nuclear Safety (KINS) to develop an evaluation model which can simulate iodine behavior in the containment following a severe accident. KINS also needs to make up its methodology for radiological consequence analysis, based on MELCOR-MACCS2 calculation, by coupling a simple iodine model which can conveniently deal with organic iodides. In the long term, such a model can contribute to develop an accident source term, which is one of urgent domestic needs. Our strategy for developing the model is as follows; 1. Review the existing methodologies, 2. Develop a simple stand-alone model, 3. Validate the model using ISTP-EPICUR (Experimental Program on Iodine Chemistry under Radiation) and OECD-BIP (Behavior of Iodine Project) experimental data.

In this paper we present the context of development and validation of our model named RAIM (**R**adio-**a**ctive **i**odine chemistry **m**odel).

2. Experimental and Model Development

2.1 EPICUR experiments

In recent years, IRSN (Institute for Radiological Protection and Nuclear Safety, France) has operated the ISTP (International Source Term Program) research project. EPICUR is a part of the project setup to analyze the chemical behavior and chemical speciation of irradiated radioiodine. Among the EPICUR experiments, fourteen tests representing the three types of experiments are selected for validation of RAIM: S1 inorganic iodide, S1 organic iodide and S2 organic iodide experiment. S1 inorganic iodide tests were performed to test the measurement system and measure radiolysis of iodide. S1 organic iodide experiments were carried out to analyze radiolysis of iodide and formation of organic iodide. S2 organic iodide tests were conducted to measure the formation of organic iodide and iodine oxide and desorption of iodine. However, simulation of two experiments, S2-6-9 and S1-9, is presented in this paper. Fig. 1 shows the EPICUR experimental system [2]. NaI(Tl) counters of May-pack system measure the concentration of volatile iodide species.

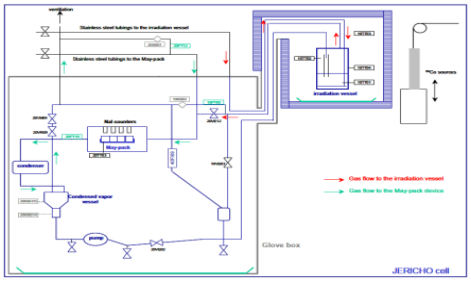

Fig.1. EPICUR experimental system.

2.2 Development of the RAIM code

In order to develop a model for chemical behavior of volatile iodine, the existing methodologies have been reviewed. There are two types of iodine behavior models: mechanistic and semi-empirical models. Mechanistic models such as LIRIC, INSPECT and MELCOR are based on detailed data established through extensive analysis of iodine reactions; and it includes mechanisms and intermediate products of iodine reactions. However, they have disadvantages such as low practicality due to their huge size and deficiencies in phase division. On the other hand, semi-empirical models such as IMOD include reactants and products only. Due to their simplified scheme, they are practical, even though they may need additional reaction calculation and result in inaccurate calculation for organic iodide at high pH [1].

Through the above review it was decided to develop our own model based on the LIRIC methodology [3,4], but seeking a smaller and simpler model like IMOD. Reactions in the aqueous phase and volatile species for mass transfer were reduced. For simplification of reactions and rates, single species were used in the gas and aqueous phases for dissolution of organic solvents from paints and adsorption of volatile iodine onto paints. The rate constants of iodine reactions were empirically determined using the EPICUR experimental data. Fig. 2 shows the behavior of iodine that is treated in RAIM.

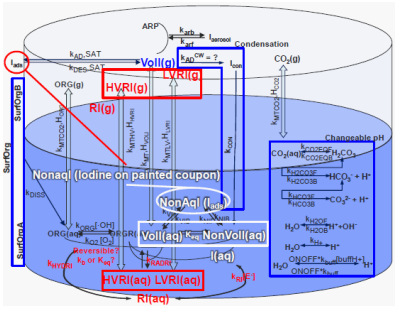

Fig.2. Behavior of iodine treated in the RAIM code.

Collectiveness and convenience of its application were considered.

3. Results and Discussion

3.1 Simulation of the EPICUR Experiments with RAIM

In order to validate the developed model, chemical behaviors of iodine species in the selected EPICUR experiments are simulated with RAIM. Experimental conditions are described in the parametric input file.

During the S2-6-9 experiment which represents the S2 organic iodide tests, volatile organic iodide was produced from iodine molecules deposited on the organic painted coupon under 80°C, 20% relative humidity [5]. Fig. 3 shows the concentrations of gaseous volatile iodine species as a function of time. The calculated concentration yield errors are estimated to be 8% for volatile inorganic and 3% for organic iodide. Calculated reaction kinetics of volatile inorganic iodide is much faster than measurement until 4 hours and reaches at the equilibrium whereas volatile iodide was consistently produced in the measurement after 4 hours (left of Fig. 3). Meanwhile, the calculated reaction kinetics of volatile organic iodide is slower than measurement until 4 hours and similar with measurement after 4 hours (right of Fig. 3). This comparison shows that the RAIM code calculates the yields with high accuracy; however, the reaction kinetics is estimated with relatively low accuracy. The errors for reaction kinetics seem to be due to the rate constants of RAIM. Therefore, the improvement of reaction kinetics in the RAIM code is recommended.

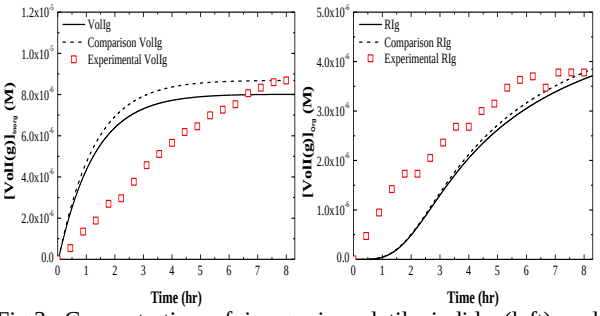

Fig.3. Concentration of inorganic volatile iodide (left) and organic volatile iodide (right) as a function of time for S2-6-9 experiment. Comparison of calculated results (line) with measurements (red box symbol).

3.2 Validation of the RAIM Code

In Fig. 4, the calculated concentration yields of volatile iodine are presented in comparison with those measured. The calculation yield errors for almost all of the experiments are less than 60% except for volatile inorganic iodide of S1-9 experiment (about 500-600%). It is not clear yet why the calculated yields of S1-9 experiment are exceptionally quite different from the measurements. Meanwhile, the calculation yield errors of volatile organic iodide are less than those of volatile inorganic iodide for all the experiments. Thus, the examination and modification of the rate constants for inorganic iodide reaction are necessary to improve the calculation accuracy for inorganic iodide.

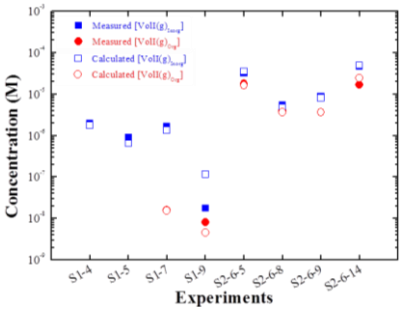

Fig.4. Comparison of calculated (open symbol) volatile iodide yields (red symbol) and inorganic iodide yields (blue symbol) with measurements (closed symbol).

4. Conclusions

Based on the LIRIC methodology, a simple iodine behavior model, RAIM has been developed and validated by the EPICUR experimental data. A reasonable estimation of volatile iodide was made by RAIM for most of the EPICUR experiments. However, relatively large yield errors were found for non-gaseous iodide and chemical kinetics. Therefore, further efforts to improve the accuracy for chemical kinetics should be required.

Acknowledgments

This work is based on the results from the International Source Term Programme operated by the IRSN, SARNET2 WP8 and Long-Term Nuclear Research and Development program of Korea Ministry of Education, Science and Technology.

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