Current Status of a Module for Iodine Behavior in Containment

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1. Introduction

In the case of a severe accident, the molten corium can be relocated to the lower plenum of the reactor vessel. If the reactor vessel is breached, the molten corium will be discharged into the containment environment. The discharge of corium would lead to substantial releases of fission products. A good understanding of iodine behavior is required for the analysis of severe accident consequences because iodine is a major contributor to the potential source term to the environment. Qualified tools for the calculation of the iodine source term are also needed to perform meaningful risk analyses and make decisions in the field of accident management, mitigation measures and emergency procedures. [1]

There exist two basic approaches to modelling of iodine interactions, either mechanistic models [2] or empirical models [3, 4, 5, and 6]. The mechanistic models are important as a scientific tool for studying the basic principles. Currently, the empirical models are those of the practical use since they are being incorporated into big containment codes. For the treatment of the aqueous phase models and mass transfer, there is a great deal of similarities in each codes. [7, 8] However, a different situation exists for the modelling of volatile iodine formation, behavior in the gas phase, and the pH value, etc. Therefore, further efforts in the area of supporting experimental programs and accompanying analytical efforts are necessary.

The final goal of this project is the development of the domestic code based on the current status of iodine chemistry knowledge and tools used for source term prediction. Here, it is described that the current status of the developing a module for iodine behavior in containment and the models.

2. Description of Model

2.1 Construction of chemical kinetics system equation

The processes included in the kinetics model are in two categories which are chemical reactions and non-reaction processes which are not expressed by chemical reaction equation such as adsorption, and desorption, etc.

In the chemical reaction processes, a reaction is can be written as follows,

$$r_1R_1 + r_2R_2 + \cdots + r_nR_n \xrightarrow{k_r} p_1P_1 + p_2P_2 + \cdots + p_mP_m \quad (1)$$

where, k_r is the rate constant of the reaction, R and P are the reactant and product species, k_i, p_i are corresponding stoichiometric coefficients, respectively. Based on the this reaction, the concentration change rate of R_i and P_j can be written as follows,

$$-\frac{d[R_i]}{dt} = \cdots + r_i k_r [R_1]^{r_1}[R_2]^{r_2} \ldots [R_n]^{r_n} + \cdots \quad (2)$$

$$\frac{d[P_j]}{dt} = \cdots + p_j k_r [R_1]^{r_1}[R_2]^{r_2} \ldots [R_n]^{r_n} + \cdots \quad (3)$$

where, *i* is one of 1,2, … , n, and *j* is one of the 1, 2, .., m, respectively. Here [A] means the concentration of A species, the unit is mol/m^3.

In a non-reaction process, the concentration change rate of species Xi is expressed as Eq.4.

$$\frac{d[X_i]}{dt} = \cdots + k_n[X_{n,1}][X_{n,2}] \ldots [X_{n,l}] + \cdots \quad (4)$$

Since all the process in the system in two categories, the equation for the concentration change of a species X_i is expressed as Eq. 5

$$\frac{d[X_i]}{dt} = \sum_r -r_i k_r [R_1]^{r_1}[R_2]^{r_2} \ldots [R_n]^{r_n} + \sum_r p_i k_r [R_1]^{r_1}[R_2]^{r_2} \ldots [R_n]^{r_n} + \sum_n k_n [R_1]^{r_1}[R_2]^{r_2} \ldots [R_n]^{r_n} \quad (5)$$

$$\frac{dX_i}{dt} = f_k(X_1, X_2, \ldots, X_M) \quad (6)$$

The first two terms indicate the chemical reaction processes and the last term indicates the non-reaction processes. Therefore, the change of every concentration is expressed as Eq. 6. Here, M is the number of species included in the system.

2.2 Numerical solution

There are two numerical method which are explicit and implicit method to solve the ordinary differential equation for the system, Eq.6.

In the explicit method, the concentration of the species X_i is obtained by Eq. 7.

$$\frac{X_i^{n+1} - X_i^n}{t^{n+1} - t^n} = f_k(X_1^n, X_2^n, \ldots, X_M^n) \rightarrow X_i^{n+1} = f_k^n dt + X_i^n \quad (7)$$

In the implicit method, the Eq. 8 can be obtained by the 1st order Taylor expansion around the intermediate values.

$$\frac{X_i^p + \delta X_i - X_i^n}{t^{n+1} - t^n} = f_k^p + \sum_{i=1}^{M} \left(\frac{\partial k_k}{\partial X_i}\right)^p \delta X_i \quad (8)$$

In the current states, the solver for the explicit method is developed and the implicit method solver is developing.

2.3 Reaction model

The concept of the iodine behavior modeling is illustrated in Fig. 1. There are aqueous reaction, the gas phase reaction, the gas-aqueous mass transfer, and the aqueous iodine and surface interaction such as wall adsorption/desorption of iodine are involved.

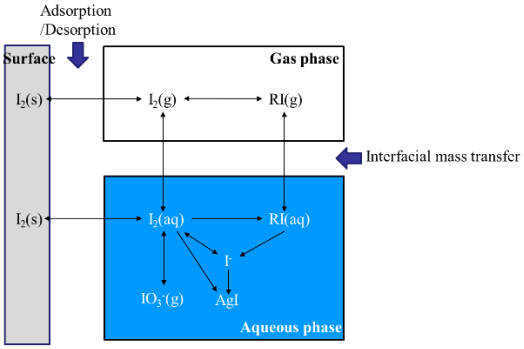

Fig. 1. Concept of the iodine behavior model

In the current, we are dealing with the aqueous phase reactions, because it is the primary reaction field to produce volatile iodine. Also, it is hard to obtain the iodine reactions in gas phase.

In a module of iodine behavior in containment, there are 52 reactions for water radiolysis reaction, 17 reactions for non-reaction, 28 reactions for iodine reaction, and 21 reactions for miscellaneous reactions.

2.4 Compared with experimental results

Jung's[9] experiment used a high gamma radiation dose source with a source activity of ~ 280 kCi. The gamma dose rate was controlled to be within a range of 2.0 to 10 kG.h-1. The I- solutions had various concentration from 1.0 mM to 10.0 mM were exposed to high intensity gamma radiation during the experiment. Separate irradiation experiments were performed to investigate the effect of the gamma dose rate on the oxidation rate of I-. The changing of pH during irradiation time in Jung's experiment was described in Table 1.

Table 1. Change of pH value of NaI solution after gamma irradiation experiments with 2 $kGyh^{-1}$ [9]

Irradiation Time (h)	pH values of NaI solutions after irradiation		
	0.2 mM NaI	1.0 mM NaI	5.0 mM NaI
0	3.04	3.04	3.04
1	3.20	3.20	3.29
2	3.15	3.28	3.50
4	3.16	3.50	5.92
7	3.16	3.93	6.04

We selected 5.0 mM NaI test as a benchmark problem. The experimental results shows in Fig. 2. We are currently conducting numerical analysis. It will be compared with the experimental results.

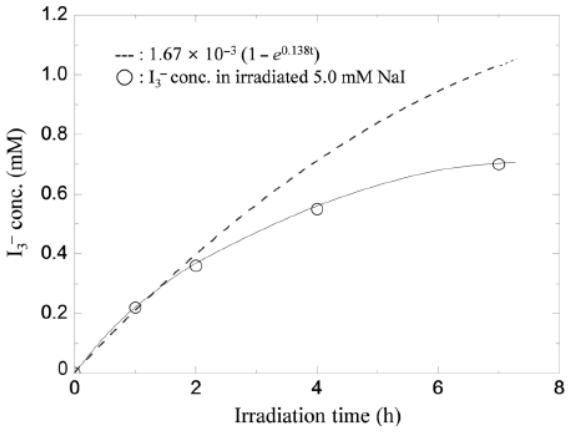

Fig. 2. I^{3-} concentration [9]

2.5 Future works

The current module is the initial version. A lot of effort is needed to make improvements. Now the solver is modifying to save the calculation time. The organic iodine reaction and the reactions for gas phase will be added. Also, some rate constant which are the surface interaction or mass transfer will be modified through experiments if it is needed.

3. Conclusions

A simulation code MIC(Module for Iodine behavior in Containment) is developing for analysis of chemical kinetics in the containment during a severe accident. There are aqueous reaction, the gas phase reaction, the gas-aqueous mass transfer, and the aqueous iodine and surface interaction such as wall adsorption/desorption of iodine are involved. Currently, more than 100 reactions are included.

The current module is the initial version. A lot of effort is needed to make improvements. Among them, some rate constant which are the surface interaction or mass transfer will be modified through experiments if it is needed.

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