

Estimation of the Gaseous Iodine Concentration in the Containment of Phebus FPT-1 using MELCOR1.8.5 with a Revised Organic Iodide Model

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1. Introduction

Under a severe accident, organic iodide starts to be generated in the late phase of a severe accident. Due to its difficulties in a removal by a spray system or a filtering, it is important to estimate the level of the equilibrium concentration of the gaseous iodine in the containment. The purpose of this study is to predict the gaseous iodine concentration in the containment using MELCOR and validate the calculation results against the Phebus FPT-1 data [1].

The organic iodide formation is mainly due to the reaction of I_2 with organic radical ($\bullet R$) produced from the reaction of dissolved organic impurities with water radiolysis products. The organic iodide starts to be generated in the late phase of a severe accident. But it cannot be removed easily either by a spray operation or a filtering due to its physical features. Therefore, in terms of the source terms, it is important to predict the amount of organic iodide concentration in the containment well.

The purpose of this study is to predict the variation of the gaseous iodine concentration in the containment for the Phebus FPT-1 test with MELCOR1.8.5. But the current MELCOR1.8.5 does not consider the production of organic iodide from the pool. Therefore, 43 relevant equations that were mentioned in MELCOR manuals were implemented [2]. In the previous study, the implemented MELCOR under-predicted the gaseous iodine in the containment for the Phebus FPT-1 test [3]. Especially the amount of organic iodide was largely under-predicted compared to the Phebus FPT-1 experimental data.

However, in a previous study, there were two problems for simulating the production of organic iodide from Phebus FPT-1 test. The first one was that the vapor pressure for CH_3I was not applied. The second one was that only CH_3I was considered as gaseous iodine. In this study, these two problems was revised and how to use the buffer material to control the pH in the pool was introduced for an accident management.

2. Methods and Results

In this section, the gaseous iodine concentration in the containment being predicted using MELCOR is shown and how to use the buffer material to control the pH in the pool is introduced.

2.1 Implementation of Organic model into MELCOR

The current MELCOR1.8.5 does not include the equations for an organic iodine generation from the pool. Therefore, 43 relevant equations were implemented into the pool chemistry model in the MELCOR. The exponential increasing mode of the HCl generation in the original MELCOR was modified to increase as a linear mode. Also a new function so that the new species in the pool can be printed out and plotted was implemented into the MELCOR. Figure 1 shows the variation of the concentration versus time for the new species of $CH_3I_{(aq)}$ dissolved in the pool.

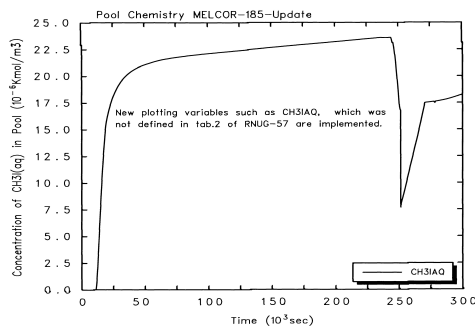


Fig. 1. Concentration of the new species 'CH3IAQ'

2.2 Update of the vapor pressure curve for the methyl iodide (CH_3I)

The methyl iodide vapor pressure curve from the ChERIC was incorporated into the MELCOR using a sensitivity card. Figure 2 shows the vapor pressure curve for the methyl iodide [4].

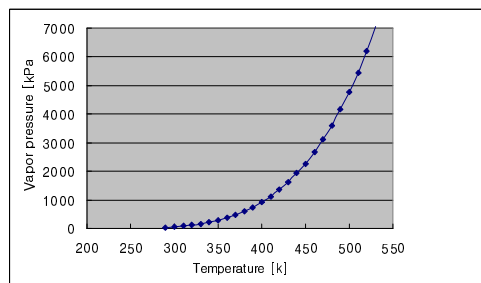


Fig. 2. Vapor Pressure for the methyl iodide (CH_3I)

2.3 Revised chemical form of the methyl iodide

The organic iodide formation is mainly due to the reaction of I_2 with organic radical ($\bullet R$) produced from the reaction of dissolved organic impurities with water radiolysis products. The rate constant of the reaction of I_2 with organic radical ($\bullet R$) and RI with e^- in water are reasonably well established but the hydrolysis rate depends on the type of RI , temperature and pH. Also the partition coefficient between the aqueous and gas phases varies considerably with the type of RI and temperature [5]. In a previous study, only CH_3I was considered as organic iodide that can be partitioned between the water and gas phases. But it is expected that other types of organic iodide may exist.

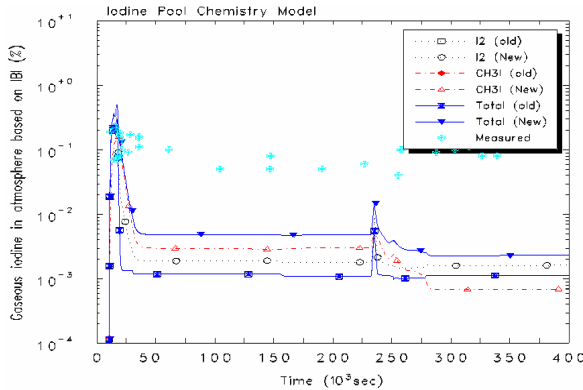


Fig. 3. Gaseous iodine (I_2+RI) concentration in the containment for Phebus FPT-1 test

Therefore, all the RI was considered as organic iodide. Although a different partition coefficient should be applied for every type of RI , only the partition coefficient for CH_3I was assumed as the representative coefficient.

Figure 3 showed the percentage of gaseous iodine mass (new) based on the initial bundle inventory (1.12×10^{-3} kg) were predicted closer to the measured data.

2.4 Buffer material use to control pH

A buffer is a solution that is resistant to changes in the pH. The desired pH level determines what compounds are used to make the buffer. In the Phebus FPT-1 experiment, the initial pH was set as 5.62. Therefore, to make a buffer solution with an acidic pH, the solution was made by adding boric acid of 0.388 M (molarity) as a weak acid and sodium hydroxide of 2.225×10^{-4} M as a soluble salt to the pool of 105 L. The initial pH ($= -\log [H^+]$) was predicted by the following cubic equation and the initial pH of 5.62 was predicted under the initial pool temperature of 362 K [6].

$$[H^+]^3 + [H^+]^2(K_a + [B]) + [H^+](B)K_a - [HA]_0K_a - K_w - K_aK_w = 0$$

$[B]$, $[HA]$ are the concentration of a strong base and weak acid respectively. K_a , K_w are the dissociation constant of a weak acid and water.

Figure 4 shows the comparison of the variation of the pH in the pool. For the case of not using the buffer solution (base), the amplitude of pH change was almost 5. But although the strong acid was introduced during the experiment, the case of using the buffer solution (buffer), the amplitude of pH change was only less than 1.

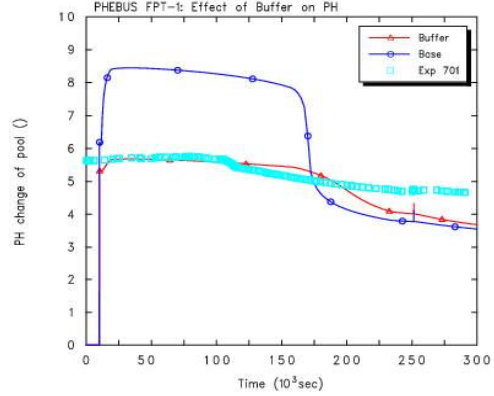


Fig. 4. pH change in the pool with/without the buffer solution

3. Conclusions

The vapor pressure curve for CH_3I was incorporated into the MELCOR. All the RI (organic iodide) such as CH_3I^+ , CH_3OI were considered as organic iodide. The partition coefficient for the CH_3I was assumed as the representative coefficient for all the types of RI .

Consequently, the predicted total amount of gaseous iodine becomes closer to the measured data. For the case of not using the buffer solution, the amplitude of the pH change was almost 5. But in the case of using the buffer solution, although a strong acid was introduced, the amplitude of the pH change remained within only 1.

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