Sensitivity Analyses of Gaseous Iodine Retention in Pool Scrubber

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1. Introduction

The amount of iodine created inside the fuel during a power plant operation is relatively small with regards to other fission products; however, the total iodine activity is one of the most important among other the fission products, that is, iodine isotopes produce a large amount of decay heat (roughly 50% of the total decay heat) [1]. During a severe accident, it is possible that the radioactive materials including the iodine may be released into the containment, and finally into the environment. The iodine, especially in a vapor form, is a major contributor to the potential source term that can be released into the environment.

The iodine interacts variously with other materials such as solid, liquid, and radiation under severe accident conditions, and the interaction processes are very complicated. Temperature and other controlling thermal-hydraulic parameters govern the transfer of iodine between the aqueous and gas phase and to the surfaces. Several homogeneous and heterogeneous reactions have a significant dependence on the gas and water temperatures. The I_2 adsorption on the surfaces additionally depends on the gas flow velocity along the walls. Under wet conditions, the wall condensation rate drives the I_2 diffusion toward the surfaces. Other important boundary conditions for many reactions are indirectly dependent on the thermal hydraulics and aerosol behavior. A crucial parameter directly affecting most of the reactions is the local or bulk dose rate that depends on the fission product concentrations in the containment atmosphere, on the walls and in the sump [1].

For the gaseous iodine retention in a pool such as a wet-type FCVS (Filtered Containment Venting System), the sump was evaluated. A pool scrubbing code was prepared using the current iodine retention models, and sensitivity studies on the iodine retention were performed, such as the pool temperature, vapor velocity in a bubble, and iodine concentration.

2. Description of Models and Results

Once the gaseous iodine has been injected into the water pool, the retention capacity of iodine species in an aqueous environment is evaluated through a decontamination factor (DF), which is defined as the ratio of the mass entering to the mass exiting the pool. On the other hand, the concentration of iodine in the liquid phase may be changed by several chemical reactions and physical processes (diffusion and mass transfer). Consequently, it is affected by the boundary conditions for the iodine removal processes in the pool.

2.1 Pool Scrubbing Phenomena

When the gaseous iodine passes through a water pool from the submerged inlet hole to the pool surface, the hydrodynamic processes can be modeled in the pool scrubbing code, as shown in Fig. 1. Three difference regions are considered, that is, an injection region at the outlet of the injection, bubble rise, and surface region.

In the injection region, the entry gaseous flow through the vent rapidly reaches equilibrium to the pool temperature, and is broken into small bubbles. Under this condition, it may lead to a part of the water vapor being condensed, and therefore some of the iodine would be captured in this region. The decontamination factor of the iodine owing to the early condensation is defined as the ratio of non-condensable gas molar fraction under equilibrium conditions to the entry gas [2, 3, 4].

In the bubble rise region (swarm region), a uniform bubble size and spherical shape are assumed and no interaction between bubbles is allowed. The iodine transfer process is taken place in a single bubble, and this region is considered to calculate gaseous iodine retention and perform sensitivity analyses using the developed pool scrubbing code.

As the bubbles reach the pool surface, most of them break up and this break-up process produces droplets, and small iodine gases in the pool might be carried away into the environment. However, this region has not been considered in this study.
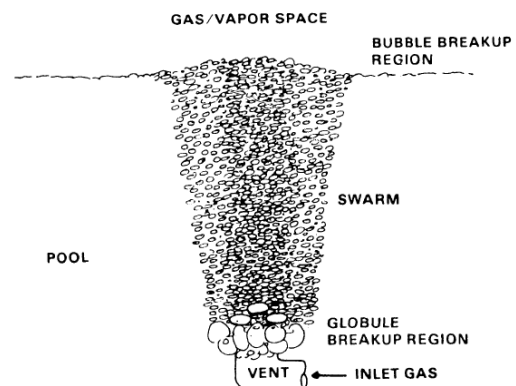
Fig. 1. Schematic of suppression pool during scrubbing of inlet gases [2]

2.2 Model on the Iodine Pool Scrubbing

The iodine mass transfer during a bubble rise is a recurrent problem because this parameter has a direct effect on the concentration of the gaseous iodine species at the gas-water interface. However, the iodine mass transfer at the interface is described by the two-film theory (this concept is used in most of the codes, such as the SPARC and BUSCA codes [3, 4]). The two-film theory supposes that the rate of absorption of iodine species is controlled by the diffusion rate of the iodine concentration through the surface film on the gas-liquid boundary, which is governed by a difference in the partial pressure of iodine species between outside the film (at the gas side) and inside (at the liquid side). In addition, the equilibrium concentration of the iodine species at the interface is considered [5]. Thus, the overall diffusion rate of the species, V_d, is evaluated through equation (1) [6].

$$V_d = V_{d,g} \frac{[I_2]_g - [I_2]_g^{int}}{[I_2]_g} \tag{1a}$$

$$V_{d,g} = \left(\frac{D_g V_r}{\pi D_b}\right)^{\frac{1}{2}} \tag{1b}$$

Under the assumption of mass conservation in equation (2a), the concentration of gaseous I_2 on the gas-liquid interface can be estimated, as shown in equation (2b) [6].
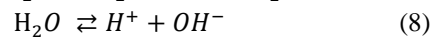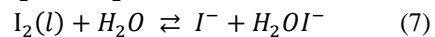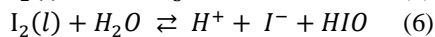
$$\sum[I_{2(aq)}] = 0.5[I^-] + 1.5[I_3^-] + [I_{2(aq)}] + 0.5[HIO] + 0.5[H_2OI^+] \tag{2a}$$

$$[I_2]_g^{int} = \frac{[I_2]_l^{eq} V_{d,l}^i + [I_2]_g V_{d,g}^i}{H_{I_2} V_{d,l}^i + V_{d,g}^i} \tag{2b}$$

With Henry's constant (or a partition coefficient), H_{I_2}, is a ratio of the gas concentration to the liquid concentration at the interface.

$$H_{I_2} = \frac{\sum[I_{2(aq)}]_{eq}}{[[I_{2g}]_{int}} \tag{3}$$

Considering the equilibrium concentration of I_2 species inside the water phase, a limited set of following chemical reactions is used, as shown in equations (4) through (8).

$$I_2(g) \rightleftarrows I_2(l) \tag{4}$$
$$I_2(l) + I^- \rightleftarrows I_3^- \tag{5}$$
$$I_2(l) + H_2O \rightleftarrows H^+ + I^- + HIO \tag{6}$$
$$I_2(l) + H_2O \rightleftarrows I^- + H_2OI^- \tag{7}$$
$$H_2O \rightleftarrows H^+ + OH^- \tag{8}$$

And the decontamination factor in the bubble rise region is defined by equation (9) [6].

$$DF_i = \exp\left(\frac{\Delta t}{V_b}\int S_b V_d dA\right) \tag{9}$$

Using the above models, the iodine pool scrubbing code was developed by the C++ language to calculate the DF of the gaseous iodine in a bubble rising zone of the pool. (Fig. 2)
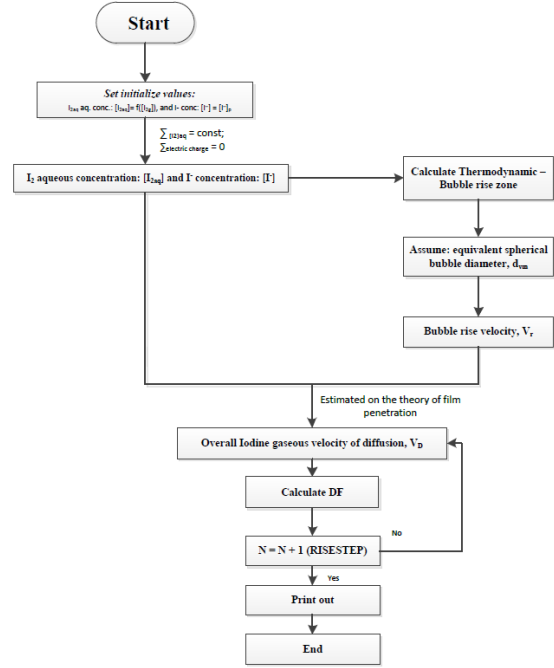

Fig. 2. Flow chart for DF calculation of iodine gas

2.3 Sensitivity Analyses of Gaseous Iodine Retention in Pool

Sensitivity analyses on the LACE-ESPANA experiments [3] were performed to verify the developed iodine pool scrubbing code. Table I shows initial conditions of the experiments. Sensitivity analyses of DFs were performed according to the pool temperature, vapor velocity in a bubble, and injected iodine concentration.

The calculated DFs of the LACE-ESPANA experiments are summarized in Table II. The DF results obtained by the current study are half of the values by the SPARC code. This discrepancy seems to be due to the differences of the bubble shape and vapor velocity in the codes. That is, the SPARC code assumes that the bubble has an ellipsoidal shape, but our code is assumed as a spherical bubble. Compared with the BUSCA code, the calculated DFs were smaller than the BUSCA results; this is why BUSCA considers no chemical consideration and assumes a null iodine concentration in the water phase.

Table I: Initial conditions for sensitivity analysis on the LACE-ESPANA experiments

Inlet gas temperature (^{o}C)	150
Inlet gas pressure (atm)	2.25
Pool Temperature(^{o}C)	25; 100
Pool Surface pressure (atm)	1.973
Pool volume (m^{3})	3.53
Volumetric inlet flow (cc/s)	400
Submergence (cm)	200
bubble diameter (cm)	0.8
Air flow(no steam) (g/s)	0.752
Iodine flow and final concentration: 9.8x10^{-7} mol/s (10^{-6} M) and 9.8x10^{-5} mol/s (10^{-4} M)	

Fig. 3 shows the gaseous iodine DF according to the pool iodine concentration. As shown in Fig. 3, in the case of a low iodine concentration (10^{-6}M) in the pool, the concentration of iodine in the aqueous phase is reduced, and this low concentration of iodine in a pool leads to an increase in the coefficient of partition at the liquid-gas interface, and this finally leads to a reduction of the gaseous iodine concentration of the interface. This means that the decontamination factor increases. However, in the case of a high concentration (10^{-4} M), because the amount of I$_2$ in the pool is large, the transport of iodine from the interface to the pool is not very effective. This leads to a larger accumulation of gaseous iodine on the interface and finally to a lower decontamination factor.

As shown in Figures 3 through 5, the gaseous iodine DF decreases as the vapor velocity in the bubble increases. The vapor velocity has an opposite direction compared with the diffusion velocity of gaseous iodine. Therefore, the vapor velocity increases, and the net diffusion velocity of gaseous iodine from the center to the interface of the bubble decreases, and the DF then finally decreases.

Table II: Results for iodine retention analysis

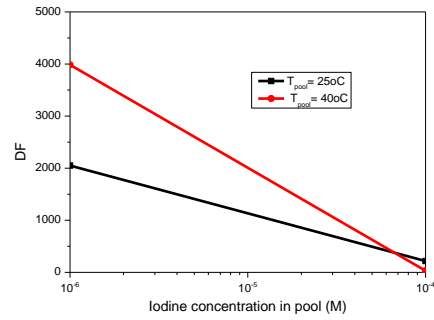| Vapor velocity (cm/s)	T$_{pool}$ (^{o}C)	Pool Iodine Conc. (M)	Cal. DF	SPARC DF[3]	BUSCA DF[3]
0.2	25	10^{-4}	216.25	550.93	12970
	100	10^{-4}	31.64	32.86	3371
	25	10^{-6}	2049	3800	12880
	100	10^{-6}	inf	56796	3369
10.0	25	10^{-4}	197.1	550.93	12970
	100	10^{-4}	29.1	32.86	3371
	25	10^{-6}	1413.0	3800	12880
	100	10^{-6}	inf	56796	3369

Fig. 3. Gaseous iodine DF according pool iodine concentration (vapor velocity=0)
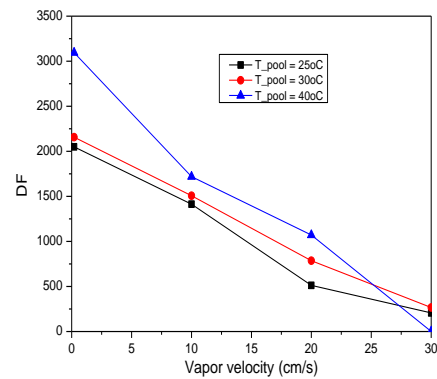

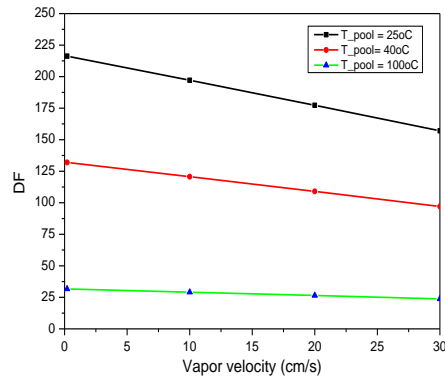Fig. 4. Gaseous iodine DF according to vapor velocity ([I$_{2(aq)}$] = 10^{-6} M)

Fig. 5. Gaseous iodine DF according to vapor velocity ([I$_{2(aq)}$] = 10^{-4} M)

The pool temperature is also considered as one of the sensitivity parameters of the DF because the pool temperature affects the chemical reactivity of iodine in the pool, the partition coefficient, and the diffusion velocity. As shown in Figs. 3 and 4, the efficiency of the hydrolysis increases, and finally the decontamination factor increases at the low iodine concentration (10^{-6}M) of the pool as the pool temperature increases. However, as shown in Figs. 3

and 5, the efficiency of hydrolysis is reduced owing to the reduction of the partition coefficient, and finally the decontamination factor decreases at a high concentration (10^{-4} M) as the pool temperature increases [3].

3. Conclusions

A pool scrubbing code was prepared using the iodine retention models, which was similar to the SPARC code, and sensitivities studies on the iodine retention were performed such as iodine pool concentration, vapor velocity in a bubble, and pool temperature.

The pool temperature is considered one of the sensitivity parameters of the DF because the pool temperature affects the chemical reactivity of iodine in the pool, the partition coefficient, and the diffusion velocity. The decontamination factor increases at a low iodine concentration (10^{-6}M) of the pool as the pool temperature increases. However, the decontamination factor decreases at a high concentration (10^{-4} M) as the pool temperature increases.

NOMENCLATURE

DF	decontamination factor
D_g	Gas diffusion coefficient (cm^2/s)
D_b	Bubble diameter (cm)
H_{I2}	Partition coefficient
V_r	Bubble rising velocity (cm/s)
V_b	Volume of bubble (cm^3)
$V_{d,g}$	Diffusion rate of species I$_2$ across the gas in the bubble (cm/s)
V_d	Overall diffusion velocity (cm/s)
S_b	Surface of bubble (cm^2)
$[I_2]_g$	Concentration of species I$_2$ in the bubble [M]
$[I_2]_g^{int}$	Concentration of species I$_2$ on the gaseous side of the gas-liquid interface [M]
$[I_2]_l^{eq}$	Concentration of species I$_2$ in the aqueous phase [M]
$\sum[I_{2(aq)}]$	Total liquid molar concentration of iodine at equilibrium pool [M]

ACKNOWLEDGMENTS

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