CHARACTERISTICS OF FISSION PRODUCT RELEASE FROM A MOLTEN POOL

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ABSTRACT

The volatile fission products are released from the debris pool, while the less volatile fission products tend to remain as condensed phases because of their low vapor pressure. The release of noble gases and the volatile fission products is dominated by bubble dynamics. The release of the less volatile fission products from the pool can be analyzed based on mass transport through a liquid with the convection flow. The physico-numerical models were orchestrated from existing submodels in various disciplines of engineering to estimate the released fraction of fission products from a molten pool. It was assumed that the pool has partially filled hemispherical geometry. For the high pool pressure, the diameter of the bubbles at detachment was calculated utilizing the Cole and Shulman correlation with the effect of system pressure. Sensitivity analyses were performed and results of the numerical calculations were compared with analysis results for the TMI-2 accident.

INTRODUCTION

During a severe accident in the nuclear power plant, as the reactor core material melts and relocates, the molten debris pool may be formed in the lower core or lower head of the reactor pressure vessel. The pool is heated by the radioactive decay of the fission products contained in the melt. Increase of the pool temperature will activate the fission product gas and result in release from the debris pool. The release of noble gases and the volatile fission products is dominated by bubble dynamics in the molten pool, while that of involatile fission products is controlled by mass transport through the pool. Results of the TMI-2 accident analysis indicate that very little fission product release is expected from the rods and debris bed during formation of the molten pool. Rather, fission products can mostly be released from the molten pool. Although cesium and iodine were almost completely released from consolidated region as shown Table 1, small fractions of these fission products remained in molten fuel debris. The reason probably is the existence of chemical forms that are relatively stable at high temperature and very low rate of bubble nucleation due to high pressure. The gas bubbles in intact fuel matrix are very small ($\sim 0.01\,\mu m$). If the matrix is suddenly disrupted, however, these volatile fission gases

can form large bubbles since the restraint of the UO_2 matrix is no longer available to restrict the fission gas volume. These bubbles that are produced prior to formation of the pool initially exist in the pool. The ratio of volatile fission product mass in the initial bubble to pool inventory may be strongly related with released fraction of the volatile fission product as a function of time.

If the pressure and temperature in the pool are high enough, as is the case of a station blackout sequence (TMLB`) for instance, the rate of nucleation and growth of bubbles are lower than at atmospheric system pressure. Thus, depressurization of the pool may significantly affect the nucleation, growth, and escape of the gas bubbles.

Table 1. Fission product distribution in the TMI-2 accident

Fission Product Repositories	Estimated Mass of Region (kg)	Total Core (%)	Fission Product Distribution Percent of Initial Core Inventory				
			^{137}Cs	^{129}I	^{85}Kr	^{106}Ru	^{154}Eu
Initial core[1]	133250	100.0	100	100	100	100	100
Intact fuel rods	44500	33.4	30	30	30	30	30
Upper core debris[2]	26600	19.9	4.3-5.3	5.3-5.9	~ 0-6	0.8-3.8	19-30
Consolidated region[3]	32700	24.5	0.77	2.1	~ 0	2.2-9.0	32
Upper CSA[4]	4200	3.2	0.46	0.12	~ 0	0.22	4.5
Lower CSA	5800	4.3	0.63	0.16	~ 0	0.32	6.3
Lower RPV head	19100	14.3	2.1	0.54	~ 0	1.1	21
Outside the RPV	100	0.1	55	55	54	0.5	~ 0

[1] Initial core mass equals total mass of post-accident core materials
[2] Fission product distribution percent in upper core debris region was detected from two samples
[3] Value of ^{106}Ru was detected in ceramic and metallic regions
[4] CSA signifies the core support assembly

Volatile fission products such as Cs, I, Kr and Xe are insoluble in either solid or liquid UO_2, as well as noble metals such as Mo, Rh and Ru. They all behave as gases in the pool and are released from the pool by means of nucleation of bubble or diffusion to a bubble. Actinide oxides and rare earth oxides are soluble in either solid or liquid UO_2. Materials such as Sr and Ba are soluble in the liquid phase and are able to form eutectic alloys. Release of these materials from the molten pool is controlled by mass transport.

BUBBLE DYNAMICS

The release of noble gases (Xe and Kr) and the volatile fission products (I and Cs) is dominated by bubble dynamics in the molten pool because these highly volatile fission products are gaseous at the high temperature (>2850 K) in the pool. As the pool heats up, fission product molecules are activated and thus the vapor pressure of the volatile fission products increases. If the vapor pressure exceeds the pool

pressure, nucleation of the bubbles occurs. Nucleation of the bubbles may be explained by the homogeneous and heterogeneous nucleation mechanisms. A simple configuration for modeling is shown in Figure 1.

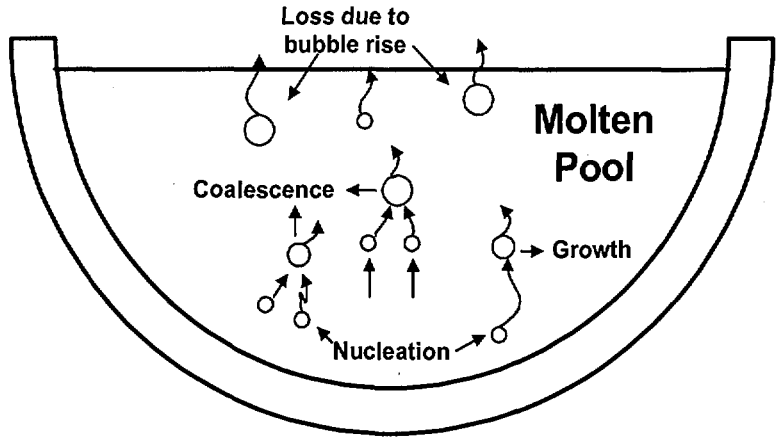

Figure 1. Bubble dynamics in a molten pool

The nucleated bubbles have very small sizes and follow the natural convection flows. These bubbles will grow by coalescence diffusion of vapor molecules to bubbles. Small bubbles coalesce into larger bubbles by turbulence and differential bubble rise in the pool. Bubbles can be released from the pool as they sufficiently grow up. Bubble dynamics in the pool is thus characterized by bubble nucleation, coalescence, growth and rise.

Nucleation of a Bubble

Heterogeneous nucleation of a volatile fission product species will occur when the vapor pressure of the species minus the pool pressure exceeds the surface tension in the bubble-liquid surface:

$$p_g - p_m > (\gamma / R) \tag{1}$$

The number of nucleation sites can be assumed to be proportional to the number of solid particles in the melt. McClure et al. (1993) proposed that the total number of nucleation sites could be represented by summation of temperature-dependent nucleation sites and permanent nucleation sites. The permanent nucleation sites were assumed to exist on temperature-independent surfaces. The number of solid particles in the molten material is assumed to be proportional to the pool mass. The number of sites may be expressed as

$$s_n = (m_p s_{n,t} + s_{n,p}) \tag{2}$$

For small cavity sizes, the bubble size at departure is dictated mainly by a balance between the buoyancy and liquid inertial forces. But, for larger cavity sizes, the bubble size at departure is calculated by a balance of the surface tension and buoyant forces. A well-known equation was proposed by Fritz and Ende (1936) as

$$R_F = 0.0104\theta_0\sqrt{\gamma / g(\rho_l - \rho_v)} \quad (3)$$

This agreed well with the experimental data at atmospheric pressure, but did not concur with the experimental data at super- and subatmospheric pressures. Cole and Shulman (1966) found that, if $R_F = 0.5\sqrt{\gamma / g(\rho_l - \rho_v)}$ for a contact angle of 48°, $\overline{R}_d = R_d / R_F$ is a function of pressure. According to the experimental data, they obtained the following formula

$$\overline{R}_d = \frac{1000}{p} \quad (4)$$

where p is in mm Hg. The nucleation sites emit bubbles with a constant frequency. The frequency can be obtained by the time to grow to the departure diameter by diffusion. The time can be estimated by solving the multi-component diffusion equation. Scriven (1959) proposed the following formulation for diffusion of a species to a sphere of changing radius

$$\frac{\partial C}{\partial t} = D\left(\frac{\partial^2 C}{\partial r^2} + \frac{2}{R}\frac{\partial C}{\partial r}\right) - \frac{R^2}{r^2}\frac{dR}{dt}\frac{\partial C}{\partial r} \quad (5)$$

Solution to Equation (5) is obtained as (Martins and Szekely, 1969)

$$\varsigma = -a\varphi(\beta_b)$$

where

$$\varphi(\beta_b) = 2\beta_b{}^3 \exp(3\beta_b{}^2) \times \int_S^\infty \xi^{-2} \exp(-\xi^2 - 2\beta_b{}^3\xi^{-1}) d\xi \quad (6)$$

$$\zeta = \frac{C - C_m}{C_m} \quad \text{and} \quad a = D\frac{A}{C_m} \quad (7)$$

The growth constant β_b can be obtained from Scriven's useful expression of solutions to Equation (6)

$$\varphi(\beta_b) \sim \sqrt{(\pi/3)}[\beta_b - 4/9 + 0(\beta_b^{-1})] \quad (8)$$

Once β_b is known, the bubble detachment frequency can be calculated as

$$f_d = \frac{1}{t} = \frac{4\beta_b{}^2 D}{R^2} \quad (9)$$

The product of the bubble detachment frequency and the number of nucleation sites determines the rate at which bubbles are formed, viz.

$$\frac{dn_{i,nucl}}{dt} = f_d \cdot s_n \quad (10)$$

Diffusion to a Bubble

Diffusion to a bubble is governed by Equation (5) used to calculate nucleation rate of the bubble. The rate of change of number density for bubble size R_i can be determined by the time to grow from bubble size R_{i-1} to size R_i. The rate of change of a discrete bubble radius R_i is the sum of loss term and production term. The loss term equals the number density of bubbles of size R_i divided by the time to grow from size R_i to size R_{i+1} and the production term is the number density of bubbles of size R_{i-1} divided by the time to grow from size R_{i-1} to size R_i. Therefore, the rate of change of number density for bubble size R_i is represented as

$$\frac{dn_{i,diff}}{dt} = \frac{n_{i-1}}{t_i - t_{i-1}} - \frac{n_i}{t_{i+1} - t_i} \tag{11}$$

Coalescence of Bubbles

Bubbles interact due to their motion and grow by coalescence. The rate of coalescence of a bubble of radius R_k is given by Olander (1976) as

$$dn_k / dt = 0.5 \sum_{i+j=k} B_{ij} n_i n_j - \sum_{i=1} B_{ik} n_i n_k \tag{12}$$

The rate of change of number density for bubble size R_k can also be obtained by summation of production term and loss term. The production term is represented by the rate of formation of bubble size R_k due to collisions of bubbles of sizes R_i and R_j. The loss term is represented by the rate of disappearance of bubble size R_k due to coalescence with bubbles of other sizes. It is assumed that bubble coalescence is caused by two mechanisms, i.e. turbulence in the pool and differential rise velocity of bubbles. For the turbulence process, a correlation for aerosol agglomeration in turbulent pipe flow is used as presented by Friedlander (1977)

$$B_{ij,turb} = 1.3(R_i + R_j)^3 (\varepsilon_d / \nu)^{1/2} \tag{13}$$

where

$$\varepsilon_d / \nu = (2/R_p)(f/2)^{1.5} v_{conv}^3 \tag{14}$$

The convective velocity in the pool can be obtained from an energy balance and is given by

$$v_{conv} = 2Q/(\rho A_{conv} c_p \Delta T), \tag{15}$$

For coalescence due to differential bubble rise velocity, the frequency function is also given by Friedlander (1977) as

$$B_{ij,rise} = \pi(R_i + R_j)^2 |v_i - v_j| \quad (16)$$

For all the fission product release calculations, the molten pool is assumed to be a partially filled hemisphere. The material is (U, Zr)O_2 with a melting point of ~2850 K. It is assumed that the pool is homogeneous.

Loss of Bubbles due to Bubble Rise

The rate at which bubbles leave the pool is proportional to the bubble number density and the rise velocity. The residence time of bubbles of size R_k is given by the pool height divided by the rise velocity. It is assumed that the rate at which bubbles leave the pool equals the number density of bubbles divided by the residence time in the pool. The rise velocity of a spherical gas bubble is found by balancing the drag and buoyant forces on the bubble. Hence,

$$v = 2\rho g R^2 / 9\mu \quad (17)$$

The rate of the loss of bubbles due to buoyant rise can be calculated by:

$$\frac{dn_{i,loss}}{dt} = \frac{v_i n_i}{z} \quad (18)$$

The time rate of change for the bubble concentration may be represented as follows

$$\frac{dn_i}{dt} = \frac{dn_{i,nucl}}{dt} + \frac{dn_{i,coal}}{dt} - \frac{dn_{i,loss}}{dt} + \frac{dn_{i,diff}}{dt} \quad (19)$$

Less-Volatile Fission Product Release

The less volatile fission products tend to remain as condensed phases in the melt because of their low vapor pressures. The chemical forms of the less volatile fission products in the melt are determined by the oxygen potential in the melt. It is assumed that mass transport governs release of the less volatile fission product from the pool. The transported mass to bubbles of involatile fission products would be very low because of their low partial pressures. Rare earth elements such as Eu and Ce exist as oxides, Sr is present as SrO, and Ru and Sb are present as metals immiscible in the molten pool (Petti et al., 1989). Partial pressures of the fission product oxides Eu_2O_3, Ce_2O_3 and SrO in the melt are quite low due to their low vapor pressures and their low mole fractions (<1%) in the pool. Metallic fission products (Ru and Sb) have moderate vapor pressures at 3000 K. Their partial pressures in the pool are also very low because these species are expected to have alloyed with other metallic components.

The rate of mass transport of a species in a liquid is given by

$$M = k_m A_{up} (C_\infty - C_{surf}) \quad (20)$$

The mass transfer rate can be estimated by

$$dC/dt = -M/V_p = -(k_m A_{up}/V_p)(C_\infty - C_{surf}) \quad (21)$$

Diffusion can be estimated by means of a heat and mass transfer analogy. The mass transfer correlations for the top of the pool can be obtained as follows

$$Sh_{up} = 0.36Ra^{0.23}(Sc/\mathrm{Pr})^{0.23} \quad (22)$$

Heat Transfer and Flow in a Molten Pool

The pattern of flow in the pool having heat-generating liquid is depicted by natural convection being governed by a Rayleigh (Ra) number characterizing the relationship between the forces of buoyancy and viscous friction. It is assumed that heat transfer in the pool can be treated with lumped parameter methods without introducing a significant error in the estimation of the pool temperature. Natural convection phenomena can be scaled in terms of the Grashof (Gr), and Praldtl (Pr) numbers. The presence of volumetric heating necessitates use of the Dammköhler (Da) number. Using the best-known correlations, heat transfer is calculated at the curved bottom and the top of the pool. The correlations are summarized below (Mayinger et al., 1976).

$$Nu_{up} = 0.36Ra^{0.23} \quad (23)$$

$$Nu_{down} = 0.54Ra^{0.18}(H/R_p)^{0.26} \quad (24)$$

Decay heat of fission products in the pool can be obtained from mass concentration and conversion factor of each fission product as

$$\dot{Q} = \sum_i M_i(t)\eta_i \quad (25)$$

RESULTS AND DISCUSSION

In this work, the parameters obtained from the analysis report of the TMI-2 accident were used for the calculations of fission product release (Akers et al., 1989). The pool geometry was assumed to be a partially filled hemisphere. The change of pool geometry during the numerical calculation was neglected. Previous study indicates that the diffusion coefficients for Xe, Kr, I, Cs are approximately equal (Iglesias et al., 1978). It was assumed that the diffusion coefficients in the molten pool are also similar in magnitude. Therefore, one single diffusion equation was used for all the volatile fission products.

Sensitivity analyses for release model of the fission product were performed utilizing values and ranges of parameters listed in Table 2. For the variation in the number of temperature-dependent nucleation sites between 1,000 and 30,000, the results for time to release 10% of the inventory are shown in Figure 2(a). Figure 2(b) indicates that variation in the diffusion coefficient yields similar trend in the release time. In a previous study, McClure et al. (1993) determined the parameters for which variations in their values have the greatest impact on the calculated rate of fission product release. These turned out to be the number of nucleation sites in the pool and the diffusivity of the pool. The results presented in Figure 2 agree with their findings.

Table 2. Values and ranges of parameters

Parameter	Value
Pool mass , M_p [kg]	32,700
Pool radius, R_p [m]	1.45
Pool pressure, p [MPa]	0.1 ~ 10.0
Pool velocity, V_{conv} [m/s]	0.13
Number of permanent nucleation sites [site/kg]	500
Number of temperature-dependent nucleation sites [site/kg]	100 ~ 30,000
Diffusivity of fission product, D [m^2/s]	1×10^{-10}~1×10^{-8}
Surface tension of liquid in pool, γ [N/m]	0.5 ~ 1.6

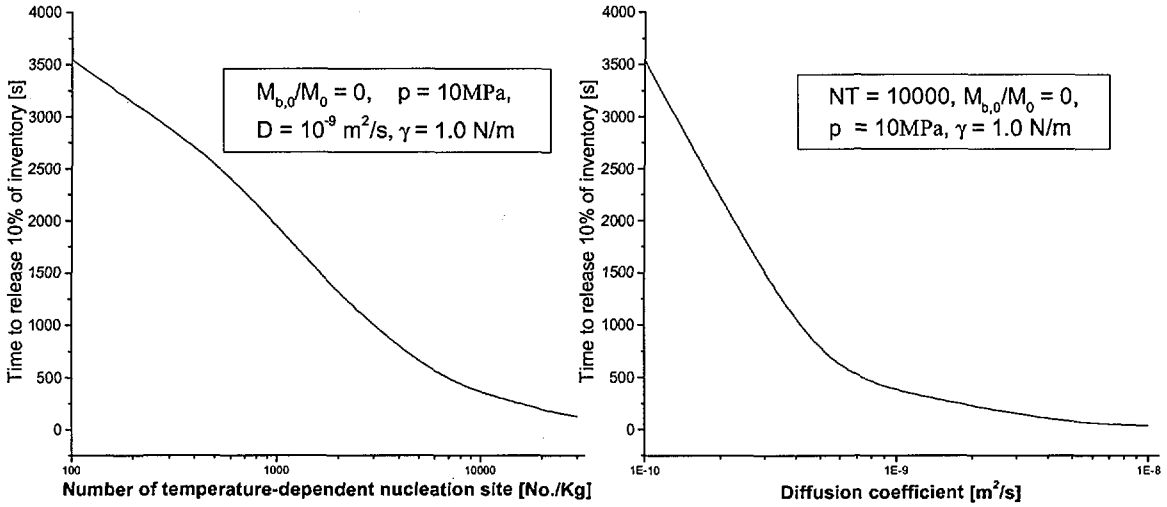

Figure 2.
(a) Release time as a function of number of temperature-dependent nucleation sites
(b) Release time as a function of the diffusion coefficient

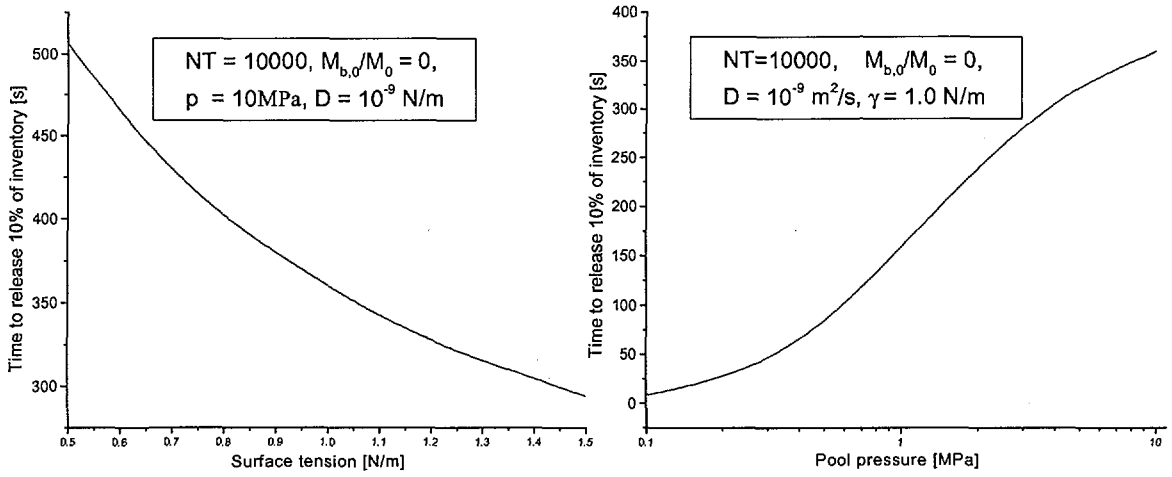

Figure 3.
(a) Release time as a function of the surface tension
(b) Release time as a function of the pool pressure

The effect of fission products on the released fraction was estimated for the surface tension of the pool and the pool pressure. The size of a nucleated bubble varies with surface tension. Pursuant to Equation (3), the bubble size at detachment R_d is proportional to $\sqrt{\gamma}$. In addition, growth to a larger bubble is affected by increase in the surface tension. Figure 3(a) shows that time to release 10% of inventory is slightly shorter than that which is proportional to $\sqrt{\gamma}$.

Bubble radius at detachment is determined with the Cole and Shulman (1966) correlation. If the pool pressure is 10 MPa, the initial bubble radius with the Fritz and Ende (1936) equation is up to 3 ~ 71 times that calculated with the Cole and Shulman correlation. Also, diffusion to a bubble is significantly reduced than at the atmospheric pressure. As depicted in Figure 3(b), the times to release 10% of the pool inventory are 8.41 s at atmospheric pressure and 360 s at 10 MPa. These results suggest that the pool pressure has the predominant impact on the fission product release.

When the initial mass ratio varies between 0 and 0.5, the times to release 90% of the pool inventory were estimated with ns_t=10^4, P_m=10 MPa, D=10^{-9} m^2/s and γ= 1 N/m. In this study, the time to release 90% of the total inventory was estimated to be greater than 1,500 s. Petti et al. (1989) suggest that most of the gas would be released from the pool very quickly, say, within 5 min, on the other hand. In Table 1, relative released fraction of Cs from the molten pool, considering the mass fraction to the total core in each region (upper debris bed and consolidated region), is about 88% of the initial inventory. Figure 4 shows that results of calculation with the initial mass ratio of 0.3 agree with the accident scenario of TMI-2. During the TMI-2 accident, the molten pool has formed in consolidated region between 180 and 224 min after reactor shutdown. After about 44 min from the beginning of pool formation, crust of the pool has failed. The result of calculation with the initial mass ratio of 0.2 indicates that 88% of Cs inventory is released at time ~2600 s.

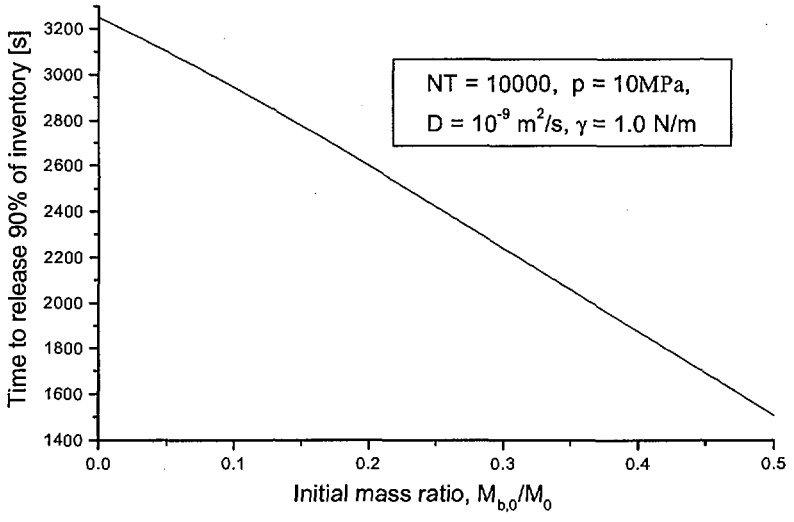

Figure 4. Release time as a function of the initial mass ratio

The rate of mass transfer due to difference of mass concentration is much smaller than that for diffusion to a bubble. The mass of less volatile fission products in the

bubbles was very small because of their low partial pressures. Therefore, very little release of the less volatile fission products was calculated because of the low partial pressures of these species in the molten pool. These results agree with the retention data of the less volatile fission products in the TMI-2 accident

CONCLUSIONS

Based on the numerical analysis of fission product behavior, following conclusions can be drawn concerning the release of fission products. A large fraction of volatile fission products would be released from the molten pool. It is concluded that time to release 90% of inventory takes more than 1,000 s. As can be seen from the sensitivity analysis results, the change in the pool pressure significantly affects the nucleated bubble radius, rate of bubble growth, and fission product release. Therefore, the change in the system pressure during formation of the molten pool should be considered for the fission product release from the pool. The release of fission products would be mainly governed by the number of nucleation sites in the pool, surface tension of liquid in the pool, diffusivity of fission products in the pool and pressure in the pool. Results of calculation with the mass ratio of 0.35 agree with the accident scenario and Cs retention data for the TMI-2 accident.

NOMENCLATURE

A	= area of bubble surface	$[m^2]$
A_{conv}	= area for convection	$[m^2]$
B	= coalescence frequency function	$[m^3/s]$
C	= fission product concentration in the pool	$[No./m^3]$
c_p	= heat capacity	$[W/mK]]$
D	= diffusivity of the fission product in the pool	$[m^2/s]$
f	= Fanning friction factor (0.004)	
f_d	= bubble detachment frequency	$[s^{-1}]$
H	= pool depth	$[m]$
k	= thermal conductivity	$[W/m \cdot K]$
n	= number density of bubble	$[No./m^3]$
ns	= total number of nucleation site	$[No.]$
ns_p	= number of permanent nucleation sites	$[No.]$
ns_t	= number density of temperature-dependent nucleation sites	$[No./kg]$
p_g	= pressure of gas bubble	$[N/m^2]$
p_m	= pressure of the pool	$[N/m^2]$
$\dot{Q}$	= volumetric heat generation rate	$[W/m^3]$
q	= average heat flux over a boundary	$[W/m^2]$
R	= radius of bubble	$[m]$
R_d	= radius of bubble at departure	$[m]$
R_P	= radius of the molten pool	$[m]$
T	= temperature of the pool	$[K]$

ΔT	= pool superheat	$[K]$
V_P	= volume of the pool	$[m^3]$
v	= velocity of bubble	$[m/s]$

Greek Letters

α	= thermal diffusivity	$[m^2/s]$
β	= thermal expansion coefficient	$[K^{-1}]$
β_b	= growth constant of diffusion to a bubble	
γ	= bubble-melt interfacial tension	$[N/m]$
η	= heat generation per unit mass	$[W/kg]$
ε_d	= eddy diffusivity	$[m^2/s^3]$
κ	= gas constant (Boltzmann constant)	$[J/K]$
θ_0	= contact angle of nucleating bubble	[degree]
ν	= kinematic viscosity	$[kg/m \cdot s]$
ρ_m	= density of the pool	$[kg/m^3]$
μ	= viscosity of the pool	$[N \cdot s/m^2]$

Subscripts

i	= bubble size bin i
j	= bubble size bin j
k	= bubble size bin k
0	= initial value or nominal value
∞	= value in the bulk
coal	= coalescence
conv	= convection
diff	= diffusion
nucl	= nucleation
loss	= loss due to bubble rise
surf	= surface

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