

Analysis of Fission Product Release and Transport in A Hemispherical Molten Pool with Crust

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Abstract

Heat transfer and fluid flow in a molten pool are influenced by internal volumetric heat generated from the radioactive decay of fission product species retained in the pool. The pool superheat is determined based on the overall energy balance that equates the heat production rate to the heat loss rate. Decay heat of fission products in the pool was estimated by product of the mass concentration and energy conversion factor of each fission product. For the calculation of heat generation rate in the pool, twenty-nine elements were chosen and classified by their chemical properties. The mass concentration of a fission product is obtained from released fraction and the tabular output of the ORIGEN 2 code. The initial core and pool inventories at each time can also be estimated using ORIGEN 2. The released fraction of each fission product is calculated based on the bubble dynamics and mass transport. Numerical analysis was performed for the TMI-2 accident. The pool is assumed to be a partially filled hemispherical geometry and the change of pool geometry during the numerical calculation was neglected. From the results of numerical calculation, peak temperature of molten pool significantly decreased and volatile fission product was almost released from the molten pool during the accidents.

1. Introduction

A severe accident management concept known as "in-vessel retention" is based on the idea that the vessel lower head will be able to arrest the downward relocation of a degraded core. Lower head integrity can be maintained through the removal process of generated heat. Heat generation in relocated material is mainly caused by radioactive decay of fission products. As the reactor core material melts and relocates, the molten debris pool may be formed in the lower plenum of the reactor pressure vessel or the lower core. If fission product is released from the molten pool, heat source in relocated material may be decreased significantly. Results of the TMI-2 accident analysis indicate that volatile (e.g., I, Cs, Xe) fission products can mostly be released from a molten pool. In the TMI-2 accident, a pool of molten core material was formed and grown within the consolidated region(Akers, et al., 1989).

The chemical state of the fission products (i.e. vapor pressure) will depend on the temperature and the oxygen potential. During the formation and growth of a pool, the release of volatile fission products would be dominated by bubble dynamics as they all behave as gases. On the other hand the release of non-volatile fission products is controlled by mass transfer because they exist as condensed phases in the pool (Iglesias, et al., 1999). The rate of fission product release is calculated using bubble dynamics and mass transfer. For the calculation of heat generation rate in the pool, twenty- nine (29) elements were chosen and classified (see Fig. 1). The change of these fission products due to decay chain can be obtained from the tabular output of ORIGEN 2. Heat transfer in the pool was treated with the lumped parameter model. The effect of fission product release

on pool temperature decrease was estimated with the energy balance. The energy balance, heat transfer and fluid dynamics within the molten pool are described in the next section.

2. Heat Transfer and Flow

The geometry of a molten pool is assumed to be a partially filled hemisphere. The material is (U, Zr)O₂ with a melting point of ~2850 K. It is assumed that the pool is well mixed. Heat transfer and fluid flow in an oxidic pool are induced by internal volumetric heat generated from the radioactive decay of fission product species retained in the pool. The pattern of flow in the pool having a heat-generating liquid is depicted by natural convection being governed by a Rayleigh number characterizing the relationship between the forces of buoyancy and viscous friction. If the pool is deep enough, a stable natural-convection current can be formed. Kulacki and Goldstein (1977) suggested that convective mixing of the fluid produces a temperature profile that is axially and radially uniform, except for thin laminar boundary layers at the top and at the bottom. Therefore, 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 Prandtl (Pr) numbers. The presence of volumetric heating necessitates use of the Dammköhler (Da) number. These numbers are expressed, respectively, as

$$Gr = \frac{g\mathbf{b}(T_{\max} - T_i)H^3}{\mathbf{n}^2}, Pr = \frac{\mathbf{n}}{\mathbf{a}}, Da = \frac{\dot{Q}H^2}{k(T_{\max} - T_i)} \quad (1)$$

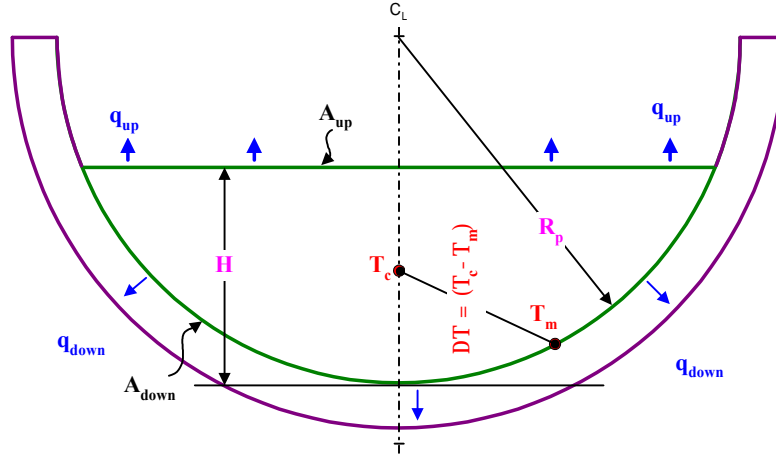


Fig. 1 Schematic of the Physical Model

The Rayleigh number is given by $Ra = Gr \cdot Pr \cdot Da$. The behavior of overall heat transfer can be characterized by a correlation in the form of

$$Nu = f(Gr, Pr, Da) = C_A Ra^{C_B} \quad (2)$$

where C_A and C_B are empirically determined constants, and

$$Ra = \frac{\mathbf{b}g\dot{Q}H^5}{\mathbf{n}ak} \quad (3)$$

Using the best-known correlations, heat transfer is calculated at the curved bottom and the top of the pool. The correlations are summarized below (Mayering et al., 1975).

$$Nu_{up} = 0.36Ra^{0.23} \quad (4)$$

$$Nu_{down} = 0.54Ra^{0.2}(H/R_p)^{0.25} \quad (5)$$

The schematic of the physical model is shown in Fig. 1. The overall energy balance that equates the heat production rate to the heat loss rate is

$$V_p\dot{Q} = A_{up}q_{up} + A_{down}q_{down} \quad (6)$$

where A_{up} and A_{down} are surface areas of the partially filled hemispherical geometry in each direction. Substituting equations (4) and (5) into equation (6) to eliminate q_{up} and q_{down} , the pool superheat ΔT may readily be calculated as

$$\Delta T = \frac{HV_p\dot{Q}}{k} [0.54A_{up}(Ra)^{0.2}(H/R)^{0.25} + 0.36A_{down}(Ra)^{0.23}] \quad (7)$$

Decrease of decay heat in the pool results from fission product release. Decay heat (i.e. heat source in the pool) of each species equals product of mass and energy conversion factor. Total decay heat in the pool is calculated as

$$\dot{Q} = \sum_i M_i(t)\mathbf{h}(i) \quad (8)$$

For twenty-nine (29) elements only, heat generation rate in the pool is calculated. Decay power fraction of the remaining elements except the 29 elements are less than 1%. At time t , the initial mass concentration of fission product i in the pool can be obtained by

$$M_{i,0}(t) = [M_{i,j} + (M_{i,j+1} - M_{i,j})\frac{(t-t_j)}{(t_{j+1}-t_j)}](1-f_c)(m_c/m_p) \quad (9)$$

In equation (9), $M_{i,j}$ and t_j can be obtained from the tabular output of the ORIGEN 2 code. Using ORIGEN 2, the initial core and pool inventories at each time can be estimated. With consideration of released fraction of fission product i at time t , the mass concentration of fission product i is estimated as follows

$$M_i(t) = M_{i,0}(t)(1-f_p(t)) \quad (10)$$

The thickness of crust surrounding the molten pool can be roughly estimated by considering the steady-state molten pool and crust model illustrated in Fig. 2.

The mini-ACOPO experiment has shown that the local heat flux varies considerably along the lower boundary of the molten pool (Theofanous et al., 1994). The local heat flux $q_{down}(\mathbf{q})$ as a function of annular position from the centerline is presented in Fig. 2. From the results of the mini-ACOPO experiment, the local heat flux $q_{down}(\mathbf{q})$ is obtained from q_{down} and the shape function as follows

$$\frac{Nu_{down}(\mathbf{q})}{Nu_{down}} = 0.1 + 1.08\left(\frac{\mathbf{q}}{j}\right) - 4.5\left(\frac{\mathbf{q}}{j}\right)^2 + 8.6\left(\frac{\mathbf{q}}{j}\right)^3 \quad (11)$$

when $0 \leq \mathbf{q}/j \leq 0.6$

$$\frac{Nu_{down}(\mathbf{q})}{Nu_{down}} = 0.41 + 0.35\left(\frac{\mathbf{q}}{j}\right) + \left(\frac{\mathbf{q}}{j}\right)^2 \quad (12)$$

when $0.6 < \mathbf{q}/j \leq 1.0$

In equation (11) and (12) above, j equals $\cos^{-1}(H/R)$ and means the maximum angle of the molten pool.

The local thickness of the crust $d(\mathbf{q})$ can be obtained by solving the one-dimensional conduction equation for the crust as

$$\frac{\dot{Q}d^2(\mathbf{q})}{2k_{cr}} + \frac{q_{down}(\mathbf{q})d(\mathbf{q})}{k_{cr}} = T_m - T_s(\mathbf{q}) \quad (13)$$

where k_{cr} is the thermal conductivity of crust. It is assumed that the heat loss from the outer surface of crust is mainly due to thermal radiation unless cooling water enter into the gap between the crust and vessel wall. Therefore, a local energy balance can be given as

$$q_{down}(\mathbf{q}) + \dot{Q}d(\mathbf{q}) = q_{cr}(\mathbf{q}) = \epsilon s T_s^4(\mathbf{q}) \quad (14)$$

where ϵ is the emissivity of the surface of the crust and s is the Stefan-Boltzman constant. The local thickness of crust can be calculated by combination of equations (11) through (14).

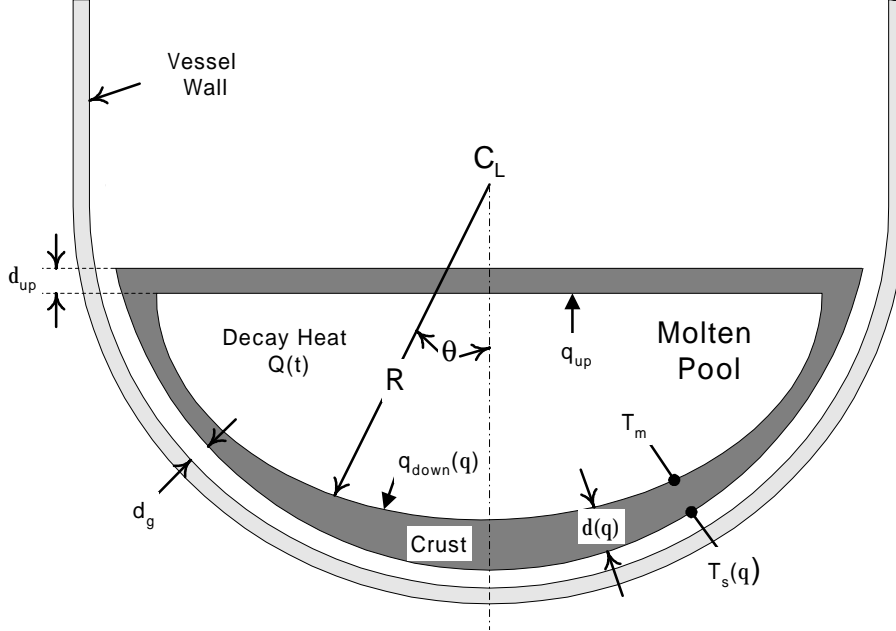


Fig. 2 Natural-Convection Model of Molten Pool Surrounded By Crust

3. Fission Product Release from Pool

Volatile fission products (Xe, Kr, Cs, I) are insoluble in liquid UO₂. Release of the volatile fission products is dominated by bubble dynamics because they all behave as gases in a molten debris pool. Bubble dynamics in the pool is thus characterized by bubble nucleation, coalescence, growth and rise. The time rate of change for the bubble concentration may be represented as follows (McClure et al., 1993).

$$\frac{dn_i}{dt} = \frac{dn_{i,nucl}}{dt} + \frac{dn_{i,coal}}{dt} + \frac{dn_{i,diff}}{dt} - \frac{dn_{i,loss}}{dt} \quad (15)$$

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

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:

$$p_g - p_m > (g/R) \quad (16)$$

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 number of sites may be expressed as

$$s_n = (m_p s_{n,t} + s_{n,p}) \quad (17)$$

For the bubble size at departure, a well-known equation was proposed by Fritz and Ende (1936) as

$$R_F = 0.0104 q_0 \sqrt{g/g(r_l - r_v)} \quad (18)$$

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 $\bar{R}_d = R_d / R_F$ is a function of pressure where $R_F = 0.5 \sqrt{g/g(r_l - r_v)}$ for a contact angle of 48°. According to the experimental data, they obtained the following formula

$$\bar{R}_d = \frac{1000}{p} \quad (19)$$

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

$$\frac{\partial C}{\partial t} = D \left(\frac{\partial^2 C}{\partial r^2} + \frac{2}{R} \frac{\partial C}{\partial r} \right) - \frac{R^2}{r^2} \frac{dR}{dt} \frac{\partial C}{\partial r} \quad (20)$$

Solution to equation (20) is obtained as (Martins and Szekely, 1969)

$$V = -aj(b_b)$$

where

$$j(b_b) = 2b_b^3 \exp(3b_b^2) \times \int_0^\infty x^{-2} \exp(-x^2 - 2b_b^3 x^{-1}) dx \quad (21)$$

$$z = \frac{C - C_m}{C_m} \quad \text{and} \quad a = D \frac{A}{C_m} \quad (22)$$

The growth constant b_b can be obtained from Scriven's useful expression of solutions to equation (20)

$$j(b_b) \sim \sqrt{(p/3)} [b_b - 4/9 + 0(b_b^{-1})] \quad (23)$$

Data of Scriven's tabulation are incorrect for especially small values of $j(b_b)$. This error is induced through the change of variable in integral equation. In this study, new tabulation is obtained, as shown in Fig. 3 and 4, by numerical integration of equation (21) using the Simpson's method. The results show that values of b_b for small $j(b_b)$ are much smaller than those of Scriven's tabulation, and values of b_b for large $j(b_b)$ are close to half values of his tabulation.

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

$$f_d = \frac{1}{t} = \frac{4b_b^2 D}{R^2} \quad (24)$$

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

$$\frac{dn_{i,nucl}}{dt} = f_d \cdot s_n \quad (25)$$

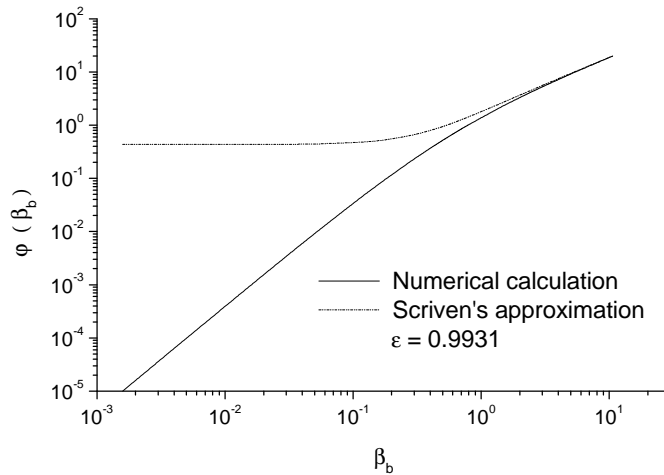


Fig. 3 Function $j(b_b)$ for Small Values of b_b

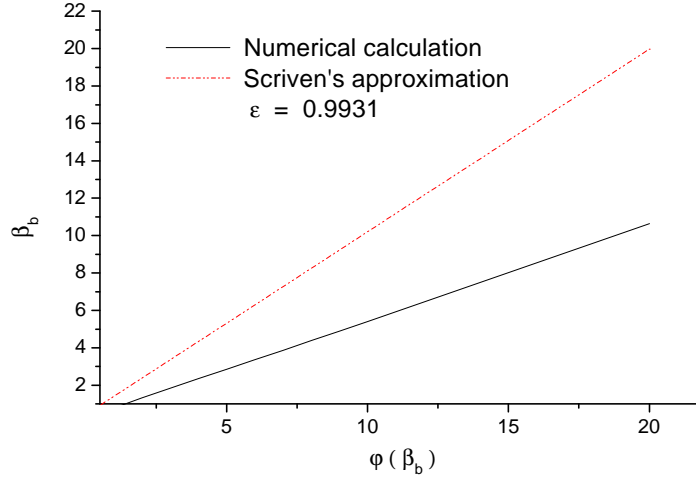


Fig. 4 Results of β_b for Large Values of $j(\beta_b)$

Diffusion to a Bubble

Diffusion to a bubble is governed by equation (20) 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

$$\frac{dn_{i,diff}}{dt} = \frac{n_{i-1}}{t_i - t_{i-1}} - \frac{n_i}{t_{i+1} - t_i} \quad (26)$$

Coalescence of Bubble

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

$$dn_k / dt = 0.5 \sum_{i+j=k} B_{ij} n_i n_j - \sum_{i=1} B_{ik} n_i n_k \quad (27)$$

The rate of change of number density for bubble size R_k can also be obtained by summation of production term and loss term. 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)

$$B_{ij,turb} = 1.3(R_i + R_j)^3 (e_d / n)^{1/2} \quad (28)$$

where

$$e_d / n = (2 / R_p)(f / 2)^{1.5} v_{conv}^3 \quad (29)$$

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

$$v_{conv} = 2Q / (rA_{conv}c_p\Delta T) \quad (30)$$

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

$$B_{ij,rise} = \mathbf{p}(R_i + R_j)^2 |v_i - v_j| \quad (31)$$

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,

$$v = 2rgR^2 / 9m \quad (32)$$

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

$$\frac{dn_{i,loss}}{dt} = \frac{v_i n_i}{z} \quad (33)$$

Fission Product Release through Mass Transport

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. It is assumed that mass transport governs release of the less volatile fission product from the pool. At high temperature (>2850 K), rare earth elements such as europium and cerium exist as oxides, strontium is present as SrO, and ruthenium and antimony are present as metals immiscible in the molten pool (Petti et al., 1989). The rate of mass transport of a species in a liquid is given by

$$\dot{m} = k_m A_{up} (C_\infty - C_{surf}) \quad (34)$$

The mass transfer correlations for the top of the pool can be obtained by means of a heat and mass transfer analogy

4. Results and Discussion

For all the fission product release calculations in this work, the main parameters were obtained from the analysis report of the TMI-2 accident (Akers et al., 1989).

The pool is assumed to be a partially filled hemisphere, 1.45 m in radius and 32,700 kg in mass. The change of pool geometry during the numerical calculation is neglected. The fission product inventories in the pool are about 24.5% of the total core inventories. The parameters used in the numerical calculations are listed in Table

1. From the numerical analysis, the height of the pool is 1.014 m and peak temperature at the pool center exceeds 3000 K.

The calculations were carried out with $p=10$ MPa, $n_s=5000$, and initial inventory = 100 %. In each cases of $M_{b,0}/M_0=0$ and 0.5, times to release 60 % of volatile gas inventory are about 4350 and 750 sec as shown in Fig. 5-(d) and 6-(d). From concentrations of the 29 elements in the pool, total heat generation rate was obtained at time t . Fig. 5-(a) and 6-(a) comparatively show the difference of volumetric heat generation rate between the volatile and non-volatile fission products. Fig. 5-(b) and (c) show decrease of decay heat by fission product release and the peak temperature in the pool. When release of the fission products from the debris pool is considered, the peak temperature of the pool decreases from 3224 K to 3166 K at 4346 sec as shown in Fig. 5-(c). In Fig. 6-(c), also, the peak temperature in the pool decreases from 3260 K to 3194 K at 745.5 sec. Because Initial bubbles interact and rapidly grow by coalescence and diffusion to a bubble, if bubbles in molten core material exist before the formation of the pool, pool peak temperature decreases faster.

When thermal properties of the pool are taken to be: $k = 4$ W/mK, $a = 9.1 \times 10^{-7}$ m²/s, $n = 5.9 \times 10^{-7}$ m²/s, and $b = 9.1 \times 10^{-7}$ K⁻¹. Pool Rayleigh number is about $3 \sim 3.5 \times 10^{15}$. Table 2 shows pool specific heat and average downward heat flux at the time to release 60% of volatile gas inventory. Under the condition which fission products release, pool specific heat and average downward heat flux are significantly reduced by removal mechanism of heat source. Therefore, fission product release may be considered to best estimate heat source of the pool and lower head integrity.

Table 1 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/sec]	0.13
Number of permanent nucleation sites [site/kg]	100 ~ 1000
Number of temperature-dependent nucleation sites [site/kg]	100 ~ 30,000
Diffusivity of fission product [m ² /sec]	1×10^{-11} ~ 1×10^{-7}
Surface tension of liquid pool σ [N/m]	0.5 ~ 1.6

Table 2 Pool specific heat and heat flux of the pool

Mass ratio	Release	Decay heat rate, \dot{Q}	Pool specific heat, ΔT	Heat flux, q_{down}
0.0	No	7.55	374.5	9.34E5
	Yes	6.37	315.6	7.61E5
0.5	No	8.27	409.8	1.04E6
	Yes	6.93	343.7	8.42E5

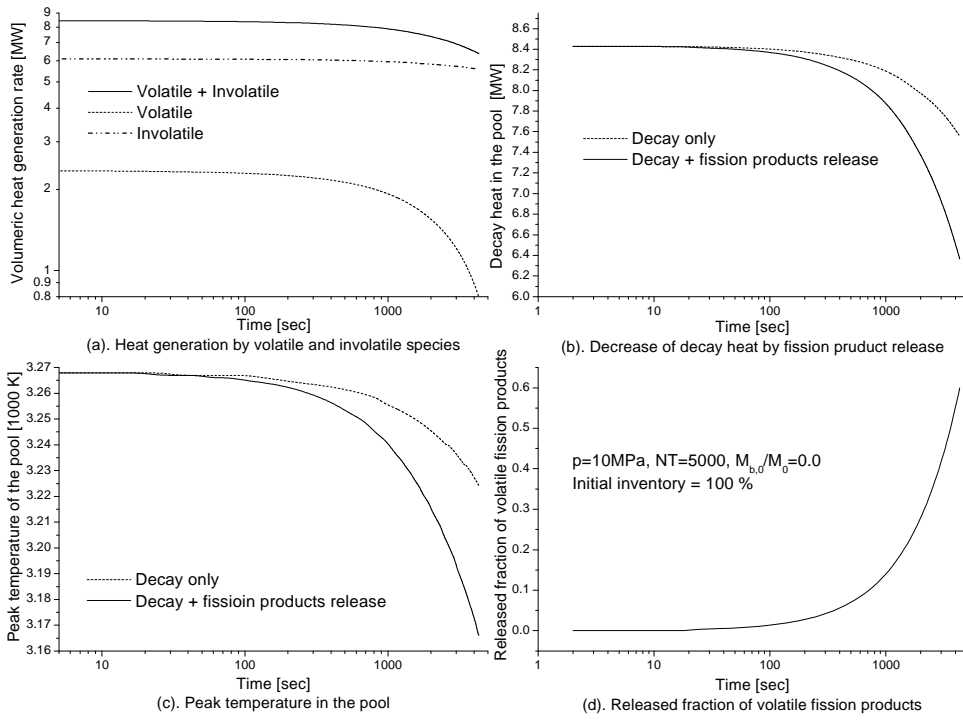


Fig. 5 Heat Generation and Transfer in Case of Mass Ratio ($M_{b,0}/M_0$) = 0.0

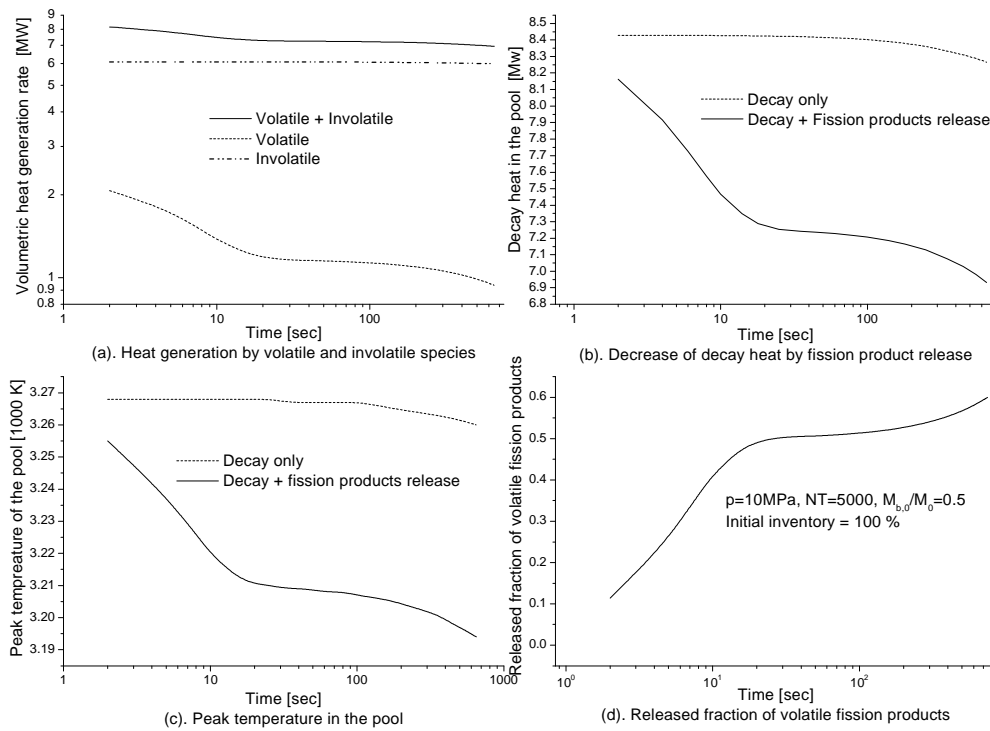


Fig. 6 Heat Generation and Transfer in Case of Mass Ratio ($M_{b,0}/M_0$) = 0.5

5. Conclusions

When fission products release from the molten pool, heat source of the pool was estimated using 29 nuclides concentration data of ORIGEN 2 code and heat flux, pool specific heat and peak temperature were calculated by overall energy balance. The heat generation rate decreases faster, if initial bubbles exist in the pool, because of release of mainly volatile fission gas. The peak temperature sizably decreased by about 60 K as the fission products are released from the pool. For both cases with $M_{b,0}/M_0 = 0.0$ and 0.5 , heat flux is reduced by ~20%. From results of numerical analysis, it is concluded that fission product release may be considered to best estimate heat source of the pool and lower head integrity

Nomenclature

A area of bubble surface (m^2)
 A_{conv} area for convection (m^2)
 A_{down} area for downward heat transfer (m^2)
 A_{up} area for upward heat transfer (m^2)
 B coalescence frequency function (m^3/s)
 C fission product concentration in the pool ($No./m^3$)
 c_p specific heat capacity (J/kgK)
 f fanning friction factor ($=0.004$)
 $f_{c,i}$ released fraction of species i before formation of the pool
 f_p released fraction of species i from the pool
 H depth of the pool (m)
 k thermal conductivity ($W/m \cdot K$)
 k_m mass transfer coefficient (m/s)
 \dot{m} rate of mass transport (kg/s)
 m_c mass of the core (kg)
 m_p mass of the pool (kg)
 $M_{b,0}$ mass of volatile species in the initial bubbles (kg)
 $M_{i,0}$ initial inventory of volatile fission product i in the pool (kg)
 M_0 initial inventory of volatile species in the pool ($=\sum M_{i,0}$) (kg)
 n number density of bubble ($No./m^3$)
 ns_p number of permanent nucleation sites ($No.$)
 ns_t number density of temperature-dependent nucleation sites ($No./kg$)
 \dot{Q} volumetric heat generation rate (W/m^3)
 p pressure of the pool (N/m^2)

p_g pressure of gas bubble (N/m^2)
 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 superheat of the pool (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})
 b_b growth constant of diffusion to a bubble
 σ bubble-melt interfacial tension (N/m)
 h heat generation per unit mass (W/kg)
 e Stefan-Boltzmann constant ($5.6E10-8$) (W/m^2K^{-4})
 e_d eddy diffusivity (m^2/s^3)
 k gas constant (Boltzmann constant) (J/K)
 q_0 contact angle of nucleating bubble ($^\circ$)
 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

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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