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Approximate model for break-up of solidifying melt particles due to thermal stresses in surface crust layer

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ABSTRACT

Fast cooling and solidification of high-temperature droplets of opaque melt is considered. The problem parameters correspond to interaction of core melt with ambient water in hypothetical severe accident in some industrial nuclear reactors. A recently suggested approximation for transient temperature profile in the particle during solidification is employed. This approach is combined with an analytical solution for quasi-steady stress–strain state of growing solid crust layer. A computational analysis showed that the resulting tensile stress on the particle surface is maximal at a certain position of solidification front. The latter is considered to be a reason of mechanical breakage of corium particles at time preceding this stress maximum. The results obtained are in qualitative agreement with recently reported observations of some fragments of thick-wall hollow spherical particles in laboratory experiments.

1. Introduction

The fragmentation of high-temperature melt droplets in ambient water is an important process in the so-called fuel–coolant interaction (FCI) which takes place in hypothetic severe accident of light-water nuclear reactors. The efforts of many researchers have been focused on hydrodynamic simulation of melt jet break-up [1–3] and specific problems of steam explosion [4–6]. It is important that fine fragmentation of melt droplets at the end of premixing stage of the process increases the probability of the steam explosion. An additional fragmentation of the melt droplets after the hydrodynamic break-up may be provided by thermal interaction of hot droplets with water. It may be a result of pressure oscillations in a steam bubble formed around the melt droplet. The model of this type has been proposed by Drumheller [7] who considered the propagation of spherical pressure wave in the droplet. In recent paper by Dombrovsky [8] the competitive processes of pressure drop in steam layer and growing solid crust on particle surface were considered. It was shown that high radiation flux from the surface of opaque corium particle leads to fast formation of the surface crust layer which can prevent from fragmentation of the corium particle.

The calculations showed that further growth of crust layer is not so fast due to low conductivity of corium [9,10]. As a result, the surface temperature appears to be much less than that in the molten core of the particle and the large temperature difference may lead to significant thermal stresses in the crust layer. In other words, the particle solidification which prevents from fragmentation by action of external forces may lead to another danger due to internal stresses produced by temperature strains.

The objective of present paper is to develop a simple model for thermal stresses in a corium particle during its solidification and to analyze conditions of possible break-up of the particle. The thermal stress fragmentation hypothesis is not new. It was considered in early papers [11–13] for possible FCI accident situations in an LMFBR system where the fuel is UO_2 and the coolant is liquid sodium. The experimental data for interaction of stimulant melts with water reported recently by Kudinov et al. [14] indicated that particulate debris bed contains some particles which look as fragments of thick-wall hollow spherical particles with clear traces of circular cracks. It was a natural motivation of the present study.

The thermal state of solidifying corium particles has been studied in recent paper by Dombrovsky and Dinh [10]. An approximate model for transient temperature profile in the particle suggested in this paper was then implemented in multiphase CFD code VAPEX-P [15]. This model is employed in present study to calculate the parameters of quasi-steady stress–strain state of crust layer during solidification of corium particle. It is assumed that the crust is an isotropic brittle material with linear elastic behavior. It allows elaborating simple analytical model for thermal stresses. The latter seems to be important for engineering estimates of possible break-up of particles and implementation of the corresponding relations into problem-oriented multiphase CFD codes.

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Nomenclature

a	particle radius
A, B, C	coefficients in analytical solution (19)
c	heat capacity
E	Young modulus
f	function defined by Eq. (4)
h	heat transfer coefficient
k	thermal conductivity
L	latent heat of melting
p	pressure
q	heat flux
r	radial coordinate
t	current time
T	temperature
u	radial displacement

Greek symbols

α	linear coefficient of thermal expansion
γ	coefficient defined by Eq. (21)
ε	hemispherical emissivity
$\varepsilon_r, \varepsilon_\theta, \varepsilon_\phi$	strain components
ν	Poisson ratio
ρ	density
σ	Stefan–Boltzmann constant
$\sigma_r, \sigma_\theta, \sigma_\phi$	stress components
σ_s	tangential surface stress
σ^*	failing strength

Subscripts and superscripts

e	external
f	front of solidification
liq	liquidus
m	melting
max	maximum
s	surface
sol	solidus
t	total
w	water

Overbar

–	dimensionless quantity

2. Model of surface crust formation

It was shown by Dombrovsky et al. [15] that radiation heat transfer between corium particles in the spectral range of water semi-transparency is usually insignificant compared to local heat transfer to surrounding water. It enables one to analyze solidification dynamics of a single particle as it was recently done by Dombrovsky and Dinh [10]. Following paper [10], we consider a transient spherically symmetric problem and assume that corium particle is totally opaque for thermal radiation. The heat conduction problem for the period of particle solidification is formulated as follows:

$$\rho[c + Lf(T)]\frac{\partial T}{\partial t} = \frac{1}{r^2}\frac{\partial}{\partial r}\left(r^2 k \frac{\partial T}{\partial r}\right) \tag{1}$$

$$t = 0, \quad T = T_{\text{liq}} \tag{2}$$

$$r = 0, \quad \frac{\partial T}{\partial r} = 0, \quad r = a, \quad -k\frac{\partial T}{\partial r} = h(T - T_{\text{w}}) + \varepsilon\sigma\left(T^4 - T_{\text{w}}^4\right) \tag{3}$$

$$f(T) = \frac{\Theta(T - T_{\text{sol}}) - \Theta(T - T_{\text{liq}})}{T_{\text{liq}} - T_{\text{sol}}} \tag{4}$$

where $T(t,r)$ is the temperature profile inside the particle of radius a, L is the latent heat of melting, h is the convective heat transfer coefficient for heat transfer from hot particle to ambient water through a steam layer, T_{liq} and T_{sol} are the liquidus and solidus temperatures of the composite melt, Θ is the Heaviside step function. Note that $T_{\text{liq}} = T_{\text{sol}} = T_{\text{m}}$ in the case of eutectic corium (70% UO_2 + 30% ZrO_2) [16]. In subsequent calculations, we consider very close values of T_{liq} and T_{sol}. It corresponds practically to the case of simple phase transition at melting temperature $T_{\text{m}} = (T_{\text{liq}} + T_{\text{sol}})/2$. We neglect also the change of density ρ during solidification of the melt.

Note that the radiative term in boundary condition (3) can be calculated more accurately on the basis of the so-called large-cell radiation model (LCRM) [17], which takes into account semi-transparency of water in the visible and near infrared spectral ranges.

The values of physical parameters used in calculations are given in Table 1. All the values are the same as those in previous papers by Dombrovsky [8–10], and one can find there the detailed references concerning these data. The use of constant values of thermal conductivity and heat capacity is justified by natural uncertainty of the known experimental data. Note that approximate value of heat transfer coefficient $h = 300$ W/(m^2 K) is in good agreement with the results of detailed numerical simulation of film boiling on a sphere reported recently by Yuan et al. [18]. Some typical temperature profiles in a corium particle are shown in Fig. 1. The surface layer of the particle is cooled very fast due to high radiation flux from the particle surface. The solidification front propagates with approximately constant velocity to the particle center.

According to the model suggested in paper [10], we consider the following approximation of temperature profiles in a spherical corium particle during solidification:

$$T(r,t) = T_{\text{liq}} - [T_{\text{liq}} - T_{\text{s}}(t)]\frac{r - r_{\text{f}}}{a - r_{\text{f}}}\Theta(r - r_{\text{f}}) \tag{5}$$

where $r_{\text{f}}(t)$ is the current radius of solidification front inside the particle, $T_{\text{s}}(t) = T(a,t)$ is the particle surface temperature. It was shown that time dependences $\bar{r}_{\text{f}}(t) = r_{\text{f}}(t)/a$ and $T_{\text{s}}(t)$ can be obtained by solving the following Cauchy problem for coupled ordinary differential equations:

$$\left[(T_{\text{liq}} - T_{\text{s}})\frac{1 + 2\bar{r}_{\text{f}} + 3\bar{r}_{\text{f}}^2}{4} + 3L\bar{r}_{\text{f}}^2/c\right]\frac{d\bar{r}_{\text{f}}}{dt} + \frac{3 - \bar{r}_{\text{f}} - \bar{r}_{\text{f}}^2 - \bar{r}_{\text{f}}^3}{4}\frac{dT_{\text{s}}}{dt} = -\frac{3q_{\text{t}}}{\rho ca} \tag{6}$$

$$\frac{dT_{\text{s}}}{dt} = \frac{q_{\text{t}}}{k/a + (h + 4\varepsilon\sigma T_{\text{s}}^3)(1 - \bar{r}_{\text{f}})}\frac{d\bar{r}_{\text{f}}}{dt},$$
$$q_{\text{t}} = h(T_{\text{s}} - T_{\text{w}}) + \varepsilon\sigma\left(T_{\text{s}}^4 - T_{\text{w}}^4\right) \tag{7}$$

Table 1
Physical parameters used in heat transfer calculations

ρ (kg/m^3)	8000
c (J/(kg K))	600
k (W/(m K))	3
L (kJ/kg)	400
ε	0.85
T_{sol} (K)	2840
T_{liq} (K)	2860
T_{w} (K)	300
h (W/(m^2 K))	300

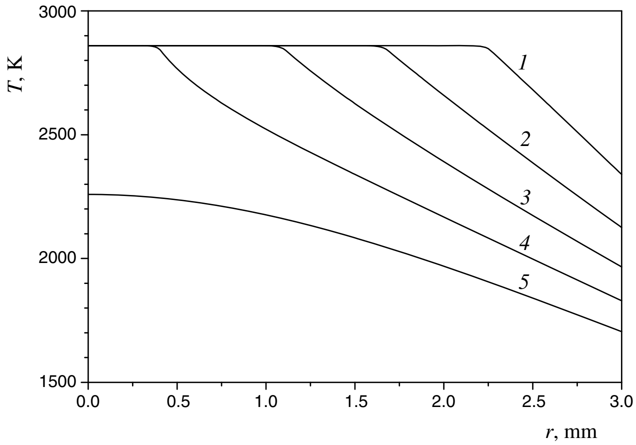

Fig. 1. Temperature profiles in corium particle of radius $a = 3$ mm during solidification: 1 – $t = 1$ s, 2 – $t = 2$ s, 3 – $t = 3$ s, 4 – $t = 4$ s, and 5 – $t = 5$ s.

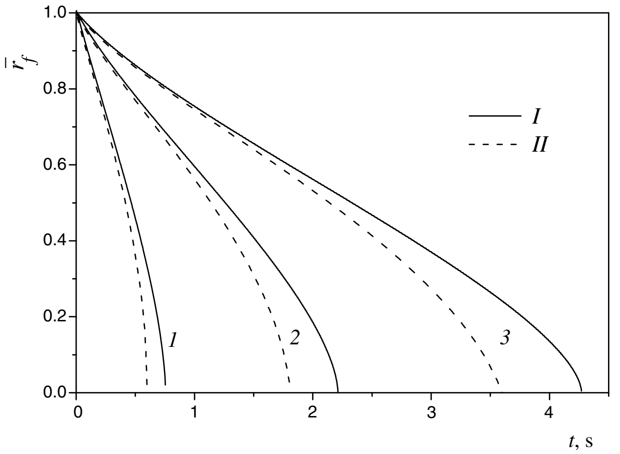

Fig. 3. Radial position of solidification front in corium particles: I – exact solution, II – approximate solution; 1 – $a = 1$ mm, 2 – $a = 2$ mm, and 3 – $a = 3$ mm.

$$\bar{r}_{\mathrm{f}}(0) = 1, \quad T_{\mathrm{s}}(0) = T_{\mathrm{liq}} \tag{8}$$

The derivation of these equations is based on assumption of relatively slow variation of heat transfer coefficient during solidification of the particle. This assumption was confirmed at real conditions of fuel–coolant interaction in paper [15].

The problems (6)–(8) should be solved from $t = 0$ to time corresponding complete solidification of the particle. The resulting dependences $\bar{r}_{\mathrm{f}}(t)$ and $T_{\mathrm{s}}(t)$ can be compared with those obtained from solution of complete heat conduction problem. One can see in Figs. 2 and 3 that approximate solution based on Eqs. (6)–(8) gives qualitatively correct results for both solidification dynamics and surface temperature of the particle. It enabled us to implement slightly modified models (6)–(8) into the multiphase code VAPEX-P for FCI calculations [15].

3. Thermoelastic stress–strain state of solid crust layer

A general analysis of stress–strain state of the growing crust layer can be based on the rigorous theory for mechanics of growing viscoelastic structures [19,20]. But the use of so detailed analysis seems to be unpractical at the moment because of unknown mechanical characteristics of corium at high temperatures. The approximate approach employed in this paper is based on the simplest quasi-steady model and assumption of elastic behavior of solid crust. As a result, the resulting expression for time variation of the maximal thermal stress appears to be very simple and convenient for potential implementation into the engineering multiphase CFD codes. It is believed that the model suggested takes into account the main special features of the process. The latter will be estimated by comparison of the model qualitative predictions concerning possible mechanical destruction of solid crust with the experimental observations.

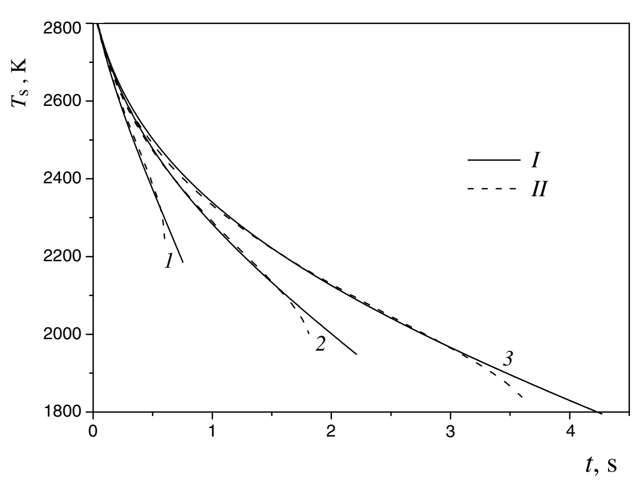

Fig. 2. Surface temperature of corium particles during solidification: I – exact solution, II – approximate solution; 1 – $a = 1$ mm, 2 – $a = 2$ mm, and 3 – $a = 3$ mm.

Let us consider the thermoelastic stress–strain state of solid crust layer on the particle surface. Due to spherical symmetry, the only non-zero displacement component is the radial displacement $u(r)$. The radial and circumferential strains are defined by equations [21]

$$\varepsilon_{\mathrm{r}} = \frac{\mathrm{d}u}{\mathrm{d}r}, \quad \varepsilon_{\theta} = \varepsilon_{\phi} = \frac{u}{r} \tag{9}$$

The strains obviously satisfy the compatibility equation

$$\varepsilon_{\mathrm{r}} = \frac{\mathrm{d}}{\mathrm{d}r}(r\varepsilon_{\theta}) \tag{10}$$

The only non-zero stress components are the radial stress σ_{r} and the circumferential stresses $\sigma_{\theta} = \sigma_{\phi}$, which satisfy the known equilibrium equation (for quasi-steady stress–strain state):

$$\frac{\mathrm{d}\sigma_{\mathrm{r}}}{\mathrm{d}r} + 2\frac{\sigma_{\mathrm{r}} - \sigma_{\theta}}{r} = 0 \tag{11}$$

For isotropic material, the thermoelastic stress–strain–temperature relations are as follows [21]:

$$\varepsilon_{\mathrm{r}} = \frac{1}{E}(\sigma_{\mathrm{r}} - 2\nu\sigma_{\theta}) - \alpha(T_{\mathrm{sol}} - T),$$
$$\varepsilon_{\theta} = \frac{1}{E}[(1-\nu)\sigma_{\theta} - \nu\sigma_{\mathrm{r}}] - \alpha(T_{\mathrm{sol}} - T) \tag{12}$$

where E is the Young's modulus, ν is the Poisson's ratio, α is the linear coefficient of thermal expansion. It is assumed that there are no strains and stresses in the spherical crust layer at temperature $T = T_{\mathrm{sol}}$.

After simple transformations, one can derive the following differential equation for the displacement:

$$\frac{1}{r^2}(r^2u')' - \frac{2}{r^2}u = \alpha\frac{1+\nu}{1-\nu}\frac{\mathrm{d}T}{\mathrm{d}r} \tag{13}$$

We assume here that physical parameters E, ν, and α do not depend on temperature in the range of $T_{\mathrm{s}} < T < T_{\mathrm{sol}}$. The presence of mushy zone $T_{\mathrm{sol}} < T < T_{\mathrm{liq}}$ and the increase in corium density after solidification makes reasonable the approximate boundary condition of zero radial stress at $r = r_f$: $\sigma_r(r_f) = 0$. Note that this condition is different from that suggested by Cao et al. [22] for the beginning of solid crust formation. As for boundary condition on surface $r = a$, it can be formulated as $\sigma_r(a) = -p_e$ (p_e is the pressure in steam blanket around the particle). But the value of p_e is expected to be small in comparison with maximal thermal stress in the crust layer, that is $p_e \ll \alpha(T_{sol} - T_s)_{max}E$. Therefore, we use the approximate boundary condition of zero radial stress: $\sigma_r(a) = 0$. To formulate both boundary conditions in terms of radial displacements we need the following relation:

$$\sigma_r = \frac{E}{1-2\nu}\left[\frac{1-\nu}{1+\nu}u' + \frac{2\nu}{1+\nu}\frac{u}{r} + \alpha(T_{sol} - T)\right] \tag{14}$$

The resulting boundary conditions are:

$$u'(r_f) = -\frac{2\nu}{1-\nu}\frac{u(r_f)}{r_f} \tag{15}$$

$$u'(a) = -\frac{1+\nu}{1-\nu}\alpha(T_{sol} - T_s) - \frac{2\nu}{1-\nu}\frac{u(a)}{a} \tag{16}$$

Eq. (13) with boundary conditions (15) and (16) can be solved numerically for arbitrary temperature profile $T(r)$. After calculation of the radial displacement $u(r)$, one can determine the stress components using Eq. (14) for σ_r and the following equation for tangential (circumferential) stress:

$$\sigma_\theta = \frac{E}{1-2\nu}\left[\frac{\nu}{1+\nu}u' + \frac{1}{1+\nu}\frac{u}{r} + \alpha(T_{sol} - T)\right] \tag{17}$$

Note that the above mathematical formulation of the problem is similar to that for homogenous spheres considered in paper [23]. In particular case of linear temperature profile in the crust layer (5), Eq. (13) is radically simplified:

$$\frac{1}{r^2}(r^2u')' - \frac{2}{r^2}u = -C, \quad C = \alpha\frac{1+\nu}{1-\nu}\frac{T_{liq} - T_s}{a - r_f} \tag{18}$$

The boundary-value problems (18), (15) and (16) have the obvious analytical solution:

$$u = -Ar + B/r^2 - Cr^2/4 \tag{19}$$

$$A = \frac{Ca}{2(1+\nu)}\left[\frac{1-2\nu-2\gamma(1-\nu)}{1+\bar{r}_f+\bar{r}_f^2} - \bar{r}_f\right] \tag{20}$$

$$B = -\frac{Ca^4}{4}\frac{\bar{r}_f^3}{1+\bar{r}_f+\bar{r}_f^2}\left(1 - 2\gamma\frac{1-\nu}{1-2\nu}\right), \quad \gamma = \frac{T_{liq} - T_{sol}}{T_{liq} - T_s} \ll 1 \tag{21}$$

It goes without saying that one can use simplified expressions for coefficients A, B, and C corresponding to the case of $T_{liq} = T_{sol} = T_m$:

$$A = \frac{Ca}{2(1+\nu)}\left(\frac{1-2\nu}{1+\bar{r}_f+\bar{r}_f^2} - \bar{r}_f\right), \quad B = -\frac{Ca^4}{4}\frac{\bar{r}_f^3}{1+\bar{r}_f+\bar{r}_f^2},$$
$$C = \frac{\alpha}{a}\frac{1+\nu}{1-\nu}\frac{T_m - T_s}{1-\bar{r}_f} \tag{22}$$

Having substituted solution (19) with coefficients (22) in Eq. (17) we obtain the tensile circumferential stress on particle surface $\sigma_s = \sigma_\theta(a)$:

$$\sigma_s(t) = \frac{\alpha(T_m - T_s)E}{4(1-\nu)}\left(1 + \bar{r}_f\frac{1+2\bar{r}_f}{1+\bar{r}_f+\bar{r}_f^2}\right) \tag{23}$$

It can be shown that it is the maximal tensile stress in the crust layer. Remember that time dependences of $T_s(t)$ and $\bar{r}_f(t)$ are determined by initial problems (6)–(8).

Note that real thermo-mechanical properties depend on temperature. Unfortunately, to the best of the author's knowledge there is no data for corium in the literature. But there is information on properties of pure uranium dioxide. It is known that the linear coefficient of thermal expansion of UO_2 increases with temperature from $\alpha = 2.2 \times 10^{-5}\,K^{-1}$ at $T = 2500$ K to $\alpha = 2.9 \times 10^{-5}\,K^{-1}$ at $T = 3000$ K [24]. According to early paper by Knapp and Todreas [12], the Young's modulus of UO_2 linearly decreases with temperature from $E = 92.5$ GPa to $E = 72.2$ GPa in the same temperature range. One can see that the resulting temperature variation of the product αE is very small: from 2.0 MPa/K at $T = 2500$ K to 2.1 MPa/K at $T = 3000$ K. Note, that the data reported in [12] are based on tests of specimens prepared by powder pressing and sintering methods and then may be not characteristic of crust formed in solidification of UO_2 melt. Nevertheless, we have no motivation at the moment to revise the model suggested by including possible temperature dependence of thermo-mechanical properties of corium.

The general assumption of brittle behavior of corium crust at high temperatures seems to be more problematic. It was mentioned by Corradini and Todreas [13] that UO_2 undergoes a brittle–ductile transition between 1000 and 1400 °C. For this reason, more complicated model was considered in [13] for UO_2 particles in liquid Na as a coolant (as applied to the liquid metal fast breeder reactors). This model is based on fracture mechanics approach using stress intensity factors and analysis of the crack propagation from the cold particle surface into the ductile zone.

Strictly speaking, one should also take into account the real morphology of solidified corium particles. According to Min et al. [16], the morphology observed by scanning electron microscopy indicated that the particles of eutectic corium had many holes, while the particles at non-eutectic mixture did not.

In all cases, the physical estimate based on simple equation (23) seems to be reasonable. It is obvious that tensile stress on particle surface is directly proportional to increasing temperature difference in the crust layer. The last factor in the right-hand side of Eq. (23) is very close to linear function $1 + \bar{r}_f$. It describes relatively small contribution of the core region typical for spherically symmetric problems.

The simplest way to decide if the particle will be fragmented due to thermal stresses is to compare the calculated value of σ_s with the failing strength of solid crust σ^*. The model suggested cannot pretend for reliable quantitative results for σ_s and there is no data for σ^*. Therefore, one should use this model in combination with experimental data to find a conventional value of failing strength σ^* for the use in semi-empirical estimates.

At the same time, the parametric study based on the above solution is expected to be important for qualitative analysis of the problem. The numerical results obtained at realistic values of physical parameters (see Tables 1 and 2) are presented in Fig. 4. According to Eq. (23), the ratio of σ_s/E does not depend on E and directly proportional to α. It means that one can determine the value of tensile stress σ_s for other parameters E and α without additional calculations. Time dependence of the tensile stress has a maximum at certain position of solidification front. This result is qualitatively clear and the error of approximate thermal model at $\bar{r}_f < 0.4$ has no considerable effect on this maximum. It is important that the maximum tensile stress σ_s^{max} increases monotonically with particle radius. It means that thermal stresses in solid crust are more dangerous for large particles of corium.

It is interesting to consider dependences of σ_s^{max} and corresponding position of solidification front $\bar{r}_f^*$ on radius of corium

Table 2
Physical parameters used in thermal stress calculations

α (1/K)	2.5×10^{-5}
E (GPa)	8
ν	0.3

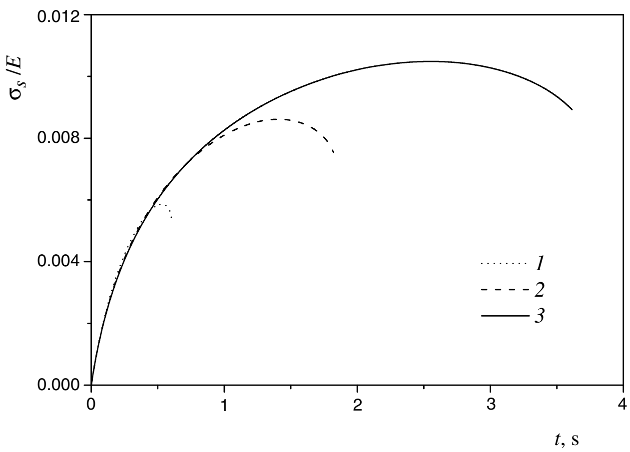

Fig. 4. Dimensionless tensile stress on surface of solidifying corium particle: 1 – a = 1 mm, 2 – a = 2 mm, and 3 – a = 3 mm.

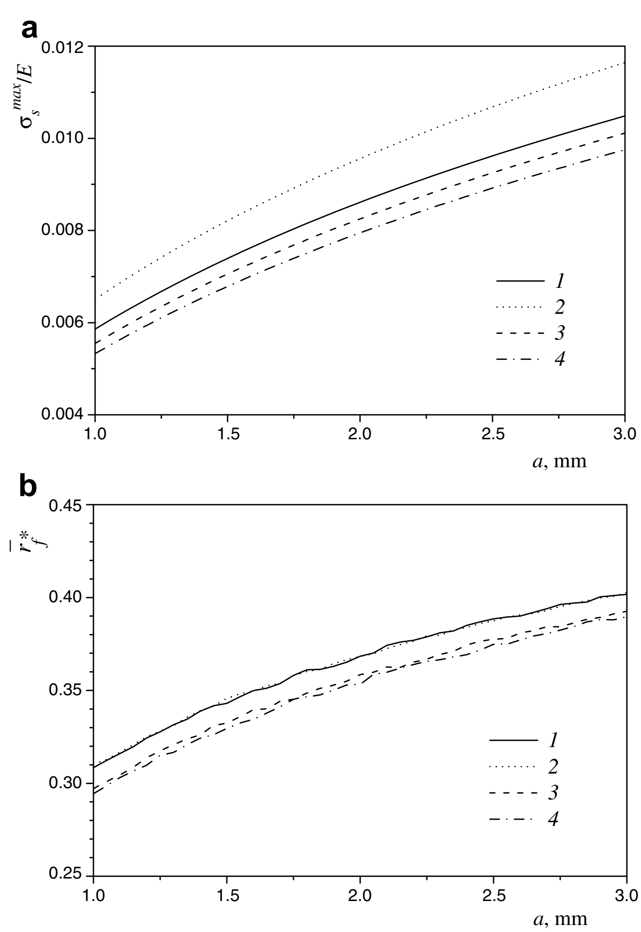

Fig. 5. The maximum value of dimensionless tensile stress on the crust layer surface (a) and the corresponding radial position of solidification front (b): 1 – calculation at adopted values of physical parameters (see Table 1), 2 – h = 500 W/(m^2 K), 3 – ε = 0.75, and 4 – k = 3.5 W/(m K).

particles. These data are presented in Fig. 5. Variants 2, 3, and 4 differ from the baseline variant 1 by only one value of physical parameter (h, ε, or k). It enables one to estimate an effect of natural uncertainty in the data of Table 1. One can see that the value of $\bar{r}_f^*$ is in narrow range of $0.3 < \bar{r}_f^* < 0.4$ for particle radii from 1 to 3 mm. This result appears to be insensitive to realistic uncertainty in physical parameters of the problem.

The above result on the range of $\bar{r}_f^*$ can be treated as a minimum value of cavity radius in fragmented particles. According to the model suggested, the value of $\bar{r}_f^*$ does not depend on mechanical properties of crust. The theoretical prediction for the minimum cavity radius in the case of fragmentation of solidified opaque particles due to thermal stresses can be compared with experimental results for morphology of debris bed particles. In the case of experimental confirmation of the model suggested, one can easily implement the derived analytical solution into multiphase codes for FCI calculations.

4. Conclusions

An approximate model for mechanical destruction of solidifying millimeter-size particles due to tensile thermal stresses in growing crust layer is suggested. The model is based on recently obtained simplified solution for transient temperature profile in the particle. The elastic behavior of the crust is assumed to derive an analytic expression for current circumferential thermal stress on the particle surface.

A computational analysis showed that this tensile stress reaches the maximum at a certain position of solidification front. The latter is considered to be a reason of mechanical destruction of corium particles at time preceding this stress maximum. The calculations showed a weak sensitivity of the minimum internal radius of destroyed hollow particle on natural uncertainty in the problem parameters. It is important that the resulting cavity radius does not depend on thermo-mechanical properties of the crust material.

The results obtained are in qualitative agreement with recently reported observations of some fragments of thick-wall hollow spherical particles in laboratory experiments. To the author's mind, it can be treated as indirect confirmation of the physical model which explains one of the typical morphologies of solid particles in the corium debris bed.

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