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FREEZING OF DROPS ON COOLED SURFACES

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The authors consider an approximate analytical solution of the problem of freezing of liquid drops on the cold surface of a semiinfinite body and a thin plate.

Freezing of liquid drops on a cooled substrate occurs in a number of contemporary and future technologies, e.g., cryodispersion technology, cryostorage of biological items in granules, and in the application of a controlled charged flux of solder drops to an electronic plate. In numerous applications it is important to know such characteristics of the freezing process as the crystallization time or the freezing rate and the temperature field in the drop during the freezing process.

The problem is formulated in these cases as follows. A drop in the form of a semiellipsoid of revolution is located on a cooled surface with temperature T at the initial time $\tau = 0$. This drop shape is the closest to the shape of granules obtained in existing technologies (Fig. 1). In most cases one can consider that the free part of the substrate and the curved surface of the drop are thermally insulated and all the heat from the drop is transferred to the substrate by heat conduction; there is no contact resistance between the drop and the substrate, and the initial temperature of the drop is the crystallization temperature (Fig. 2).

The temperature fields of the frozen part of the drop and in the substrate are described by the unsteady heat-conduction equations:

$$z >= 0, \quad \frac{\partial \Theta_d}{\partial \mathrm{Fo}_d} = \Delta\Theta_d; \tag{1}$$

$$z <= 0, \quad \frac{\partial \Theta_s}{\partial \mathrm{Fo}_s} = \Delta\Theta_s; \qquad \mathrm{Fo}_d = 0, \ \Theta_d = 1, \ \Theta_s = 0. \tag{2}$$

The temperature fields at the drop–substrate boundary are linked by the usual conjugate conditions:

$$\Theta_s = \Theta_d, \tag{3}$$

$$\left(\frac{\lambda_s}{\lambda_d}\right) \operatorname{grad}\Theta_s = \operatorname{grad}\Theta_d. \tag{4}$$

At the crystallization front in the drop we have the condition

$$\frac{\partial \Theta_d}{\partial n} = \frac{r \dfrac{\partial n}{\partial \mathrm{Fo}_d}}{[c_d(T_{cr} - T_\infty)]}. \tag{5}$$

It is scarcely possible to obtain an exact analytical solution of the Stefan problem of Eqs. (1)-(5) in the coupled form.

In [1] we proposed an approximate analytical method of solving this problem, based on introducing a curvilinear orthogonal coordinate system conforming to the shape of the object being analyzed.

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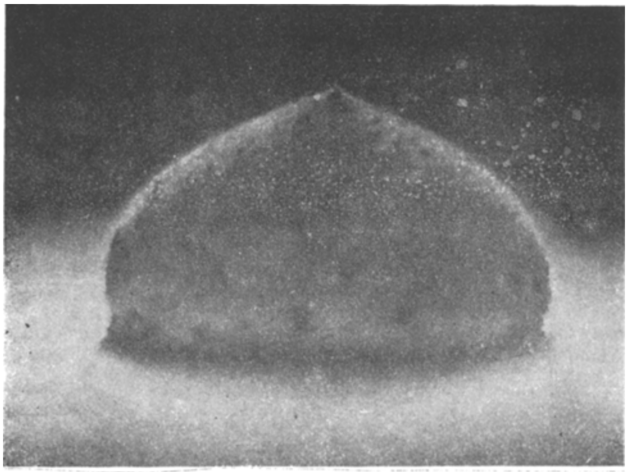

Fig. 1

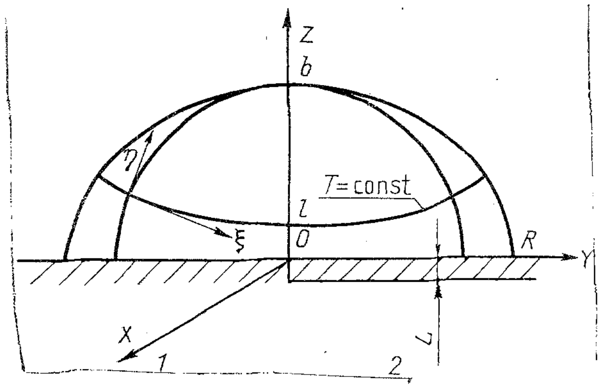

Fig. 2

Fig. 1. Characteristic shape of a granule obtained by freezing of a drop of water on a cooled substrate (R = 5 mm).

Fig. 2. Schematic of the process of freezing of a liquid drop on a surface: 1) semiinfinite body; 2) thin plate.

In the case examined, for an ellipsoidal drop, such a coordinate system is given by the equations:

$$z^2 + \frac{y^2 + x^2}{\xi^2} = 1, \tag{6}$$

$$z^2 + y^2 + x^2 - \eta^2 + 2\ln\left(\frac{\eta}{z}\right) = 1. \tag{7}$$

Figure 3 shows a photograph of a water drop cut in half prior to the end of the solidification. The shape of the crystallization front thus recorded agrees well with isotherms computed from Eq. (7). This indicates that the choice of the coordinate system is well founded.

Temperature Field of the Solidified Part of the Drop. Since the position of the isotherms is given by Eq. (7), subsequent analysis can limit the search for a solution for x = 0 and y = 0, i.e., i.e., for the central axis of the drop, taking into account that in the general case $\eta \to z$.

Using the classical quasisteady approximation, which is most correct for the cases $a_d \gg a_s$ and $r/[c_d(T_d - T_s)] \gg 1$, for the solidified part of the drop we have

$$0 \leqslant z \leqslant 1, \quad \frac{\partial^2 \Theta_d}{\partial z^2} + \frac{2z}{z^2 - 1}\frac{\partial \Theta_d}{\partial z} = 0. \tag{8}$$

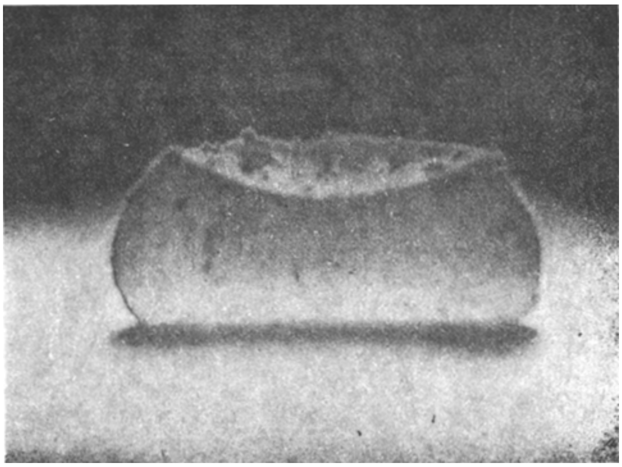

Fig. 3. Photograph of a water drop cut in half prior to the end of solidification (the unfrozen liquid during the crystallization was quickly removed using a pipette).

The boundary conditions are evidently: $z=1, \Theta_d=1; z=0, \Theta_d=\Theta_s(\mathrm{Fo_s}, 0)$. The solution of Eq. (8) has the form

$$\Theta_d(\mathrm{Fo_s}, z) = (1-\Theta(\mathrm{Fo_s}, 0))\frac{\ln[(1+z)/(1-z)]}{\ln[(1+l)/(1-l)]} + \Theta_s(\mathrm{Fo_s}, 0). \tag{9}$$

Temperature Field in the Substrate. 1. Semiinfinite Body. To describe the temperature field we used the two-dimensional unsteady equation of heat conduction in cylindrical coordinates [1]. Solutions were obtained for the two cases of ratios of thermal diffusivities a_d and a_s:

a) $a_d \gg a_s$.

The temperature of the substrate surface under the drops is

$$\Theta_s(\mathrm{Fo_s}, 0) = 1 - \frac{1}{h}\left[\mathrm{erfc}\frac{1}{2\sqrt{\mathrm{Fo_s}}} + \frac{1}{\sqrt{\pi\,\mathrm{Fo_s}}}\right], \tag{10}$$

where

$$h = \frac{2\lambda_d R}{\lambda_s b \ln[(1+l)/(1-l)]}. \tag{11}$$

The position of the crystallization front as a function of time is

$$\int_0^{\mathrm{Fo_s}} \mathrm{erfc}\frac{1}{2\sqrt{\mathrm{Fo_s}}}\,d\,\mathrm{Fo_s} + \frac{2}{\sqrt{\pi}}\sqrt{\mathrm{Fo_s}} = \frac{r\rho_d b a_s}{R\lambda_d(T_{cr}-T_\infty)}\left(l-\frac{l^3}{3}\right). \tag{12}$$

b) $a_d \ll a_s$.

The substrate temperature under the drop in this case can be considered constant:

$$\Theta_s = 0. \tag{13}$$

The position of the crystallization front as a function of time is

$$\mathrm{Fo_d} = \frac{r}{2c_d(T_{cr}-T_\infty)}\left[l\left(1-\frac{l^2}{3}\right)\ln\frac{1+l}{1-l} + \frac{2}{3}\ln(1-l^2) - \frac{l^3}{3}\right]. \tag{14}$$

We now compare the limiting cases of Eqs. (12) and (14) for computing the solidification time of a liquid drop on a cooled surface with the exact solution of Shvartz problem [2] of solidification of a semiinfinite volume of liquid with initial temperature $T = T_{cr}$ on a semi-infinite solid body having temperature T_∞:

$$\frac{\lambda_s\sqrt{a_d}\exp(-\beta^2)}{\lambda_d\sqrt{a_s}+\lambda_s\sqrt{a_d}\,\mathrm{erf}(\beta)} = \frac{\beta r\sqrt{\pi}}{c_d(T_{cr}-T_\infty)}, \tag{15}$$

where

$$\beta = \frac{l}{2\sqrt{a_d\tau_{cr}}} = \frac{1}{2\sqrt{\mathrm{Fo_d}}}. \tag{16}$$

It is evident that when $R \to \infty$ the drops degenerate into a planar layer of height b. Then for the limiting case $a_d \gg a_s$ we obtain the total crystallization time of the layer of thickness b:

$$\mathrm{Fo_d} = \frac{a_d\tau_{cr}}{b^2} = \frac{\pi}{9}a_d a_s\left(\frac{r\rho_d}{\lambda_s(T_{cr}-T_\infty)}\right)^2. \tag{17}$$

The solution of the Shvartz problem of Eq. (15) for $a_d \gg a_s$ with $\mathrm{Fo_d} \gg 1$ takes the form

$$\mathrm{Fo_d} = \frac{\pi}{4}a_d a_s\left(\frac{r\rho_d}{\lambda_s(T_{cr}-T_\infty)}\right)^2. \tag{18}$$

For the limiting case $a_d \ll a_s$ Eq. (14) is rewritten in the form

$$\mathrm{Fo_d} = 0{,}3\frac{r}{c_d(T_{cr}-T_\infty)}. \tag{19}$$

For $c_d\rho_d \approx c_s\rho_s$ and $\mathrm{Fo_d} \gg 1$, taking into account $\sqrt{(a_d)/a_s} \ll 1$, we obtain from Eq. (15)

$$\mathrm{Fo_d} = \frac{1}{2}\frac{r}{c_d(T_{cr}-T_\infty)}. \tag{20}$$

The results of comparing the expressions obtained for $a_d \gg a_s$, Eqs. (17) and (18), and for $a_d \ll a_s$, Eqs. (19) and (20), show that they differ only in the numerical coefficients. This satisfactory agreement validates our approach to solving the problem of solidification of the liquid drop. Regarding the differences of the numerical coefficients, they are explained by the fact that in our drop problem we have a finite volume, and not a semiinfinite one, and in addition, the drop surface is thermally insulated. This leads to the fact that as $R \to \infty$ the shape of the isotherms remains curvilinear, in contrast with the planar isotherms for solidification of a semiinfinite layer of liquid.

It should be noted that one should obtain perfect agreement of the solutions considered at small ℓ, as can easily be shown.

2. Plate of Thickness $L' \ll R$. We shall examine in more detail the solution for this case. For the temperature field in the plate we have

$$\frac{\partial^2\Theta_s}{\partial\rho^2} + \frac{1}{\rho}\frac{\partial\Theta_s}{\partial\rho} + \frac{\partial^2\Theta_s}{\partial z_\rho^2} = \frac{\partial\Theta_s}{\partial \mathrm{Fo}_s}. \tag{21}$$

The boundary conditions are analogous to those of the previous case, except for the condition at the lower surface of the plate, where we assign boundary conditions of the first kind:

$$z_\rho = L,\ \Theta_s = 0; \tag{22}$$

$$z_\rho = 0,\ \rho > 1,\ \frac{\partial\Theta_s}{\partial z_\rho} = 0; \tag{23}$$

$$\rho \leqslant 1,\ \frac{\partial\Theta_s}{\partial z_\rho} = h(\Theta_s - 1). \tag{24}$$

Thus, for the substrate we have a boundary problem of unsteady heat conduction with inhomogeneous mixed boundary conditions, i.e., on the top surface of the plate the heat from the drop is supplied through a circle of radius R, and the rest of the surface is thermally insulated.

We represent the desired solution $\Theta_s(\rho, z_\rho, \mathrm{Fo}_s)$ in the form of the sum (for $\rho < 1$)

$$\Theta_s(\rho,\ z_\rho,\ \mathrm{Fo}_s) = W(\rho,\ z_\rho,\ \mathrm{Fo}_s) + U(z_\rho,\ \mathrm{Fo}_s), \tag{25}$$

where the function $V(z_\rho, \mathrm{Fo}_s)$ is chosen such that it satisfies only the boundary conditions, i.e.,

$$\begin{gathered} z_\rho = 0,\ \frac{\partial U}{\partial z_\rho} = h(U - 1); \\ z_\rho = L,\ U = 0. \end{gathered} \tag{26}$$

Condition (26) is satisfied by a linear dependence of V on z of the type

$$U(z_\rho,\ \mathrm{Fo}_s) = h(L - z_\rho)/(1 + hL). \tag{27}$$

Taking account of Eq. (26) from Eq. (21) we obtain equations for determining $W(\rho, z_\rho, \mathrm{Fo}_s)$:

$$\frac{\partial W}{\partial \mathrm{Fo}_s} = \Delta W - \frac{\partial U}{\partial \mathrm{Fo}_s}; \tag{28}$$

$$\mathrm{Fo}_s = 0,\ W = z/L - 1; \tag{29}$$

$$z = 0,\ \frac{\partial W}{\partial z} = hW; \tag{30}$$

$$z = L,\ W = 0. \tag{31}$$

Because of Eq. (27) we have

$$\frac{\partial U}{\partial \mathrm{Fo}_s} = \frac{L - z_\rho}{(1 + hL)^2}\frac{\partial h}{\partial \mathrm{Fo}_s}. \tag{32}$$

If we assume that $L \ll 1$, and in addition we take into consideration that $0 \leqslant z \leqslant L$, for finite values of $\partial h/\partial \mathrm{Fo}_s$ we can neglect the quantity $\partial U/\partial \mathrm{Fo}_s$. Then

$$\frac{\partial W}{\partial \mathrm{Fo_s}} = \frac{\partial^2 W}{\partial \rho^2} + \frac{1}{\rho}\frac{\partial W}{\partial \rho} + \frac{\partial^2 W}{\partial z_\rho^2}. \tag{33}$$

To solve Eqs. (28)-(33) we use the method of integral transformations, successively applying a finite Fourier integral transformation in the variable z:

for $\rho > 1$

$$\bar{f}(n) = \int_0^L f(z_\rho)\cos\mu_n \frac{z_\rho}{L}\, dz_\rho, \tag{34}$$

where $\mu_n = \frac{\pi}{2}(2n-1),\ n = 1, 2, 3, \ldots;$

for $\rho \leqslant 1$

$$\bar{f}(n) = \int_0^L f(z_\rho)\left(\mu_n' \cos\mu_n' \frac{z_\rho}{L} + hL\sin\mu_n' \frac{z_\rho}{L}\right) dz_\rho, \tag{35}$$

where μ_n' are the roots of the equation

$$\operatorname{ctg}\mu_n' = -hL/\mu_n',$$

and the Laplace transformation in the variable $\mathrm{Fo_s}$

$$\varphi_L(s) = \int_0^\infty \varphi(\rho, z_\rho, \mathrm{Fo_s})\exp(-s\,\mathrm{Fo_s})\, d\,\mathrm{Fo_s} \tag{36}$$

to Eq. (21), written for $\rho \leqslant 1$ and $\rho > 1$, respectively.

For a thin plate we have $hL/\mu_n' \ll 1$. In this case $\mu_n' = \mu_n = (\pi/2)(2n-1)$ and the solution of Eq. (21) for the region $\rho > 1$ has the form

$$\Theta = \frac{2}{L}\sum_{n=1}^\infty \cos\frac{\mu_n z_\rho}{L}\frac{1}{2\pi i}\int_{\sigma-i\infty}^{\sigma+i\infty}\exp(s\,\mathrm{Fo_s})\,\bar{\Theta}_L ds, \tag{37}$$

where

$$\bar{\Theta}_L = \frac{hL^2}{\mu_n^2(1+hL)}\left(\frac{1}{s} - \frac{1}{\frac{\mu_n^2}{L^2}+s}\right) I_1\left(\sqrt{\frac{\mu_n^2}{L^2}+s}\right) K_0\left(\sqrt{\frac{\mu_n^2}{L^2}+s}\right)\sqrt{\frac{\mu_n^2}{L^2}+s}. \tag{38}$$

For the region $\rho \leqslant 1$ the solution is written in the form

$$\Theta = U + W = \frac{h(L-z_\rho)}{1+hL} + \frac{2}{L}\sum_{n=1}^\infty \cos\frac{\mu_n z}{L}\frac{1}{2\pi i}\int_{\sigma-i\infty}^{\sigma+i\infty}\exp(s\,\mathrm{Fo_s})\bar{W}_L ds, \tag{39}$$

where

$$\bar{W}_L = -\frac{hL^2}{\mu_n^2(1+hL)}\left(\frac{1}{s} - \frac{1}{\mu_n^2/L^2+s}\right) K_1\left(\sqrt{\frac{\mu_n^2}{L^2}+s}\right) \times$$

$$\times I_0\left(\rho\sqrt{\frac{\mu_n^2}{L^2}+s}\right)\sqrt{\frac{\mu_n^2}{L^2}+s} - \frac{hL^2}{(1+hL)\mu_n^2(\mu_n^2/L^2+s)}. \tag{40}$$

Using tables of transforms and theorems of the Laplace integral transform [3], we can obtain the temperature of interest to us, at the center of the base of the drop, i.e., with $z_\rho = 0$, $\rho = 0$

$$\Theta(\mathrm{Fo_s}, 0, 0) = \frac{hL}{1+hL} + \frac{2}{L}\sum_{n=1}^\infty W_s, \tag{41}$$

where

$$W_s = \frac{hL^2}{\mu_n^2(1+hL)}\left\{\exp\left(-\frac{\mu_n^2}{L^2}\mathrm{Fo_s}\right)(-1+\exp(-1/4\,\mathrm{Fo_s})) - \int_0^{\mathrm{Fo_s}}\exp\left(-\frac{\mu_n^2}{L^2}\mathrm{Fo_s} - \frac{1}{4\,\mathrm{Fo_s}}\right)\frac{\partial\,\mathrm{Fo_s}}{4\,\mathrm{Fo_s^2}}\right\}. \tag{42}$$

We note that the series $\sum_{n=1}^\infty W_s$ rapidly converges, since

$$W_s \sim \exp(-\mu_n^2/L^2\cdot\mathrm{Fo_s})/\mu_n^2,$$

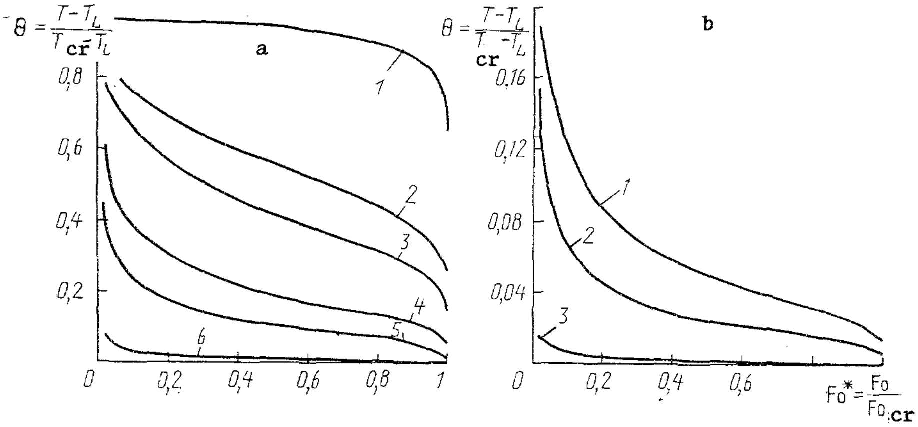

Fig. 4. Change of temperature of the base of the drop during solidification on various surfaces: a) Teflon surface [$R/b = 1$, $ra_s/(c_d(T_{cr} - T_L)a_d) = 0.05$]: 1) semiinfinite body, Eq. (10); 2-6) plate, Eq. (37) [2) $L/R = 0.1$; 3) 0.06; 4) 0.02; 5) 0.01; 6) 0.001]; b) stainless steel plate [$R/b = 1$; $ra_s/(c_d \times (T_{cr} - T_L)a_d) = 1.2$]: 1) $L/R = 0.1$; 2) 0.02; 3) 0.01.

and therefore we can restrict attention to the first term. Thus we have

$$\Theta(\mathrm{Fo_s},\ 0,\ 0) = \frac{hL}{1+hL} + \frac{2hL}{\mu_1^2(1+hL)}\left\{\exp\left(-\frac{\mu_1^2}{L^2}\mathrm{Fo_s}\right)\times\right.$$

$$\left.\times\left(-1+\exp\left(-\frac{1}{4\,\mathrm{Fo_s}}\right)\right) - \int\limits_0^{\mathrm{Fo_s}} \exp\left(-\frac{\mu_1^2}{L^2}\mathrm{Fo_s} - \frac{1}{4\mathrm{Fo_s}}\right)\frac{\partial\,\mathrm{Fo_s}}{4\,\mathrm{Fo_s^2}}\right\}. \tag{43}$$

Determination of the Crystallization Time. To determine the dependence of the position of the crystallization front in the drop on the time and the crystallization time of the drop we use condition (5). Taking account of Eqs. (9) and (43) we obtain

$$1 - \Theta(\mathrm{Fo_s},\ 0) = \frac{r}{2c_d(T_{cr} - T_L)}\ \frac{b^2 a_s}{R^2 a_d}(1 - l^2)\ln\frac{1+l}{1-l}\ \frac{\partial l}{\partial \mathrm{Fo_s}}. \tag{44}$$

We can solve Eq. (44) using the Picard method of successive approximations or using numerical methods. We use the notation

$$A = \frac{r}{2c_d(T_{cr} - T_L)}\ \frac{b^2 a_s}{R^2 a_d}. \tag{45}$$

For the first approximation we can write

$$\mathrm{Fo_s^{[1]}} = A\left[l\left(1 - \frac{l^2}{3}\right)\ln\frac{1+l}{1-l} + \frac{2}{3}\ln(1-l^2) - \frac{l^2}{3} + \frac{2L\lambda_d}{\lambda_s b}\left(l - \frac{l^3}{3}\right)\right]. \tag{46}$$

We note that the first approximation gives a value exceeding the crystallization time of Eq. (14) (the limiting case $a_d \ll a_s$ for a semiinfinite body) by the amount

$$\frac{2A\lambda_d L}{\lambda_s b}\left(l - \frac{l^3}{3}\right). \tag{47}$$

The increase of the crystallization time can be explained by the fact that in the given solution one takes account of the variation of temperature of the substrate under the drop, i.e., the plate is an added thermal resistance in removing heat from the drop.

Now, knowing the dependence $\mathrm{Fo} = \mathrm{Fo}(\ell)$, i.e., the position of the crystallization front in the drop at any time, we can compute the temperature field in the solidified part of the drop from Eq. (9).

Figures 2 and 3 show computed temperatures of the base of the drop from Eq. (43) at the time of solidification on plates of Teflon and stainless steel of different thicknesses and the data obtained for a semiinfinite cooled surface. Comparison of the data shows good qualitative agreement. The influence of the thermophysical properties of the plate material on the

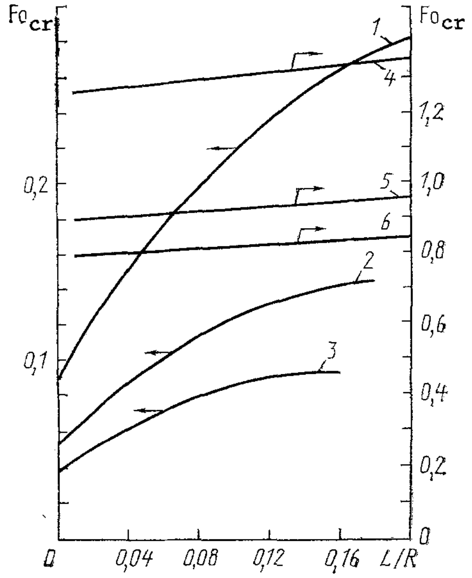

Fig. 5. Dependence of the crystallization time on the plate thickness from Eq. (44) ($R/b = 1$), $s = ra_s/[c_d(T_{cr} - T_L)a_d]$: 1, 2, 3) Teflon S = 1.6, 2.4, 3.1, respectively; 4, 5, 6) stainless steel S = 0.05, 0.07, 0.1.

variation of drop base temperature during drop solidification is shown in Fig. 4. The drop base temperature during crystallization proved to be substantially lower for the plate with higher thermal conductivity, stainless steel. This leads to greater nonuniformity of drop cooling when there is solidification on these surfaces. At the moment of completion of crystallization the drop cooling rate is a maximum, since ($T_d - T_s$) is a maximum, and there is no heat release associated with crystallization in the drop.

Figure 5 shows the computed crystallization time as a function of the plate thickness, from Eq. (44). Evidently for materials with higher thermal conductivity the crystallization time will depend less on the plate thickness, a result which we found for plates of stainless steel, in contrast to the Teflon plates.

Thus, from the relation obtained we can compute the crystallization time and the temperature field when a liquid drop solidifies on different surfaces.

NOTATION

a, thermal diffusivity, m^2/sec; b, drop height, m; c, heat capacity, J/(kg·K); R, drop radius, m; $\ell = \ell'/b$, dimensionless coordinate of the crystallization front; ℓ, coordinate of the crystallization front, m; $L = L'/R$, dimensionless plate thickness; L', plate thickness, m; r, heat of crystallization, J/kg; n, normal to the phase transition surface; $\Theta_d = (T_d - T_L)/(T_{cr} - T_L)$, $\Theta_s = (T_s - T_L)/(T_{cr} - T_L)$, dimensionless temperatures of the drop and the substrate; T_d and T_s, temperature of the drop and the substrate, K; T_L, temperature of the lower cooled surface of the plate, K; $T_L = T$, for a semiinfinite surface; $Fo_d = a_d\tau/b^2$; $Fo_s = a_s\tau/R^2$; $z = z'/b$, $y = y'/b$, $x = x'/b$, dimensionless cartesian coordinates; $z_\rho = z'/R$, $\rho = \rho'/R$, dimensionless cylindrical coordinates; ξ, η, curvilinear orthogonal coordinates; Δ, Laplace operator; erf(x), Gauss function; τ, time, sec; λ, thermal conductivity, W/(m·K); ρ, density, kg/m^3. Subscripts: d, drop; cr, crystallization; ℓ, crystallization front; s, substrate.

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