INSTABILITY OF THE STEADY-STATE SOLIDIFICATION PROCESS

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A new type of planar crystallization-front instability is found and studied: dynamic instability with respect to front-velocity perturbations, independent of morphological stability.

The features of the supermolecular structure of solids obtained by crystallization from the liquid phase depend to a significant degree on the stability of the crystallization front with respect to perturbations of various sorts. Therefore, study of the stability of the solidification process is important in determining the causes of inhomogeneity development in the solid phase and for developing methods for active interference in the solidification process to eliminate or modify such inhomogeneities.

The type of stability usually studied is so-called morphological instability of the crystallization front with respect to small perturbations of its form (see, for example, [1-4]). Such instability leads to the appearance of various cellular structures, to formation of longitudinally spatially periodic structures during solidification of eutectic alloys, etc. [5, 6].

In addition, there exist many examples of the establishment of oscillatory regimes of front motion (rhythmic crystallization), unrelated to morphological instability, but also leading to the development of periodic inhomogeneities, in particular, transverse layered structures (individual layers normal to the direction of front motion, with layer form reflecting the front configuration at the time of its formation) [7, 8].

Formation of such inhomogeneities has traditionally been ascribed to either various external factors and influences (convective motion of the melt, random temperature oscillations and other process conditions, rotation of the crystallization bath, external electromagnetic fields, etc.) [9], or to the possibility of self-excitation of oscillatory solidification regimes in cooled melts containing an impurity which is driven back from the crystallization front. The main physical cause of this self-excitation is to be found in the specific properties of the two-phase zone which may form near the front due to the appearance of dendrites or homogeneous formation of crystalline nuclei in a locally supercooled melt [10], the inclination of the solid solution to layering into isomorphous layers of various compositions [11], and certain other factors. It often remains unclear to what degree conclusions on the generation of self-oscillations and their unique features actually reflect the objective properties of real systems and solidification regimes, or to what degree they are consequences of a priori and usually poorly justified assumptions as to the structure and properties of the two-phase zone or the kinetics of the transfer processes occurring near the front.

It will be shown below that under certain conditions even the steady-state solidification process corresponding to uniform motion of a morphologically stable planar crystallization front will destabilize. The possibility of such instability, not related to loss of front morphological stability, nor the presence of a two-phase zone ahead of the front, nor to the affinity of the solid phase to layering, was predicted previously in [12], and was later subjected to undeserved criticism in a number of studies (see, for example, [5]). The results of the present study demonstrate that this assumption is completely valid.

We will consider a planar crystallization front moving with a velocity $u(\tau) = d\Sigma/d\tau$ (the melt corresponds to $\xi > \Sigma(\tau)$). Considering the thermophysical characteristics and diffusion coefficient of the impurity to be homogeneous, and neglecting the change in specific volume upon solidification and the possibility of formation of a two-phase zone ahead of

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the front, independent of whether this is caused by penetration of dendrites into the melt or formation of crystalline nuclei within the melt, we write the one-dimensional diffusion and thermal conductivity equations in the melt:

$$\frac{\partial\sigma}{\partial\tau} = D\frac{\partial^2\sigma}{\partial\xi^2}, \frac{\partial\theta}{\partial\tau} = a\frac{\partial^2\theta}{\partial\xi^2}, \ \xi > \Sigma(\tau) \tag{1}$$

and the corresponding boundary conditions:

$$u(\tau)\sigma + D\,\partial\sigma/\partial\xi = ku(\tau)\sigma, \ \xi = \Sigma(\tau);$$

$$\lambda_1\partial\theta/\partial\xi + \rho Lu(\tau) = \alpha(\theta - \theta_l), \ \xi = \Sigma(\tau). \tag{2}$$

The first condition of Eq. (2) reflects the continuity of the impurity flux; the distribution coefficient k is considered constant. The second condition on the front corresponds to continuity of the thermal flux with consideration of the heat of phase transition; for simplicity, thermal conductivity in the solid phase will not be considered, and heat loss will be described phenomenologically by introducing an effective heat-liberation coefficient α and external heat-sink temperature θ_l.

Additional conditions on the front are obtained from the requirement of equality of the front temperature to the phase transition temperature, which is dependent on impurity concentration. We write:

$$\theta = \theta_* - m\sigma, \ \xi = \Sigma(\tau), \tag{3}$$

where the coefficient m may be either positive or negative.

At some constant distance from the front in the depths of the melt the temperature and concentration will be regarded as fixed, i.e., we have the boundary conditions

$$\theta = \theta_\infty, \ \sigma = c_\infty, \ \xi = \Sigma(\tau) + H, \tag{4}$$

which can be realized physically in production of a single crystal by the Czochralski method, as well as in other cases of crystallization. Of the five boundary conditions, Eqs. (2)-(4), four are required for correct formulation of the boundary problems for Eq. (1), while one is needed to determine the unknown front velocity. Initial conditions for the temperature and concentration fields can be varied over wide limits.

For uniform motion of the crystallization front at velocity u_0 the problem of Eqs. (1)-(4) can be solved easily. We have

$$\sigma_0 = c^*\left\{1 + \frac{1-k}{k}\exp\left[-\frac{u_0}{D}(\xi - u_0\tau)\right]\right\},$$

$$\theta_0 = \theta_\infty - \theta^*\left\{\exp\left[-\frac{u_0}{a}(\xi - u_0\tau)\right] - \exp\left(-\frac{u_0H}{a}\right)\right\}, \tag{5}$$

where

$$c^* = \frac{c_\infty}{1 + k^{-1}(1-k)\exp(-h)}, \ h = \frac{u_0H}{D},$$

$$\theta^* = \frac{\alpha(\theta_\infty - \theta_l) - \rho Lu_0}{\rho Cu_0 + \alpha[1 - \exp(-\varepsilon h)]}, \ \varepsilon = \frac{D}{a}, \ C = \frac{\lambda_1}{\rho a}, \tag{6}$$

Equations (3)-(5) allow construction of a transcendental equation for determination of u_0. At sufficiently high H to the accuracy of exponentially small terms we obtain

$$u_0 = \frac{\alpha\Delta\theta_1}{\rho(L + C\Delta\theta_2)}, \tag{7}$$

$$\Delta\theta_1 = \theta_* - mc_\infty/k - \theta_l, \ \Delta\theta_2 = \theta_\infty - (\theta_* - mc_\infty/k).$$

Removal of heat from the front to the external sink corresponds to $\Delta\theta_1 > 0$, while solidification under these conditions occurs at $\Delta\theta_2 > 0$. Solidification will also occur with heat supply to the front from without $\Delta\theta_1 < 0$, if simultaneously $\Delta\theta_2 < 0$. The latter may correspond not only to a supercooled, but also to an infinitely superheated melt, if simultaneously the conditions $m > 0$, $k > 1$ or $m < 0$, $k < 1$ are fulfilled. Although usually $m > 0$, $k < 1$ or $m < 0$, $k > 1$, we will perform the analysis below for arbitrary m and k.

Similarly, fusion may occur not only with supply of heat to the front, but also with loss to an external sink. From the relationships presented it is evident that at sufficiently small k (intense removal of the impurity from the front) the real solidification process can scarcely be steady state, even if the front is morphologically stable. In connection with this, [13] reached the somewhat unexpected conclusion that the diffusion equation itself is inapplicable.

It will be convenient to introduce the following dimensionless variables and parameters:

$$t = \frac{u_0^2}{D}\tau,\ \{x,\ X(t)\} = \frac{u_0}{D}\{\xi,\ \Sigma(\tau)\},\ c = \frac{k}{1-k}\left(\frac{\sigma}{c^*} - 1\right),$$

$$\{T,\ T_l,\ T_*,\ T_\infty\} = \frac{1}{mc^*}\{\theta,\ \theta_l,\ \theta_*,\ \theta_\infty\},\ \beta = \frac{\rho L u_0}{\alpha m c^*},\ b_0 = \frac{\rho C u_0}{\alpha}. \qquad (8)$$

Equation (1), conditions (2)-(4), and relationships (5)-(7) can easily be expressed in the dimensionless variables of Eq. (8).

The study of stability of the steady-state solidification process with uniformly moving front must be based on solution of a linearized problem obtained from the dimensionless analog of Eqs. (1)-(4), with initial data for the perturbed dimensionless concentration and temperature fields [14]. Solution of such problems is quite cumbersome, so that we will use a simplified method for analysis of the evolution of small perturbations, which corresponds to specification of initial conditions for the concentration field perturbation in fully defined form. As will be shown in the appendix for the case $h \to \infty$ this has no effect on conclusions as to stability or instability of the process.

We assume that the perturbed dimensionless fields c(t, x) and T(t, x) as well as the dimensionless front coordinate can be expressed in the form

$$c = c_0 + c_1,\ T = T_0 + T_1,\ X = t + X_1,$$

$$|c_1| \ll c_0,\ |T_1| \ll T_0,\ |\dot{X}_1| \ll 1,$$

where the dot indicates differentiation with respect to dimensionless time, and the expressions for c_0 and T_0 follow from Eqs. (5) and (8).

For liquids usually $\varepsilon = D/a \leqslant 10^{-4}$, i.e., the time l^2/a for relaxation of the temperature field in a region with linear dimension l is much less than the analogous time l^2/D for the concentration field. Considering that change in the position of the crystallization front is limited by the slower diffusion process, it is natural to use the quasisteady-state approximation for defining the temperature T(t, x). This quasisteady-state hypothesis significantly simplifies calculations, but is not of principal significance in the sense that all results can easily be generalized to significantly nonsteady-state situations. Using this assumption, from heat-balance condition (2) in the variables of Eq. (8) we obtain

$$\varphi(t) = T(t,\ X) = \frac{T_l + b(\dot{X})T_\infty\dot{X} + \beta\dot{X}}{1 + b(\dot{X})\dot{X}},$$

$$b(\dot{X}) = b_0\,[1 - \exp(-\varepsilon h\dot{X})]^{-1}, \qquad (9)$$

with ε, h, and b_0 being defined in Eqs. (6) and (8). The value of the dimensionless front temperature in the constant front-velocity regime is obtained from Eq. (9) at dX/dt = 1. Therefore, perturbation of the dimensionless front temperature can be written to an accuracy of terms first order in the perturbations as

$$T_1(t,\ X) = T(t,\ X) - T_0(t,\ X) = k^{-1}(1-k)\,R\dot{X}_1, \qquad (10)$$

where

$$R = \frac{(T_\infty - T_l)(b' - b'') + \beta(1 + b'')}{(1 + b')^2}\ \frac{k}{1-k};$$

$$b' = \frac{b_0}{1 - \exp(-\varepsilon h)};\ b'' = \frac{\varepsilon h b_0 \exp(-\varepsilon h)}{[1 - \exp(-\varepsilon h)]^2}. \qquad (11)$$

To the accuracy of terms first order in the perturbations inclusive the values of c_1 and $\partial c_1/\partial x$ at $x = X(t)$ can be replaced by their values at $x = t$, which corresponds to a front moving with constant velocity u_0. To the same accuracy we may write

$$c_0|_{x=X(t)} = c_0|_{x=t} + X_1\,(\partial c_0/\partial x)_{x=t}$$

and a similar expression for $\partial c_0/\partial x$. All this permits "removal" of boundary conditions (2) and (3), transformed to dimensionless form, from the plane $x = X(t)$ to the plane $x = t$. As a result, from Eq. (3) and the first expression of Eq. (2), after calculations we obtain

$$R\dot{X}_1 - X_1 + c_1|_{x=t} = 0, \tag{12}$$
$$\dot{X}_1 + kX_1 + (1-k)\,c_1|_{x=t} + \left.\frac{\partial c_1}{\partial x}\right|_{x=t} = 0.$$

For the function $c_1(t, x)$ we have from Eqs. (1) and (4)

$$\frac{\partial c_1}{\partial t} = \frac{\partial^2 c_1}{\partial x^2},\ x > t;\ c_1 = 0,\ x = t + h, \tag{13}$$

while the value of $c_1(t, t)$ is, generally speaking, an arbitrary function of t, and $c_1(0, x)$ is an arbitrary function of x, satisfying the condition of Eq. (13) at $x = t + h$. It can be shown that the stability or instability of the uniform crystallization-front motion does not depend on the choice of these functions. We take

$$c_1(t, x) = \Psi(x - t)e^{\lambda t},\ X_1(t) = Ve^{\lambda t}. \tag{14}$$

Then

$$\Psi(z) = C\left\{\exp\left[-\frac{(1-\gamma)z}{2}\right] - \exp\left[\gamma h - \frac{(1+\gamma)z}{2}\right]\right\},$$
$$\gamma = -\sqrt{1+4\lambda}. \tag{15}$$

Using Eqs. (14) and (15) in Eq. (12), we obtain a homogeneous system of linear algebraic equations in the constants C and V, the determinant of which is

$$\Delta(\lambda, h, R) = (\lambda + k)(1 - e^{\gamma h}) + (1 - R\lambda)\left[\frac{\gamma}{2} - \frac{1}{2} + (1+\gamma)e^{\gamma h} + (1 - k)(1 - e^{\gamma h})\right]. \tag{16}$$

The eigenvalues λ, which define the behavior of the perturbations, are the roots of the equation $\Delta = 0$. In particular, for $h \gg 1$ we have from Eqs. (11) and (16)

$$\lambda \approx \frac{1}{kR_0}e^{-h},\ R_0 = \frac{(T_\infty - T_l)\,b_0 + \beta}{(1 + b_0)^2}\,\frac{k}{1-k}. \tag{17}$$

In the case of greatest practical interest $m(1 - k) > 0$ (increased capture of impurity by the solid phase, accompanied by increase in phase-transition temperature with increase in concentration, or removal of impurity by the moving front, accompanying decrease in liquidus temperature with increase in concentration). Then, as follows from Eq. (17), the steady-state solidification process with uniform front motion is unstable at any parameter values. In fact, it follows from the instability condition that $R_0 > 0$; if we write R_0 from Eq. (17) in dimensional variables with the aid of Eq. (8) we see that this quantity has the same sign (at $\theta_\infty > \theta_l$) as $m(1 - k)$. Similarly, for the case $m(1 - k) < 0$ we arrive at the conclusion that the solidification process is always stable. In principle, these conclusions remain valid even if the condition $h \gg 1$ is not satisfied, as can be seen from analysis of determinant (16).

We note that as $h \to \infty$, i.e., on transition to the problem of a melt occupying an infinite semispace, the root λ from Eq. (17) vanishes, which corresponds to neutral stability. It is possible that this is related to the fact that proof of the presence of dynamic instability has not been presented previously.

The physical cause of instability development in the cases indicated is the appearance of concentration supercooling ahead of the front moving at constant velocity. In fact, near the front supercooling increases with removal from the front, since on the front itself it is eliminated by the heat of phase transition. Upon a virtual increase in front velocity the front displaces into a region with higher "initial" supercooling, which leads to further increase in velocity. In fact, because of the nonlinearity of Eq. (3), which is not considered herein, at a certain distance from the front supercooling reaches a maximum. Therefore, the growth in velocity is replaced after some time by a decrease due to an increase in the amount of heat upon phase transition and a corresponding decrease in supercooling. As a result, self-oscillating front motion develops.

The conclusion reached above as to the possibility of dynamic instability with respect to front-velocity perturbations without loss of morphological stability has been confirmed by many experiments (see, for example, [8]) and agrees with the physical evaluation of the situation presented in [12]. Doubts of the validity of the hypothesis of [12] are based primarily on observations in which axial segregation of the impurity was accompanied by temperature fluctuations [5, 7], whence it has usually been concluded that these fluctuations are the primary cause of the segregation. Such a categorical conclusion is hardly justifiable, since the self-oscillating process which develops due to disruption of stability of uniform front motion produces oscillations not only of front velocity, but also front temperature, i.e., the latter are a result, not a cause of the instability. (This, of course, does not eliminate the possible effect on segregation of independent temperature oscillations produced by external causes. The corresponding induced front-velocity and impurity-concentration oscillations can be found by solution of Eq. (13), or for weak temperature oscillations, from solution of a system of linear inhomogeneous equations easily obtained therefrom. Temperature oscillations of the external heat sink θ_l can be regarded as periodic modulation of a parameter which can lead to change in the form of the instability regions in the parametric plane.)

Analysis reveals that, in principle, both soft and hard regimes of disruption of stability of uniform planar crystallization-front motion can be realized. In the first case front motion is established with a velocity which performs almost harmonic oscillations relative to the mean value, which may be studied by retaining terms of higher order of smallness in the perturbations in the equations.

Thus, there are two basic types of instability of the steady-state solidification regime: morphological (with respect to perturbations of front form) and dynamic (with respect to velocity perturbations), both types being, in principle, of equal significance. Morphological instability has been studied several times without consideration of dynamic instability, in particular, in [1-5], while dynamic instability has been treated without consideration of morphological in the present study. It is significant that for disruption of the stability of a uniformly moving front there is no necessity that it simultaneously become morphologically unstable, while for disruption of morphological instability it is necessary that the front become dynamically unstable. For the case $m(1 + k) > 0$ a uniformly moving front is dynamically unstable at any parameter values, which cannot be said of morphological instability.

In the general case these types of stability loss are interrelated and therefore, generally speaking, should be studied simultaneously. The criteria for onset of instability of either type obtained without consideration of the other type of instability may not be correct, since the presence of instability of the second type may cause readjustment of the temperature and concentration fields which affects development of instability of the first type.

APPENDIX

The procedure used in the present study is not completely accurate, since the stability problem was not considered as a problem with initial data [14]. However, it can be shown by the method proposed in [14] that the simplification used is justifiable. We will demonstrate this for the case where the melt occupies an infinite volume (a similar analysis can be performed for finite h).

As was shown above, to determine the temperature field the quasisteady-state approximation can be used, so that there is no need to specify initial conditions for temperature. For definiteness we will write the initial conditions for the impurity concentration field in the form $\sigma(0, \xi) = c_\infty$, which in the variables of Eq. (8) is equivalent to $c(0, x) = 0$. The material-balance condition of Eq. (2) and condition (3) in the variables of Eq. (8) appear as

$$[k + (1 - k)c]\dot{X} + \partial c/\partial x = 0,\ T_* - 1 - (1 - k)k^{-1}c = \varphi,\ x = X(t), \tag{18}$$

where φ is determined by Eq. (9).

The solution of the equation following from Eq. (1) for the dimensionless concentration satisfying the boundary condition at infinity and the initial condition c(0, x) = 0 can be written in the form of the thermal potential of a simple layer:

$$c(t,\ x)=\frac{1}{2\sqrt{\pi}}\int\limits_0^t \mu(\tau)\exp\left[-\frac{(x-X(\tau))^2}{4(t-\tau)}\right]\frac{d\tau}{\sqrt{t-\tau}}, \tag{19}$$

where $\mu(t)$ is some unknown function. Using the properties of the thermal potential and transforming in the standard manner to the limit $x \to X(t) + 0$, it is easy to obtain from Eq. (19) expressions for the quantities c and $\partial c/\partial x$ on the crystallization front. As a result, we obtain for determination of the unknown functions $\mu(t)$ and $X(t)$ the integrodifferential equations

$$\lim_{\varepsilon\to 0}\frac{1}{2\sqrt{\pi}}\int\limits_0^t \mu(\tau)\exp\left\{-\frac{[\varepsilon+(X(t)-X(\tau))]^2}{4(t-\tau)}\right\}\frac{d\tau}{\sqrt{t-\tau}}=k(1-k)^{-1}(T_*-1-\varphi), \tag{20}$$

$$\lim_{\varepsilon\to 0}\frac{1}{4\sqrt{\pi}}\int\limits_0^t \mu(\tau)[\varepsilon+(X(t)-X(\tau))]\exp\left\{-\frac{[\varepsilon+(X(t)-X(\tau))]^2}{4(t-\tau)}\right\}\frac{d\tau}{(t-\tau)^{3/2}}=k(T_*-\varphi)\dot{X},$$

following from conditions (18). In the steady-state regime at constant front-motion velocity by definition $X(t) = t$, $\mu(t) = \mu_0$. In this case after simple calculations we arrive at the steady-state impurity-concentration distribution obtained above, while

$$\mu_0=\frac{(T_*-1-\varphi_0)k}{1-k}=1,\ \varphi_0=\frac{T_l+b_0T_\infty+\beta}{1+b_0}, \tag{21}$$

where φ_0 is the temperature on the front.

Such a solution is realized asymptotically as $t \to \infty$. We assume that at a time $t_1 \gg 1$, when to a high accuracy $X(t) = t$, $\mu(t) = \mu_0$ (i.e., a solidification regime with constant front-motion velocity has been established), small perturbations appear in the front coordinate, its derivatives with respect to t, and the function $\mu(t)$. We take for $t > t_1$

$$\begin{gathered}X(t)=t+z(t),\ \dot{X}=1+v(t),\ |v(t)|=|\dot{z}(t)|\ll 1,\\ \mu(t)=1+w(t),\ |w(t)|\ll 1,\ |z(t)|\ll t-t_1.\end{gathered} \tag{22}$$

Substituting Eq. (22) into Eq. (20) and considering only terms of first order in the perturbations, we easily obtain integrals describing the perturbation $c_1(t, x)$ of the dimensionless concentration field $c(t, x)$ and $\partial c_1(t, x)/\partial x$. Transforming to the limit $x \to X(t) + 0$ and introducing as a new dimensionless variable the quantity $t - t_1$, we have

$$c_1(t,\ X(t))=\frac{1}{2\sqrt{\pi}}\int\limits_0^t\left\{w(\tau)-\frac{1}{2}[z(t)-z(\tau)]\right\}\exp\left(-\frac{t-\tau}{4}\right)\frac{d\tau}{\sqrt{t-\tau}},$$

$$\begin{aligned}-\left.\frac{\partial c_1}{\partial x}\right|_{x=X(t)}={}&\frac{1}{4\sqrt{\pi}}\int\limits_0^t[z(t)-z(\tau)]\exp\left(-\frac{t-\tau}{4}\right)\frac{d\tau}{(t-\tau)^{3/2}}+\\&+\frac{1}{4\sqrt{\pi}}\int\limits_0^t\left\{w(\tau)-\frac{1}{2}[z(t)-z(\tau)]\right\}\exp\left(-\frac{t-\tau}{4}\right)\frac{d\tau}{(t-\tau)^{1/2}}+\\&+\lim_{\varepsilon\to 0}\frac{\varepsilon}{4\sqrt{\pi}}\int\limits_0^t w(\tau)\exp\left[-\frac{\varepsilon^2}{4(t-\tau)}-\frac{t-\tau}{4}\right]\frac{d\tau}{(t-\tau)^{3/2}}.\end{aligned} \tag{23}$$

The equations for the unknowns $w(t)$ and $z(t)$ (or $v(t)$) are obtained by using Eq. (22) as the corresponding perturbations of Eq. (18). Using the relationship between temperature perturbations on the front and the crystallization-front velocity, Eq. (10), we obtain

$$c_1(t,\ X(t))=-R_0v(t)=-R_0\dot{z}(t), \tag{24}$$

$$-\left.\frac{\partial c_1(t,\ x)}{\partial x}\right|_{x=X(t)}=Sv(t)=S\dot{z}(t);\ S=-(1-k)R_0+1, \tag{25}$$

where the parameter R_0 is defined by Eq. (17), and the quantities on the left side are defined by Eq. (23). Taking the Laplace transform of Eq. (24), expanding in a Taylor series

$$z(t)-z(\tau)=\sum_{n=1}^{\infty}\frac{1}{n!}\,z^{(n)}(\tau)\,(t-\tau) \tag{26}$$

and assuming that the integrands of Eq. (23) possess all the properties required in order to interchange the order of summation and integration, we arrive at representations of the integrals of Eq. (23) in the form of infinite sums of the convolutions of various functions. Using theorems on the representation of derivatives and convolutions and tabular representations, we obtain for the terms of these sums:

$$\frac{1}{2\sqrt{\pi}}\int_0^t w(\tau)\exp\left(-\frac{t-\tau}{4}\right)\frac{d\tau}{\sqrt{t-\tau}}\div\frac{W(p)}{\sqrt{1+4p}},$$

$$\lim_{\varepsilon\to 0}\frac{\varepsilon}{4\sqrt{\pi}}\int_0^t w(\tau)\exp\left[-\frac{\varepsilon^2}{4(t-\tau)}-\frac{t-\tau}{4}\right]\frac{d\tau}{(t-\tau)^{3/2}}\div\frac{W(p)}{2},$$

$$I_j=\int_0^t [z(t)-z(\tau)]\exp\left(-\frac{t-\tau}{4}\right)\frac{d\tau}{(t-\tau)^{j+1/2}}\div$$

$$\div\sum_{n=1}^{\infty}\frac{1}{n!}\frac{\Gamma(n-j+1/2)}{(p+1/4)^{n-j+1/2}}\left[p^nZ(p)-\sum_{i=1}^{n}p^{n-i}z^{(i-1)}(0)\right],$$

where $W(p)$ and $Z(p)$ are representations of $w(t)$ and $z(t)$; $j = 0, 1$.

It is possible to sum the series appearing in the representations of the integrals of Eq. (23) by using the fundamental integral concept for the gamma-function, changing the order of summation and integration, and summing the series appearing in the integrand. As a result, we obtain

$$I_j\div Z(p)\left(p+\frac{1}{4}\right)^{j-1/2}\int_0^{\infty}\left[\exp\frac{4px}{1+4p}-1\right]e^{-x}x^{-j-1/2}dx+$$

$$+8\sqrt{\pi}J_j(p), \tag{27}$$

where $J_j(p)$ are some series in powers of p with coefficients dependent on the values of the derivatives of $z(t)$ of various order at $t = 0$. With consideration of Eq. (27) we write the Laplace transforms of Eq. (24) as

$$-\frac{W(p)}{\gamma(p)}+\left[R_0p-\frac{1}{2}\left(1+\frac{1}{\gamma(p)}\right)\right]Z(p)=R_0z(0)+2J_0(p),$$

$$\frac{W(p)}{2}\left(1-\frac{1}{\gamma(p)}\right)+\left[(1-k)R_0p-p-\frac{1}{4}\left(\gamma(p)+2+\right.\right. \tag{28}$$

$$\left.\left.+\frac{1}{\gamma(p)}\right)\right]Z(p)=-(1-(1-k)R_0)z(0)-2J_1(p)+J_0(p),\ \gamma(p)=-\sqrt{1+4p}.$$

It can easily be seen that the determinant $\Delta(p, h)$ of this system coincides with $\Delta(\lambda, \infty, R)$ when λ is replaced by p. A similar correspondence occurs at finite h. The unknown representations $W(p)$ and $Z(p)$ can be expressed from a system analogous to Eq. (28) as fractions obtained by dividing certain polynomials in p by the determinant $\Delta(p, h)$. In the corresponding asymptotic representations of the originals $w(t)$ and $z(t)$ as $t \to \infty$ there appear terms containing the exponential factor $\exp\omega t$, where ω is the root of the equation $\Delta(\omega, h, R) = 0$. In view of the unique correspondence of the determinant $\Delta(\lambda, h, R)$ to the determinant $\Delta(p, h)$ the stability analysis performed above proved justifiable. We note that an analogous result is also obtained in the case in which in place of expansion (26) we use exponential representations of $w(t)$ and $z(t)$ in Eq. (23).

It can be shown that the conclusion of system stability or instability does not depend on whether polynomials $J_j(p)$ with form determined by initial conditions for the time derivatives of the front coordinate are considered or not considered in Eq. (28). Such time derivatives depend on the initial condition imposed on the concentration perturbation in the region $x > X(t)$, i.e., the exact form of the initial condition does not affect the appearance of instability, as was proposed above.

NOTATION

a, thermal diffusivity coefficient in melt; b_0, b', b'', parameters introduced in Eqs. (8) and (11), respectively; C, specific heat of melt; c, c_1, dimensionless concentration and its perturbation; c^*, θ^*, parameters introduced in Eq. (6); D, diffusion coefficient; H, length of region occupied by melt; h, dimensionless length of region occupied by melt; J, function introduced in Eq. (27); k, impurity distribution coefficient; L, specific heat of phase transition; m, slope of liquidus line; R, R_0, S, parameters introduced in Eqs. (11), (17), (25); T, dimensionless temperature; t, dimensionless time; u, crystallization-front velocity; v, w, perturbations of dimensionless velocity and quantity μ; x, dimensionless coordinate; X, z, dimensionless coordinate and its perturbation; α, effective heat-transfer coefficient; β, parameter introduced in Eq. (8); γ, function introduced in Eq. (15); Δ, characteristic determinant of Eq. (12); ε, parameter introduced in Eq. (6); θ, θ_*, θ_l, temperature, temperature on phase-transition front of pure melt, and external heat-sink temperature; $\Delta\theta_1$, $\Delta\theta_2$, temperature differentials introduced in Eq. (7); λ_1, thermal conductivity coefficient in melt; λ, parameter introduced in Eq. (14); μ, function introduced in Eq. (19); ξ, coordinate; ρ, melt density; σ, impurity concentration by weight; τ, time, φ, temperature at front; Ψ, function introduced in Eq. (14). Subscripts: 0, steady-state solidification regime; ∞, value far from front.

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