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THEORY OF THE HARDENING OF BINARY MELTS WITH AN EQUILIBRIUM TWO-PHASE ZONE

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Descriptions of processes involved in directed crystallization is normally accomplished on the basis of concepts dealing with the existence of a clearly defined phase-transition front and reduces to the solution of various versions of the Stefan problem [1-3]. If the liquid that is subjected to hardening is one consisting of numerous components, the motion of the front is accompanied by a redistribution of the composition of the phases, and in addition to the equations of heat conduction, it is necessary also to deal with the equations of diffusion and the relationship between the temperature of the phase transition and the composition of the melt, or its dependence on the composition of the solution near the front. Under specific conditions the effective temperature of the liquidus ahead of the front proves to be higher than the temperature of the liquid phase, i.e., a metastable supercooled zone is formed [4]. The same situation is encountered in the hardening of supercooled single- or multicomponent liquids.

In the metastable region conditions prevail for the growth of the solid phase nuclei, generated spontaneously or in the impurity crystallization cores. Moreover, the front becomes morphologically unstable, which may lead to the development of a system of dendrites. Either mechanism enhances the appearance of a transition two-phase zone (in which the liquid and solid phases coexist) ahead of the front, as well as to the partial removal of the supercooling. In the general case this zone is thermodynamically in a state of nonequilibrium, and its characteristics determine the relationship between the kinetic processes of formation and the growth of the solid-phase elements, as well as the velocity at which the front is displaced. Supercooling ranging from the very lowest to several tens of degrees has been experimentally established [5-8].

The traditional frontal formulation describes approximately the situation in which the two-phase zone is almost entirely absent, which is characteristic of pure liquids under conditions in which the morphological instability stimulates development of cellular structures, but no dendrites (the majority of semiconductor and certain metal melts). In the opposite extreme case (melts with nuclei or crystallization catalysts, liquid seals, true aqueous solutions) it is permissible to use an approximation of an equilibrium two-phase zone in which supercooling has been entirely removed [9-12]. Then, in connection with the fact that the literature is full of an excess of categorically extreme assertions either as to the significance of concentration supercooling in those cases in which it should appear

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in accordance with frontal theory [13], or in terms of the physical inadequacy of models which tolerate this supercooling in any form [14, 15]. There is no basis for such assertions, if for no other reason than that they contradict experimental facts (see, for example, [5-8]).

Hardening processes with a two-phase zone result in extremely complex problems, and in most of the research cumbersome numerical methods have therefore been used. These produce results that are poorly predictable, thus hindering generalization and the derivation of a clear concept as to the manner in which the physical and regime parameters exert their influence (see, for example, the review in [12]). The few attempts to analytically analyze the structure of a two-phase zone have usually been based on extreme and seriously disputed assumptions [11, 16, 17]. Thus, in [11] diffusion in this zone is neglected, and this leads to conclusions that are in poor agreement with experiment [18].

The processes of heat and mass transfer in an equilibrium two-phase zone are dealt with in the following with the aid of the small parameter method. If the thickness of the zone is small in comparison to the linear scales of the process, it can be replaced by a discontinuity surface with boundary conditions defined by the structure of the zone and differing substantially from the conditions specified at a conventional phase transition front.

1. Basic Equations. Let us examine regions 1 and 2, occupied, respectively, by a binary melt or a solution and a solid. The boundaries of these regions consist of the external parts Γ_1 and Γ_2 and the surfaces Σ_1 and Σ_2, between which this two-phase zone is situated. For purposes of simplification, the densities, specific heat capacities, coefficients of thermal conductivity and diffusion, the specific heat of fusion, and the coefficient of impurity distribution among the phases, are held to be independent of temperature and concentration. We assume the phase densities to be identical, which offers us the opportunity of ignoring the convective transfer of heat and mass. In approximate terms, we assume the two-phase zone to be in thermodynamic equilibrium. This means that the temperatures of the phases in this zone are identical and related to the impurity concentration by a relationship which determines the liquidus line.

The processes of heat and mass transfer are described by the following equations: in regions 1 and 2.

$$\partial\theta_j/\partial\tau = a_j\Delta\theta_j,\ \partial\sigma_j/\partial\tau = D_j\Delta\sigma_j,\ j = 1, 2 \tag{1.1}$$

(θ_j and σ_j are the temperature and concentration, and a_j and D_j are the coefficients of thermal diffusivity and diffusion) while in the two-phase zone we have

$$\rho\frac{\partial(C\theta)}{\partial\tau} = \nabla(\lambda\nabla\theta) + \rho L\frac{\partial\varphi}{\partial\tau},\quad \frac{\partial}{\partial\tau}[(1-\varphi)\sigma] = \nabla(D\nabla\sigma) - k\sigma\frac{\partial\varphi}{\partial\tau}. \tag{1.2}$$

Here σ φ, and θ denote the concentration of the impurities in the liquid phase of the zone, the volumetric concentration of the solid phase, and temperature; C and ρ represent the mean specific heat capacity and density; λ and D are the effective coefficients of thermal conductivity and diffusion, dependent on φ; k is the coefficient of impurity distribution; L is the specific heat of fusion.

We will write the equation for the liquidus line in linearized form:

$$\theta = \theta_0 - m\sigma, \tag{1.3}$$

where θ_0 is the crystallization temperature for a single-component solvent, and the parameter m may carry any sign. For the sake of definiteness, we will assume it to be positive in the following. Generalization of the theory for negative m, as well as for situations in which the linear relationship (1.3) is not suited, raises no fundamental difficulties.

At the boundary of separation Σ_1 between the two-phase zone and the melt we have

$$\begin{gathered}\varphi = 0,\ \theta = \theta_1,\ \sigma = \sigma_1,\ \lambda\mathbf{n}_1\nabla\theta = \lambda_1\mathbf{n}_1\nabla\theta_1,\\ D\mathbf{n}_1\nabla\sigma = D_1\mathbf{n}_1\nabla\sigma_1.\end{gathered} \tag{1.4}$$

At the surface of separation Σ_2 between the two-phase zone and the solid alloy the following conditions must be satisfied:

$$\theta_2 = \theta,\ \sigma_2 = k\sigma,\ \lambda_2 \mathbf{n}_2 \nabla \theta_2 - \lambda \mathbf{n}_2 \nabla \theta = \\ = \rho L(1-\varphi)u,\ D_2 \mathbf{n}_2 \nabla \sigma_2 - D \mathbf{n}_2 \nabla \sigma = (1-k)(1-\varphi)\sigma u \tag{1.5}$$

($\mathbf{n}_1$ and $\mathbf{n}_2$ are the unit vectors of the normal to Σ_1 and Σ_2, directed in the direction of the melt and u represents the local displacement velocity for Σ_2 in the direction of $\mathbf{n}_2$). The two last conditions in (1.5) represent the equations of balance for heat and mass at the alloy surface.

The boundary conditions determined by the specific manner in which the hardening process has been organized must be specified at the surfaces Γ_1 and Γ_2. These, as well as the initial conditions imposed on the unknowns of system (1.1), (1.2) do not affect the aims of this study and are therefore not dealt with below. The solution of the formulated problem must determine both the indicated unknowns and the surfaces Σ_1 and Σ_2.

If the two-phase zone is not present, then Σ_1 and $\mathbf{n}_1$ coincide with Σ_2 and $\mathbf{n}_2$. In this event we have to solve only Eq. (1.1) with the boundary conditions at the surface of phase separation

$$\theta_1 = \theta_2 = \theta_0 - m\sigma_1,\ \sigma_2 = k\sigma_1,\ \lambda_2 \mathbf{n} \nabla \theta_2 - \lambda_1 \mathbf{n} \nabla \theta_1 = \rho L u, \\ D_2 \mathbf{n} \nabla \sigma_2 - D_1 \mathbf{n} \nabla \sigma_1 = (1-k)\sigma_1 u, \tag{1.6}$$

which follow out of (1.3)-(1.5), i.e., we arrive at a standard formulation of the frontal problem related to the directed hardening of a binary melt.

Substantial simplification in the formulated problem can be achieved by taking note, first of all, that D_2 is usually many orders of magnitude lower than D_1. This indicates that in the majority of cases there is absolutely no sense in dealing with the diffusion of the impurity in the solid material. Second, the coefficients of thermal diffusivity in each of the cases are several orders of magnitude higher than the diffusion factor for the impurity in the melt and the relaxation times of the temperature fields are consequently smaller than the relaxation times of the concentration fields. Therefore, with a high degree of accuracy, we can limit ourselves to an analysis of the processes that are quasisteady in terms of heat transfer, neglecting the time derivatives in the first of the equations in (1.1) and (1.2). The simplified formulation of the problem follows directly out of (1.1)-(1.5) and is therefore not written out here.

2. Equations for the Two-Phase Zone. Let us assume that the curvature radii (~R) of the surfaces Σ_1 and Σ_2 are considerably greater than the distance δ between them, and let us represent the temperature and concentration fields in the melt near these surfaces in the following form:

$$\theta_1 = \theta_{10} + g_1\xi,\ \theta_2 = \theta_{20} + g_2(\xi + \delta),\ \sigma_1 = \sigma_{10} + h_1\xi, \tag{2.1}$$

where ξ is a coordinate reckoned along the normal to the surface Σ_1, on which $\xi = 0$, in the direction of the melt. It follows from (1.3) and (1.4) that

$$\theta_{10} = \theta_0 - m\sigma_{10},\ \theta_{20} = \theta_0 - m\sigma|_{\xi=-\delta},\ h_1 = -g_1/m. \tag{2.2}$$

The quantities θ_{j0}, σ_{10}, and g_j depend on the tangential coordinates with scale R and on time. In view of the assumed inequalitity $R \gg \delta$ we can neglect the coordinate dependence of these quantities in the transformation of Eqs. (1.2). As a result, in the accompanying system of coordinates, from (1.2) and (1.3) we derive the equations for the two-phase zone:

$$\rho L\left(\frac{\partial}{\partial\tau} - u\frac{\partial}{\partial\xi}\right)\varphi = m\frac{\partial}{\partial\xi}\left(\lambda\frac{\partial\sigma}{\partial\xi}\right), \\ \left(\frac{\partial}{\partial\tau} - u\frac{\partial}{\partial\xi}\right)[(1-\varphi)\sigma] = \frac{\partial}{\partial\xi}\left(D\frac{\partial\sigma}{\partial\xi}\right) - k\sigma\left(\frac{\partial}{\partial\tau} - u\frac{\partial}{\partial\xi}\right)\varphi, \tag{2.3}$$

while from (1.4) and (1.5), with consideration of (2.1) and (2.2), we derive the pertinent boundary conditions

$$\varphi = 0,\ \sigma = \sigma_{10},\quad \partial\sigma/\partial\xi = -g_1/m,\quad \xi = 0; \\ \lambda_2 g_2 + m\lambda_*\partial\sigma/\partial\xi = \rho L(1-\varphi_*)u,\quad \xi = -\delta; \\ D_*\partial\sigma/\partial\xi = -(1-k)(1-\varphi_*)\sigma u,\quad \xi = -\delta. \tag{2.4}$$

Here we have introduced $\lambda_* = \lambda(\varphi_*)$ and $D_* = D(\varphi_*)$ (φ_* is the value of φ when $\xi = -\delta$) and it has been taken into consideration that $D(0) = D_1$ and $\lambda(0) = \lambda_1$.

Problem (2.3), (2.4) describes the structure of the two-phase zone in one-dimensional processes of directed crystallization under specified flows of heat and impurity mass. In the most general case expression (2.1) should be treated as an internal asymptotic expansion of the fields of temperature and concentration in the liquid and solid phases.

We now introduce the characteristic frequency ω for change in parameters in (2.1) and the natural scale for the duration of the hardening process, i.e., D_1/u_0 (u_0 is the characteristic value for the velocity of the alloy surface displacement), as well as the following dimensionless variables and parameters:

$$t = \omega\tau, \quad x = \frac{\xi}{\delta}, \quad c = \frac{\sigma}{\sigma_0}, \quad c_{10} = \frac{\sigma_{10}}{\sigma_0}, \quad \varepsilon = \frac{u_0}{D_1}\delta, \tag{2.5}$$

$$P = \frac{m\sigma_0}{D_1}\frac{\lambda_2}{\rho L}, \quad G_j = \frac{D_1 g_j}{m\sigma_0 u_0}, \quad \gamma = \frac{D_1\omega}{u_0^2}, \quad s = \frac{u}{u_0}$$

(σ_0 is the characteristic concentration value). In these variables from (2.3) and (2.4) we derive the problem

$$\begin{gathered}
\varepsilon^2\gamma\frac{\partial\varphi}{\partial t} = P\frac{\partial}{\partial x}\left(\frac{\lambda}{\lambda_2}\frac{\partial c}{\partial x}\right) + \varepsilon(s + \varepsilon\gamma f x)\frac{\partial\varphi}{\partial x}, \quad f = \frac{d\ln\delta}{dt}, \\
\varepsilon^2\gamma\left[(1-\varphi)\frac{\partial c}{\partial t} - (1-k)c\frac{\partial\varphi}{\partial t}\right] = \frac{\partial}{\partial x}\left(\frac{D}{D_1}\frac{\partial c}{\partial x}\right) + \\
+ \varepsilon(s + \varepsilon\gamma f x)\left[(1-\varphi)\frac{\partial c}{\partial x} - (1-k)c\frac{\partial\varphi}{\partial x}\right]; \\
\varphi = 0,\ c = c_{10},\ \partial c/\partial x = -\varepsilon G_1,\ x = 0; \\
P\frac{\lambda_*}{\lambda_2}\frac{\partial c}{\partial x} + \varepsilon P G_2 = \varepsilon(1-\varphi_*)s, \quad \frac{D_*}{D_1}\frac{\partial c}{\partial x} = -\varepsilon(1-k)(1-\varphi_*)sc, \quad x = -1
\end{gathered} \tag{2.6}$$

(the specification of the initial conditions, not written out here, is also understood).

An analysis of the experimental data shows that in the growth of monocrystals the dimensionless thickness ε of the two-phase zone is usually small in comparison to unity. This is obviously also valid for any processes of directed hardening, provided that the parameter values remain close to the critical, where the condition of metastability is first encountered. At the same time, there exist processes (for example, the continuous casting of steel [18]) in which ε is several orders of magnitude greater than unity. It is therefore expedient to examine the extreme cases of small and large ε, using the small parameter method.

In order to close (2.6) we have to find the relationship between the transfer coefficients λ/λ_2 and D/D_1 in their dependence on φ, and these are determined by the topological structure of the two-phase zone. Here we will use the simplest formulas

$$\lambda/\lambda_2 = (1-\varphi)\varkappa + \varphi,\ D/D_1 = 1 - \varphi,\ \varkappa = \lambda_1/\lambda_2, \tag{2.7}$$

corresponding to the familiar rule of mixtures. These formulas must correspond nearly to reality, if it is dendrites that the zone contains predominantly. For a zone with discrete crystals it is, of course, more correct to use the results from transfer theory in disperse mixtures.

3. Structure of a Narrow Two-Phase Zone.

Let us examine the solutions of problem (2.6) with the closing relationships (2.7) for the case in which $\varepsilon \ll 1$. We will assume that

$$c = c_{10} + \varepsilon(\psi_0 + \varepsilon\psi_1 + \varepsilon^2\psi_2 + \ldots),\ \varphi = \varepsilon\varphi_1 + \varepsilon^2\varphi_2 + \ldots \tag{3.1}$$

Substituting this into (2.6) and separating the terms of various exponents of ε, we obtain the problems for ψ_i and φ_i. Assuming, for the sake of definiteness, that $\gamma \lesssim 1$, $s \sim 1$, from the relationship of the first order with respect to ε we have

$$\psi_0 = -G_1 x. \tag{3.2}$$

The second-order equations subsequent to the single integration involving the use of the boundary conditions for x = 0 and (2.7) assume the form

$$P\varkappa\partial\psi_1/\partial x + [s - P(1-\varkappa)G_1]\varphi_1 = 0,$$
$$\partial\psi_1/\partial x + [-sc_{10}(1-k) + G_1]\varphi_1 = (\gamma dc_{10}/dt + sG_1)x.$$

Hence it follows that

$$\psi_1 = Ax^2/2, \quad \varphi_1 = Bx, \quad A = \frac{P(1-\varkappa)G_1 - s}{P\varkappa}B, \qquad B = \frac{P\varkappa(\gamma\, dc_{10}/dt + sG_1)}{P\varkappa[G_1 - (1-k)sc_{10}] + P(1-\varkappa)G_1 - s}. \tag{3.3}$$

The process of deriving the subsequent terms of the series in (3.1) is easily extended; the use of the small-parameter method in this case involves no singularities.

With accuracy to terms on the order of ε^2 we will write out the conditions for x = −1 inclusively. Using (2.7) and (3.1)-(3.3), from (2.6) we have

$$P(G_2 - \varkappa G_1) + \varepsilon P(-\varkappa A + (1-\varkappa)G_1B) = (1+\varepsilon B)s, \qquad G_1 + \varepsilon A = (1-k)(c_{10} + \varepsilon G_1)s. \tag{3.4}$$

Additionally assuming that $s = s_0 + \varepsilon s_1 + \ldots$ and bearing in mind in the first of the relationships (3.4) only the principal terms (on the order of ε^0), we have

$$s_0 = P(G_2 - \varkappa G_1). \tag{3.5}$$

From the second of the relationships in (3.4) we have

$$\varepsilon = \frac{G_1 - (1-k)s_0c_{10}}{(1-k)(s_0G_1 + s_1c_{10}) - A}.$$

Using this in (3.3), within the limits of the assumed accuracy, we write

$$A = \gamma dc_{10}/dt + s_0G_1, \; B = \varkappa A/(G_1 - G_2). \tag{3.6}$$

In the first of the relationships in (3.4) we will examine the terms with the order of ε and this yields $s_1 = 0$. This is quite natural, since the rate of the process is limited by the removal of heat from the crystallization zone, which is determined by the temperature gradients g_1 and g_2, but not by the structure of this zone. Therefore,

$$\varepsilon = \frac{(1-k)s_0c_{10} - G_1}{\gamma\, dc_{10}/dt + ks_0G_1}, \tag{3.7}$$

and here, in this case, and in (3.6), s_0 can be replaced by s. Returning to the dimensional variables with the aid of (2.5), from (3.1)-(3.7) we derive

$$u = \frac{\lambda_2 g_2 - \lambda_1 g_1}{\rho L}, \quad \sigma = \sigma_{10} - \frac{g_1}{m}\xi + \frac{1}{2D_1}\left(\frac{d\sigma_{10}}{d\tau} + \frac{g_1}{m}u\right)\xi^2,$$
$$\varphi = \frac{\varkappa}{D_1}\frac{m\, d\sigma_{10}/d\tau + ug_1}{g_1 - g_2}\xi, \quad \xi \in (-\delta, 0), \quad \delta = \frac{(1-k)u\sigma_{10} - D_1g_1/m}{d\sigma_{10}/d\tau + kug_1/m}. \tag{3.8}$$

As $\varepsilon \to 0$, from (3.4) we could derive the expression in (3.8) for u and the following relationship [taking (2.2) into consideration]:

$$\sigma_{10} = \sigma_1|_{\xi=0} = \frac{1}{1-k}\frac{D_1}{u}\frac{g_1}{m} = -\frac{1}{1-k}\frac{D_1}{u}\frac{\partial\sigma_1}{\partial\xi}\bigg|_{\xi=0},$$

describing the quasisteady motion of the crystallization front without the two-phase zone.

We can see from (3.8) that the nonsteadiness of the hardening process may have a significant effect on the characteristics of the two-phase zone. In particular, when m > 0 and k < 1, the establishment of a steady regime is accompanied by progressive expulsion of the impurity in the direction of the melt ($d\sigma_{10}/d\tau > 0$), which corresponds to an increase in the thickness of the zone from the initial zero to some steady value. When $m < 0$ (and $k > 1$) the surface concentration diminishes monotonically, and the derivative $d\sigma_{10}/d\tau < 0$ declines in absolute value, i.e., an analogous conclusion regarding the change in the thickness of the two-phase zone follows out of (3.8).

The quantities δ, u, σ, and φ must, of course, be positive. In the steady regime, together with the requirement of smallness for the dimensionless thickness ε of the two-phase zone, this corresponds to satisfaction of the inequalities

$$0 < m(1-k)u\sigma_{10} - D_1 g_1 \ll kD_1 g_1, \tag{3.9}$$

whose significance will be clarified below.

4. The Broad Two-Phase Zone. Let us now examine the opposite extreme case $\varepsilon \gg 1$, assuming for the sake of simplicity that $\varepsilon^2\gamma \lesssim 1$, i.e., that the hardening process is nearly steady. Then, in the second of the equations in (2.6) retaining the higher-order terms with respect to ε, we formally obtain $(1-\varphi)\partial c/\partial x - (1-k)c\partial\varphi/\partial x = 0$ and also

$$\varphi = 1 - (c_{10}/c)^n,\ n = 1/(1-k). \tag{4.1}$$

This corresponds to the model put forth by V. T. Borisov [10-12], in which the diffusion of the impurity in the two-phase zone is neglected. According to (4.1) and the boundary condition for $\partial c/\partial x$ in (2.6) for P, s, and $c_{10} \sim 1$, we have $\partial c/\partial x \sim \varepsilon G_1$ (we will drop the modulus notation here and below). From the first of the equations in (2.6) it follows that $\partial^2 c/\partial x^2 \sim \varepsilon\partial\varphi/\partial x \sim \varepsilon^2 G_1$. Now evaluating the terms in the second equation, we write

$$\varepsilon s\left[(1-\varphi)\frac{\partial c}{\partial x} - (1-k)c\frac{\partial\varphi}{\partial x}\right] \sim \varepsilon^2 G_1, \quad \frac{\partial}{\partial x}\left(\frac{D}{D_1}\frac{\partial c}{\partial x}\right) \sim \varepsilon^2 G_1,$$

i.e., the diffusion term which was neglected in the derivation of (4.1) in actuality has the same order of magnitude as the terms that we retain. Thus, the procedure used to derive (4.1) and this relationship itself cannot be held to be correct and we cannot neglect diffusion in the two-phase zone.

It is also highly probable that when the zone is in a state of equilibrium, its dimensionless thickness cannot possibly be greater than unity. In order to demonstrate this contention from its qualitative aspects, let us examine the second of the equations in (2.6), using (2.7), assuming that $s \approx 1$ and replacing $(1-\varphi)^{-1}\partial\varphi/\partial x$ by the zone-averaged value of $T = \ln(1-\varphi_*) \sim 1$. Then

$$d^2c/dx^2 + (\varepsilon - T)dc/dx + \varepsilon(k-1)Tc \approx 0.$$

When $\varepsilon \gg 1$ the solution of this equation, satisfying the conditions with $x = 0$ in (2.6), has the form

$$c \approx G_1 e^{-\varepsilon x} + (c_{10} - G_1)e^{(1-k)Tx}.$$

Substituting this into the boundary condition for $x = -1$ in (2.6), describing the balance of mass and the surface of the alloy, we obtain

$$\varepsilon \approx -\ln\frac{kG_1}{(c_{10}-G_1)(1-k)} - (1-k)T. \tag{4.2}$$

Hence it follows that for real processes $\varepsilon \sim 1$. Apparently, broad two-phase zones, such as those that are observed, for example, in the continuous pouring of steel [18], can hardly be regarded as ones that are in equilibrium. It is possible that the primary cause for the divergence between experiment and theory, as discussed in [18], is to be found in this fact.

5. Formulation of Problems Dealing with Directed Hardening in an Equilibrium Two-Phase Zone. If the thickness δ of the two-phase zone is small in comparison to the linear scales of the process and, in particular, with the curvature radii of the alloy surface, it is natural to replace this zone by the discontinuity surface at which not only the concentration but temperature undergo discontinuities.

When diffusion in the melt and the relaxation effect accompanying the establishment of quasisteady fields of temperature on both sides of the two-phase zone are neglected, for the unknown functions θ_1, θ_2, and σ_1 from (1.1) we obtain the following equations:

$$\Delta\theta_1 = \Delta\theta_2 = 0, \partial\sigma_1/\partial\tau = D_1\Delta\sigma_1. \tag{5.1}$$

The boundary conditions at the discontinuity surface replacing the zone follow out of (1.3)-(1.5), (2.2) and the contents of Sec. 3. Thus we have

$$\begin{gathered} \theta_1 = \theta_2 + \theta_{12} = \theta_0 - m\sigma_1, \ \mathbf{n}\nabla\theta_1 = -m\mathbf{n}\nabla\sigma_1, \\ \lambda_2\mathbf{n}\nabla\theta_2 - \lambda_1\mathbf{n}\nabla\theta_1 = \rho L u, \end{gathered} \tag{5.2}$$

where the jump in the temperature in the zone, provided that the latter is narrow, is given by

$$\begin{gathered} \theta_{12} = -m\sigma_{12} = \left[\mathbf{n}\nabla\theta_1\left(1 + \frac{u\delta}{2D_1}\right) + \frac{m\delta}{2D_1}\frac{\partial\sigma_1}{\partial\tau}\right]\delta, \\ \delta = [m(1-k)u\sigma_1 - D_1\mathbf{n}\nabla\theta_1](ku\mathbf{n}\nabla\theta_1 + m\partial\sigma_1/\partial\tau)^{-1}. \end{gathered} \tag{5.3}$$

If there is no two-phase zone, the same boundary conditions as in (5.2) are obtained from (1.6), but with the conditions for the derivatives of θ_1 and σ_1 with respect to the normal coordinate written in (1.6) replaced by the condition of material balance.

Relations (5.3) correspond to the model of a narrow two-phase zone, but the form of the condition (5.2) does not depend on this model: the structure of the zone affects only the magnitude of the temperature discontinuity θ_{12}. If we are dealing with processes that have specified flows of heat to the external boundaries of the system, this discontinuity does not affect the gradients of the temperature fields. The solution of problem (5.1), (5.2), given appropriate initial conditions, then fully describes the macrokinetics of such processes for an arbitrary value of ε, i.e., with an arbitrary structure for the equilibrium zone.

The condition of absence of concentration supercooling ahead of a clearly defined phase-transition front obviously involves satisfaction of the inequalities $-m\mathbf{n}\nabla\sigma_1 < \mathbf{n}\nabla\theta_1$ [or $-mh_1 < g_1$, see (2.1)]. If this condition is violated in some segment of the separation surface, a two-phase zone is formed ahead of such a segment, and under the conditions of thermodynamic equilibrium we have $-m\mathbf{n}\nabla\sigma_1 = \mathbf{n}\nabla\theta_1$. Thus, in the general case discontinuous boundary conditions are specified for this surface.

For purposes of illustration let us examine the steady-state hardening process for given temperature gradients (and flows of heat) at some distance from the surface of separation. It follows from the conventional frontal formulation that

$$\sigma_1 = \sigma_0\left[1 + \frac{1-k}{k}\exp\left(-\frac{u\xi}{D_1}\right)\right], \tag{5.4}$$

where σ_0 is the concentration of the melt at some distance from the front, as well as the expression in (3.8) for the velocity u. The condition for the validity of (5.4) reduces to the requirement that concentrated supercooling be absent, the latter occurring when $m\sigma_0(1 - k)u(kD_1)^{-1} > g_1$. When we take (5.4) into consideration, we see that this inequality is identical to the left-hand inequality in (3.9). Under these conditions, in the place of (5.4) we obtain

$$\sigma_1 = \sigma_0 + \frac{D_1 g_1}{mu}\exp\left(-\frac{u\xi}{D_1}\right). \tag{5.5}$$

The significant difference from (5.4) of the field in (5.5), dependent on the temperature gradient in the melt, is obvious. The condition of narrowness for the equilibrium zone [the right-hand inequality in (3.9)] can be represented as follows, taking into consideration (5.5):

$$m(1-k)u\sigma_0 \ll 2kD_1g_1. \tag{5.6}$$

As the velocity u is increased as a consequence of the intensification of the removal of heat from the alloy, provided that all of the remaining parameters have been fixed, a frontal regime corresponding to the field in (5.4) is initially realized. When the velocity surpasses the critical value of $u^* = kD_1g_1/(1 - k)m\sigma_0$ this regime is replaced by a regime with a two-phase zone, and the field from (5.5) is established in this case. The formation of such a zone is facilitated by a reduction in the coefficient of impurity distribution and by the flow of heat to the melt, and with an increase in the concentration of the melt and in the slope of the liquidus line. The strong inequality (5.6) under these critical conditions reduces to the requirement that $1 \ll 2$ (not satisfied, in the strict sense). Therefore, the concept of the narrow two-phase zone introduced earlier apparently has only a methodological significance, while relationships (5.3) should be regarded as tentative estimates. If we also take into consideration the conclusions of Sec. 4, we find that if the two-phase zone is in a state of equilibrium, its dimensionless thickness $\varepsilon \sim 1$. Analysis of the structure of such a zone and the subsequent calculation of the discontinuity in θ_{12}, which plays a role in (5.2), therefore requires numerical solution of Eqs. (2.6), in which we can detect the important problem of the subsequent studies.

In conclusion, let us note that this analysis is directly applicable to melts or solutions of comparatively low concentrations, provided that the linearized relationship (1.3) is approximately valid. For concentrated systems (particularly for small k, i.e., with strong expulsion of the impurity from the separation surface) it is necessary to take into consideration the nonlinearity of the liquidus equation and, what is most important, the presence of singular points on the phase-equilibrium diagrams, corresponding, in particular, to the eutectic or peritectic. The latter may alter the nature of the hardening process, even from a qualitative standpoint, and they therefore deserve separate attention.

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ABSORPTION OF SOUND NEAR A SEMIINFINITE RIGID PLANE

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1. Research was conducted into the absorption of sound in a viscous heat-conducting compressible fluid (or gas) in the familiar work by Konstantinov [1] for the reflection of a plane sound wave from an infinite rigid plane. The absorption factor in this case, defined as the ratio of the absorbed energy to the incident energy, given small angles of incidence ($\alpha \ll 1$), is equal to

$$d = 4M/(1 + 2M + 2M^2), \tag{1.1}$$

where $M = k_0(\nu/2\omega)^{1/2}/\alpha$; $k_0 = \omega/c$ (ω is the angular frequency of the oscillations and c is the speed of sound); ν is the coefficient of the kinematic viscosity of the fluid (for the sake of brevity, here and below we will assume that the dissipation of the sonic energy is governed exclusively by the viscosity of the medium); moreover, it is assumed that $k_0(\nu/\omega)^{1/2} \ll 1$. Of particular interest is the behavior of the coefficient d in the angles-of-incidence region $\alpha \lesssim k_0(\nu/\omega)^{1/2}$: it changes sharply with respect to the angle and when $\alpha = k_0(\nu/\omega)^{1/2}$ attains a maximum equal to $2(\sqrt{2} - 1)$, and is it not dependent on the properties of the fluid and on the frequency of oscillation (the Konstantinov effect [2]).

Let us note that for a real case of a finite plate in precisely this area of angles of incidence formula (1.1) accurately reflects the process which takes place at such a great distance from the edge of the plate, where the incident wave itself is virtually attenuated owing to absorption in free space. Indeed, we know from the theory of diffraction that a reflected wave near the surface of a plate may be regarded as plane (as was assumed in [1]) at a distance x from the edge of the plate such that the condition $\alpha^2 k_0 x \gg 1$ is satisfied. The coefficient of sound absorption in free space, i.e., $\gamma = 2k_0^3\nu/\omega$ [3], so that if $\alpha \sim k_0(\nu/\omega)^{1/2}$, the indicated condition assumes the form of $\gamma x \gg 1$, which indicates the strong attenuation of the incident wave at a distance x along the plate.

In order to investigate the sound absorption near a finite plate in the case of any small angle of incidence and at such distances from the edge of the plate that the incident wave has not yet been attenuated, it is necessary to drop the assumption that the "reflected" field is a plane wave. This study has been undertaken in the present paper for the case of a semiinfinite plane. It has been demonstrated that the effect of viscosity and the condition of adhesion at the plane leads to a unique "waveguide" effect which consists of the

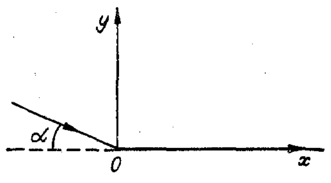

Fig. 1

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