MATHEMATICAL MODELING OF TWO-PHASE ZONE ORIGINATION IN DIRECTIONAL CRYSTALLIZATION PROCESSES

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Abstract: *Directional crystallization of binary melts is one of the ways of obtaining solid materials, the quality of which is completely determined by the physical and regime parameters of the solidification process. It is well known that, under certain conditions, the solid-liquid interface becomes morphologically unstable and the constitutional supercooling occurs ahead of the planar solidification front, which leads to the appearance of a metastable region. In this supercooled region, crystals can grow in the form of dendrites, grow on impurity inclusions, etc. As this zone is transitional between the already formed crystal and the melt, it is called a two-phase zone. In this paper, we present the results of numerical experiments carried out in accordance with the model taking into account the formation of a two-phase zone. In particular, qualitative and quantitative patterns of the time dependence of the formation of a two-phase zone in the Fe-Ni melt are obtained under two different regimes of active and passive cooling. Based on the results of calculations, it can also be concluded that, in both cases, there are such values of the cooling parameters, under which a two-phase zone is formed very close to the right boundary of the sample, i.e. practically the whole sample remains homogeneous. Choosing the appropriate cooling mode and its parameters makes it possible to optimize the directional crystallization process both in the quality of the obtained alloys and in the speed of their production.*

Keywords: SOLIDIFICATION, TWO-PHASE ZONE, CONSTITUTIONAL SUPERCOOLING

1. Introduction

Directional crystallization of binary melts is one of the ways of obtaining solid materials, the quality of which is completely determined by the physical and regime parameters of the solidification process. It is well known that, under certain conditions, the solid-liquid interface becomes morphologically unstable [1-3] and the constitutional supercooling occurs ahead of the planar solidification front [4], which leads to the appearance of a metastable region [5-9]. In this supercooled region, crystals can grow in the form of dendrites, grow on impurity inclusions, etc. As this zone is transitional between the already formed crystal and the melt, it is called a two-phase zone.

In the case of the formation of a two-phase zone, the crystallization pattern changes radically. In particular, the classical Stefan thermodiffusion model becomes inapplicable. To describe the process of crystallization, one can use one of the mathematical models [10-17]. However, it should be noted that in the modern scientific literature there are many other models of the solidification process with a two-phase zone, taking into account the different features of the latter. One of the goals of this study was to determine the moment of origination of a two-phase zone and the rate of solidification at various thermophysical parameters characterizing the crystallization process. The determination of the formation time of the two-phase zone also defines the time frame for the applicability of the Stefan model with a planar solid-liquid interface.

In addition to a purely theoretical study, the study of the solidification processes of binary melts has a great practical importance. This is due to the fact that two-component alloys are widely used in industry. The inclusion of a small amount of impurity in some metals can significantly change the mechanical properties of materials. Examples include the well-known Ti-Al alloy, which combines the strength of titanium with the cheapness of aluminum, or the iron-nickel alloys (Fe-Ni).

The problem is that the formation of a two-phase zone, frequently occurring during the crystallization processes of binary melts, in most cases adversely affects the quality of the resulting material. Investigations of the structure of alloy samples show that a sharp deviation of the impurity concentration from the mean value over the sample is observed in places where the metastable zone is supposed to be formed. This effect was called the layered liquation, because the impurity is usually concentrated by layers parallel to the surface of the crystallization front. The damage of layer segregation is obvious, because it leads to a strong heterogeneity of mechanical and strength properties of products obtained from alloys and, consequently, to a decrease in product quality and production efficiency. In this connection, we must know when and where the two-phase zone appears.

In this paper, we present the results of numerical experiments carried out in accordance with the model taking into account the formation of a two-phase zone. In particular, qualitative and quantitative patterns of the time dependence of the formation of a two-phase zone in the Fe-Ni melt are obtained under two different cooling regimes. Based on the results of calculations, it can also be concluded that, in both cases, there are such values of the cooling parameters, under which a two-phase zone is formed very close to the right boundary of the sample, i.e. practically the whole sample remains homogeneous. Choosing the appropriate cooling mode and its parameters makes it possible to optimize the metallurgical process both in the quality of the obtained alloys and in the speed of their production.

2. Governing Equations

Let us consider the process of directional crystallization in a mold of length L. The regions $0 < \xi < \Sigma(\tau)$ and $\Sigma(\tau) < \xi < L$ are filled with the solid and liquid (binary melt) phases, respectively (here $\Sigma(\tau)$ represents the position of the crystallization front at the time τ). The front moves from the cooled wall from the position $\xi = \Sigma(0)$ towards the opposite wall $\xi = L$. The process of solidification in the solid phase and the melt is described by the following equations of heat conduction and diffusion (diffusion in the solid phase is neglected)

$$\partial \theta_s / \partial \tau = a_s \partial^2 \theta_s / \partial \xi^2, \quad 0 < \xi < \Sigma(\tau), \tag{1}$$

$$\partial \theta_l / \partial \tau = a_l \partial^2 \theta_l / \partial \xi^2, \quad \partial \sigma / \partial \tau = D \partial^2 \sigma / \partial \xi^2, \quad \Sigma(\tau) < \xi < L, \tag{2}$$

where θ_s and θ_l are the temperatures of the crystal and the melt, a_s and a_l are the corresponding thermal diffusivity coefficients, σ and D are the impurity concentration and the diffusion coefficient in the melt.

Let's consider two cooling modes for the left mold wall.

1. The heat flux at the cold boundary increases linearly with time (forced or active cooling)

$$\lambda_s \partial \theta_s / \partial \xi - \lambda_l g_l = \alpha_a \tau, \quad \xi = 0. \tag{3}$$

2. Heat exchange with the environment through the left wall is carried out according to Newton's law (natural or passive cooling)

$$\lambda_s \partial \theta_s / \partial \xi = \alpha_p (\theta_s - \theta_E), \quad \xi = 0. \tag{4}$$

Here α_a is the cooling coefficient, α_p is the coefficient of heat exchange, θ_E is the ambient temperature, λ_s and λ_l are the coefficients of thermal conductivity in the solid phase and melt, and g_l is the temperature gradient in the melt.

At some time $\tau=\tau_*$ the constitutional supercooling ahead of the planar crystallization front occurs, i.e. the gradient of the impurity concentration exceeds the temperature gradient at the crystallization front as a result of displacement of the impurity by the moving front

$$-m\partial\sigma/\partial\xi>\partial\theta_l/\partial\xi,\ \xi=\Sigma(\tau),\qquad(5)$$

where m is the liquidus slope.

This leads to the formation of a two-phase zone at the front, where the nucleation processes and the growth of dendrite-like structures can occur [18-25]. The heat and mass transfer processes in the two-phase zone are described by equations different from the aforementioned model (1)-(4) with a planar front. Therefore, the models for active and passive solidification regimes, strictly speaking, are only valid for times $\tau<\tau_*$. The time of two-phase zone incipience should be determined from the condition

$$-m\partial\sigma/\partial\xi=\partial\theta_l/\partial\xi,\ \xi=\Sigma(\tau).\qquad(6)$$

The other boundary and initial conditions are given in ref. [26].

3. Origination of the Two-Phase Zone

The above-described model is a problem with unknown moving boundaries. Methods for solving such problems are described in [27]. To reduce this problem to the standard form (with fixed boundaries), the coordinate transformation was used separately for the crystal and melt regions. Thus, the original problem was reduced to a system of parabolic partial differential equations with variable coefficients defined on fixed domains. Similarly, the initial and boundary conditions were transformed. The system solution was based on an implicit four-point finite-difference scheme with accuracy $O(h^2+t)$ [27]. Here h and t are the steps of the spatial and temporal grids. The advantages of this scheme are the ease of implementation and the weak dependence of the convergence on the ratio for calculations over small time intervals. The numerical solution of the system of differential equations at the i-th step $t=t_i$ is reduced to solving a system of linear algebraic equations of order $N=1/h$. Note that the system matrix has a tridiagonal form and has a diagonal predominance, which makes it possible to solve the system of linear equations by the sweep method in time $O(N)$. In the calculations, the results of which are given in the article, a uniform spatial-temporal grid was used.

Fig. 1 shows that the rate of solidification is a linear function of time until the formation of a two-phase zone. The position of the crystallization front, respectively, varies according to a parabolic law. Large values of the coefficient α_a of active cooling correspond to a faster crystallization process, because the heat removal through the left mold wall is increased. This leads to an earlier origination of a two-phase zone. A similar effect on the two-phase zone origination time has an increase of the temperature gradient in the melt (in fact, the heat flux going to the sample through the right wall): with increasing g_l (for fixed α_a), the time of origination increases (see Fig. 2). This is explained by the fact that for large values of the gradient g_l, the growing crystal needs to displace more impurities in order to satisfy the condition of constitutional supercooling (6) and, therefore, to go for this longer distance and spend more time. In addition, Fig. 2 demonstrates that when the cooling rate of the sample decreases, the position of the resulting two-phase zone shifts more and more toward the right wall of the mold. The crystallization time is also increased. Based on this, it is possible to select process parameters that will correspond to obtaining a homogeneous sample of a given length. At the same time, for a given position of the two-phase zone, the crystallization time also depends on the temperature gradient in the melt, which provides additional possibilities for optimizing the technological process from the point of view of energy costs.

Figs. 3 and 4 show the results of calculations for the case of passive cooling. It is easy to see that the crystallization rate slowly decreases with increasing time, as the mold cools down at a constant ambient temperature. Fig. 4 illustrates the time τ_* of two-phase zone incipience and the position of the crystallization front at $\tau=\tau_*$ versus the heat transfer coefficient α_p. In general, the physical situation here is analogous to the regime of active cooling.
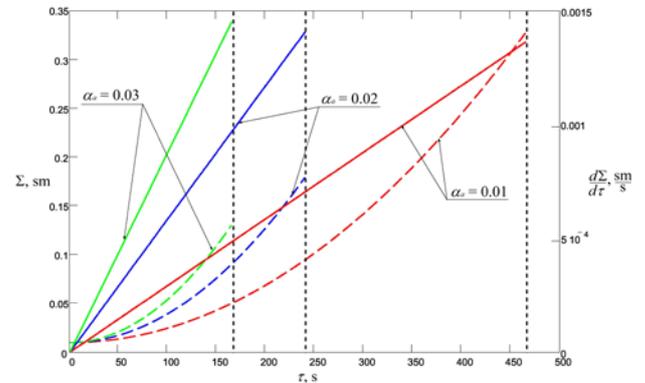

Fig. 1 The frontal coordinate Σ (dashed lines) and crystallization velocity $d\Sigma/d\tau$ (solid lines) as functions of time τ at different coefficients of active cooling α_a (measured in cal/($s^2\ cm^2$)).
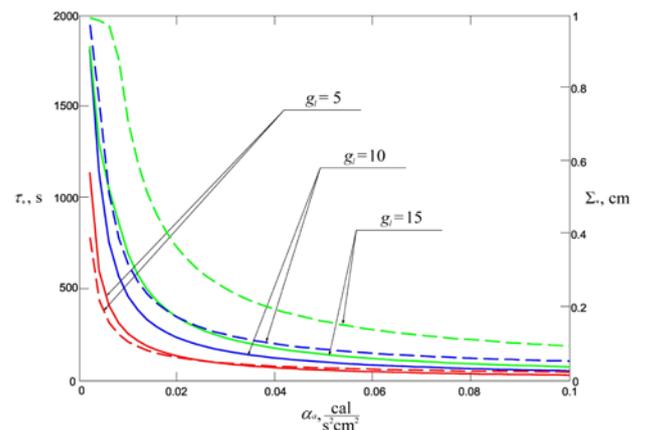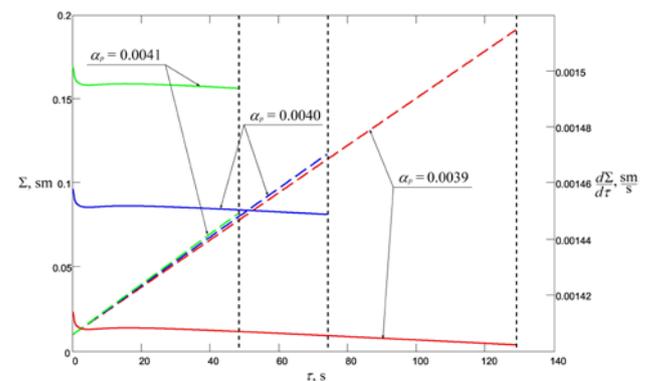

Fig. 2 The time τ_* and coordinate Σ_* of two-phase zone incipience as functions of the coefficient α_a of active cooling at different temperature gradients g_l (measured in $^OC/cm$).

Fig. 3 The frontal coordinate Σ (dashed lines) and crystallization velocity $d\Sigma/d\tau$ (solid lines) as functions of time τ at different coefficients of passive cooling α_p (measured in cal/($s\ cm^2\ ^OC$)).
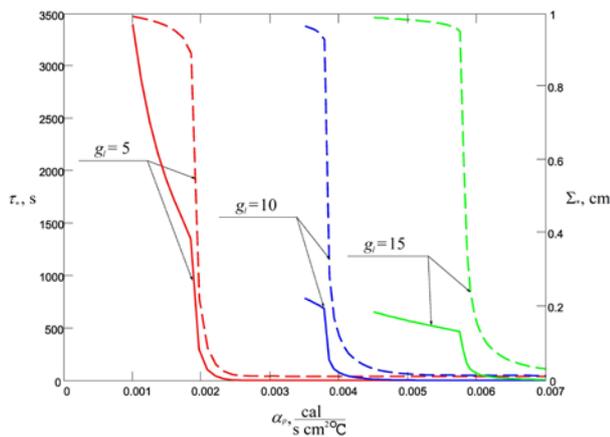
Fig. 4 *The time τ_* and coordinate Σ_* of two-phase zone incipience as functions of the coefficient α_p of passive cooling at different temperature gradients g_l (measured in $^OC/cm$).*

4. Conclusion

The calculations carried out in the present work show that when the constitutional supercooling condition is satisfied, a two-phase zone forms ahead of the planar solidification front. Physically, this means that the mathematical model describing the crystallization process is changing. In a two-phase zone, processes such as nucleation and crystal growth, evolution of dendrites, coalescence and coagulation, and the like can occur.

Acknowledgements

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