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Analytical solutions of two-phase region equations in the presence of an incoming flow

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Abstract. This paper describes theory behind the process of directional crystallization with a two-phase region, taking into account the presence of a two-phase region (a layer of mixed solid and liquid phases), as well as a weak outgoing or incoming flow. The analytical solution to high non-linear heat and mass transfer model is found analytically in a parametric form. The impurity concentration profile, temperature distribution, the solid phase portion, the solidification velocity, and the thickness of the two-phase region are determined analytically. The dependencies of impurity concentration versus the solid phase portion are plotted and compared with experimental data. In addition, the permeability of the two-phase region is plotted versus the spatial coordinate, and the interdendritic distance is plotted versus the crystallization rate and compared with the experimental data. In all cases, the theory discussed in this paper is in agreement with the experimental data.

Keywords: directional solidification, two-phase region, weak flow, heat and mass transfer, precise analytical solutions, nonlinear systems of equations

Introduction

A controlled and regulated crystallization process allows the creation of materials with a given structure, desired characteristics, and properties (Alexandrov et al. 2024; Batchelor 1974; Buyevich, Natalukha 1994; Ivanov et al. 2021; Kelton, Greer 2010; Makoveeva 2023; 2024; Makoveeva, Ivanov 2024; Slezov 2009; Vollmer, Raisch 2001). It is well known that the crystallization process can be directional or volumetric. Directional crystallization is caused by a temperature or concentration gradient that determines the direction of crystallization in space. Volumetric crystallization involves the nucleation and growth of crystals within a supercooled bulk of liquid. In the case of directional solidification, a curvilinear interphase boundary between solid and liquid phases moves in a certain spatial direction. Sometimes, such a boundary is not the solid-liquid interface of indefinitely small thickness, and it represents a zone of finite thickness filled with mixed solid and liquid phases. This zone is called the two-phase region.

Note that the heat and mass transfer processes in the two-phase region are influenced by various factors, and the portion of the solid and liquid phase plays a significant role. This portion affects the density, heat capacity, thermal conductivity, and diffusion coefficient of a given material (Buyevich et al. 2000; Worster 1986). Since heat and mass transfer are determined by the phases, among other things, transport equations should be written for each of the regions to describe the crystallization process.

As a result, transport equations are formulated for a purely solid phase, a two-phase region, and a purely liquid phase. To ensure the closure of the mathematical model, one should consider the boundary conditions at both boundaries of the two-phase region with a purely solid phase and a purely liquid phase. The initial and boundary conditions far from these boundaries should also be considered. Thus, we obtain a problem with heat and mass transport equations in each zone supplemented with boundary and initial conditions. This problem of heat and mass transfer is non-linear, with two moving phase transition boundaries (Aseev, Alexandrov 2006b; Buevich et al. 2000; Fowler 1985; Makoveeva et al. 2023). Such problems cannot be solved by standard methods, each problem requiring an original approach (see, for example, some previous studies (Alexandrov 2018; 2001a; Alexandrov, Alexandrova 2019; Mansurov 1990; Toropova et al. 2022)). A model of quasi-steady solidification with a two-phase region, without convection, was solved analytically by Alexandrov et al. (Alexandrov 2001b; Aseev, Alexandrov 2006a) using the transition to a new variable — the solid-phase portion. Subsequently, the nucleation and growth of particles in a two-phase region were taken into account, and this problem was solved analytically in Refs. (Alexandrov, Malygin 2006; Toropova et al. 2022). Worster was the first to consider a self-similar process, analyzing it numerically (Buevich et al. 2000). Subsequently, this process was analytically studied using a series expansion of the impurity concentration, temperature, and fraction of the solid phase in Refs. (Alexandrov et al. 2007). The LeadEx experiment on the freezing of sea ice is described using a model of non-steady-state directional solidification, which is caused by variations in the atmospheric temperature as a function of time. This problem was approximately solved in Refs. (Alexandrov, Nizovtseva 2008b; Alexandrov et al. 2008). Later, AIDJEX and SHEBA experiments on freezing sea ice from below with a false bottom formed were also described in Refs. (Alexandrov, Malygin 2011; Alexandrov, Nizovtseva 2008a; Worster 1991).

This paper examines the directional crystallization process with a two-phase region. Weak convection, i. e., a flow directed away from or toward the two-phase region, is also included in the model. The two-phase region solidifies at a constant rate. In addition, the present analysis is based on the methods of Refs. (Alexandrov 2001b; Aseev, Alexandrov 2006a).

Directed steady-state crystallization with a two-phase region

We consider a directed crystallization process along the spatial axis ζ . The crystallization rate v is constant. The process is determined by given temperature gradients in the solid and liquid phases, g_s and g_l , respectively (the process model is shown in Fig. 1). We introduce the following notations: τ is time, and δ is the thickness of the two-phase region. Shown at the top in Fig. 1 is the region with a purely solid phase, at $\zeta < v\tau$. Next, from top to bottom, is the two-phase region for $v\tau < \zeta < v\tau + \delta$. In the last region, there is a purely liquid phase, $\zeta > v\tau + \delta$.

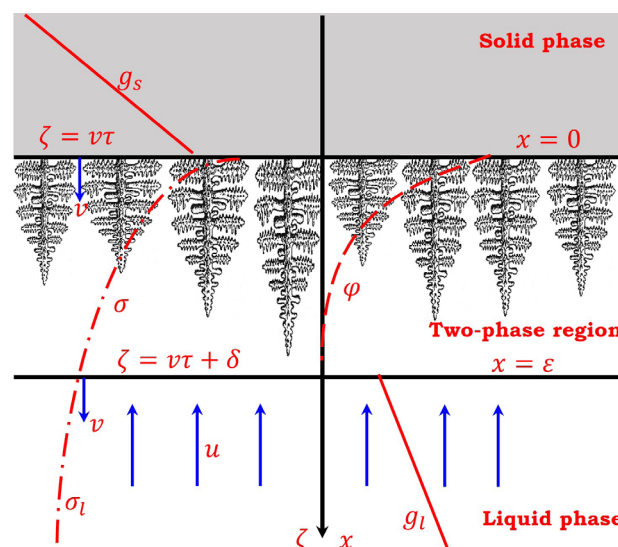


Fig. 1. A scheme of steady-state directed crystallization with constant velocity v along the ζ -axis. The solid phase — two-phase region and two-phase region — liquid phase boundaries are respectively located at $\zeta = v\tau$ ($x = 0$) and $\zeta = v\tau + \delta$ ($x = \varepsilon$). The solid, dash-dotted, and dashed lines schematically illustrate the temperature, impurity concentration, and solid phase portion distributions, respectively

The two-phase zone is a mixed zone consisting of both phases. The solid phase consists of dendritic structures, between which there is a liquid phase, represented by a molten melt. We also take into account that weak convection may occur in the liquid phase, the influence of which leads to a change in the distribution of impurities in the two-phase region and in the interdendritic space. This change in impurity distribution influences how the solid-phase impurity is absorbed by the solidifying liquid phase.

To present the process described above, heat and mass transfer equations will be required in the following form (Alexandrov et al. 2023; Kaviani, Kanury 2002):

$$\rho(\varphi)c(\varphi)\frac{\partial\vartheta}{\partial\tau} + \rho_l c_l(1-\varphi)\mathbf{u} \cdot \nabla\vartheta = \nabla(\lambda(\varphi)\nabla\vartheta) + L_v \frac{\partial\varphi}{\partial\tau}, v\tau < \zeta < v\tau + \delta, \quad (1)$$

$$\frac{\partial}{\partial\tau}((1-\varphi)\sigma) + (1-\varphi)\mathbf{u} \cdot \nabla\sigma = \nabla(D(\varphi)\nabla\sigma) - k\sigma \frac{\partial\varphi}{\partial\tau} - 2\alpha k\varphi \frac{\partial\sigma}{\partial\tau}, \quad (2)$$

$$v\tau < \zeta < v\tau + \delta,$$

where ϑ , σ , and φ are the temperature, impurity concentration, and portion of the solid phase in the two-phase region, $\mathbf{u}$ is the velocity of the liquid, L_v is the latent heat parameter, k is the distribution coefficient determining the portion of impurity absorbed by the solid phase, and α is the back diffusion in the solid state (for more details, see Appendix 12 in (Mullins, Sekerka 1964)). The functions φ can be written as a superposition of the transport coefficients in the solid and liquid phases (Buevich et al. 2000; Worster 1986). The functions of φ are: density ρ , heat capacity c , thermal conductivity λ , and diffusion coefficient D of the impurity

$$\begin{aligned} \rho(\varphi)c(\varphi) &= \rho_s c_s \varphi + \rho_l c_l(1-\varphi), \\ \lambda(\varphi) &= \lambda_s \varphi + \lambda_l(1-\varphi), \end{aligned} \quad (3)$$

$$D(\varphi) = D_l(1-\varphi),$$

where the indices s and l denote the properties of solid and liquid, respectively. The theory outlined in this paper takes into account that the heat and mass transfer coefficients are arbitrary functions of φ .

Note that the characteristic values of the relaxation times of the temperature (τ_ϑ) and concentration (τ_σ) fields are known. As a result, we can simplify models (1) and (2) by introducing a characteristic length scale: $\tau_\vartheta = l^2/\kappa_l$ and $\tau_\sigma = l^2/D_l$, where $\kappa_l = \lambda_l/(\rho_l c_l)$ is the thermal diffusivity. We know the characteristic values of τ_ϑ and τ_σ ; that is, their ratio is also known: $\tau_\vartheta/\tau_\sigma \sim 10^{-3} - 10^{-4}$, so the left-hand side of equation (1) is significantly smaller than the right-hand side.

The crystallization process is directed along the ζ axis, while the impurity concentration gradient is directed in the opposite direction due to the displacement of impurities by the growing solid phase. Since we consider flows predominantly in one direction of the ζ axis, we can neglect heat transfer in perpendicular directions. Thus, equations (1) and (2), taking all estimates into account, will look as follows:

$$\frac{\partial}{\partial\zeta}\left(\lambda(\varphi)\frac{\partial\vartheta}{\partial\zeta}\right) + L_v \frac{\partial\varphi}{\partial\tau} = 0, v\tau < \zeta < v\tau + \delta, \quad (4)$$

$$\begin{aligned} \frac{\partial}{\partial\tau}((1-\varphi)\sigma) + (1-\varphi)u \frac{\partial\sigma}{\partial\zeta} &= \frac{\partial}{\partial\zeta}\left(D(\varphi)\frac{\partial\sigma}{\partial\zeta}\right) - \\ &- k\sigma \frac{\partial\varphi}{\partial\tau} - 2\alpha k\varphi \frac{\partial\sigma}{\partial\tau}, v\tau < \zeta < v\tau + \delta, \end{aligned} \quad (5)$$

where the positive value of the fluid velocity ($u > 0$) is chosen against the ζ axis, as shown in Fig. 1.

Let us take into account the relationship between temperature ϑ and impurity concentration σ using the liquidus equation of the form:

$$\vartheta = \vartheta_0 - m\sigma, v\tau < \zeta < v\tau + \delta, \quad (6)$$

where ϑ_0 is the phase transition temperature for the pure melt ($\sigma = 0$), and m is the slope of the liquidus line.

Taking into account that $\tau_g/\tau_\sigma \ll 1$, we obtain the following equations of heat and mass transfer in the liquid and solid phases, respectively:

$$\frac{\partial^2 \vartheta_l}{\partial \zeta^2} = 0, \frac{\partial \sigma_l}{\partial \tau} + u \frac{\partial \sigma_l}{\partial \zeta} = D_l \frac{\partial^2 \sigma_l}{\partial \zeta^2}, \zeta > v\tau + \delta, \quad (7)$$

$$\frac{\partial^2 \vartheta_s}{\partial \zeta^2} = 0, \zeta < v\tau, \quad (8)$$

where ϑ_l is the temperature of the liquid phase, ϑ_s is the temperature of the solid phase, and σ_l is the concentration of impurities. Equations (7) and (8) mean that the first derivatives of the temperatures in the liquid and solid phases define the temperature gradients $\partial \vartheta_l / \partial \zeta = g_l$ and $\partial \vartheta_s / \partial \zeta = g_s$; i. e., the temperature gradients are fixed.

Let us supplement equations (4)–(8) with boundary conditions at both boundaries of the two-phase region:

$$\varphi = \varphi_*, \lambda_s g_s - \lambda(\varphi_*) \frac{\partial \vartheta}{\partial \zeta} = L_V(1 - \varphi_*)v, \zeta = v\tau, \quad (9)$$

$$(1 - k)(1 - \varphi_*)\sigma v + D(\varphi_*) \frac{\partial \sigma}{\partial \zeta} = 0, \zeta = v\tau, \quad (10)$$

$$\varphi = 0, \lambda(0) \frac{\partial \vartheta}{\partial \zeta} = \lambda_l \frac{\partial \vartheta_l}{\partial \zeta} = \lambda_l g_l, D(0) \frac{\partial \sigma}{\partial \zeta} = D_l \frac{\partial \sigma_l}{\partial \zeta}, \zeta = v\tau + \delta, \quad (11)$$

where φ_* is the portion of the solid phase at the boundary between the solid phase and the two-phase region.

Let us set the concentration of impurities in the liquid far from the boundary $\zeta = v\tau + \delta$, that is,

$$\sigma_l \rightarrow \sigma_{l\infty}, \zeta \rightarrow \infty. \quad (12)$$

As a result, we obtained a nonlinear model (4)–(12) describing the stationary directed crystallization process with a two-phase region. The resulting model will be solved analytically in the next section.

Analytical solutions

We will analyze the mathematical model from the viewpoint of a moving reference frame. We will introduce dimensionless variables and parameters for the transition to a moving coordinate system:

$$\begin{aligned} x &= \frac{v}{D_l}(\zeta - v\tau), C = \frac{\sigma}{\sigma_{l\infty}}, C_l = \frac{\sigma_l}{\sigma_{l\infty}}, \varepsilon = \frac{v\delta}{D_l}, \\ G_l &= \frac{D_l g_l}{vm\sigma_{l\infty}}, \Lambda_1 = \frac{\lambda_l}{\lambda_s}, \Lambda_0(\varphi) = \frac{\lambda(\varphi)}{\lambda_s}, \\ P &= \frac{\lambda_s m \sigma_{l\infty}}{L_V D_l}, D_0(\varphi) = \frac{D(\varphi)}{D_l}, M(\varphi) = \frac{D_0(\varphi)}{\Lambda_0(\varphi)}. \end{aligned} \quad (13)$$

Let us first consider equation (6) — the liquidus equation. Using it, we can eliminate the temperature field from the heat transfer equations. As a result, in the new moving reference frame, equations (4) and (5) take the following form:

$$\frac{d}{dx} \left(\Lambda_0(\varphi) \frac{dC}{dx} \right) + \frac{1}{P} \frac{d\varphi}{dx} = 0, 0 < x < \varepsilon, \quad (14)$$

$$\begin{aligned}
& -\frac{d}{dx}((1-\varphi)C) + ((1-\varphi)\omega - 2\alpha k\varphi)\frac{dC}{dx} = \\
& = \frac{d}{dx}\left(D_0(\varphi)\frac{dC}{dx}\right) + kC\frac{d\varphi}{dx}, 0 < x < \varepsilon,
\end{aligned} \tag{15}$$

where $\omega = u/v$.

We obtain the impurity concentration in the two-phase region by integrating equation (14) and also taking into account the boundary conditions (11). Then the concentration derivative becomes

$$\frac{dC}{dx} = -\frac{\varphi + PG_l\Lambda_1}{P\Lambda_0(\varphi)}, 0 < x < \varepsilon. \tag{16}$$

We next combine equations (6), (9), and (16) to find the steady-state crystallization rate v

$$v = \frac{\lambda_s g_s - \lambda_l g_l}{L_v}. \tag{17}$$

An important note is that the velocity (17) during directed crystallization with a two-phase zone coincides with the velocity of a flat crystallization front (Alexandrov 2001b; Alexandrov, Galenko 2024; Brody 1965).

Let us now derive a differential equation for $C(\varphi)$. To do this, we substitute the concentration (16) into equation (15) and multiply the resulting expression by $dx/d\varphi$. This gives us the following equation for $C(\varphi)$:

$$(1-k)C + [(1-\varphi)(\omega-1) - 2\alpha k\varphi]\frac{dC}{d\varphi} + \gamma(\varphi) = 0, \tag{18}$$

where

$$\gamma(\varphi) = \frac{1}{P}\left(M(\varphi) + (\varphi + PG_l\Lambda_1)\frac{dM}{d\varphi}\right).$$

Equation (18) demonstrates that the concentration of impurities and the temperature are functions that depend only on the solid phase portion.

We now obtain concentration distribution in the liquid. We integrate equation (7) for the liquid phase and also take into account the boundary conditions (11) and (12). Then the concentration in the liquid is as follows:

$$C_l(x) = 1 + \frac{G_l}{1-\omega} \exp[-(1-\omega)(x-\varepsilon)], x > \varepsilon. \tag{19}$$

Let us find the final expression for the concentration in the two-phase zone. We integrate equation (18) taking into account the boundary condition $C_0 = C_l(\varepsilon)$, obtaining the concentration distribution in the two-phase zone:

$$\begin{aligned}
C(\varphi) = & \left[\left(1 + \frac{G_l}{1-\omega}\right) (1-\omega)^{1-s} + \int_0^\varphi \frac{\gamma(\phi)d\phi}{[(1-\omega)(1-\phi) + 2\alpha k\phi]^s} \right] \cdot \\
& \cdot [(1-\omega)(1-\varphi) + 2\alpha k\varphi]^{s-1}, 0 < x < \varepsilon,
\end{aligned} \tag{20}$$

where

$$s = \frac{(1-2\alpha)k - \omega}{1-2\alpha k - \omega}.$$

We now consider the resulting equation (20). This equation shows that the concentration of impurities is only a function of the solid phase portion. Equation (20) includes previously found solutions and generalizes them so that we can obtain limit transitions to previously found theories. To move

to the Brody–Flemings distribution (Scheil 1942), we remove the diffusion and liquid flows and let them tend to zero ($D_l \rightarrow 0$ and $\omega \rightarrow 0$). We can also obtain the classical Scheil formula (Alexandrov, Malygin 2006; Galenko 2023) with $\alpha \rightarrow 0$. Galenko obtained the distribution of the impurity concentration with weak convection and reverse diffusion (Dehnavi et al. 2015). To move on to this solution, we let the diffusion in the liquid phase tend to zero, i. e., $D_l \rightarrow 0$.

Thus, equation (20) is a solution that unifies many previous theories. The resulting dependence takes into account impurity diffusion, weak convection in the liquid, and the movement of the solidifying region during directed crystallization. It is important to note that in equation (20), the concentration of impurities completely determines the temperature in the two-phase region, in accordance with equation (6).

By differentiating $C(\varphi)$ with respect to x , we obtain $dC/dx = (dC/d\varphi)/(d\varphi/dx)$. Thus, we arrive at the distribution of the solid phase portion in the two-phase region as an inverse function of $x(\varphi)$

$$x(\varphi) = \varepsilon - P \int_0^\varphi \frac{(dC/d\phi)\Lambda_0(\phi)d\phi}{\phi + PG_l\Lambda_1}, 0 < x < \varepsilon, \quad (21)$$

where $dC/d\varphi$ is determined by expression (20).

The dimensionless thickness of the two-phase region follows from expression (21) taking into account $\varphi = \varphi_*$ at $x = 0$ and has the form:

$$\varepsilon = P \int_0^{\varphi_*} \frac{(dC/d\phi)\Lambda_0(\phi)d\phi}{\phi + PG_l\Lambda_1} \quad (22)$$

Taking this into account, we determine from (10) the solid phase portion φ_* at the interface between the solid phase and the two-phase region:

$$M(\varphi_*)(\varphi_* + PG_l\Lambda_1) = P(1 - k)(1 - \varphi_*)C(\varphi_*), \quad (23)$$

where $C(\varphi_*)$ is defined by formula (20), and φ_* is the root of the transcendental equation (23).

Thus, expressions (17), (19)–(23) represent a complete analytical solution of the stationary model of directed crystallization with weak convection.

Fig. 2 shows the solid phase concentration $C_s = kC$, where C is defined by expression (20). The Brody–Flemings model (Scheil 1942) characterizes the absence of a liquid phase, $\omega = 0$, and is indicated by a transparent solid surface. The figure also shows two possible solutions: incoming and outgoing flows. For the incoming flow, the blue line, $\omega > 0$, indicates that the concentration of impurities exceeds the concentration in the absence of a weak flow. This occurs because copper also enters the two-phase region with the flow; it affects the growth of dendrites within the two-phase region.

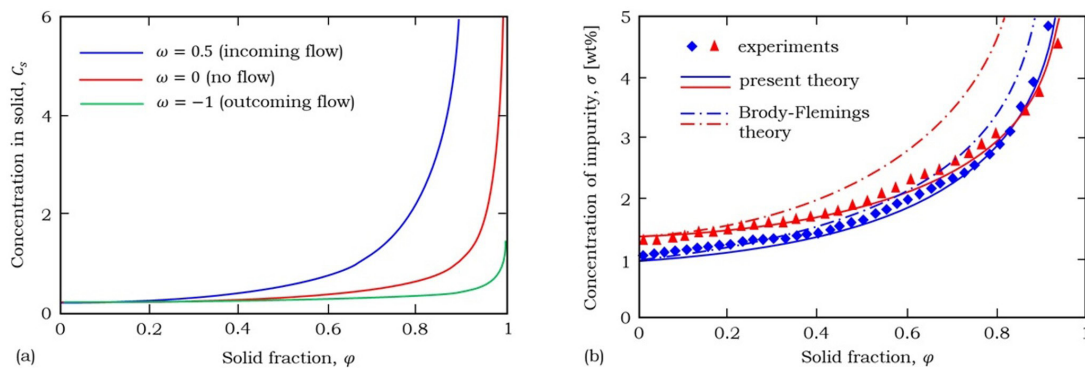


Fig. 2. (a) Dimensionless concentration in solid $C_s = kC$ for various convective flows: $\omega = 0.5$ (the incoming flow is shown in Fig. 1), $\omega = 0$ (stagnant melt), and $\omega = -1$ (outcoming flow); $\sigma_{\infty} = 5\text{wt\%}$. (b) Dimensional concentration of impurities for various convective flows: $\omega = -0.2$ ($\sigma_{\infty} = 1\text{wt\%}$, blue line) and $\omega = -0.55$ ($\sigma_{\infty} = 1.3\text{wt\%}$, red line) is compared with the experimental data (Dehnavi et al. 2015) (symbols). Dashed lines illustrate the Brody–Flemings theory (Brody 1965.) with $\omega = 0$. The physical parameters of the Al–Cu melt are presented in Table 1 with $g_s = 5 \cdot 10^2 \text{ }^\circ\text{C m}^{-1}$, $g_l = 10^2 \text{ }^\circ\text{C m}^{-1}$

Table 1. Physical parameters

Parameters	Al–Cu	Sn–Pb
λ_s [kg m s ⁻³ °C ⁻¹]	211.18	65
λ_l [kg m s ⁻³ °C ⁻¹]	90.92	33
D_l [m ² s ⁻¹]	$5 \cdot 10^{-9}$	$4.5 \cdot 10^{-9}$
L_v [kg s ⁻² m ⁻¹]	$1.08 \cdot 10^9$	$4.425 \cdot 10^8$
m [°C wt% ⁻¹]	2.5	2.2
ϑ_0 [°C]	660	505
ν [m ² s ⁻¹]	$5 \cdot 10^{-7}$	$8 \cdot 10^{-7}$
κ_l [m ² s ⁻¹]	10^{-6}	10^{-8}
d_0 [m]	10^{-9}	$5 \cdot 10^{-12}$
k	0.17	0.0656
α	0.03	0.03
n	4	4
A_4	1	1
σ_{04}	40	70

The opposite is also possible (outgoing flow). At $\omega < 0$, the concentration of impurities is lower compared to the red curve. The outgoing flow washes out copper, lowering the concentration inside the two-phase zone. The liquid flow can act in both directions, increasing or decreasing the concentration of impurities. The results obtained are consistent with the phase field calculations of dendritic crystallization performed in Refs. (Deguen et al. 2007).

We also compare expression (20) for the concentration of impurities with the experimental data. The lines differ as a result of various initial conditions and concentrations. The solid lines describe the current theory with a flow. The dashed lines describe the theory without a flow, the Brody–Flemings solution. As follows from the graph, both solid curves agree with the experimental data much better than the solutions without a flow. Although the flow was assumed to be weak, the differences are significant.

During solidification, the dendritic mushy zone can be considered as a permeable medium for a fluid flow. Permeability is defined as a quantitative parameter indicating how difficult it is for the fluid to pass through the mushy zone. Fig. 3a shows a plot of permeability K versus solid phase portion φ , calculated using the formula:

$$K(\varphi) = K_0 (1 - \varphi)^3, \quad (24)$$

where K_0 is the permeability at $\varphi = 0$. Next, we compare expression (24) for permeability with the experimental data; as we can see from the graph, the curve agrees well with the experimental data. Fig. 3b shows the dependence of permeability K on the coordinate x at different flow velocities; expressions (21), (22), and (24) were used to plot this dependence. The graph shows that for an incoming flow $\omega > 0$, the permeability of the two-phase region is lower than the permeability in the absence of a flow. This is due to the absorption of impurities brought about by the flow by growing dendrites, due to which the volume of the solid phase in the two-phase region increases and the permeability decreases.

The situation with the outgoing flow, at $\omega < 0$, is associated with a decrease in the concentration of impurities due to their washing by the outflow, which leads to a decrease in the volume of the solid material in the two-phase region and an increase in its permeability.

During the solidification process, the microstructural characteristics are influenced by various quantities: the portion of the solid phase φ , the impurity concentration at given temperature gradients g_s and g_l , and consequently the crystallization rate ν , which is determined by the temperature gradients, equation (17), as well as the flow rate u . As a microstructural parameter, we will consider the interdendritic distance λ_1 . Considering the axisymmetric shape of the dendrites, we have the expression for the interdendritic distance in the following form (Quaresma et al. 2000):

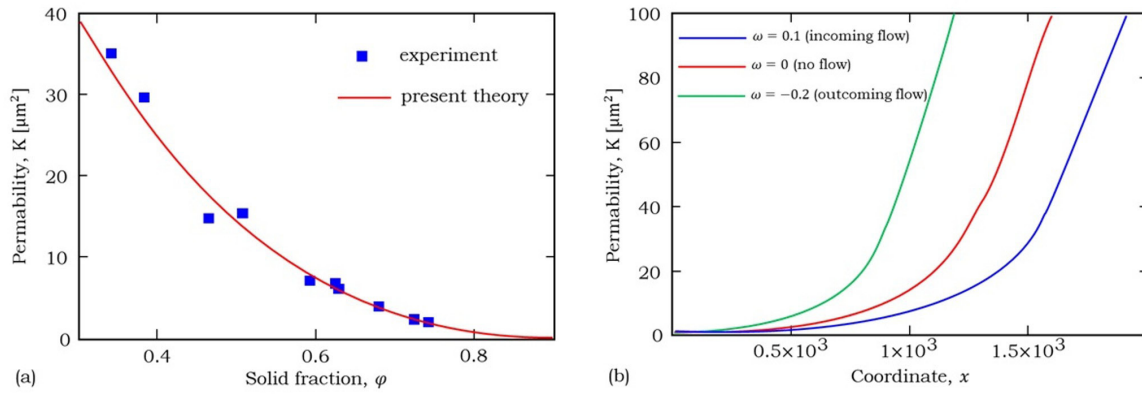


Fig. 3. (a) The permeability of the two-phase region (24) is compared with the experimental data (Murakami et al. 1984) (blue squares). (b) The permeability of the two-phase region for different convective flows: $\omega = 0.1$ (incoming flow, as shown in Fig. 1), $\omega = 0$ (stagnant melt), and $\omega = -0.2$ (outcoming flow); the physical parameters of the Al–Cu melt are presented in Table 1 with $\sigma_{\infty} = 5 \text{ wt\%}$, $g_s = 5 \cdot 10^2 \text{ }^\circ\text{C m}^{-1}$, $g_l = 10^2 \text{ }^\circ\text{C m}^{-1}$

$$\lambda_1 = \sqrt{\frac{2\pi p}{q(\partial\phi/\partial\zeta)}}, \zeta = v\tau, \quad (25)$$

where p is the diameter of the dendrite tip, $q = 1$ and $q = 0.86$ for cubic and hexagonal dendritic arrays, respectively.

Taking into account the moving frame of reference, we can rewrite equation (25) for the interdendritic distance, which describes the stationary crystallization process with a quasi-equilibrium two-phase layer, in the following form:

$$\lambda_1(v, u) = \sqrt{\frac{2\pi D_l p(v, u)}{qv} \frac{dx}{d\phi}}, x = \varepsilon, \phi = \phi_*, \quad (26)$$

at the boundary between the purely solid phase and the two-phase region, where $\phi = \phi_*$, $dx/d\phi$ is determined by expression (21) and takes the form

$$\frac{dx}{d\phi} = -\frac{P\Lambda_0(\phi_*)}{\phi_* + PG_l\Lambda_1} \frac{dC}{d\phi}, \phi = \phi_*,$$

where $dC/d\phi$ is from expression (20). In this case, the diameter of the dendrite tip p is determined by the theory of stable growth of dendrites

$$\frac{2d_0\kappa_l}{p^2v} = \sigma_*(p, v, u), \phi = \phi_*, \quad (27)$$

where d_0 is the capillary length and σ_* is a function of p , v , and u given in Appendix A. The selection criterion (27) for stable dendritic growth is a nonlinear equation to determine the dendrite tip diameter p as a function of the solidification rate v and the flow rate u . By substituting $p(v, u)$ into (26), we obtain the primary interdendritic distance as a function of velocity and flow rate $\lambda_1 = \lambda_1(v, u)$.

Fig. 4 shows the dependence of the interdendritic distance on the crystallization rate, according to equation (25). The graph represents three experiments with binary melts. The purple squares represent the aluminum-copper melt with a constant inflow of $\omega = -2$. The orange circles and pink diamonds represent the Sn-5wt%Pb and Sn-2.5wt%Pb melts, respectively. In the case of tin-lead melts, the convective currents in the liquid are minimized, so the flow velocity in the liquid phase is $\omega = 0$. All curves lie within the experimental data and are consistent, taking into account the experimental error.

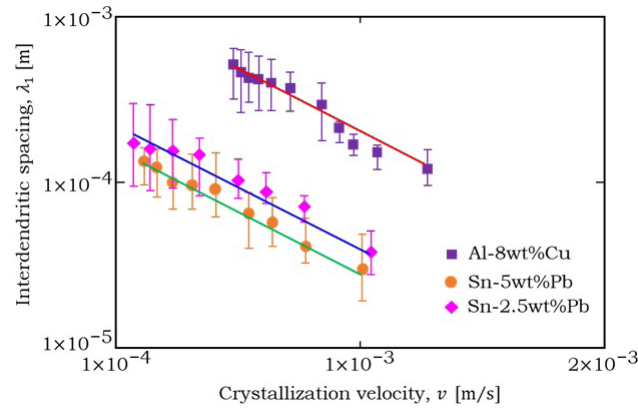


Fig. 4. Primary interdendritic distance λ_1 as a function of crystallization rate v . The theory (expressions (25)–(27), red line for Al–Cu ($\sigma_{iso} = 8$ wt%), blue line for Sn–Pb ($\sigma_{iso} = 2.5$ wt%), green line for Sn–Pb ($\sigma_{iso} = 5$ wt%)) is compared with experimental data for Al–Cu (Barros et al. 2015) and Sn–Pb (Rocha et al. 2003) melts. The physical parameters for the melts are given in Table 1 with $g_s = 140^\circ\text{C m}^{-1}$, $g_l = 45^\circ\text{C m}^{-1}$, $b = 0.5$, $\alpha_d = 0.8$ for Al-8wt%Cu; $g_s = 90^\circ\text{C m}^{-1}$, $g_l = 20^\circ\text{C m}^{-1}$, $b = 0.7$, $\alpha_d = 2.9$ for Sn-5wt%Pb; $g_s = 90^\circ\text{C m}^{-1}$, $g_l = 20^\circ\text{C m}^{-1}$, $b = 0.8$, $\alpha_d = 0.8$ for Sn-2.5wt%Pb

Conclusions

This paper presents a theory describing the crystallization process with a two-phase region. The model also takes into account weak inflow and outflow currents. To find solutions, relaxation times of temperature and concentration fields were considered, on the basis of which some estimates were made to simplify the model. The heat and mass transfer equations were then integrated, taking into account the fixed temperature gradients in the solid and liquid phases, to determine the solidification rate. This solidification rate of the two-phase layer coincides with the rate of directed solidification with a flat solid–liquid interface.

It is important to note that an extension of the Scheil–Brody–Flemings law was derived. This law allows one to determine the impurity concentration in a two-phase layer as a function of the solid phase portion for the steady-state solidification scenario. The law takes into account the effects of back diffusion and weak convection in the two-phase layer. This law has the form of an exact solution to the heat and mass transfer equations for the process described above. In this paper, the following parameters were also analytically determined: impurity concentration, temperature, and solid phase fraction in the two-phase region, as well as the impurity concentration in the liquid. The thickness of the two-phase region was determined and, by combining theory with the selection criterion, a relationship was found for the interdendritic distance.

To test the theory developed, some comparisons with experiments were conducted. Figs. 2 and 3 show the dependence of concentration on the solid phase fraction and the dependence of permeability on the coordinate, respectively. The graphs present data with different values of the incoming and outgoing fluxes. In each case, it is clear that even the weak flow influences the crystallization process, and the agreement is much better when the weak flow is taken into account. Fig. 4 shows the dependence of interdendritic spacing on the crystallization rate. The dependence is plotted for three binary melts; in all the three cases, there is good agreement with the experimental data.

The method developed in this paper can be applied to the solution of the problem of non-stationary crystallization with a two-phase region in the presence of weak convection in the liquid phase. Such a theory can be developed on the basis of the mathematical algorithm described in detail in Refs. (Alexandrov et al. 2008; Alexandrov, Nizovtseva 2008b; Galenko et al. 2024). Another direction of research is the theory of combined directional and volumetric crystallization, taking into account the slow flow, both outgoing and incoming. Such a problem can be considered in conjunction with theories of volumetric and directed crystallization (Alexandrov, Britousova 2015; Alexandrov, Galenko 2017; Makoveeva, Alexandrov 2024).

Appendices A: Selection criterion of stable dendritic growth

Considering crystal growth in the case of weak forced convection, one can derive the following criterion of stable growth for a dendrite tip, which connects the tip diameter p , tip velocity v , and velocity u of the fluid flow as follows (Galenko et al. 2024):

$$\sigma_*(p, v, u) = \frac{\sigma_{0n} \alpha_d^{7/n} A_n^{7/n}}{1 + b \tau_n^{v_n}} \cdot \left[\frac{1}{(1 + a_{1n} \alpha_d^{2/n} A_n^{2/n} P_g)^2} + \frac{2m\sigma_i(1-k)\lambda_l}{(1 + a_{2n} \alpha_d^{2/n} A_n^{2/n} P_c)^2 L_V D_l} \right], \quad (28)$$

where

$$\begin{aligned} A_n &= 2^{-3n/4} \sum_{k=0}^n \binom{n}{k} i^{n-k} \cos \frac{(n-k)\pi}{2}, \\ \tau_n &= \alpha_d^{-3/n} A_n^{-3/n} \frac{a u d_0}{4 p v P} \left(1 + \frac{2\kappa_l}{D_l} \right), \\ v_n &= \frac{n+7}{2(n+3)}, P = 1 + \frac{2m\sigma_i(1-k)\lambda_l}{L_V D_l}, \\ a &= \begin{cases} \sqrt{\frac{Re \exp(-Re/2)}{2\pi \operatorname{erfc}(\sqrt{Re/2})}}, & \text{2D case,} \\ \frac{\exp(-Re/2)}{E_1(Re/2)}, & \text{3D case,} \end{cases} \\ E_1(y) &= \int_y^\infty \frac{\exp(-x)}{x} dx, a_{2n} = \sqrt{2} a_{1n}, \\ a_{1n} &= \sqrt{\frac{8\sigma_{0n}}{7} \left(\frac{3}{56} \right)^{3/8}} A_n^{3/(2n)} \alpha_d^{(12-3n)/(8n)}, \end{aligned}$$

$P_g = pv/(2\kappa_l)$ and $P_c = pv/(2D_l)$ are the growth and chemical Péclet numbers, $Re = pu/v$ is the Reynolds number, ν is the kinematic viscosity, n is the order of crystalline symmetry ($n = 4$ for rectangles, $n = 6$ for ice crystals and snowflakes; see Ref. (Alexandrov, Galenko 2017.) for more details), σ_{0n} and b are the selection constants, α_d is the anisotropy constant, d_0 is the capillary constant, and $\sigma_i = \sigma_{i\infty} C(\varphi)$.

Substitution of σ_* from (28) into (27) leads to a single non-linear equation for $p(v, u)$. Note that this equation can be solved analytically in the case of small solidification velocities in the absence of convection (for details, see Refs. (Alexandrov, Britousova 2015; Makoveeva, Alexandrov 2024)).

Conflict of Interest

The author declares that there is no conflict of interest, either existing or potential.

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