

## Article

# Selection Criterion of Stable Dendritic Growth for a Ternary (Multicomponent) Melt with a Forced Convective Flow

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**Abstract:** A stable growth mode of a single dendritic crystal solidifying in an undercooled ternary (multicomponent) melt is studied with allowance for a forced convective flow. The steady-state temperature, solute concentrations and fluid velocity components are found for two- and three-dimensional problems. The stability criterion and the total undercooling balance are derived accounting for surface tension anisotropy at the solid-melt interface. The theory under consideration is compared with experimental data and phase-field modeling for Ni<sub>98</sub>Zr<sub>1</sub>Al<sub>1</sub> alloy.

**Keywords:** dendrites; stable growth; selection criterion; solvability theory; morphological stability; forced convective flow; undercooling balance; multicomponent melt



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## 1. Introduction

Solidification phenomena have a very important role in metallurgy and in some cases fully determine the physical and mechanical features of a solidified substance. Mathematical models of such phenomena stem from the classical Stefan model that describes crystallization with a flat boundary [1–3]. However, such a crystallization scenario is rarely realized in practical conditions. Usually, a flat boundary is destroyed due to the thermal or concentration undercooling [4–8]. Concentration undercooling, which appears at the crystallization interface (when the concentration gradient exceeds the temperature gradient), leads to favorable conditions for the growth of individual solid phase ridges, which create an extended area for dendritic growth ahead of the solid-liquid phase boundary known as a mushy layer [9–12]. In this layer, the evolution of dendrite-like structures, nucleation and growth of crystals are possible to observe [13–17].

The problem for the selection of a stable dendrite growth regime in the melt has arisen from the analysis of Ivantsov solutions [18–20] and experiments for a parabolic needle-shaped crystal [21–27]. These comparisons and tests have led to the conclusion that the continuous family of isotropic Ivantsov thermal and solute concentration distributions is unstable: the needle crystal loses its initial parabolic shape in the steady-state growth regime (see, among others, [28]). Therefore the Ivantsov solution is used as the main approximation for the analysis of stable growth, where the role of a small parameter is played by the surface tension anisotropy or the growth kinetics anisotropy [29,30]. After establishing the criterion for steady growth of a dendrite vertex in a single-component medium [28,29], the problem has been extended to the case of convective motion of the medium [31], to the case of dendrite growth in a binary system without convection [32] and to the case of dendrite growth in a binary system with convection [33]. Then, this theory was developed for rapid dendritic growth phenomena [34,35]. In many circumstances, however,

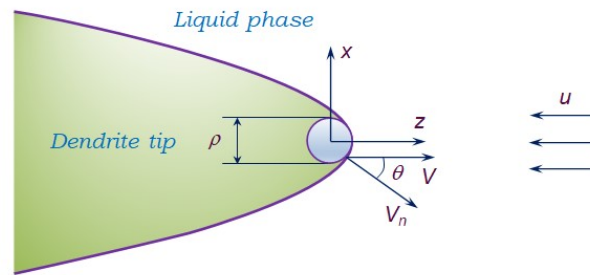
a comparative analysis of dendrite growth should be carried out taking into account the multi-component nature of the system [36,37]. In the present paper, which continues and develops the aforementioned studies, the effect of three-component (multicomponent) melt when considering the the stable mode of dendritic growth with a forced flow is investigated.

## 2. The Model of Dendritic Growth

The growth of dendritic tip moving deep into the undercooled ternary melt is characterized by a nonlinear moving-boundary problem with a curvilinear solid/liquid interface (a sketch of the process is demonstrated in Figure 1). As this takes place, the interfacial temperature  $\theta_{\text{int}}$  for a one-component melt is defined by the interface local curvature  $\mathcal{K}$ , the phase transition temperature of planar boundary  $\theta_*$ , the adiabatic temperature  $\theta_Q = Q/\kappa_p$  and the capillary length  $d$

$$\theta_{\text{int}} = \theta_* - \theta_Q d \mathcal{K}, \quad (1)$$

where  $Q$  represents the latent heat of solidification,  $\kappa_p$  is the specific heat, and  $d$  is a function of polar angle  $\theta$  shown (Figure 1) defined as [20,29]



**Figure 1.** A sketch of dendritic tip with incoming melt flow.

$$d(\theta) = d_0 \{1 - \chi_d \cos[n(\theta - \theta_d)]\}, \quad (2)$$

where  $d_0$  is the capillary constant. It should be especially emphasized that Equation (2) takes place for  $n$ -fold crystal symmetry. The constant parameter  $\chi_d$  is estimated to be significantly less than one. In addition, it should be mentioned that the solid/liquid interface curvature  $\mathcal{K}$  is  $2/\rho$  and  $4/\rho$  in the two- and three-dimensional dendrite growth geometries, respectively [38] ( $\rho$  represents the crystal tip diameter). An important point is that the misalignment angle  $\theta_d$  is negligibly small [20,29].

The temperature field in the solid ( $\theta_s$ ) and liquid ( $\theta_l$ ) phases with allowance for melt convection is described by equations

$$\frac{\partial \theta_s}{\partial \tau} = D_\theta \nabla^2 \theta_s \quad (\text{solid phase}), \quad (3)$$

$$\frac{\partial \theta_l}{\partial \tau} + (\mathbf{v} \cdot \nabla) \theta_l = D_\theta \nabla^2 \theta_l, \quad (\text{liquid phase}). \quad (4)$$

Here,  $\tau$  is the process time,  $D_\theta$  is the temperature diffusivity, and  $\mathbf{v}$  is the melt velocity. Let us especially underline that the temperature diffusivities in expressions (3) and (4) are chosen to be equal. This is traditionally used in the selection theory of a stable mode of dendritic growth [20,28–33]. If we would choose different temperature diffusivities in Equations (3) and (4), this would greatly complicate the mathematical transformation, but would not change the final selection criterion, because only the constant  $\sigma_{0n}$  in expression (26) would change.

The solute concentrations  $\sigma_1$  and  $\sigma_2$  of dissolved impurities in a ternary melt satisfy the diffusion equations [39–43]

$$\frac{\partial \sigma_1}{\partial \tau} + (\mathbf{v} \cdot \nabla) \sigma_1 = D_1 \nabla^2 \sigma_1, \quad \frac{\partial \sigma_2}{\partial \tau} + (\mathbf{v} \cdot \nabla) \sigma_2 = D_2 \nabla^2 \sigma_2 \quad (\text{liquid phase}), \quad (5)$$

where  $D_1$  and  $D_2$  are the diffusion coefficients of corresponding dissolved impurities. Let us especially highlight that the diffusion coefficients in Equation (5) should be different because both dissolved impurities in the melt diffuse at different rates.

At the moving dendrite interface with a constant velocity  $v$ , we have the equality of temperatures in solid and liquid phases to the phase transition temperature dependent of solute concentrations as well as the heat and mass balances [40–43], i.e.,

$$\theta_s = \theta_l = \theta_{\text{int}} - m_1 \sigma_1 - m_2 \sigma_2, \quad (\text{at the interface}), \quad (6)$$

$$\frac{Q}{D_\theta \kappa_p} \mathbf{V} \cdot \mathbf{n} = (\nabla \theta_s - \nabla \theta_l) \cdot \mathbf{n}, \quad (\text{at the interface}), \quad (7)$$

$$(1 - k_{1,2}) \sigma_{1,2} \mathbf{V} \cdot \mathbf{n} + D_{1,2} \nabla \sigma_{1,2} \cdot \mathbf{n} = 0, \quad (\text{at the interface}), \quad (8)$$

where  $m_1$  and  $m_2$  represent the liquidus slopes determined from the phase diagram,  $\mathbf{V} \cdot \mathbf{n}$  is the normal growth velocity,  $k_1$  and  $k_2$  are the partition coefficients corresponding to the solute concentrations  $\sigma_1$  and  $\sigma_2$ .

In addition, we assume that the temperature and solute concentrations at infinity are known, i.e.,

$$\theta_l \rightarrow \theta_\infty, \quad \sigma_1 \rightarrow \sigma_{1\infty}, \quad \sigma_2 \rightarrow \sigma_{2\infty} \quad (\text{far from the crystal}). \quad (9)$$

Accounting for the melt viscosity and simplifying the matter, we use the Oseen hydrodynamic model as follows [44–46]

$$u \frac{\partial \mathbf{v}}{\partial z} = -\frac{1}{\rho_m} \nabla p + \mu \Delta \mathbf{v}, \quad \nabla \cdot \mathbf{v} = 0, \quad (\text{liquid phase}), \quad (10)$$

where  $u$  is the velocity of undercooled melt far from the dendrite at  $z \rightarrow \infty$  (Figure 1),  $\rho_m$  is the density of undercooled ternary melt,  $p$  is the liquid-phase pressure, and  $\mu$  is the melt viscosity.

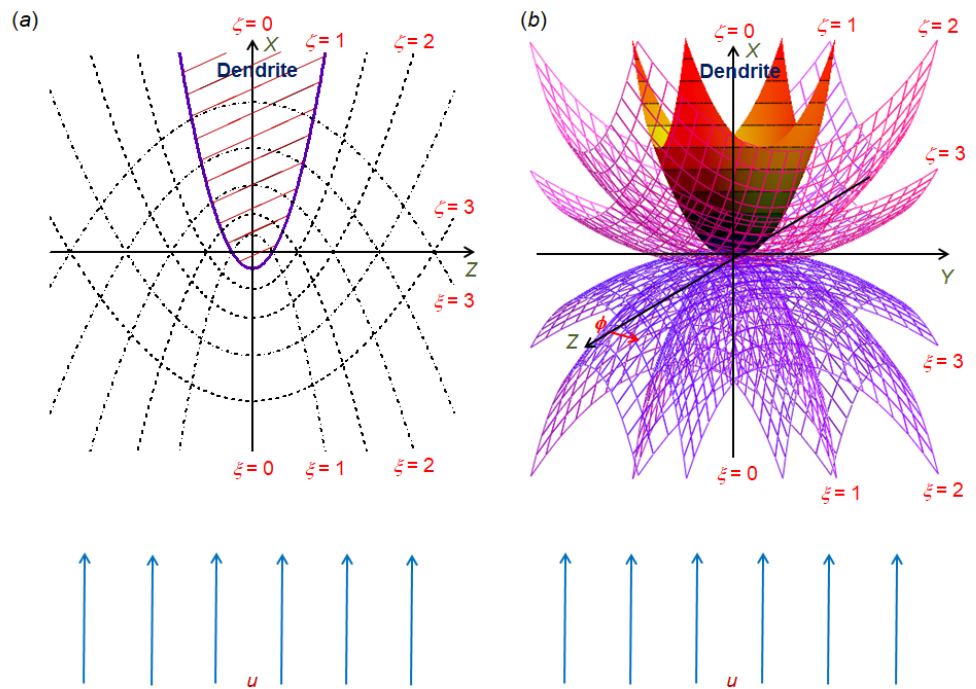
Equations (10) should be supplemented with the adhesion condition of liquid particles to the dendrite surface. Note that the hydrodynamic problem based on Equation (10) has been previously solved in Refs. [47,48]. Below, we use this solution, referring the interested reader to the original solution developed in Refs. [47,48].

### 3. Analytical Solutions for Steady-State Dendritic Growth

The aforementioned governing equations and boundary conditions can be solved in parabolic (paraboloidal) coordinates, which follow isothermal (isoconcentration) surfaces  $\zeta = \text{const.}$  around the dendritic vertex (Figure 2).

Thus, instead of Cartesian coordinates  $x$ ,  $y$  and  $z$ , we use the curvilinear coordinates  $\zeta$ ,  $\xi$  (and  $\phi$  in 3D).

$$\begin{aligned} z &= \frac{\rho(\zeta - \xi)}{2}, \quad x = \rho(\xi \zeta)^{1/2} \quad \text{in 2D}, \\ z &= \frac{\rho(\zeta - \xi)}{2}, \quad x = \rho(\xi \zeta)^{1/2} \cos \phi, \quad y = \rho(\xi \zeta)^{1/2} \sin \phi \quad \text{in 3D}. \end{aligned} \quad (11)$$



**Figure 2.** A scheme of a dendritic vertex swept by an undercooled melt (2D in (a) and 3D in (b)). The figure shows the dendrite vertex and the isothermal (isoconcentration) surfaces  $\zeta = \text{const.}$  around it.

We seek for a steady-state solution for the dendrite tip moving with a constant velocity  $v$  upstream of the melt flow (Figure 1). The process is established and temperature and solute concentrations are dependent only on  $\zeta$ , i.e.,  $\theta_l = \theta_l(\zeta)$ , and  $\sigma_{1,2} = \sigma_{1,2}(\zeta)$ . Assuming that the temperature in solid does not change, we arrive at the steady-state temperature and solute concentration profiles in the melt phase

$$\begin{aligned} \theta_l(\zeta) &= \theta_i + \frac{(\theta_\infty - \theta_i)I_\theta(\zeta)}{I_\theta(\infty)}, \\ \sigma_1(\zeta) &= \sigma_{1i} + \frac{(\sigma_{1\infty} - \sigma_{1i})I_{1\sigma}(\zeta)}{I_{1\sigma}(\infty)}, \quad \sigma_2(\zeta) = \sigma_{2i} + \frac{(\sigma_{2\infty} - \sigma_{2i})I_{2\sigma}(\zeta)}{I_{2\sigma}(\infty)}, \end{aligned} \tag{12}$$

where  $\theta_i$ ,  $\sigma_{1i}$  and  $\sigma_{2i}$  represent the interfacial temperature and solute concentrations (at  $\zeta = 1$ ) and have the form

$$\begin{aligned} \theta_i &= \theta_\infty + \theta_Q P_\theta \exp(P_0) I_\theta(\infty), \\ \sigma_{1i} &= \frac{\sigma_{1\infty}}{1 - (1 - k_1) \exp(P_0 D_\theta / D_1) P_\theta I_{1\sigma}(\infty) D_\theta / D_1}, \\ \sigma_{2i} &= \frac{\sigma_{2\infty}}{1 - (1 - k_2) \exp(P_0 D_\theta / D_2) P_\theta I_{2\sigma}(\infty) D_\theta / D_2}, \\ I_\theta(\zeta) &= \int_1^\zeta \exp \left[ (k - 1) P_u \int_1^{\zeta_1} \frac{G(\zeta_2) d\zeta_2}{\sqrt{\zeta_2}} - P_0 \zeta_1 \right] \frac{d\zeta_1}{\zeta_1^{(k-1)/2}}, \\ I_{1\sigma}(\zeta) &= \int_1^\zeta \exp \left[ (k - 1) P_u \frac{D_\theta}{D_1} \int_1^{\zeta_1} \frac{G(\zeta_2) d\zeta_2}{\sqrt{\zeta_2}} - P_0 \frac{D_\theta}{D_1} \zeta_1 \right] \frac{d\zeta_1}{\zeta_1^{(k-1)/2}}, \\ I_{2\sigma}(\zeta) &= \int_1^\zeta \exp \left[ (k - 1) P_u \frac{D_\theta}{D_2} \int_1^{\zeta_1} \frac{G(\zeta_2) d\zeta_2}{\sqrt{\zeta_2}} - P_0 \frac{D_\theta}{D_2} \zeta_1 \right] \frac{d\zeta_1}{\zeta_1^{(k-1)/2}}, \end{aligned}$$

where  $k = 2$  in the case of two-dimensional growth and  $k = 3$  in three dimensional space. Here,  $G(\zeta)$  follows from the Oseen hydrodynamic equations and has the form [47,48]

$$G(\zeta) = \frac{\operatorname{erfc}\sqrt{\zeta\Re/2}}{\operatorname{erfc}\sqrt{\Re/2}}\sqrt{\zeta} + \frac{\sqrt{2}}{\sqrt{\pi\Re}\operatorname{erfc}\sqrt{\Re/2}}\left[\exp\left(-\frac{\Re}{2}\right) - \exp\left(-\frac{\zeta\Re}{2}\right)\right] \text{ in 2D,}$$

$$G(\zeta) = \frac{\sqrt{\zeta}E_1(\zeta\Re/2)}{2E_1(\Re/2)} + \frac{\exp(-\Re/2) - \exp(-\zeta\Re/2)}{\sqrt{\zeta\Re}E_1(\Re/2)} \text{ in 3D,}$$

where  $\Re = \rho u/\mu$  represents the Reynolds number, and  $E_1(b) = \int_b^\infty \gamma^{-1} \exp(-\gamma)d\gamma$ . In addition,  $P_\theta$  and  $P_u$  represent the thermal and flow Péclet numbers

$$P_\theta = \frac{\rho V}{2D_\theta}, \quad P_u = \frac{\rho u}{2D_\theta} \text{ and } P_0 = P_\theta + P_u.$$

#### 4. The Total Melt Undercooling

The total melt undercooling  $\Delta\theta = \theta_* - \theta_\infty - m_1\sigma_{1\infty} - m_2\sigma_{2\infty}$  is the reason of dendritic growth and solidification phenomenon. This undercooling contains several contributions stemming from the temperature difference ( $\Delta\theta_\theta$ —thermal undercooling), displacement of impurities ( $\Delta\theta_\sigma$ —concentration undercooling) and curvilinear crystal shape ( $\Delta\theta_\rho$  shape undercooling)

$$\Delta\theta = \Delta\theta_\theta + \Delta\theta_\sigma + \Delta\theta_\rho. \tag{13}$$

These undercooling contributions are dependent on crystal tip diameter  $\rho$  and its velocity  $V$ . So, the first of them is given by

$$\Delta\theta_\theta(\rho, V) = \theta_i - \theta_\infty = \theta_Q \operatorname{Iv}_\theta^*(\rho, V). \tag{14}$$

The second one reads as

$$\begin{aligned} \Delta\theta_\sigma(\rho, V) &= m_1(\sigma_{1i} - \sigma_{1\infty}) + m_2(\sigma_{2i} - \sigma_{2\infty}) \\ &= \frac{m_1\sigma_{1\infty}(1 - k_1)\operatorname{Iv}_{\sigma_1}^*(\rho, V)}{1 - (1 - k_1)\operatorname{Iv}_{\sigma_1}^*(\rho, V)} + \frac{m_2\sigma_{2\infty}(1 - k_2)\operatorname{Iv}_{\sigma_2}^*(\rho, V)}{1 - (1 - k_2)\operatorname{Iv}_{\sigma_2}^*(\rho, V)}. \end{aligned} \tag{15}$$

Here, the thermal and concentration Ivantsov integrals are

$$\begin{aligned} \operatorname{Iv}_\theta^*(\rho, V) &= P_\theta(\rho, V) \exp[P_0(\rho, V)] I_\theta(\infty), \\ \operatorname{Iv}_{\sigma_1}^*(\rho, V) &= P_\theta(\rho, V) \frac{D_\theta}{D_1} \exp\left[\frac{P_0(\rho, V)D_\theta}{D_1}\right] I_{1\sigma}(\infty), \\ \operatorname{Iv}_{\sigma_2}^*(\rho, V) &= P_\theta(\rho, V) \frac{D_\theta}{D_2} \exp\left[\frac{P_0(\rho, V)D_\theta}{D_2}\right] I_{2\sigma}(\infty). \end{aligned}$$

The third contribution has the form

$$\Delta\theta_\rho(\rho) = \frac{2d_0\theta_Q}{\rho} \text{ (2D) and } \Delta\theta_\rho(\rho) = \frac{4d_0\theta_Q}{\rho} \text{ (3D)}. \tag{16}$$

The total undercooling (13) is a function of  $\rho$  and  $V$ . To obtain  $\rho(\Delta\theta)$  and  $V(\Delta\theta)$ , we need one more condition that connects these the variables. Such a condition (selection criterion) follows from the solvability theory and morphological stability analysis developed below.

#### 5. Selection Criterion for Stable Dendritic Growth

To derive the selection criterion we shall use the microscopic solvability condition. By substituting the marginal mode of the wavenumber  $\kappa_m$  into this condition, we obtain a

criterion for the selection of a stable growth mode of dendrite vertex. Let us write down this condition as [2,49]

$$\int_{-\infty}^{\infty} A[\Xi_0(\ell)]\wp_m(\ell)d\ell = 0, \quad \wp_m(\ell) = \exp\left[i\int_0^{\ell}\kappa_m(\ell_1)d\ell_1\right], \quad (17)$$

where  $A$  stands for the curvature operator,  $\Xi_0(\ell)$  represents the solution of moving-boundary problem, and  $i$  is the imaginary unit. It is significant to note that expression (17) represents an additional condition connecting three main growth parameters: dendrite tip diameter  $\rho$ , dendrite tip velocity  $V$  and melt undercooling  $\Delta\theta$ .

Our subsequent analysis builds on the theory developed by Bouissou and Pelcé [31]. According to this theory, we expand in Taylor series the components of fluid velocity in the neighbourhood of the dendrite surface  $\zeta = 1$

$$u_{\zeta} = -V(\zeta + 1)^{-1/2}, \quad u_{\xi} = \left(\frac{\xi}{\zeta + 1}\right)^{1/2} (V + b(\Re)u(\zeta - 1)), \quad (18)$$

where

$$b(\Re) = \left(\frac{\Re}{2\pi}\right)^{1/2} \frac{\exp(-\Re/2)}{\operatorname{erfc}(\sqrt{\Re}/2)} \text{ in 2D and } b(\Re) = \frac{\exp(-\Re/2)}{E_1(\Re/2)} \text{ in 3D.}$$

in addition, we introduce the orthogonal coordinate system  $(x_l, y_l)$  linked to the moving dendrite surface [31] ( $x_l$  and  $y_l$  are the tangent and normal axes, respectively). Taking this into account we represent expressions (18) in the form of

$$\tilde{v} = -V \cos \theta, \quad \tilde{u} = -V \sin \theta - \frac{b(\Re)u \sin(2\theta)}{2\rho} y_l, \quad (19)$$

where  $\tilde{u}$  and  $\tilde{v}$  stand for the tangent and normal velocities in the vicinity of growing crystal.

Expressions (7) and (8) lead to the temperature and solute concentrations derivatives at the crystal boundary  $y_l = 0$

$$\frac{d\tilde{\theta}_l}{dy_l} = -\frac{\theta_Q V \cos \theta}{D_{\theta}}, \quad \frac{d\tilde{\sigma}_1}{dy_l} = \frac{(k_1 - 1)V\sigma_{1i} \cos \theta}{D_1}, \quad \frac{d\tilde{\sigma}_2}{dy_l} = \frac{(k_2 - 1)V\sigma_{2i} \cos \theta}{D_2}, \quad y_l = 0. \quad (20)$$

Here, the symbol "tilde" designates the stationary solutions. Bearing this in mind we obtain the temperature and solute concentrations in the vicinity of dendritic boundary

$$\tilde{\theta}_l = \theta_i - \frac{\theta_Q V \cos \theta}{D_{\theta}} y_l, \quad \tilde{\sigma}_1 = \sigma_{1i} + \frac{(k_1 - 1)V\sigma_{1i} \cos \theta}{D_1} y_l, \quad \tilde{\sigma}_2 = \sigma_{2i} + \frac{(k_2 - 1)V\sigma_{2i} \cos \theta}{D_2} y_l. \quad (21)$$

Let the solid/liquid interface undergoes morphological perturbations  $\Sigma'$ . By this is meant that the temperature and concentration fields also undergo perturbations  $\theta'_s, \theta'_l, \sigma'_1$  and  $\sigma'_2$ . Perturbations also take place in the fluid velocity components  $u'$  and  $v'$  and pressure  $p'$ . All of these perturbations are considered to be sufficiently small as compared to the stationary solutions.

The heat and mass transfer Equations (3)–(5) lead us to the equations for perturbations, which read as

$$\begin{aligned} \frac{\partial \theta'_s}{\partial \tau} + \tilde{u} \frac{\partial \theta'_s}{\partial x_l} + \tilde{v} \frac{\partial \theta'_s}{\partial y_l} + v' \frac{d\tilde{\theta}_s}{dy_l} &= D_{\theta} \nabla^2 \theta'_s, & \frac{\partial \theta'_l}{\partial \tau} + \tilde{u} \frac{\partial \theta'_l}{\partial x_l} + \tilde{v} \frac{\partial \theta'_l}{\partial y_l} + v' \frac{d\tilde{\theta}_l}{dy_l} &= D_{\theta} \nabla^2 \theta'_l, \\ \frac{\partial \sigma'_1}{\partial \tau} + \tilde{u} \frac{\partial \sigma'_1}{\partial x_l} + \tilde{v} \frac{\partial \sigma'_1}{\partial y_l} + v' \frac{d\tilde{\sigma}_1}{dy_l} &= D_1 \nabla^2 \sigma'_1, & \frac{\partial \sigma'_2}{\partial \tau} + \tilde{u} \frac{\partial \sigma'_2}{\partial x_l} + \tilde{v} \frac{\partial \sigma'_2}{\partial y_l} + v' \frac{d\tilde{\sigma}_2}{dy_l} &= D_2 \nabla^2 \sigma'_2, \\ & & \nabla p' - \mu \rho_m \Delta \mathbf{v}' &= 0, \quad \nabla \cdot \mathbf{v}' = 0. \end{aligned} \quad (22)$$

Here, the symbol ' designates the perturbations.

Expanding the boundary conditions (6)–(8) in series in the vicinity of crystal boundary  $y_l = 0$ , we come to

$$\begin{aligned} \theta'_s &= \frac{\theta_Q V \cos \theta}{D_\theta} \Sigma' - \theta'_l, \\ \theta'_s + \frac{m_1 \sigma_{1i}(1 - k_1) V \cos \theta}{D_1} \Sigma' + \frac{m_2 \sigma_{2i}(1 - k_2) V \cos \theta}{D_2} \Sigma' &= \theta_Q d(\theta) \frac{\partial^2 \Sigma'}{\partial y_l^2} + m_1 \sigma'_1 + m_2 \sigma'_2, \\ v' = -\frac{\partial \Sigma'}{\partial \tau}, \quad u' = 0, \quad \frac{\theta_Q}{D_\theta} \frac{\partial \Sigma'}{\partial \tau} + \frac{\partial \theta'_l}{\partial y_l} + \frac{\theta_Q V^2 \cos^2 \theta}{D_\theta^2} \Sigma' &= \frac{\partial \theta'_s}{\partial y_l}. \end{aligned} \tag{23}$$

An important point is that the morphological perturbations represent an exponential function of the form

$$\exp(\omega \tau + i \kappa x_l - \varepsilon \kappa y_l) \tag{24}$$

when dealing with the linear stability analysis. Note that  $\omega$  and  $\kappa$  represent the frequency and wavenumber, whereas  $|\varepsilon| = 1$  describes the decay of perturbations at infinity, i.e., at  $y_l \rightarrow \infty$ .

Below, we assume that the morphological perturbations have the exponential form (24). Substituting them into conditions (23) we arrive at equations for their amplitudes. By equating the determinant of the system of equations for the perturbation amplitudes to zero, we construct a non-trivial solution. This leads us to the equation for the marginal wavenumber  $\kappa = \kappa_m$

$$\begin{aligned} \kappa_m^3 - \left( \frac{V}{2D_\theta d(\theta)} + \frac{m_1(1 - k_1)\sigma_{1i}V}{\theta_Q D_1 d(\theta)} + \frac{m_2(1 - k_2)\sigma_{2i}V}{\theta_Q D_2 d(\theta)} \right) \exp(i\theta)\kappa_m - \frac{ib(\Re)u \sin(2\theta)}{16\rho D_\theta} \kappa_m \\ - \frac{ib(\Re)u \sin(2\theta)}{8\rho D_1} \kappa_m - \frac{ib(\Re)u \sin(2\theta)}{8\rho D_2} \kappa_m + \frac{iV \sin \theta}{2D_\theta} \kappa_m^2 - \frac{V^2 \cos \theta \exp(i\theta)}{4D_\theta^2 d(\theta)} = 0. \end{aligned} \tag{25}$$

Here, we assume that the stationary dendrite evolves with the rate  $-i\kappa V \sin \theta$ . By this is meant that the perturbation grows with the rate  $\omega(\kappa)$ . Thus, the rate of morphological perturbation behaves as a sum of these functions, i.e.,  $\omega(\kappa) - iV\kappa \sin \theta$ . It is significant that we assume here the constant dendritic rate  $V$ . This rate can be attained quickly enough in real growth processes [50]. In addition, expression (25) has been derived for the neutral stability curve  $\omega = 0$  with allowance for  $\varepsilon = -1$  (also,  $i$  was replaced by  $-i$ , [31]). The marginal mode of the wavenumber (25) contains all limiting transitions to previous studies [2,31–33,49,51–56] devoted to simpler cases of dendritic growth.

Now, substituting the wavenumber from expression (25) into the solvability condition (17), we obtain

$$\begin{aligned} \sigma^* \equiv \frac{2d_0 D_\theta}{\rho^2 V} = \frac{\sigma_{0n} \chi_d^{7/n} B_n^{7/n}}{1 + \beta \bar{v}_n^{s_n}} \left\{ \frac{1}{\left[ 1 + b_{1n} \chi_d^{2/n} B_n^{2/n} P_\theta \right]^2} \right. \\ \left. + \frac{2(1 - k_1)m_1 \sigma_{1i} D_\theta}{\left[ 1 + b_{2n} \chi_d^{2/n} B_n^{2/n} P_{1\sigma} \right]^2 D_1 \theta_Q} + \frac{2(1 - k_2)m_2 \sigma_{2i} D_\theta}{\left[ 1 + b_{2n} \chi_d^{2/n} B_n^{2/n} P_{2\sigma} \right]^2 D_2 \theta_Q} \right\}, \end{aligned} \tag{26}$$

where

$$\bar{v}_n = \left( \frac{b(\Re)ud_0}{4V\rho R} + \frac{b(\Re)ud_0 D_\theta}{2V\rho R D_1} + \frac{b(\Re)ud_0 D_\theta}{2V\rho R D_2} \right) \chi_d^{-3/n} B_n^{-3/n}, \quad s_n = \frac{n + 7}{2(n + 3)},$$



$$R = 1 + \frac{2(1-k_1)m_1\sigma_{1i}D_\theta}{D_1\theta_Q} + \frac{2(1-k_2)m_2\sigma_{2i}D_\theta}{D_2\theta_Q}, \quad b_{2n} = b_{1n}\sqrt{2},$$

$$b_{1n} = B_n^{3/2n} \chi_d^{3/2n-3/8} \left(\frac{8\sigma_{0n}}{7}\right)^{1/2} \left(\frac{3}{56}\right)^{3/8}, \quad B_n = 2^{-3n/4} \sum_{j=0}^n \binom{n}{j} i^{n-j} \cos \frac{\pi(n-j)}{2},$$

where  $\sigma_{0n}$  is the selection constant characterizing the stable evolution of dendrites without forced convection,  $\beta$  is the selection constant if considering convection in a pure system,  $P_\theta = \rho V / (2D_\theta)$  is the thermal Péclet number,  $P_{1\sigma} = \rho V / (2D_1)$  and  $P_{2\sigma} = \rho V / (2D_2)$  are the concentration Péclet numbers.

Expression (26) represents the selection criterion in the case of ternary melt with convection. It defines an additional relation between  $\rho$ ,  $V$  and  $\Delta\theta$  so that the undercooling balance (14) and selection criterion (26) represent two nonlinear equations leading to  $\rho(\Delta\theta)$  and  $V(\Delta\theta)$ . It is significant that criterion (26) has limiting transitions to previous theories [2,31–33,49,51–56] devoted to simpler cases. As a special note, we note that the selection criterion (26) coincides with the previously developed criterion for a ternary system with four-fold crystalline symmetry without convection [57]. Moreover, a generalized selection criterion (26) for the  $N$ -component dilute melt is presented in Appendix A.

## 6. A Test of Theory against Experimental Data and Phase-Field Modeling for Ni<sub>98</sub>Zr<sub>1</sub>Al<sub>1</sub> Alloy

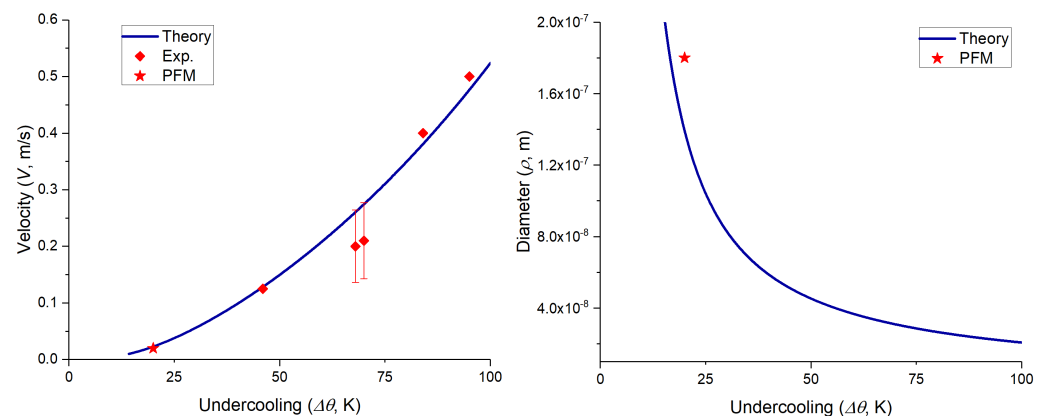
The experiments for detecting dendritic growth velocities of Ni<sub>98</sub>Zr<sub>1</sub>Al<sub>1</sub> alloy [58] were carried out using an electromagnetic levitation facility [56]. The sample temperature during solidification was measured with a two-color pyrometer, recorded using a transient recorder, and, in addition, the solidification process was observed by a high-speed camera. Further, growth velocity measurements were evaluated in solidified samples described by steady-state dendritic growth where the growth front is depicted by the intersection of the envelope of all dendrites with the sample surface. Figure 3 represents a good match of calculated theoretical results together with the obtained experimental data [58] of dendritic growth velocity as a function of melt undercooling (material and calculation parameters for the alloy Ni<sub>98</sub>Zr<sub>1</sub>Al<sub>1</sub> are given in Table 1). Even though the dendritic growth velocity is predicted for the whole range of undercoolings (see Figure 3), the theoretical curve overestimates the experimental data within the experimental margin of error in the region of diffusion-limited growth with  $\Delta\theta \approx 70$  K. One reason for such a disagreement could be the dependency of the solute partitioning and the liquidus slope on the temperature and composition in the multicomponent alloy system.

**Table 1.** Material and calculation parameters for the alloy Ni<sub>98</sub>Zr<sub>1</sub>Al<sub>1</sub>. Subscripts Zr and Al correspond to subscripts 1 and 2 in formulas (lines marked by \*).

Parameter	Symbol	Value	Units
Solute partition coefficient *	$k_{Zr}/k_{Al}$	0.013/1.133	-
Liquidus slope *	$m_{Zr}/m_{Al}$	-18.58/0.18	K/at.%
Solute diffusion coefficient *	$D_{CZr}/D_{CAI}$	$2.1 \times 10^{-9}/2.1 \times 10^{-9}$	m <sup>2</sup> s <sup>-1</sup>
Initial composition *	$\sigma_{Zr}/\sigma_{Al}$	1/1	at.%
Thermal diffusivity	$D_\theta$	$1.2 \times 10^{-5}$	m <sup>2</sup> s <sup>-1</sup>
Hypercooling	$\theta_Q$	656	K
Liquidus temperature	$\theta_*$	1728	K
Capillary constant	$d_0$	$3.28 \times 10^{-10}$	m
Specific heat	$\kappa_p$	41	J mol <sup>-1</sup> K <sup>-1</sup>
Surface energy stiffness	$\chi_d$	0.021	-
Solvability constant	$\sigma_{0n}$	0.3	-
Order of crystalline symmetry	$n$	4	-



In addition to the experimental data, phase-field is often used to test the accuracy of theoretical models. Nowadays, the application of the phase-field method to predict the formation and evolution of microstructures is undergoing a remarkable development. For instance, the phase-field modeling (PFM) can be used to study the multi-controlling factors of dendrite growth in directional solidification [59] and for describing the evolution of complex typical and atypical types of dendrite morphology observed during solidification of Al-Zn alloys [60]. Here, a general multicomponent multiphase field model was applied for the direct simulation of dendritic growth structures. This model and its implementation allow online coupling to thermodynamic databases and, consequently, make it possible to take into account the temperature and compositional dependence of the solute distribution at the interface (see for details [61]). Figure 3 compares the model predictions of the dendritic tip velocity and dendritic diameter as a function of undercooling and the star indicates the result of three-dimensional phase-field modeling [58]. As it can be easily seen a good agreement between our calculated results and PFM modeling was achieved for  $\text{Ni}_{98}\text{Zr}_1\text{Al}_1$  alloy.



**Figure 3.** Dendrite tip velocity  $V$  and diameter  $\rho$  as functions of the melt undercooling  $\Delta\theta$  for the alloy  $\text{Ni}_{98}\text{Zr}_1\text{Al}_1$ . The curves represent the theory under consideration. Experimental and PFM points are taken from Galenko et al. [58].

## 7. Conclusions

In summary, a new selection theory for a stable growth mode of dendritic crystals is developed with allowance for dissolved impurities and forced convection in an undercooled melt. The theory is developed for laminar viscous melt stream flowing around a dendrite, taking conductive heat and mass transfer on the crystal surface into account. Such crystal growth conditions correspond to the melt undercooling less than or of the order of  $10^2$  K. At a greater undercooling, the kinetic effects of atoms attachment to the dendrite solid-liquid interface should be taken into account. This, in particular, leads to the kinetic undercooling in the undercooling balance condition and essential corrections in the selection criterion (see, among others, Ref. [20], where these contributions have been considered for a two-component system). For even more undercooling greater than  $2 \times 10^2$  K, the effects of local-nonequilibrium crystallization play a decisive role (the diffusion equations become of a hyperbolic type, the partition coefficients and liquidus slopes become dependent on the growth velocity  $V$ , and the undercooling balance and selection criterion essentially change (see, among others, refs. [20,35])). Keeping this in mind, we expect to extend the present analysis for large undercoolings ( $\gtrsim 10^2$  K) in the nearest future.

As a special note let us underline that the theory under consideration describes the growth of dendritic tip, which preserves its shape and evolves with the constant velocity  $V$ . As this takes place, the whole dendritic interface can propagate in unsteady manner whose velocity satisfies the boundary integral equation [38].

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## Appendix A

Let us now generalize the selection criterion (26) for  $N$ -component dilute melt. Keeping in mind that contributions from additional impurities are additive within the framework of linear theory, we obtain

$$\sigma^* = \frac{2d_0 D_\theta}{\rho^2 V} = \frac{\sigma_{0n} \chi_d^{7/n} B_n^{7/n}}{1 + \beta \bar{v}_n^{5/n}} \left\{ \frac{1}{\left[1 + b_{1n} \chi_d^{2/n} B_n^{2/n} P_\theta\right]^2} + \sum_{j=1}^N \frac{2(1 - k_j) m_j \sigma_{ji} D_\theta}{\left[1 + b_{2n} \chi_d^{2/n} B_n^{2/n} P_{j\sigma}\right]^2 D_j \theta_Q} \right\}, \quad (\text{A1})$$

where

$$\bar{v}_n = \left( \frac{b(\Re) u d_0}{4V\rho R} + \sum_{j=1}^N \frac{b(\Re) u d_0 D_\theta}{2V\rho R D_j} \right) \chi_d^{-3/n} B_n^{-3/n},$$

$$R = 1 + \sum_{j=1}^N \frac{2(1 - k_j) m_j \sigma_{ji} D_\theta}{D_j \theta_Q}, \quad P_{j\sigma} = \frac{\rho V}{2D_j}$$

with  $D_j$  being the diffusion coefficient of  $j$ -component of impurity.

An important point is that the selection criterion (A1) coincides with the previously developed criterion for a multicomponent system with four-fold crystalline symmetry without convection [57].

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