

Towards the Stable Evolution of Dendrites in the Case of Intense Convection in the Melt

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Abstract: The solid-phase pattern in the form of a dendrite is one of the frequently met structures produced from undercooled liquids. In the last decades, an analytical approach describing the steady-state crystal growth in the presence of conductive heat and mass transport has been constructed. However, experimental works show that crystal patterns frequently grow in the presence of convection. In this paper, a theoretical description based on convective heat and solute concentration transport near the solid/liquid phase interface is developed. The stable regime of crystallization in the presence of vigorous convection near the steady-state crystal vertex is studied. The stability analysis, determining the stable growth mode, and the undercooling balance law have been applied to deduce the stable values for the growth rate and tip diameter. Our analytical predictions (with convective transport) well describe experimental data for a small melt undercooling. Moreover, we compare both convective and conductive mechanisms in the vicinity of the crystal vertex. Our theory shows that convective fluxes substantially change the steady-state growth of crystals.

Keywords: Phase transformations, Dendrites, undercooled materials.

1. Introduction

Dendritic melts and supersaturated solutions [1-8] (figure 1). Their surface anisotropy and mechanisms of heat and impurity transport near the phase transition boundaries are the main features influencing the crystallization process [9-11]. An important point is that the solid phase microstructure highly depends on the melt undercooling, crystallization rate, and the size of crystal vertices [12-14]. Also, note that vigorous convection can completely change the growth conditions through convective-type heat and mass transfer mechanisms. This in turn leads to mechanical deformations of solid-phase structures [15-17].

So, for instance, convection intensifies liquid motions increasing the rate of solidification, compressing the boundary layers for heat and mass transport, and increasing the temperature and solute concentration gradients in front of the solid/liquid interfaces [18-20]. These mechanisms are usually omitted in mathematical models of heat and mass transport phenomena. As an example of this, we can pay attention to the classical model of steady-state crystal growth. This model is written with conductive-type



boundary conditions, which completely ignore any type of convective motions in the melt (see, among others, [13, 21-28]). Such a classical model (within the framework of laminar flow in the liquid phase or in the absence of flow) can explain different experiments in a wide diapason of Péclet and Reynolds numbers. However, these models cannot explain a lot of experiments with vigorous convection or turbulent flows near growing solid/liquid crystal surfaces [29, 30]. It is significant that the classical Fourier heat and mass fluxes at the phase interfaces should be changed by the corresponding convective fluxes following Newtonian heat exchange law [29, 30].

Below we study the influence of these changed fluxes on the main characteristics of crystal growth. Namely, we compare the stability theory of steady-state crystal growth in the presence of both convective and conductive fluxes with experiments on the undercooled $\text{Ti}_{45}\text{Al}_{55}$ melt [19].

patterns represent frequently met types of solid microstructure originating from undercooled

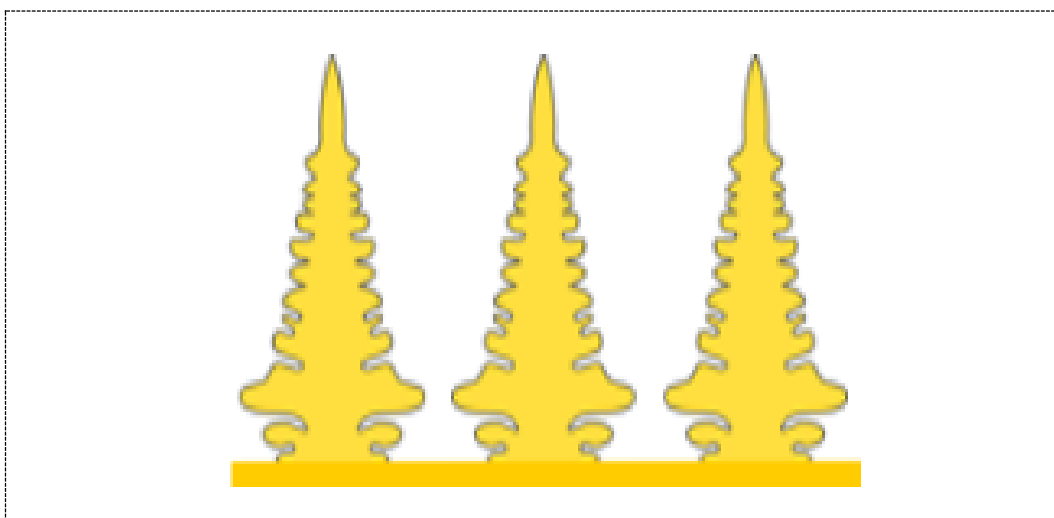


Figure 1. Dendrites growing in a metastable melt.

2. Stability Law with Allowance for Convective Fluxes

The present study is concerned with slow crystallization phenomena in the presence of vigorous hydrodynamic flow in the vicinity of the dendritic surface (figure 2). If this is really the case, transport processes for heat and dissolved impurity near the solid-liquid boundary are of convective type. This means that the boundary conditions at the solid-liquid interface should be written with allowance for this convective heat and mass transport [29-31]. The aim of this study is to derive a stability (selection) law for the steady motion of crystal tips. To do this, we assume that convective-type boundary conditions take place at the solid-liquid interface of a growing crystal [31]. Note that such nonlinear heat and mass transport happens as a result of fluid turbulization near the crystal interface [29, 30] or thermo-electrical mechanism [12, 18].

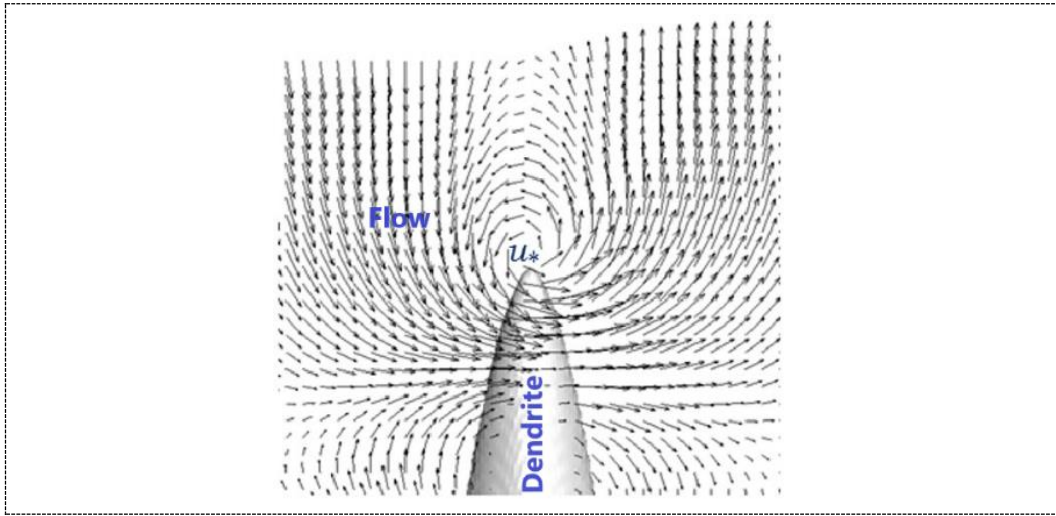


Figure 2. Isolated crystal evolving in a metastable melt with high-level convection, illustrated by the arrows.

The stability law characterizing a steady solidification scenario and (a relationship between the crystal growth rate v and its tip diameter R) can be written down as follows [32]:

$$s^* = \frac{2d_0D}{R^2v} = \frac{2s_0\sqrt{\alpha}Db_1}{R} + \frac{s_0\alpha^{5/4}(1+BDb_1)(1+lz^{3/2})^2}{\left[1+n_1\left(RB\alpha^{3/4} + \frac{3\alpha^{1/4}pb_1D}{2^{1/4}d_0}\right)\right]^2}. \quad (1)$$

Here

$$z = \frac{\alpha^{1/4}RB^2d_0}{2^{1/4}p(1+BDb_1)}, \quad b_1 = b_0 + \frac{m\sigma_i(1-k)}{\theta_Q a_s u_*},$$

$$B = \frac{a_h \rho_l C_l u_*}{2\lambda_s}, \quad n_1^2 = \frac{2^{9/2}25s_0}{27}, \quad \sigma_i = \frac{a_s u_* \sigma_{l\infty}}{a_s u_* - (1-k)v}, \quad p = \frac{Rv}{2D},$$

where s_0 is the fitting constant, d_0 is the capillary length parameter, ρ_l is the density of melt, k is the impurity segregation parameter, α is the surface energy stiffness, b_0 is the kinetic parameter, l is the stability constant, m is the equilibrium slope of liquidus equation, C_l is the heat capacity, u_* is the friction rate, θ_Q is the adiabatic temperature, and λ_s is the heat conductivity of solid material. Here a_h and a_s are the parameters defining intensity of convective fluxes, $a_h/a_s = (D/D_C)^r$ with $2/3 < r < 4/5$ (D and D_C represent the thermal diffusivity and diffusion coefficients).

3. Contributions to the Total Undercooling

An additional law to find v and R is defined by different contributions to the total melt undercooling $\Delta\theta$, which reads as [13]:

$$\Delta\theta = \Delta\theta_T + \Delta\theta_C + \Delta\theta_R + \Delta\theta_K. \quad (2)$$

The first summand in the r.h.s. of Eq. (2), thermal term $\Delta\theta_T$, can be presented as

$$\Delta\theta_T = \theta_i - \theta_{l\infty} = \frac{\theta_Q v \lambda_s}{a_h \rho_l C_l u_* D}, \quad (3)$$

where θ_i and $\theta_{l\infty}$ stand for the temperatures on the solid-liquid boundary and far from this boundary in the melt.

The next summand $\Delta\theta_C$ describing the impurity concentration effect is given by:

$$\Delta\theta_C = m(\sigma_i - \sigma_{l\infty}) = \frac{(1-k)vm\sigma_{l\infty}}{a_s u_* - (1-k)v}, \quad (4)$$

where σ_i and $\sigma_{l\infty}$ represent the impurity concentrations on the dendrite surface and far from this surface in liquid.

The contributions $\Delta\theta_R$ and $\Delta\theta_K$ defining the Gibbs-Thomson shift and atomic kinetics on the crystal surface are given by

$$\Delta\theta_R = \frac{4d_0\theta_Q}{R}, \quad \Delta\theta_K = \frac{v}{l_k}, \quad (5)$$

where l_k is the kinetic parameter.

Expressions (2)-(5) enable us to find the following explicit dependence

$$R(v) = \frac{4d_0\theta_Q}{\Delta\theta - \Delta\theta_T(v) - \Delta\theta_C(v) - v/l_k}. \quad (6)$$

Now substituting Eq. (6) into Eq. (1), we find the following equation for v

$$\frac{R^2(v)v}{2d_0D} s^*(R(v), v) = 1, \quad (7)$$

where s^* is determined by Eq. (1). We see that Eq. (7) connects the crystal growth rate v and the total melt undercooling $\Delta\theta$. As this takes place, R as a function of ΔT is defined by Eq. (6).

Let us now write out the stability growth law for conductive-type boundary conditions [7, 32, 33]

$$s^* = \frac{s_0 \alpha^{7/4}}{1 + \tilde{B}(A\alpha^{-3/4})^{11/14}} \left\{ \frac{1}{\left[1 + A_1 \sqrt{\alpha} p \left(1 + \frac{wD b_0}{d_0}\right)\right]^2} + \frac{2m\sigma_i(1-k)D}{\left[1 + A_2 \sqrt{\alpha} p_c \left(1 + \frac{wD_C b_0}{d_{0CD}}\right)\right]^2 \theta_Q D_C} \right\}. \quad (8)$$

Here the following notations are introduced

$$A_1 = \left(\frac{8s_0}{7}\right)^{1/2} \left(\frac{3}{56}\right)^{3/8}, \quad p_0 = p + p_f, \quad p_c = \frac{Rv}{2D_C}, \quad A_2 = \sqrt{2}A_1,$$

$$A = \frac{qd_0u}{4Rvp} + \frac{qd_0uD}{2RvQD_C}, \quad Q = 1 + \frac{2m\sigma_i(1-k)D}{D_C\theta_Q},$$

$$\sigma_i = \frac{\sigma_{l\infty}D_C}{D_C - (1-k)\exp(p_0D/D_C)pI_C(\infty)D},$$

$$d_{0CD} = \frac{\theta_Q d_0}{2m\sigma_i(1-k)}, \quad q(Rey) = \sqrt{\frac{Rey \exp(-Rey/2)}{2\pi \operatorname{erfc}\sqrt{Rey/2}}} \text{ in 2D,}$$

$$q(Rey) = \frac{\exp(-Rey/2)}{E_1(Rey/2)} \text{ in 3D,}$$

$$w = 20 \sqrt{\frac{7}{24} \left(\frac{56}{3}\right)^{3/8}}, \quad E_1(s) = \int_s^\infty \frac{\exp(-u)}{u} du.$$

Here u is the hydrodynamic flow far from the crystal, $p_f = Ru/(2D)$, \tilde{B} is the fitting constant, $Rey = Ru/\nu$ is the Reynolds number, ν is the kinematic viscosity, and the integral $I_C(\infty)$ is found in [32]. Note that formula (8) should be calculated together with the undercooling balance (Eq. (2)), which contains $\Delta\theta_T$, $\Delta\theta_C$, $\Delta\theta_R$, and $\Delta\theta_K$ determined in ref. [32] for 2D and 3D crystals.

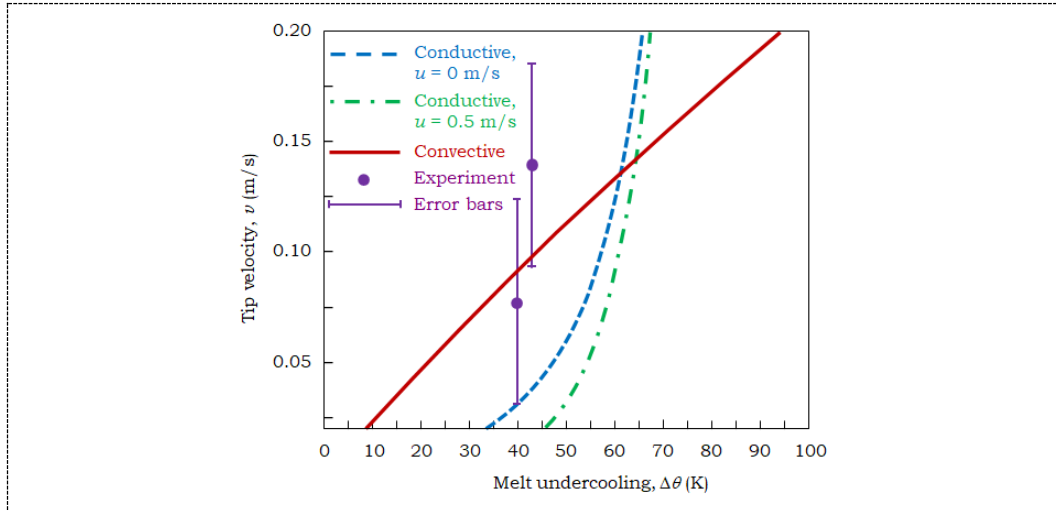


Figure 3. Crystal growth rate as a function of system undercooling for $Ti_{45}Al_{55}$. The dashed and dash-dotted curves show conductive transport without ($u = 0$ m/s) and with ($u = 0.5$ m/s) convection [34].

The solid line is illustrated accordingly to Eq. (7). Theoretical predictions are compared with experiments [19] for small $\Delta\theta$. The model parameters are $s_0 = 1.17$, $d_0 = 9.28 \cdot 10^{-10}$ m, $D = 2.5 \cdot 10^{-6}$ m² s⁻¹, $\rho_l = 2.46 \cdot 10^3$ kg m⁻³, $k = 0.86$, $\sigma_{l\infty} = 55$ at%, $\alpha = 0.3$, $b_0 = 1.88 \cdot 10^{-2}$ s m⁻¹, $l = 10^{-3}$, $m = 8.78$ K at%⁻¹, $C_l = 12370$ J kg⁻¹ K⁻¹, $a_h = 3.55$, $u_* = 4$ m s⁻¹, $\theta_Q = 272.64$ K, $\lambda_s = 29.22$ W m⁻¹ K⁻¹.

Figure 3 illustrates the difference between classical and conductive mechanisms of heat and impurity transport as well as experimental points [19] for low melt undercooling. Note that the theory well agrees with experiments only in the range of moderate and large melt undercooling (see, for details, [34]). If we pay our attention to small growth rates, the classical theory (conductive-type boundary conditions) does not describe experiments (dashed and dash-dotted lines in figure 3 lie outside of experimental error bars). This figure also shows that convective-type boundary conditions give $V(\Delta T)$ (solid line in figure

3) that satisfies experimental data for small ΔT . An important question is that we might expect a transition from the classical (laminar) growth scenario to the turbulent one in electromagnetic levitators [35] in the case of vigorous hydrodynamic fluxes near the crystals. Such a transition probably occurs due to the fact that vigorous hydrodynamic curls mean the appearance of convection near crystal vertices, where the flow is of turbulent-type [35].

Figure (4) shows how convection changes the crystal tip rate and diameter. Note that convection leads to greater values of v and R (compare the dotted and solid lines in Fig. 4(a)). Moreover, these values also grow with increasing the friction rate (Fig. 4(b)). In other words, the growing crystals are thinner in the absence of convection.

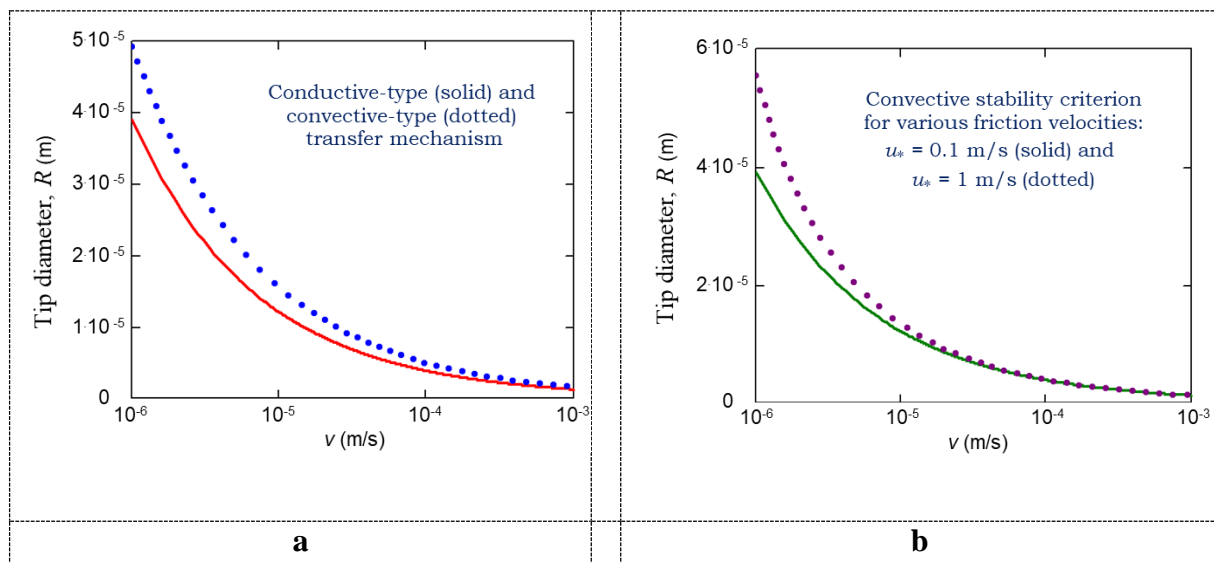


Figure 4. The diameter of crystal vertex as a function of the growth rate. Physical parameters are $a_h = 0.0095$, $\lambda_s = 2.03 \text{ W m}^{-1} \text{ C}^{-1}$, $\rho_l = 10^3 \text{ kg m}^{-3}$, $C_l = 4187 \text{ W s kg}^{-1}$, $d_0 = 2.8 \cdot 10^{-10} \text{ m}$, $\alpha = 0.35$, $s_0 = 0.17$, $D = 1.17 \cdot 10^{-7} \text{ m}^2 \text{ s}^{-1}$, $l = 0$, $b_1 = b_0 = 0$.

4. Conclusions

In this paper, we extend the analytical approach to study the steady regime of crystal growth in undercooled liquids in the presence of vigorous convection near the surfaces of evolving dendrites. Also, we compare our new analytical approach with the classical case of conductive boundary conditions and experiments for small undercooling. Note that convection plays a significant role in a small surface layer, where melt particles are disordered and migrate randomly. Consequently, the heat and solute concentration fluxes should be changed by the corresponding Newtonian laws. Here the main mechanism is that these fluxes are dependent on the friction rate u_* .

To compare convective theory with experiments and classical conductive fluxes, we use two nonlinear relationships for crystal growth rate and its diameter: stability criterion for the steady-state growth and balance law for the melt undercooling. This nonlinear system of algebraic equations is analytically solved to obtain an implicit dependence $v(\Delta\theta)$. Then this dependence is compared with experiments for small values of $\Delta\theta$. We show that the function $v(\Delta\theta)$ in the presence of convection well agrees with experiments ($\text{Ti}_{45}\text{Al}_{55}$) for small $\Delta\theta$.

An important question is that the real heat and mass fluxes near the surfaces of growing crystals can be of mixed type, i.e. they can contain convective and conductive parts. Such a theory of mixed boundary

conditions might be based of the present study and existing approaches [36-39]. Such a study represents a challenging problem for future works.

5. Acknowledgments

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