

## EFFECT OF A FORCED FLOW ON 3D DENDRITIC GROWTH IN BINARY SYSTEMS

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## ABSTRACT

The effects of a forced flow on dendritic growth rate in binary systems are studied theoretically. By using the Oseen approximation of the Navier-Stokes equation, the linear stability analysis and the solvability criterion, one determines a scaling factor  $\sigma^* = 2d_0 D_T / \rho^2 V$  as a function of the velocity of the forced flow and the impurity concentration in the melt in the three-dimensional model. The criterion obtained rallies analytic results for dendrite growth under forced convection in a pure system [1] and dendrite growth in a stagnant binary system [2].

## INTRODUCTION

It is well-known that the mechanisms of dendrite growth determine the microstructure evolution during solidification of melts and solutions [3-5]. These mechanisms are dependent, in particular, on the tip kinetics during free dendritic growth into undercooled liquid [6-8] as well as on effects of liquid convection [9] and diffusion of impurities partially rejected by the moving interface [2]. One of the theoretically and practically important problems consists in obtaining stable crystallization mode of the growing dendrite. The problem of finding a stability criterion for the growing dendrite comes from the linear and nonlinear stability theories [10-18]. After establishing robust stable conditions for the dendritic tip growing into a one-component stagnant liquid [19,20], these were extended to the one-component dendritic growth under forced flow [1,21,22] as well as to the binary dendritic growth in a stagnant media [2]. However, in many cases, knowledge of stable dendritic growth in binary system mode is a crucial question in evaluation and verification of the dendritic theory

predictions in comparison with experimental data [23]. Therefore, the present analytical investigation extending the boundaries of the two-dimensional analysis of ref. [24] is devoted to the problem of a selection criterion for the crystalline dendritic tip growing into a binary liquid under forced convective flow within the framework of the three-dimensional model.

## NOMENCLATURE

|                       |                       |   |
|-----------------------|-----------------------|---|
| $c_p$                 | [J/m <sup>3</sup> K]  | Heat capacity   |
| $C_i$                 | [at%]                 | Concentration at the solid-liquid interface                     |
| $C_l$                 | [at%]                 | Concentration of impurity                                       |
| $C_\infty$            | [at%]                 | Concentration in the liquid far from the solid-liquid interface |
| $d$                   | [m]                   | Capillary length  |
| $d_0$                 | [m]                   | Capillary constant  |
| $D_C$                 | [m <sup>2</sup> /s]   | Diffusive coefficient for concentration                         |
| $D_T$                 | [m <sup>2</sup> /s]   | Diffusive coefficient for temperature                           |
| $i$                   | [-]                   | Imaginary unit  |
| $k$                   | [1/m]                 | Wave-number of the perturbations                                |
| $k_0$                 | [-]                   | Equilibrium partition coefficient                               |
| $m$                   | [K/at%]               | Liquidus slope  |
| $\vec{n}$             | [-]                   | Unit normal at the interface pointing into the liquid           |
| $p$                   | [kg/ms <sup>2</sup> ] | Pressure  |
| $P_f = \rho U / 2D_T$ | [-]                   | Peclet number associated to the flow                            |
| $P_g = \rho V / 2D_T$ | [-]                   | Peclet number associated to the crystal growth                  |
| $Q$                   | [J/m <sup>3</sup> ]   | Latent heat per unit volume of solid                            |
| $1/R$                 | [1/m]                 | Local curvature   |
| $\Re = \rho U / \nu$  | [-]                   | Reynolds number   |
| $t$                   | [s]                   | Time  |
| $T_i$                 | [K]                   | Temperature at the solid-liquid interface                       |
| $T_l$                 | [K]                   | Temperature in the liquid phase                                 |
| $T_s$                 | [K]                   | Temperature in the solid phase                                  |

|                         |       |   |
|-------------------------|-------|---|
| $T_0$                   | [K]   | Crystallization temperature of the pure liquid                            |
| $T_\infty$              | [K]   | Temperature in the liquid far from the solid-liquid interface             |
| $u_\xi, u_\eta, u_\phi$ | [m/s] | Parabolic components of the fluid velocity                                |
| $U$                     | [m/s] | External flow velocity far ahead of the crystal                           |
| $V$                     | [m/s] | Growth velocity   |
| $\vec{v}$               | [m/s] | Solidification velocity directed along the normal vector to the interface |
| $\vec{w}$               | [m/s] | Fluid velocity  |
| $x, y$                  | [m]   | Spatial coordinates directed perpendicular to the crystal growth          |
| $z$                     | [m]   | Spatial coordinate directed along the crystal growth                      |

#### Special characters

|                   |                      |  |
|-------------------|----------------------|--|
| $\beta$           | [-]                  | Anisotropic factor                                 |
| $\xi, \eta, \phi$ | [-]                  | Parabolic coordinates                              |
| $\nu$             | [m <sup>2</sup> /s]  | Kinematic viscosity of the liquid                  |
| $\rho$            | [m]                  | Tip radius   |
| $\rho_l$          | [kg/m <sup>3</sup> ] | Density of the liquid phase                        |
| $\sigma$          | [J/m <sup>2</sup> ]  | Effective anisotropic liquid-solid surface tension |
| $\sigma^*$        | [-]                  | Scaling factor                                     |
| $\omega$          | [1/m]                | Frequency of the perturbations                     |

#### Subscripts

|     |  |
|-----|--|
| $i$ | Designates the physical parameters at the solid-liquid interface |
| $l$ | Designates the physical parameters in the liquid phase           |
| $s$ | Designates the physical parameters in the solid phase            |

## THE MODEL OF DENDRITE GROWTH

We consider a forced convection heat and momentum transfer problem when the crystal interface is assumed to be rough so that the Gibbs-Thomson relation determines the interface temperature as a function of the local curvature  $1/R$  of the solid-liquid interface:

$$T_i = T_0 - \frac{T_0 \sigma}{QR} \quad (1)$$

The temperature field satisfies the diffusion equation in the solid and liquid phases:

$$\frac{\partial T_s}{\partial t} = D_T \Delta T_s, \quad \frac{\partial T_l}{\partial t} + (\vec{w} \cdot \nabla) T_l = D_T \Delta T_l \quad (2)$$

and the concentration of impurity satisfies:

$$\frac{\partial C_l}{\partial t} + (\vec{w} \cdot \nabla) C_l = D_C \Delta C_l \quad (3)$$

in the liquid only. Here the advection of heat and mass by the fluid velocity field  $\vec{w}$  is taken into account.

At the interface, the temperature continuity holds. Also, the conservation of heat and mass must be satisfied, i.e.:

$$T_l = T_i - m C_l, \quad T_s = T_l, \quad Q(\vec{v} \cdot \vec{n}) = D_T c_p (\nabla T_s - \nabla T_l) \cdot \vec{n} \quad (4)$$

$$C_l(1 - k_0)(\vec{v} \cdot \vec{n}) + D_C(\vec{n} \cdot \nabla) C_l = 0 \quad (5)$$

The velocity field  $\vec{w}$  in the case of the small Reynolds number limit can be described by the so-called Oseen and mass conservation equations:

$$U \frac{\partial \vec{w}}{\partial z} = -\frac{\nabla p}{\rho_l} + \nu \Delta \vec{w}, \quad \nabla \cdot \vec{w} = 0 \quad (6)$$

The three-dimensional solid-liquid interface of the growing dendrite is assumed parabolic, of tip radius  $\rho$ , and moves at the velocity  $V$  along the  $z$ -direction. The external flow at infinity is parallel to the  $Oz$  axis and is directed to the crystal. Consequently, we consider a case of the so-called ‘‘up-stream branch forced flow’’. The Cartesian coordinates are connected with the parabolic coordinates as:

$$x = \rho \sqrt{\xi \eta} \cos \phi, \quad y = \rho \sqrt{\xi \eta} \sin \phi, \quad z = \rho(\eta - \xi)/2 \quad (7)$$

where the crystal surface  $\eta = 1$  has a tip radius  $\rho$ . Solution for the velocity field can be found in the Oseen approximation under consideration. Omitting tedious and combersome mathematical manipulations (for details, see ref. [25]), one can find the following expressions for the parabolic components of the fluid velocity:

$$u_\eta = -\frac{f(\eta)}{\sqrt{\xi + \eta}}, \quad u_\xi = \sqrt{\frac{\xi}{\xi + \eta}} \frac{d}{d\eta} (\sqrt{\eta} f(\eta)), \quad u_\phi = 0 \quad (8)$$

where:

$$f(\eta) = (U + V)\sqrt{\eta} - 2Ug(\eta), \quad (9)$$

$$g(\eta) = \frac{\exp(-\Re/2) - \exp(-\Re\eta/2)}{\Re\sqrt{\eta}E_1(\Re/2)} + \frac{\sqrt{\eta}E_1(\Re\eta/2)}{2E_1(\Re/2)} \quad (10)$$

Here  $E_1(\tau)$  is the exponential integral and is given by:

$$E_1(\tau) = \int_\tau^\infty \frac{\exp(-\psi)}{\psi} d\psi$$

Using Eqs. (7) and (8), one can integrate the Eqs. (2) and (3) of heat and mass transport in the liquid in their steady-state approximation. Seeking for a simple solution depending on  $\eta$  only and rewriting Eqs. (2) and (3) supplemented by the boundary conditions (4) and (5) in parabolic coordinates (see ref. [21]), we arrive at:

$$u_\eta \frac{\partial T_l}{\partial \eta} = \frac{2D_T}{\rho\sqrt{\eta(\xi + \eta)}} \frac{d}{d\eta} \left( \eta \frac{\partial T_l}{\partial \eta} \right) \quad (11)$$

$$u_\eta \frac{\partial C_l}{\partial \eta} = \frac{2D_C}{\rho\sqrt{\eta(\xi + \eta)}} \frac{d}{d\eta} \left( \eta \frac{\partial C_l}{\partial \eta} \right) \quad (12)$$

$$\left( \frac{\partial T_l}{\partial \eta} \right)_{\eta=1} = -\frac{Q}{c_p} P_g, \quad \left( \frac{\partial C_l}{\partial \eta} \right)_{\eta=1} = -(1 - k_0) C_l \frac{P_g D_T}{D_C}$$

From Eqs. (11) and (12), solutions for temperature and concentration fields are:

$$T_i(\eta) = T_i + (T_\infty - T_i) \frac{I(\eta)}{I(\infty)}, \quad C_i(\eta) = C_i + (C_\infty - C_i) \frac{I_1(\eta)}{I_1(\infty)} \quad (13)$$

with:

$$\begin{aligned} I(\eta) &= \int_1^\eta \exp \left[ 2P_f \int_1^{\sigma_1} \frac{g(\gamma_1) d\gamma_1}{\sqrt{\gamma_1}} - (P_f + P_g) \sigma_1 \right] \frac{d\sigma_1}{\sigma_1} \\ I_1(\eta) &= \int_1^\eta \exp \left[ 2P_f \frac{D_T}{D_C} \int_1^{\sigma_1} \frac{g(\gamma_1) d\gamma_1}{\sqrt{\gamma_1}} - (P_f + P_g) \frac{D_T}{D_C} \sigma_1 \right] \frac{d\sigma_1}{\sigma_1} \\ T_i &= T_\infty + \frac{QP_g}{c_p} \exp(P_g + P_f) I(\infty) \\ C_i &= \frac{C_\infty}{1 - (1 - k_0) \exp \left[ (P_g + P_f) \frac{D_T}{D_C} \right] P_g I_1(\infty) \frac{D_T}{D_C}} \quad (14) \end{aligned}$$

It is noteworthy that the Ivantsov parabolas are no longer solutions of the free-boundary problem when surface tension effects are taken into account. However, in the case of weak surface tension effects, steady solutions can be found close to an Ivantsov parabola if a solvability condition is satisfied [1,26]. We use here the solvability condition previously derived by Pelce and Bensimon [26] as the vanishing of an oscillating integral in the form:

$$\int_{-\infty}^{+\infty} G[X_0(l)] Y_m(l) dl = 0, \quad Y_m(l) = \exp \left[ i \int_0^l k_m(l_1) dl_1 \right] \quad (15)$$

The main interest of this formulation is that it is adaptable to other kinds of fronts, e.g. to the Saffman-Taylor finger [22]. One needs the curvature operator  $G$  and a continuum of solutions  $X_0(l)$  from which the function  $k_m(l)$  of the local non-zero marginal mode of the conjugate dispersion equation for the perturbations is deduced. The condition of application of the solvability criterion is that  $k_m(l)$  is large compared to the inverse of the scale of the unperturbed solution. It is obtained using Wentzel-Kramers-Brillouin approximation which has been applied to the flame propagation [27] and dendritic growth [28]. In other related problems, e.g., in description of the Saffman-Taylor fingers [29,30], it has been shown that one can get non-trivial solvability condition by considering elements  $Y_m(l)$  (they vary on a wavelength scale  $\lambda$ , which is small compared to the tip radius of the parabola).

## LINEAR STABILITY ANALYSIS

We use the linear stability theory provided by Bouissou and Pelce [1] in which the growth rate of a perturbation has a wavelength small compared to the characteristic spatial scale of the unperturbed solution. We consider that a perturbation disturbs the fluid on a distance of the order of  $\lambda$ . The latter enables us to expand the stationary velocity components (8) in a series in  $\eta - 1$  around the parabola  $\eta = 1$ . Taking into account only the main contributions, we arrive at:

$$u_\xi = \sqrt{\frac{\xi}{1 + \xi}} [V + a(\Re)U(\eta - 1)], \quad u_\eta = -\frac{V}{\sqrt{1 + \xi}} \quad (16)$$

where:

$$a(\Re) = \frac{\exp(-\Re/2)}{E_1(\Re/2)} \quad (17)$$

Hence, from Eqs. (16) and (17) it follows that only the tangent fluid velocity  $u_\xi$  is dependent of the forced flow close to the tip of the growing dendrite.

For the following analysis, we introduce new local Cartesian coordinates  $(x_c, y_c)$  fixed to the crystal, where  $x_c$  and  $y_c$  are, respectively the tangent and normal axes to the solid-liquid interface at a point where the normal to the interface has an angle  $\theta$  with the growing axis. These coordinates enable us to rewrite the velocity components (16) in the form of a shear flow whose magnitude is a function of  $\theta$  as:

$$\bar{u} = -V \sin \theta - \frac{aU}{\rho} \sin \theta \cos \theta y_c, \quad \bar{v} = -V \cos \theta \quad (18)$$

where  $\bar{u}$  and  $\bar{v}$  designate the tangent and normal velocity components to the interface. Let us express temperature and concentration derivatives from (4) and (5) as:

$$\frac{dT_i}{dy_c} = \frac{Q\bar{v}}{D_T c_p}, \quad \frac{dC_i}{dy_c} = \frac{C_i(1 - k_0)\bar{v}}{D_C} \text{ at } y_c = 0 \quad (19)$$

A similar expansion in series for the temperature and concentration fields in the liquid is obtained as follows:

$$\bar{T}_l = T_0 - \frac{QV}{D_T c_p} \cos \theta y_c, \quad \bar{C}_l = C_i - \frac{C_i(1 - k_0)V}{D_C} \cos \theta y_c \quad (20)$$

Let us now pay our attention to the linear stability theory of the aforementioned problem. Let  $u'$ ,  $v'$  and  $T'$  designate the perturbations of the stationary field,  $\xi'$  corresponds to the perturbation of the steady solid-liquid interface with a wavelength  $\lambda$  assumed very small compared to  $\rho$ . The solutions of the perturbed temperature conductivity Eq. (2) in the solid and hydrodynamic Eq. (6) within the framework of the Oseen approximation can be written in the form:

$$\begin{aligned} u' &= -i\varepsilon\omega\Sigma \exp(\omega t + ikx_c - \varepsilon ky_c), \quad v' = -\omega\Sigma \exp(\omega t + ikx_c - \varepsilon ky_c) \\ \xi' &= \Sigma \exp(\omega t + ikx_c - \varepsilon ky_c), \quad T'_s = T_{s0} \exp(\omega t + ikx_c - \varepsilon ky_c) \end{aligned} \quad (21)$$

where a relation  $v' = -\partial\xi'/\partial t$  at the solid-liquid interface between perturbations is taken into consideration. Here parameter  $\varepsilon$  has the same sign as real part of  $k$  since all perturbations cannot diverge as  $y_c$  goes to  $+\infty$ ,  $\Sigma$  and  $T_{s0}$  are the perturbation amplitudes of the interface and temperature field in the solid.

Consider the perturbed form of nonlinear equation for the temperature in the liquid. Keeping in mind only linear terms in perturbations, one obtains:

$$\frac{\partial T'_l}{\partial t} + \bar{u} \frac{\partial T'_l}{\partial x_c} + \bar{v} \frac{\partial T'_l}{\partial y_c} + v' \frac{dT'_l}{dy_c} = D_T \left( \frac{\partial^2 T'_l}{\partial x_c^2} + \frac{\partial^2 T'_l}{\partial y_c^2} \right) \quad (22)$$

If the forced flow is negligible, the solution has the similar form to  $T'_s$  at large  $k$  consistent with the well-known Mullins-Sekerka criterion [11] for  $k$  within the framework of the thermal problem of solidification of a pure melt (see, among others, [1]). Substituting:

$$T'_l = g(y_c) \exp(\omega t + ikx_c - \varepsilon ky_c) \quad (23)$$

into Eq. (22) and taking into account Eq. (20), one can get the following equation for the new amplitude function  $g(y_c)$ :

$$\frac{d^2 g}{dy_c^2} - 2\epsilon k \frac{dg}{dy_c} = L(g(y_c), y_c) \quad (24)$$

where:

$$L = \left[ \omega + kV\epsilon \exp(-i\epsilon\theta) - \frac{aUk \sin\theta \cos\theta}{\rho} y_c \right] \frac{g(y_c)}{D_T} + \frac{\omega QV \cos\theta}{c_p D_T^2} \Sigma \quad (25)$$

We search for a solution of Eq. (24) around the Mullins-Sekerka solution [11] with a constant amplitude  $g(y_c) = T_{10} = \text{const}$ . Substitution  $T_{10}$  in the right-hand side of (24) gives the first order approximation for  $g(y_c)$ . The result is:

$$g(y_c) = T_{10} - \left[ \left( \frac{\omega}{2\epsilon k} + \frac{V}{2} \exp(-i\epsilon\theta) - \frac{aU \sin\theta \cos\theta}{4k\rho} i \right) \frac{T_{10}}{D_T} + \frac{\omega QV \cos\theta}{2D_T^2 c_p \epsilon k} \Sigma \right] y_c + \frac{aU \sin\theta \cos\theta}{4\epsilon\rho D_T} i T_{10} y_c^2 \quad (26)$$

where the strong inequality  $V/D_T \ll k$  is taken into account (we estimate  $k$  from the Mullins-Sekerka theory as  $10^7 \text{ m}^{-1}$  [11] and  $V/D_T$  as  $10^2 \text{ m}^{-1}$  for metallic binary alloys).

Eq. (3) written for the concentration perturbations  $C'_i$  in the liquid can be solved in the same manner. The result is:

$$C'_i = h(y_c) \exp(\omega t + ikx_c - \epsilon ky_c) \quad (27)$$

$$h(y_c) = C_{10} - \frac{aUk \sin\theta \cos\theta}{2\rho D_c (V \cos\theta/D_c - 2\epsilon k)} i C_{10} y_c^2 + \left\{ \left[ \omega + V\epsilon k \exp(-i\epsilon\theta) + \frac{aUk \sin\theta \cos\theta}{V \cos\theta/D_c - 2\epsilon k} \frac{i}{\rho} \right] \frac{C_{10}}{D_c} + \frac{\omega C_i (1-k_0) V \cos\theta}{D_c^2} \Sigma \right\} \frac{y_c}{V \cos\theta/D_c - 2\epsilon k}$$

Now, expanding the boundary conditions (4) and (5) in series, we arrive at the following set of conditions at the solid-liquid interface  $y_c = 0$ :

$$T'_l = \frac{QV \cos\theta}{D_T c_p} \xi' - mC'_l + \frac{mC_i (1-k_0) V \cos\theta}{D_c} \xi' - \frac{Qd}{c_p} \frac{\partial^2 \xi'}{\partial y_c^2}$$

$$T'_s = mC'_l - \frac{mC_i (1-k_0) V \cos\theta}{D_c} \xi' + \frac{Qd}{c_p} \frac{\partial^2 \xi'}{\partial y_c^2}$$

$$\frac{Q}{c_p} \frac{\partial \xi'}{\partial t} = D_T \left( \frac{\partial T'_s}{\partial y_c} + \frac{\partial T'_l}{\partial y_c} - \frac{QV^2 \cos^2\theta}{D_T^2 c_p} \xi' \right)$$

$$\frac{1-k_0}{D_c} (C_i v' - V \cos\theta C'_l) = \frac{\partial C'_l}{\partial y_c} + \frac{C_i k_0 (1-k_0) V^2 \cos^2\theta}{D_c^2} \xi' \quad (28)$$

where  $d = \sigma_p T_0 / Q^2$  stands for the capillary length. Substitution of perturbations (21), (23) and (27) into the boundary conditions (28) gives four linear relations for the perturbation amplitudes  $\Sigma$ ,  $T_{10}$ ,  $T_{s0}$ , and  $C_{10}$ .

Let us consider a frame with the normal axis and tangent axis to the interface whose origin moves normally to the solid-liquid boundary at the velocity  $V \cos\theta$ . Because of the rotational symmetry of the system, a perturbation of wave number  $k$  grows with the rate  $\omega(k)$ . If now the origin of the

frame moves in the  $z$ -direction with the constant velocity  $V$ , the growth rate of the same perturbation is  $\omega(k) + iVk \sin\theta$  due to the tangential velocity of the new frame  $V \sin\theta$  [2]. Therefore, replacing  $\omega(k)$  by  $-iVk \sin\theta$  at the neutral stability curve and eliminating the perturbation amplitudes from expressions (28), we arrive at the following equation for the wave number  $k$ :

$$k^4 = \frac{Vk^2}{2dD_T} \left( \exp(i\theta)P + \frac{Ndi}{4V} \right) - \frac{mC_i V (1-k_0) N \cos\theta}{4dD_c^2 Q/c_p} i \quad (29)$$

$$N = \frac{aU \sin\theta \cos\theta}{\rho}, \quad P = 1 + \frac{2mC_i (1-k_0) D_T}{D_c Q/c_p}$$

where, in accordance with our estimates  $k \sim 10^7 \text{ m}^{-1}$ ,  $V/D_T \sim 10^2 \text{ m}^{-1}$ ,  $V/D_c \sim 10^6 \text{ m}^{-1}$ ,  $U \sim V$ ,  $d \sim 10^{-10} \text{ m}$ ,  $\rho \sim 10^{-5} \text{ m}$ . Here we write down only the terms corresponding to the solution in the absence of a forced flow and terms describing the influence of this flow for pure thermal and impurity problems. The first term in the right-hand side of Eq. (29) represents the solution of the thermal problem without impurities and external flow [1,22,26], the first two summands give the solution of the thermal problem complicated by impurity transport in the liquid without external flow [2], the first and third summands describe the solution with a forced flow in the absence of the solute transport [1]. As a result, solution of the complete Eq. (29) gives the critical wave number with which perturbations neither grow or decay at the dendrite tip growing into the binary system under convective flow.

Let us consider the case of a fourfold symmetry of the crystal. Then, the capillary length can be written as  $d(\theta) = d_0(1 - \beta \cos 4\theta)$ , where  $\beta = 15\epsilon_c \ll 1$  is the anisotropic factor,  $\epsilon_c$  is the strength of anisotropy of the surface energy at the solid-liquid interface.

Expanding the solution of equation (29) in series in  $U$ , we can get an approximate expression for the wave-number in the form:

$$k = k_{TC} \sqrt{\frac{\exp(i\theta) + i\alpha_0 Z(\theta)(1 - \beta \cos 4\theta) \sin\theta \cos\theta}{1 - \beta \cos 4\theta}} \quad (30)$$

where:

$$k_{TC} = -\sqrt{\frac{VP}{2d_0 D_T}}, \quad \alpha_0 = \frac{aUd_0}{4P\rho V}, \quad (31)$$

$$Z(\theta) = 1 - \alpha_1 \cos\theta \exp(-i\theta), \quad Z(\theta) = \alpha_1 = \frac{4(1-k_0)mC_i D_T^2}{PD_c^2 Q/c_p}$$

Let us compare expressions (30) and (31) with known theories. Setting  $U = 0$  and  $C_i = 0$ , we arrive at the Mullins-Sekerka solution [11,22,26]. If  $U = 0$ , we have the Ben Amar-Pelce solution [2]. In the case of  $C_i = 0$ , we come to the Bouissou-Pelce solution [1].

Taking into account that:

$$l = -\frac{\rho}{2} \left[ \frac{\tan\theta}{\cos\theta} + \ln \left( \tan\theta + \frac{1}{\cos\theta} \right) \right]$$

(see, among others, ref. [26]), let us rewrite the solvability condition (15) by analogy with Bouissou and Pelce [1] in the form:

$$\int_{-\infty}^{+\infty} G(\chi) \exp[\sqrt{C} \Psi_\alpha(\chi)] d\chi = 0, \quad \chi = \tan \theta \quad (32)$$

where:

$$\Psi_\alpha(\chi) = \frac{i}{2} \int_0^\chi \frac{[(1+i\chi')(1+\chi'^2)^{5/2} + i\alpha\chi'B(\chi')]^{1/2}}{\sqrt{B(\chi')}} d\chi' \quad (33)$$

$$B(\chi) = (1+\chi^2)^2(1-\beta) + 8\beta\chi^2, \quad \alpha(\chi') = \alpha_0 \left(1 + \frac{i\alpha_1\chi'}{1+i\chi'}\right)$$

and constant  $C$  is normalized by a factor  $VP\rho^2/(2d_0D_T)$ .

Let us evaluate this integral in the limit of small anisotropy by means of the method developed by Bouissou and Pelce [1]. The numerator of the integrand vanishes for  $\chi$  close to  $\chi=i$  (stationary phase point) and the denominator for  $\chi=i(1-\sqrt{2\beta})$  (point of singularity). As the dominant contribution to the integral is determined by the neighborhood of  $\chi=i$ , function  $\Psi_\alpha(\chi)$  can be approximated by:

$$\Psi_\alpha(\varphi) = 2^{9/8} \beta^{7/8} \int_{1/\sqrt{2\beta}}^\varphi \frac{[\varphi'^{7/2} - \tau\Phi(\varphi')(\varphi'^2-1)]^{1/2}}{\sqrt{\varphi'^2-1}} d\varphi' \quad (34)$$

with

$$\chi = i(1-\sqrt{2\beta}\varphi), \quad \tau = 2^{-5/4} \beta^{-3/4} \alpha, \quad \Phi = 1 - \frac{i\alpha_1}{\sqrt{2\beta}\varphi'}$$

The integral (34) can be approximately calculated by analogy with the similar integral met in the problem of dendritic growth in a pure (one-component) system [1]. Following the result of this analysis, only two dominant contributions to the integral (34) exist: the contribution from the loop and the contribution from the stationary phase points. The first of them should be calculated between a distance  $\sim \tau^{2/7}$  (a splitting distance of the stationary phase points) at the intersection of the steepest descent path and the real axis and  $\varphi=1$ . It gives an oscillating factor to the exponentially small value of the integral which behaves as  $\cos[A_1\sqrt{C}\beta^{7/8}(1+B_1\tau^{11/10})]$ . Each stationary phase point contributes by a term with oscillating part of the form  $\cos[A_2\sqrt{C}\beta^{7/8}(1+B_2\tau^{11/10})]$ , where  $A_1$ ,  $A_2$ ,  $B_1$  and  $B_2$  are constants. The cancelation of the sum of these contributions in Eq. (30) gives the following selected values of  $C$ :

$$C = \frac{n^2}{\beta^{7/4}} [1 + b(\beta^{-3/4}\alpha_0)^{1/14}] \quad (35)$$

where  $n$  is an arbitrary interger and  $b$  a numerical constant.

Substitution of the normalization requirement into Eq. (34) leads to the expression for the scaling factor  $\sigma^*$  in the form:

$$\sigma^* \equiv \frac{2d_0D_T}{\rho^2V} = \frac{\sigma_0\beta^{7/4}}{1+b(\beta^{-3/4}\alpha_0)^{1/14}} \left(1 + \frac{2D_T(1-k_0)mC_i}{D_C Q/c_p}\right) \quad (36)$$

where  $\sigma_0$  stands for a numerical constant which can be found from the asymptotic analysis [20] or from the fitting of the model predictions to experimental data.

Eq. (36) gives a criterion for the stable mode of a dendritic tip in the presence of anisotropy of surface energy and for the non-isothermal binary systems under forced flow in the liquid phase. This criterion joins results obtained from the dendritic model of Bouissou and Pelce [1] and Ben Amar and Pelce [2].

With infinite solute diffusion,  $D_C \rightarrow \infty$ , the parameters  $P$  and  $\alpha_0$  in Eqs. (30) and (31) are equal to  $P=1$  and  $\alpha_0 = aUd_0/(4\rho V)$ , respectively. In this case criterion (36) transforms into the result extracted from analysis of Bouissou and Pelce (Eq. (45) in ref. [1]). Note that with  $U=0$  and  $\alpha_0=0$ , Eq. (36) further transforms to the case of dendritic growth in a pure stagnant system [20,22].

In Eqs. (30) and (31), the wave-number  $k_{TC}$  being consistent with those one given by Ben Amar and Pelce (see Eq. (50) in ref. [2]). With the absence of convection, i.e., at  $U=0$  and  $\alpha_0=0$ , the system of Eqs. (30) and (31) completely merges with the results of ref. [2].

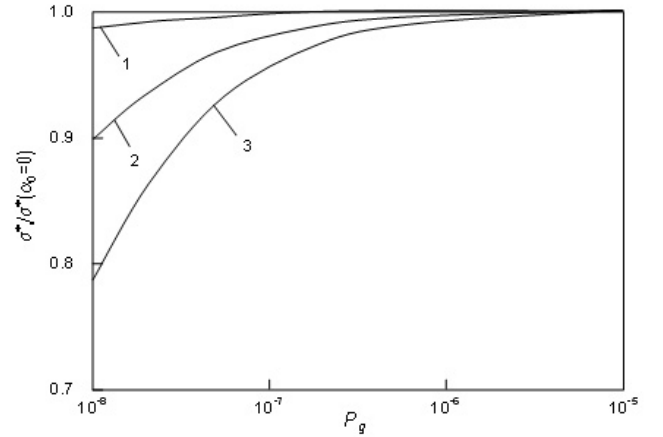
The complete stability criterion (36) can be rewritten in the following form:

$$\frac{\sigma^*}{\sigma^*_{\alpha_0=0}} = \frac{1}{1+b(\beta^{-3/4}\alpha_0)^{1/14}} \quad (37)$$

where:

$$\sigma^*_{\alpha_0=0} = \sigma_0\beta^{7/4} \left(1 + \frac{2D_T(1-k_0)mC_i}{D_C Q/c_p}\right)$$

Figure 1 shows the influence of Peclet numbers on stability criterion. As is seen, with the increasing the flow Peclet number  $P_f$  and decreasing of the growth Peclet number  $P_g$  the contribution of the convection into the stability of the dendrite tip gradually increases.



**Figure 1** Ratio  $\sigma^*/\sigma^*_{\alpha_0=0}$  as a function of the growth Peclet number  $P_g$  for different values of the flow Peclet number  $P_f$  in accordance with criterion (37). Physical parameters used in calculations are closely related to parameters for metallic binary systems:

$$D_T/\nu = 10, \quad d_0/\rho = 10^{-5}, \quad \beta = 0.195, \\ D_T/D_C = 5 \cdot 10^3, \quad k_0 = 0.5, \quad c_\infty = 1 \text{ at\%}, \quad m = 10 \text{ K/at\%}, \\ Q/c_p = 300, \quad P_f = 10^{-5} \text{ (1)}, \quad 5 \cdot 10^{-5} \text{ (2)}, \quad P_f = 10^{-4} \text{ (3)}.$$

## CONCLUDING REMARKS

The three-dimensional problem of a steady-state dendritic growth with forced convective flow was taken up in a binary system. For the axi-symmetric crystal shape with anisotropy of surface energy, the analysis of a stable mode for tip of parabolic dendrite is performed. The critical wave-number is found as a marginal state at which perturbations neither grow or decay. The solvability condition gives a stability criterion (36) for a binary system under convective flow.

As a final note, the criterion (36) is not valid for large Reynolds numbers and, as a result, for large growth and flow Peclet numbers. This is connected with the Oseen approximation of the Navier-Stokes equation used in the aforementioned theory.

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## REFERENCES

- [1] Bouissou Ph., and Pelce P., Effect of a forced flow on dendritic growth, *Phys. Rev. A*, Vol. 40(1), 1989, pp. 6673-6680.
- [2] Ben Amar M., and Pelce P., Impurity effect on dendritic growth, *Phys. Rev. A*, Vol. 39(8), 1989, pp.4263-4269.
- [3] Flemings M.C., Solidification Processing, McGraw-Hill, New York, 1974.
- [4] Chernov A.A., In Modern Crystallography III, in M. Cardona, P. Fulde, H.J. Queisser (Eds.), Springer Series in Solid-State Science, Vol. 36, Springer, Berlin, 1984.
- [5] Kurz W., and Fisher D.J., Fundamentals of Solidification, third ed., Trans. Tech., Aedermannsdorf, 1992.
- [6] Galenko P.K., Funke O., Wang J., and Herlach D.M., Kinetics of dendritic growth under the influence of convective flow in solidification of undercooled droplets, *Mat. Sci. Eng.*, Vols. 375-377, 2004, pp. 488-492.
- [7] Funke O., Phanikumar G., Galenko P.K., Chernova L., Reutzel S., Kolbe M., and Herlach D.M., Dendrite growth velocity in levitated undercooled nickel melts, *J. Crystal Growth*, Vol. 297, 2006, pp. 211-222.
- [8] Herlach D.M., Galenko P.K., and Holland-Moritz, Metastable Solids from Undercooled Melts, Elsevier, Amsterdam, 2007.
- [9] Asta M., Beckermann C., Karma A., Kurz W., Napolitano R., Plapp M., Purdy G., Rappaz M., and Trivedi R., Solidification microstructures and solid-state parallels: recent developments, future directions, *Acta Mater.*, Vol. 57(4), 2009, pp. 941-971.
- [10] Ivantsov G.P., *Dokl. Akad. Nauk USSR*, Vol. 58, 1947, pp. 567, Translation by Horvay G. in General Electric Report No 60-RL-2511M, 1960.
- [11] Mullins W.W., and Sekerka R.F., Stability of a planar interface during solidification of a dilute binary alloy, *J. Appl. Phys.*, Vol. 35, 1964, pp. 444-451.
- [12] Langer J.S., Müller-Krumbhaar H., Theory of dendritic growth – I. Elements of stability analysis; II. Instabilities in the limit of vanishing surface tension; III. Effect of surface tension, *Acta Metall.*, Vol. 26, 1978, pp.1681-1708.
- [13] Stuart J.T., Nonlinear stability theory, *Ann. Rev. Fluid Mech.*, Vol. 3, 1971, pp. 347-370.
- [14] Wollkind D.J., and Segel L.A., A nonlinear stability analysis of the freezing of a dilute binary alloy, *Philos. Trans. Roy. Soc. A*, Vol. 268, 1970, pp. 351-380.
- [15] Alexandrov D.V., and Ivanov A.O., Dynamic stability analysis of the solidification of binary melts in the presence of a mushy region: changeover of instability, *J. Crystal Growth*, Vol. 210, 2000, pp. 797-810.
- [16] Alexandrov D.V., A nonlinear instability analysis of crystallization processes with a two-phase zone, *J. Metastable and Nanocrystalline Materials*, Vols. 20-21, 2004, pp. 468-475.
- [17] Aleksandrov D.V., Mansurov V.V., and Galenko P.K., Morphological stability of the plane phase boundary in a binary melt for processes of high-rate crystallization, *Physics-Doklady*, Vol. 41(1), 1996, pp. 511-513.
- [18] Alexandrov D.V., Self-similar solidification: morphological stability of the regime, *Int. J. Heat Mass Transfer*, Vol. 47, 2004, pp. 1383-1389.
- [19] Kessler D.A., Koplik J., and Levine H., Pattern selection in fingered growth phenomena, *Adv. Phys.*, Vol. 37(3), 1988, pp. 255-339.
- [20] Brener E.A., and Mel'nikov V.I., Pattern selection in two-dimensional dendritic growth, *Adv. Phys.*, Vol. 40(1), 1991, pp. 53-97.
- [21] Benamar M., and Bouissou Ph., An exact solution for the shape of a crystal growing in a forced flow, *J. Crystal Growth*, Vol. 92, 1988, pp. 97-100.
- [22] Pelce P., Dynamics of Curved Fronts, Academic Press, Boston, 1988.
- [23] Galenko P.K., and Herlach D.M., in D.M. Herlach (Ed.) Phase Transformations in Multicomponent Systems, Wiley-VCH, Weinheim, 2008, pp.353-373.
- [24] Alexandrov D.V., Galenko P.K., and Herlach D.M., Selection criterion for the growing dendritic tip in a non-isothermal binary system under forced convective flow, *J. Crystal Growth*, Vol. 312, 2010, pp. 2122-2127.
- [25] Dash S.K., and Gill W.N., Forced convection heat and momentum transfer to dendritic structures (parabolic cylinders and paraboloids of revolution), *J. Heat Mass Transfer*, Vol. 27(8), pp. 1345-1356.
- [26] Pelce P., and Bensimon D., Theory of dendrite dynamics, *Nuclear Phys.*, Vol. B 2, 1987, pp. 259-270.
- [27] Zel'dovich Ya.B., Istratov A.G., Kidin N.I., and Librovich V.B., Flame propagation in tubes: hydrodynamics and stability, *Combust. Sci. Technol.*, Vol. 24, 1980, pp. 1-13.
- [28] Caroli B., Caroli C., Roulet B., and Langer J.S., Solvability condition for needle crystals at large undercooling in a nonlocal model of solidification, *Phys. Rev. A*, Vol. 33(1), 1986, pp. 442-452.
- [29] Shraiman B.I., Velocity selection in the Saffman-Taylor problem, *Phys. Rev. Lett.*, Vol. 56, 1986, pp. 2028-2031.
- [30] Dombre T., Hakim V., and Pomeau Y., Finger widths selection in the Saffman-Taylor instability, *C.R. Acad. Sci.*, Vol. 302, 1986, pp. 803.