

# On the Theory of Dendritic Growth: Soret and Temperature-Dependent Diffusion Effects

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**Abstract**—An analytical solution is found for the problem of the growth of an isolated dendrite in a convective binary melt with allowance for the Soret and temperature-dependent diffusion effects. Nonlinear impurity transport is shown to radically change the impurity concentration in front of the growing crystal and, correspondingly, the concentration supercooling, which is responsible for the condition of choosing the dendrite tip growth rate.

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## INTRODUCTION

The evolution of dendrites is known to control the structure formation in the materials formed during the solidification of melts [1, 2]. Apart from experimental observation of the dendritic growth dynamics, qualitatively new materials have recently been obtained by mathematical simulation to test the basic concepts of crystal morphology formation (see review [3] and Refs. therein). Among the important problems, we distinguish the problem of stable growth of the tip of a freely growing dendrite crystal and the problem of the effect of a convective flow on the mechanism of choosing its growth mode. These problems have theoretical and practical importance [4, 5].

The problem of choosing the stable growth mode of an isolated crystal appeared from an analysis of the Ivantsov solutions [6, 7] and the experimental data on the growth of a needlelike crystal of a parabolic shape [8–14]. These comparison and tests led to the conclusion that the continuous family of isotropic Ivantsov solutions is unstable: a needlelike crystal loses its initial parabolic shape in a steady-state growth mode [15]. Therefore, the Ivantsov solution is only used as a zeroth approximation to solve the problem of stable growth, where the role of a small parameter is played by surface tension anisotropy or growth kinetics anisotropy [16]. After finding the criterion of stable dendrite tip motion in a single-component unmovable medium [15, 16], the problem was extended to the cases of a convective medium flow [17–19], crystal growth in a binary (chemically two-component) convective-free system [20], and dendritic growth in a binary liquid with convection [21, 22].

Directional solidification is known to be controlled by changing the temperature gradients or temperatures in solid and liquid phases. The temperature gradients and temperatures set far from the phase-transi-

tion temperature can be rather high. This circumstance can result in the thermodiffusion effect (Soret effect), when the difference in the temperatures in various melt regions leads to substance transport into them; that is, a temperature gradient changes the concentration (see, e.g., [23]). As a rule, a thermodiffusion flow depends on the substance concentration. For example, it is proportional to the impurity concentration in dilute solutions and melts [23–25]. Therefore, we can write the substance flow in the form

$$\bar{J} = -D\nabla C - D_T(\beta_0 + \beta_1 C)\nabla T, \quad (1)$$

where  $C$  and  $T$  are the concentration and temperature fields, respectively;  $D$  and  $D_T$  are the diffusion and thermodiffusion coefficients, respectively; and  $\beta_0$  and  $\beta_1$  are constants. The first term in Eq. (1) is responsible for conventional substance diffusion, and the second term, for thermodiffusion. We will consider the following cases:  $\beta_0 = C_\infty$ ,  $\beta_1 = 0$  ( $C_\infty$  is the initial concentration, or the concentration at infinity) and  $\beta_0 = 0$ ,  $\beta_1 = 1$ . The first case was discussed in [26]: it corresponds to small concentration changes as a function of the spatial coordinate in a melt. If the change in the concentration cannot be neglected, the second case is better to describe the process [27].

The inverse Soret effect (so-called Dufour effect), where a concentration gradient causes temperature changes, is also known. However, when solutions and melts are studied, this effect may be neglected (as a rule, it is important in gas mixtures (see, e.g., [28])).

To facilitate calculations, the diffusion coefficient is usually taken to be constant; however, generically speaking, this is not the case, since the diffusion coefficient depends on temperature. By analogy with

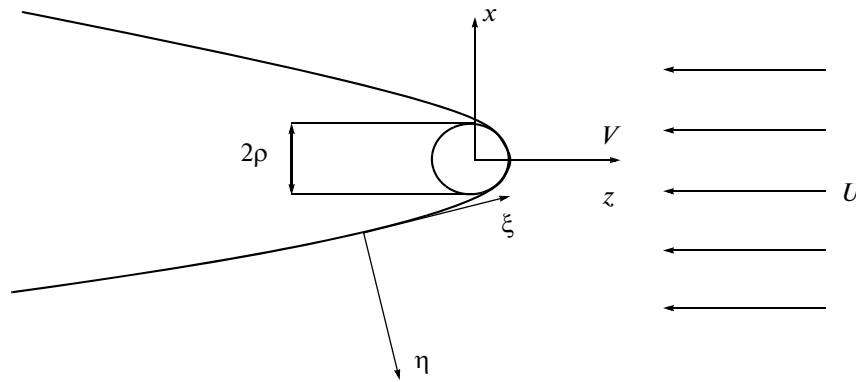


Fig. 1. Schematic diagram of dendrite growth in an incident liquid flow.

[27, 29], we consider this temperature dependence in the linear form

$$D(T) = D_0 + \frac{\partial D}{\partial T}(T - T_0), \quad (2)$$

where  $D_0$  is the diffusion coefficient at melting temperature  $T_0$  and  $\partial D/\partial T = D'$  is the temperature coefficient (see, e.g., [29]).

It is known from experiments that the growth of dendritic structures completely characterizes the properties of solids. Solidification usually proceeds in the mode of “dendritic forest” formation in the supercooled region in a melt, which is often called a mushy zone [30, 31]. Therefore, to study the specific features of dendritic growth is important from the standpoint of structure–phase transitions in the mushy zone. The rejection of impurities by a growing crystal (dendrites) is known to result in a concentration supercooling [32], which favors the growth of a solid phase deep into the melt. The supercooling that appears during the solidification of metallic alloys is often compensated by intense formation of latent heat of solidification. In this case, the mushy zone having appeared during solidification is quasi-equilibrium [33–35].

The authors of [36–43] developed methods for solving nonlinear heat- and mass-transfer equations in a quasi-equilibrium mushy zone solidifying at a constant rate. This process takes place in many experiments and corresponds to the pulling of a crystal from a melt. Solidification often occurs from a cooling boundary, the temperature of which is maintained at a constant level or is a function of time. In the former case, solidification corresponds to a self-similar mode. The theory of mushy zone solidification for this situation was developed in [44–52]. In the latter case, the system dynamics depends substantially on the time variation of the cooling boundary temperature. To describe the processes that take place under these conditions, the authors of [53–65] developed methods for analytical solution of quasi-equilibrium mushy zone equations.

Thermodiffusion and the temperature dependence of a diffusion coefficient during the heat and mass transfer in the mushy zone were taken into account in [66, 67]. In those works, however, the mushy zone was considered without taking into account the specific features of the growth of individual dendritic structures; therefore, it is important to perform such an analysis. Although dendritic growth was studied in many works, the temperature dependence of a diffusion coefficient and the possibility of thermodiffusion substance transport have not yet been considered. The purpose of this work is study the effect of Eqs. (1) and (2) on the impurity distribution created by the growth of an isolated dendrite in an incident melt flow.

The problem is formulated for an extended Stefan model with a frontal interfacial dendrite surface (parabolic crystal–liquid interface) when thermodiffusion and the temperature dependence of a diffusion coefficient are taken into account. The problem for a forced flow is solved in the Oseen approximation.

## MODEL FOR DENDRITIC GROWTH

Figure 1 shows the dendritic growth in an incident viscous melt flow. The temperature field in the liquid phase is described by the convective heat conduction equation

$$\frac{\partial T}{\partial t} + (\bar{w}, \nabla)T = a\Delta T, \quad (3)$$

where  $a$  is the thermal diffusivity,  $\bar{w}$  is the liquid flow velocity,  $t$  is the time, and  $\Delta$  is the Laplace operator.

The impurity distribution in the liquid part of the system obeys the convective diffusion equation

$$\frac{\partial C}{\partial t} + (\bar{w}, \nabla)C = -\nabla \bar{J}, \quad (4)$$

and flow  $\bar{J}$  is determined by Eq. (1).

The heat and mass balance conditions

$$Q\bar{v} \cdot \bar{n} = ac_p(\nabla T_s - \nabla T)\bar{n}, \quad (5)$$

$$(1 - k_0)C\bar{v} \cdot \bar{n} - \bar{J}\bar{n} = 0. \quad (6)$$

hold true at the mobile interface. Here,  $Q$  is the latent heat of solidification,  $T_s$  is the (isothermal) dendrite temperature,  $\bar{v}\vec{n}$  is the normal velocity of the dendrite surface,  $c_p$  is the heat capacity of the melt, and  $k_0$  is the equilibrium impurity distribution coefficient. Following [19], we consider the liquid flow at small Reynolds numbers. In this case, the velocity distribution in the melt obeys the Oseen equation and the continuity equation [68]

$$U \frac{\partial \bar{w}}{\partial z} = -\frac{1}{\rho_l} \nabla p + \nu \Delta \bar{w}, \quad \nabla \bar{w} = 0, \quad (7)$$

where  $U$  is the incident liquid flow velocity far from the growing crystals and  $\rho_l$  and  $\nu$  are the liquid density and viscosity, respectively. Note that the Oseen approximation used in motion equation (7) can only take into account the most important inertial terms and yields rather accurate calculation results (see, e.g., the classical problem of the motion of a sphere in a viscous liquid [69, 70]).

### ANALYTICAL SOLUTION

Let a two-dimensional parabolic dendrite grow at a constant rate  $V$  along spatial axis  $z$  (Fig. 1). The liquid flow far from the crystal is parallel to axis  $z$  and is opposite to the dendrite growth direction (so-called incident flow). We introduce parabolic coordinates  $\xi$  and  $\eta$  connected to Cartesian coordinates  $x$  and  $z$  by the relations

$$x = \rho \sqrt{\xi \eta}, \quad z = \frac{\rho}{2} (\eta - \xi), \quad (8)$$

where  $\rho$  is the dendrite tip radius and the interface is located at the level  $\eta = 1$ .

With Eq. (7), we can determine liquid velocity components  $u_\eta$  and  $u_\xi$  in parabolic coordinates (8). Following [19], we write the result in the form

$$u_\eta = -\frac{f(\eta)}{2\sqrt{\xi + \eta}}, \quad u_\xi = \frac{\sqrt{\xi \eta}}{\sqrt{\xi + \eta}} \frac{df}{d\eta}, \quad (9)$$

where we introduced the defining functions

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

$$g(\eta) = \sqrt{\eta} \frac{\operatorname{erfc}\sqrt{\eta \operatorname{Re}/2}}{\operatorname{erfc}\sqrt{\operatorname{Re}/2}} + \frac{\sqrt{2/(\pi \operatorname{Re})}}{\operatorname{erfc}\sqrt{\operatorname{Re}/2}}$$

$$\times [\exp(-\operatorname{Re}/2) - \exp(-\eta \operatorname{Re}/2)],$$

which take into account the flow intensity using Reynolds number  $\operatorname{Re} = \rho_l U/\nu$ .

Equation (3) can easily be integrated in parabolic coordinates (8). Having found its solution (which

depends only on variable  $\eta$ ), we rewrite the formulation of the problem in the form (see [18])

$$u_\eta \frac{dT}{d\eta} = \frac{2a}{\rho \sqrt{\xi + \eta}} \left( \sqrt{\eta} \frac{d^2 T}{d\eta^2} + \frac{1}{2\sqrt{\eta}} \frac{dT}{d\eta} \right), \quad (10)$$

$$\left( \frac{dT}{d\eta} \right)_{\eta=1} = -\frac{Q \rho V}{c_p 2a},$$

where the temperature in the melt at infinity is considered to be specified; that is,  $T \rightarrow T_\infty$  at  $\eta \rightarrow \infty$ .

The integration of Eq. (10) gives the temperature distribution in the liquid phase of the system,

$$T(\eta) = T_i + (T_\infty - T_i) \frac{I(\eta)}{I(\infty)}. \quad (11)$$

Here, we introduced the following designations:

$$I(\eta) = \int_1^\eta \exp \left[ P_f \int_1^{\eta'} \frac{g(\eta'')}{\sqrt{\eta''}} d\eta'' - (P_f + P_g) \eta' \right] \frac{d\eta'}{\sqrt{\eta'}},$$

$$T_i = T_\infty + \frac{Q}{c_p} P_g \exp(P_f + P_g) I(\infty), \quad P_g = \frac{\rho V}{2a}, \quad P_f = \frac{\rho U}{2a},$$

where  $P_g$  and  $P_f$  are the growth and flow Peclet numbers, respectively, which are defined in terms of dendrite growth rate  $V$  and liquid flow velocity  $U$ .

Equation (4) depends only on coordinate  $\eta$  and is written as

$$-\frac{f(\eta)}{2} \frac{dC}{d\eta} = \frac{2}{\rho} \frac{d}{d\eta} \times \left[ \sqrt{\eta} \left( D(T) \frac{dC}{d\eta} + D_T (\beta_0 + \beta_1 C) \frac{dT}{d\eta} \right) \right]. \quad (12)$$

We introduce designations  $y(\eta) = \beta_0 + \beta_1 C(\eta)$  and  $dy/d\eta = y(\eta)/q(\eta)$  and reduce the order of Eq. (12),

$$\frac{dq}{d\eta} \cong F(q, \eta), \quad (13)$$

where

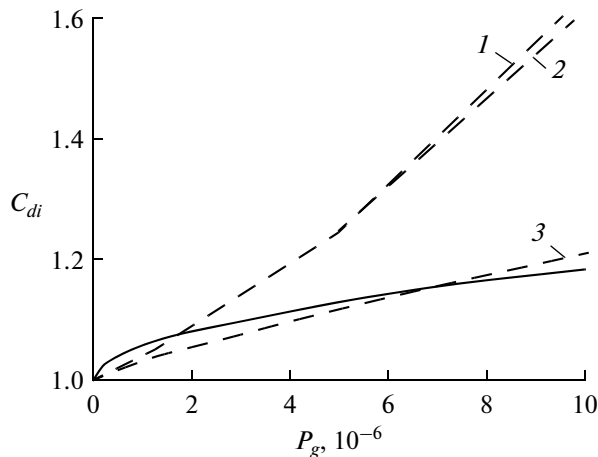
$$F(q, \eta) = -\frac{D_T \beta_1}{D(T)} \left[ \frac{1}{2\eta} \frac{dT}{d\eta} + q \frac{dT}{d\eta} + \frac{d^2 T}{d\eta^2} \right] - q^2$$

$$- \frac{D_T P_f + P_g - P_f g(\eta)/\sqrt{\eta}}{D_0 [1 + (T(\eta) - T_0) D'/D_0]} q - \frac{q D'}{D(T)} \frac{dT}{d\eta} - \frac{q}{2\eta}.$$

Here, the temperature and its derivative are known and determined with Eq. (11), and the ratios of coefficients  $D_T/D(T)$  and  $D'/D(T)$  are

$$\frac{D_T}{D(T)} = \frac{D_T/D_0}{1 + (T - T_0) D'/D_0},$$

$$\frac{D'}{D(T)} = \frac{D'/D_0}{1 + (T - T_0) D'/D_0}.$$



**Fig. 2.** Impurity concentration on the dendrite surface  $C_{di}$  (at  $\eta = 1$ ) vs. Peclet number  $P_g$ . (solid line) Solution to the problem in the absence of the Soret and temperature-dependent diffusion effects ( $D_T = D' = 0$ ,  $D = D_0 = \text{const}$ ). (dashed lines) Calculation at (1)  $D_T/D_0 = -10^{-2} \text{ K}^{-1}$ ,  $D'/D_0 = 10^{-1} \text{ K}^{-1}$ ; (2)  $D_T/D_0 = 10^{-1} \text{ K}^{-1}$ ,  $D'/D_0 = 10^{-1} \text{ K}^{-1}$ ; and (3)  $D_T/D_0 = -10^{-2} \text{ K}^{-1}$ ,  $D'/D_0 = 1 \text{ K}^{-1}$ .

Boundary condition (6) on the dendrite surface  $\eta = 1$  is now written in the situation where  $\beta_0 = 0$  and  $\beta_1 = 1$  [27],

$$q = -\frac{D_T/D_0}{1 + (T - T_0)D'/D_0} \frac{dT}{d\eta} - \frac{(1 - k_0)P_g D_T/D_0}{1 + (T - T_0)D'/D_0}. \quad (14)$$

Equations (13) and (14) represent the Cauchy problem the solution to which determines function  $q(\eta)$ . Using the found dependence  $q(\eta)$ , we can determine the impurity concentration distribution in the melt,

$$C(\eta) = \frac{1}{\beta_1} \left[ (\beta_0 + \beta_1 C_\infty) \frac{\exp\left(\int_1^\eta q(\lambda) d\lambda\right)}{\exp\left(\int_1^\infty q(\lambda) d\lambda\right)} - \beta_0 \right], \quad (15)$$

where  $C_\infty$  is the impurity concentration in the melt far from the dendrite. Thus, Eqs. (11) and (13)–(15) represent the solution to the heat-and-mass transfer problem of dendritic growth when the thermodiffusion and temperature-dependent diffusion effects are taken into account.

## CONCLUSIONS

Figure 2 shows the impurity concentration on the dendrite surface versus the growth Peclet number. The calculation parameters of the system were borrowed from [21, 22, 66, 67]:  $a/D_0 = 5 \times 10^3$ ,  $k_0 = 0.5$ ,  $C_\infty = 1$ ,  $Q/c_p = 300$ ,  $a/v = 10$ ,  $P_f = 10^{-6}$ ,  $T_\infty = 1000^\circ\text{C}$ ,  $\beta_0 = 0$ ,

and  $\beta_1 = 1$ . An analysis of the calculated curves demonstrates that the presence of nonlinear impurity transport effects in the melt radically changes the impurity distribution. In this case, the role of coefficient  $D'$  is more substantial than the role of coefficient  $D_T$  in the system parameter ranges under study.

The change in the surface impurity concentration (and the concentration profile in the melt) results in a change in the concentration supercooling in the space in front of the dendrite tip. The calculations show that the impurity concentration near the dendrite tip can both increase and decrease. As a result, the concentration supercooling increases or decreases, respectively, and the dendrite tip growth rate changes respectively. To study this problem in detail, it is necessary to analyze the stability of dendritic growth with allowance for the Soret and temperature-dependent diffusion effects in terms of works [18–22].

In conclusion, note that the effects of nonlinear impurity transport considered in this work will change the solutions to the equations of a weakly nonequilibrium mushy zone [71–73] and will affect the interface stability [74–84].

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