Selection criterion of stable dendritic growth at arbitrary Péclet numbers with convection

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A free dendrite growth under forced fluid flow is analyzed for solidification of a nonisothermal binary system. Using an approach to dendrite growth developed by Bouissou and Pelcé [Phys. Rev. A **40**, 6673 (1989)], the analysis is presented for the parabolic dendrite interface with small anisotropy of surface energy growing at arbitrary Péclet numbers. The stable growth mode is obtained from the solvability condition giving the stability criterion for the dendrite tip velocity V and dendrite tip radius ρ as a function of the growth Péclet number, flow Péclet number, and Reynolds number. In limiting cases, the obtained stability criterion presents known criteria for small and high growth Péclet numbers of the solidifying system with and without convective fluid flow.

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It is well known that solidification processes from undercooled melts or saturated solutions are characterized by the growth of a complex and highly branched crystal structure called a dendrite [1]. A first step towards a modern theory of dendritic growth was Ivantsov's solution of the needle crystal problem in the limit of zero surface tension [2,3]. For this analytic solution of the thermal problem, only the growth Péclet number, $P_g = \rho V/(2D_T)$, giving the product of the dendrite tip velocity V and its tip radius ρ, is determined by the undercooling $\Delta = (T_0 - T_\infty)c_p/Q$. Here D_T is the thermal diffusivity, T_0 is the crystallization temperature of the pure liquid, T_∞ is the temperature in the liquid phase far from the growing dendrite, c_p is the specific heat, and Q is the latent heat released per unit volume of solid.

As the Ivantsov solution does not provide information on the tip velocity V and tip radius ρ separately, the criterion for the stable growth mode can be obtained as a second condition for both these parameters [1]. This criterion is obtained from a solvability theory which predicts the marginal mode of the dispersion relation for perturbations on the dendrite interface [4,5]. The solvability theory predicts the second combination of parameters as $\sigma^* = 2d_0D_T/(\rho^2V)$, where d_0 is the capillary length and σ^* is the dimensionless scaling factor ($\sigma^* \propto \beta^{7/4}$ for the thermal problem in the limit of $P_g \ll 1$, where β is the small anisotropy parameter [4–6]). When an external flow and impurities are introduced, a family of the Ivantsov paraboloids can still be used as a solution of the Stefan problem [7–9], either in the large Reynolds-number limit (potential flow approximation) [10], or in the small Reynolds-number limit (Oseen approximation) [11,12]. As in the case of the pure thermal problem, the solidification mode is determined only by the growth and flow Péclet numbers, which are related to thermal and solute transport by the molecular and convective mechanisms. These numbers give the product ρV to which an additional expression should be found again in a form of stability criterion.

Using a solvability condition, different scaling ratios σ^* were obtained [7–9] for a stable dendrite growth mode at small growth Péclet numbers in one-component (pure), binary, and multicomponent systems. The opposite limit of large Péclet numbers was considered in Refs. [13–15], where the scaling ratio is obtained as $\sigma^* \propto \beta^{3/4}/P_g^2$. However, theoretical results connecting these limiting cases are still absent (except approximate interpolation for the scaling parameter σ^* formally postulated in Ref. [16]). Therefore, the main goal of the present study is to develop a unified theory for the stable mode of the nonisothermal dendrite growing in a chemically binary system with convective flow effects and anisotropy of surface energy. The theory presents a case of arbitrary growth Péclet numbers that is considered as a generalization of the aforementioned limiting cases.

We consider a two-dimensional parabolic dendrite growing in the opposite direction to a forced convective flow of one-component undercooled liquid. The starting point of the present analysis is the dispersion law previously derived by Bouissou and Pelcé [8]. In their expression (30), the dispersion law at the neutral stability curve (having the zero perturbation frequency) gives the following cubic equation for the wave-number marginal mode k_m:

$$k_m^3 = \frac{V\,\exp(i\theta)}{2dD_T}k_m + \frac{iaU\,\sin\theta\,\cos\theta}{8\rho D_T}k_m - \frac{iVk_m^2\sin\theta}{2D_T} + \frac{V^2\cos\theta\,\exp(i\theta)}{4dD_T^2}. \quad (1)$$

Here U is the flow velocity far from the growing dendrite, i is the imaginary unit, and θ is the angle between the normal to the dendrite interface and its growth direction. The capillary length d in the case of a fourfold symmetry of the crystal is expressed as $d(\theta) = d_0[1 - \beta\cos(4\theta)]$ and the parameter a characterizes the hydrodynamic solution of the spatially two-dimensional problem as [8]

$$a(\mathrm{Re}) = \sqrt{\frac{\mathrm{Re}}{2\pi}}\frac{\exp(-\mathrm{Re}/2)}{\mathrm{erfc}(\sqrt{\mathrm{Re}/2})}, \quad \mathrm{Re} = \frac{\rho U}{\nu},$$

where Re is the Reynolds number and ν is the kinematic viscosity. Note that the only difference between the two-dimensional

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and three-dimensional models consists in different functions of $a(\mathrm{Re})$ [8,17].

The first summand on the right-hand side of Eq. (1) corresponds to the usual Mullins-Sekerka growth rate. The first line of this relation describes the growth rate in the presence of convection analyzed by Bouissou and Pelcé [8]. Their solution is

$$k_{m\mathrm{BP}} = -\left(\frac{V}{2dD_T}\right)^{1/2}\left[\exp(i\theta) + i\frac{aUd}{4\rho V}\sin\theta\,\cos\theta\right]^{1/2}. \tag{2}$$

Let us estimate the main contributions in Eq. (1) in accordance with the Mullins-Sekerka or Bouissou-Pelcé solution as $V/D_T \sim 10^2\,(\mathrm{m}^{-1})$, $d \sim 10^{-9}\,(\mathrm{m})$, and $k_m \sim 10^6\,(\mathrm{m}^{-1})$. Then the summands entering in the second line of this expression are much smaller than the first summand. Physically, the second line represents perturbations to the Mullins-Sekerka and Bouissou-Pelcé solutions. This is why the second line can be neglected in comparison with the first line in the case of a small Péclet number limit. The role of the two last summands in Eq. (1) increases with increasing the growth Péclet number $P_g = \rho V/(2D_T)$, and Eq. (1) can be solved using Cardano's formula for cubic equations. Taking into account the aforementioned numeric estimations, one obtains

$$k_m = k_{m\mathrm{BP}} + \frac{V\,\exp(-i\theta)}{4D_T}, \tag{3}$$

with $k_{m\mathrm{BP}}$ given by Eq. (2). As would be expected, the solution (3) transforms to the Bouissou-Pelcé wave number in the limit of small growth velocity V.

The solvability condition previously derived and used by Pelcé, Bensimon, and Bouissou [4,5,8] as the vanishing of an oscillating integral is given by

$$\int_{-\infty}^{+\infty} dl\; G[X_0(l)]\exp\left[i\int_0^l k_m(l')dl'\right] = 0. \tag{4}$$

Here G is the curvature operator, $k_m(l)$ is the function of the local nonzero marginal mode of the conjugate dispersion equation for the perturbations, and $X_0(l)$ represents a continuum of solutions from which the function $k_m(l)$ is deduced. Substituting now Eq. (3) into Eq. (4), one can rewrite the solvability condition as

$$\int_{-\infty}^{+\infty} d\eta\, G[X_0(\eta)]\exp\left\{-i\int_0^\eta\left[\frac{P_g}{2}(1-i\eta) - \sqrt{\frac{(1+i\eta)(1+\eta^2)^{5/2} + i\alpha\eta B(\eta)}{\sigma^* B(\eta)}}\right]d\eta\right\} = 0, \tag{5}$$

where

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

$$B(\eta) = (1+\eta^2)^2(1-\beta) + 8\beta\eta^2, \quad \alpha = \frac{aUd_0}{4\rho V}.$$

Equation (5) corresponds to the result obtained by Bouissou and Pelcé for small growth Péclet numbers [see Eq. (39) in Ref. [8]].

The solvability integral (5) can be calculated in analogy with the theory developed by Bouissou and Pelcé [8], where two dominant contributions to the integral (5) exist: the contribution from the loop and the contribution from the stationary phase points. The first of them should be calculated between a distance $\sim i(1-\sqrt{2\beta}\tau^{2/7})$ (which is a splitting distance of the stationary phase points, $\tau = 2^{-5/4}\beta^{-3/4}\alpha$) at the intersection of the steepest descent path and the real axis and $\eta = i(1-\sqrt{2\beta})$. As a result, the first contribution gives an oscillating factor to the exponentially small value of the integral which behaves as

$$\cos\left[\frac{A_1\beta^{7/8}}{\sqrt{\sigma^*}}(1 + B_1\tau^{11/14}) - P_g\sqrt{2\beta}(1-\tau^{2/7})\right].$$

Each stationary phase point contributes by a term with an oscillating part of the form

$$\cos\left[\frac{A_2\beta^{7/8}}{\sqrt{\sigma^*}}(1 + B_2\tau^{11/14}) - P_g\sqrt{2\beta}\tau^{2/7}\right].$$

Here A_1, A_2, B_1, and B_2 are constants, which transform to corresponding expressions found in the case of small growth Péclet numbers [8].

The cancellation of the sum of these contributions gives the following selection criterion:

$$\sigma^* = \frac{\sigma_0\beta^{7/4}}{(1+a_1\sqrt{\beta}P_g)^2}\left[1 + b\left(\frac{\alpha}{\beta^{3/4}}\right)^{11/14}\right]^{-1}, \tag{6}$$

where σ_0, b, and a_1 are constants. With $a_1\sqrt{\beta}P_g \ll 1$, Eq. (6) transforms to the selection criterion in the limit of small Péclet numbers [8].

Substituting

$$\sigma^* = \frac{2d_0D_T}{\rho^2 V} = \frac{d_0 V}{2D_T P_g^2}$$

into the left-hand side of Eq. (6), the selection criterion for the thermal problem with convection (expressed in terms of the dendrite tip velocity V, the growth Péclet number P_g, and the anisotropy parameter β) reads

$$V = \frac{D_T\beta^{3/4}}{d_0}\frac{2\sigma_0\beta P_g^2}{(1+a_1\sqrt{\beta}P_g)^2}\left[1 + b\left(\frac{\alpha}{\beta^{3/4}}\right)^{11/14}\right]^{-1}. \tag{7}$$

Expression (7) completely corresponds to the previous theory developed by Brener and Mel'nikov [15] in the absence of external flow ($U = 0$ and $\alpha = 0$), which predicts the dendrite velocity at arbitrary Péclet numbers as

$$V \propto \frac{D_T\beta^{3/4}}{d_0} f(\sqrt{\beta}P_g).$$

Considering now the case of large Péclet numbers ($a_1\sqrt{\beta}P_g \gg 1$) in the absence of convection ($U = 0$), Eq. (7) predicts the following relation:

$$V = \frac{D_T\beta^{3/4}}{d_0}\frac{2\sigma_0}{a_1^2}, \tag{8}$$

where $V \sim \beta^{3/4}$. The limiting case (8) was studied earlier by Langer and Hong [13] and Barbieri [14]. To obtain the unknown constant a_1 in Eq. (8) one can use the asymptotic formula (19) of Brener and Mel'nikov [15], which, in the limit of large Péclet numbers, gives

$$V = \frac{D_T \beta^{3/4}}{d_0} \frac{7}{4} \left(\frac{56}{3}\right)^{3/4}. \tag{9}$$

Combining Eqs. (8) and (9), one can easily get

$$a_1 = \left(\frac{8\sigma_0}{7}\right)^{1/2} \left(\frac{3}{56}\right)^{3/8} \approx 0.381 \sigma_0^{1/2}. \tag{10}$$

An interesting example can be formulated for the growth of a so-called "chemical dendrite,"which is considered as a dendrite growing in a binary system with convection at a constant temperature. The concentration problem for a chemical dendrite can be well formulated as a one-sided model in which the diffusion coefficient is negligible in the solid phase. Therefore, multiplying the result by a scale factor of 2, we rewrite the selection criterion (6) as

$$\sigma^*_{\mathrm{CD}} \equiv \frac{2 d_0 D_C}{\rho^2 V} = \frac{d_0 V}{2 D_C P^2_{\mathrm{CD}}} = \frac{\sigma_0 \beta^{7/4}}{(1 + a_2 \sqrt{\beta} P_{\mathrm{CD}})^2} \times \frac{2 m C_i (1 - k_0)}{(Q/c_p)} \left[1 + b \left(\frac{\alpha_{\mathrm{CD}}}{\beta^{3/4}}\right)^{11/14}\right]^{-1}, \tag{11}$$

where D_C is the diffusion coefficient in the liquid phase, m is the liquidus slope, C_i is the concentration of impurity at the dendrite surface, k_0 is the equilibrium partition coefficient, $P_{\mathrm{CD}} = \rho V/(2D_C) = P_g D_T/D_C$, and $\alpha_{\mathrm{CD}} = a U d_{0\mathrm{CD}}/(2\rho V)$ with the rescaled capillary length,

$$d_{0\mathrm{CD}} = \frac{(Q/c_p) d_0}{2 m C_i (1 - k_0)},$$

defined for chemical dendrite. The factor $m C_i (1 - k_0) c_p/Q$ arises due to the symmetry of Stefan models for temperature and concentration [7]. Rewriting the selection criterion (11) in the limiting case of large Péclet numbers P_{CD}, we arrive at the expression ($U = 0$)

$$V = \frac{D_C \beta^{3/4}}{d_{0\mathrm{CD}}} \frac{2\sigma_0}{a_2^2}.$$

Equating this expression for V to its asymptotics [15] (factor 2 in denominator appears due to the use of the one-sided model for solute diffusion),

$$V = \frac{D_C \beta^{3/4}}{2 d_{0\mathrm{CD}}} \frac{7}{4} \left(\frac{56}{3}\right)^{3/4},$$

we determine constant a_2 in the form

$$a_2 = \left(\frac{16\sigma_0}{7}\right)^{1/2} \left(\frac{3}{56}\right)^{3/8} \approx 0.505 \sigma_0^{1/2}. \tag{12}$$

Now generalizing expressions (6) and (11), the selection criterion for the simultaneously solved thermal and concentration problem with convection at small anisotropy β and arbitrary growth Péclet numbers looks like

$$\sigma^* = \frac{\sigma_0 \beta^{7/4}}{1 + b(\bar{\alpha}\beta^{-3/4})^{11/14}} \left[\frac{1}{(1 + a_1 \sqrt{\beta} P_g)^2} + \frac{1}{(1 + a_2 \sqrt{\beta} P_{\mathrm{CD}})^2} \frac{2 m C_i (1 - k_0) D_T}{(Q/c_p) D_C}\right], \tag{13}$$

where constants a_1 and a_2 are determined by Eqs. (10) and (12), respectively, and

$$\bar{\alpha} = \frac{a U d_0}{4 \rho V P} + \frac{a U d_0 D_T}{2 \rho V P D_C}, \quad P = 1 + \frac{2 m C_i (1 - k_0) D_T}{(Q/c_p) D_C}.$$

Note that the presence of factor P in the denominator of $\bar{\alpha}$ is caused by renormalization of the scaling factor σ^* in the solvability condition (4). The two constants σ_0 and b entering in Eq. (13) can be found from the asymptotic analysis, experimental data [18] or the phase-field simulations [19,20].

Several limiting cases following from Eq. (13) can be outlined. At small growth Péclet numbers ($a_1 \sqrt{\beta} P_g \ll 1$ and $a_2 \sqrt{\beta} P_{\mathrm{CD}} \ll 1$) and in the absence of convection ($U = 0$ and $\alpha = 0$) expression (13) transforms to the criterion earlier obtained by Ben Amar and Pelcé [7]. The analogues limiting case with convection was studied in Ref. [21] for a chemically diluted system with the far-field concentration C_∞ in the liquid phase and leading to the corresponding σ^* from Eq. (13). Interpolation for the dendrite growing in a stagnant system at small and large Péclet numbers was done by Müller-Krumbhaar with coauthors [16]. Equation (13) tends to their expression (3) in the limit of large velocities V when $U = 0$. Thus the selection criterion (13) generalizes previously obtained results on dendritic growth with anisotropy of surface energy.

To illustrate the effect of large growth Péclet numbers on the scaling factor σ^* one needs to know the concentration C_i at the surface of the dendrite entering in the criterion (13). The concentration is determined from the steady-state solution for two-dimensional and three-dimensional geometries [8,17,21]. More specifically, for a two-dimensional geometry, a part of this solution required for the solute concentration C_i is given by [21]

$$C_i = \frac{C_\infty}{1 - (1 - k_0) \exp(P_0 D_T/D_C) P_g I_C(\infty) D_T/D_C},$$

$$I_C(\eta) = \int_1^\eta \exp\left[P_f \frac{D_T}{D_C} \int_1^{\eta'} \frac{g(\eta'')}{\sqrt{\eta''}} d\eta'' - \frac{D_T}{D_C} P_0 \eta'\right] \frac{d\eta'}{\sqrt{\eta'}},$$

$$g(\eta) = \frac{\sqrt{2/(\pi\,\mathrm{Re})}}{\mathrm{erfc}\sqrt{\mathrm{Re}/2}} \left[\exp\left(-\frac{\mathrm{Re}}{2}\right) - \exp\left(-\frac{\mathrm{Re}\eta}{2}\right)\right] + \sqrt{\eta} \frac{\mathrm{erfc}\sqrt{\mathrm{Re}\eta/2}}{\mathrm{erfc}\sqrt{\mathrm{Re}/2}}, \quad P_0 = P_g + P_f,$$

and $P_f = \rho U/(2D_T)$ is the flow Péclet number. Substituting C_i into the right-hand side of Eq. (13) we illustrate in Fig. 1 the selection criterion (13) obtained for arbitrary growth Péclet numbers and its limiting case valid for low values of P_g [8,23]:

$$\sigma^* = \frac{\sigma_0 \beta^{7/4} P}{1 + b(\bar{\alpha}\beta^{-3/4})^{11/14}}. \tag{14}$$

It is seen that the two curves plotted in Fig. 1 practically coincide at the small values of the growth Péclet number. With

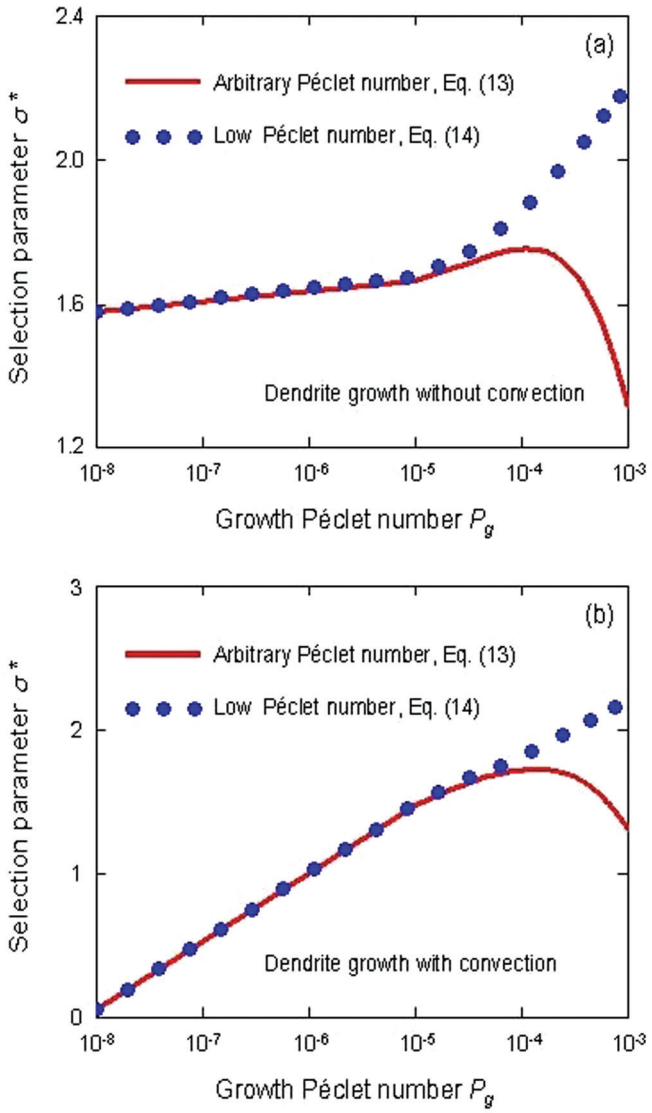

FIG. 1. (Color online) Scaling ratio $\sigma^* = 2d_0 D_T/(\rho^2 V)$ as a function of the growth Péclet number. Parameters $\sigma_0 = 2.1$ and $b = 10$ of the selection theory are estimated using the phase-field simulations [19]. Physical parameters used for calculations are typical for metallic alloys [8,21,22]: $\rho = 10^{-5}$ m, $D_T = 5 \times 10^{-6}$ m^2 s^{-1}, $D_C = 5 \times 10^{-9}$ m^2 s^{-1}, $mC_\infty = 10$ K, $Q/c_p = 660$ K, $d_0 = 2 \times 10^{-9}$ m, $k_0 = 0.6$, $\beta = 0.195$, (a) $P_f = 0$, Re $= 0$, and (b) $P_f = 10^{-4}$, Re $= 10^{-3}$.

$P_g \geqslant 10^{-5} - 10^{-4}$ the difference between these functions becomes significant. Moreover, in the case of high Péclet numbers these curves behave differently: the presently obtained selection criterion (13) decreases with the increase of P_g, contrary to predictions of the previously obtained criterion (14). Such difference in predictions is explained by the presence of quadratic terms ($\sim P_g^2$ at high Péclet numbers, which are already not negligible in comparison with unity) in denominators of Eq. (13). Finally, the influence of convective flow is clearly visible in Fig. 1 with the growth Péclet number $P_g \leqslant 10^{-5}$, where the selection criterion σ^* decreases for the growth with convection [compare figures (a) and (b) in Fig. 1 at small values of P_g]. This shows that the dendritic tip grows more rapidly in

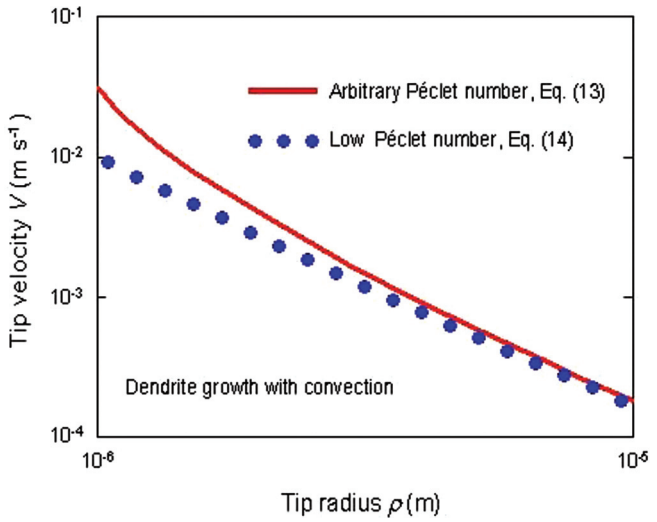

FIG. 2. (Color online) Dendritic growth velocity as a function of tip radius. Parameters correspond to Fig. 1 and, additionally, $\nu = 10^{-7}$ m^2 s^{-1} and $U = 10^{-3}$ m s^{-1}.

the presence of convective flow than in stagnant liquid (see also results from Ref. [24] on the influence of forced convection of viscous fluid on a stable mode of growing dendrite).

Substitution $\sigma^* = 2d_0 D_T/(\rho^2 V)$ into the left-hand sides of selection criteria (13) and (14) leads to direct dependencies between the dendrite velocity and tip radius as is shown in Fig. 2. An important point is that the tip velocity V obtained for arbitrary Péclet numbers sufficiently differs from its low Péclet number limit in the case of high growth rates when $\rho \sim 10^{-6}$ m. Our calculations clearly demonstrate that V selected in accordance with the presently obtained criterion (13) is approximately three times greater than V obtained from expression (14) at $\rho \sim 10^{-6}$ m (see Fig. 2). Such essential difference in the growth velocity might be verified in natural or computational experiments.

As a final note, the solvability theory is able to predict the criterion for the stable mode of dendritic growth with convection at arbitrary Péclet numbers. The obtained criterion of stability generalizes earlier known results for the cases of stable growth mode with anisotropy of surface energy and with the negligible atomic kinetics on the phase interface. The demonstrated numeric examples explicitly show an essential difference in the prediction of dendrite velocity using criteria following from the theory which takes into account small Péclet numbers and from the theory which takes the arbitrary value Péclet numbers into account.

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