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Generalised 'Jackson–Hunt' model for eutectic solidification at low and large Peclet numbers and any binary eutectic phase diagram

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Abstract

Based on the generalised 'Jackson–Hunt' model from Donaghey and Tiller for eutectic solidification of binary alloys, a relation between growth velocity, lamellar spacing and interface undercooling for any binary eutectic phase diagrams and any Peclet number is derived. This relation is then used to propose a generalised scaling law for eutectic growth. The predictions of this generalised scaling law are discussed for extremely different eutectic phase diagrams and compared with predictions made by the TMK-model for symmetrical phase diagrams.

Keywords: Eutectic solidification; Diffusion; Multi-phase alloys

1. Introduction

The most well-known theory for the eutectic solidification is the classical work by Jackson and Hunt (JH model) [1]. Fig. 1 is a schematic diagram of a lamellar eutectic structure which forms under steady-state directional solidification conditions. To estimate the diffusion field ahead of such planar eutectic interface solidifying with a constant growth rate V, Jackson and Hunt suggested a Fourier series approach of the following form

$$C(x,z) = C_\infty + B_0 e^{-2z/l_C} + \sum_{n=1}^{\infty} B_n e^{-2F_n z/l_C} \cos\left(2n\pi \frac{x}{\lambda_E}\right) \quad (1a)$$

$$\text{with } F_n := \frac{1}{2}\left[1 + \sqrt{1 + \left(\frac{2n\pi}{P_E}\right)^2}\right] \quad \text{for } n = 1, \ldots, \infty \text{ and } P_E \neq 0. \quad (1b)$$

Here, C_∞ is the solute concentration in liquid far from the interface;[1] $l_c := 2D/V$ the diffusion length; D the diffusion coefficient in the liquid; $P_E := \lambda_E/l_c$ the Peclet number; and λ_E the interlamellar spacing. B_i with $i = 0, \ldots, \infty$ are constants which have to be determined by further conditions. The frame of reference is considered to move at constant velocity, V, in the z-direction with the planar eutectic front at $z = 0$. Eq. (1) fulfils both, the solute conservation differential equation and the far field condition. Corresponding to the general properties of Fourier coefficients Jackson and Hunt estimated the B_n coefficients by multiplying Eq. (1a) and the two solute flux balances with $\cos(2i\pi x/\lambda_E)$ and then integrating over an interlamellar spacing. They further assume that the differences in liquid and solid compositions at the α/liquid and the β/liquid interfaces, $\Delta C_{L,\alpha}$ and $\Delta C_{L,\beta}$, are independent of x and equal to the corresponding thermodynamic equilibrium quantities at the eutectic composition: $\Delta C_{L,\alpha} = \Delta C_\alpha^E$ and $\Delta C_{L,\beta} = \Delta C_\beta^E$. As further restriction they assume $P_E \ll 1$.

With the estimated B_n coefficients, Jackson and Hunt determined the average compositions, $\bar{C}_{L,\alpha}$ and $\bar{C}_{L,\beta}$, in the liquid at the interface in front of the α phase and β phase. With respect to the average constitutional and capillary undercoolings they got the following expressions for the relation between average undercooling at the front, ΔT_E, interlamellar spacing, λ_E and growth velocity, V

$$\Delta T_E = K_1 V \lambda_E P(f_\alpha) + \frac{K_2}{\lambda_E} \quad (2a)$$

$$\text{with } K_1 := \Delta C_E \frac{\bar{m}}{D}\left(\frac{1}{f_\alpha} + \frac{1}{f_\beta}\right) \quad \text{and } K_2 := 2\bar{m}\left[\frac{\Gamma_\alpha \sin\theta_\alpha}{f_\alpha |m_\alpha|} + \frac{\Gamma_\beta \sin\theta_\beta}{f_\beta |m_\beta|}\right] \quad (2b)$$

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[1] Here, $0 < C_\infty < 1$ is taken as dimensionless parameter.

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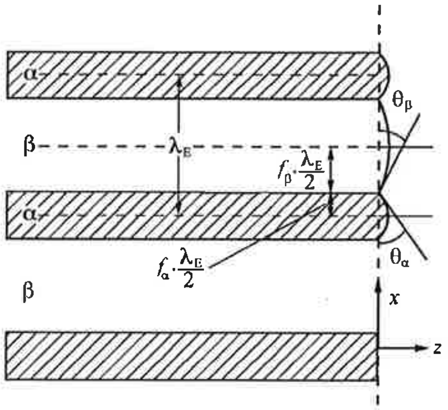

Fig. 1. A schematic diagram of a lamellar structure.

plus $\bar{m} := \frac{|m_\alpha| \cdot |m_\beta|}{|m_\alpha| + |m_\beta|}$ and $P(f_\alpha) := \sum_{n=1}^{\infty} \frac{\sin^2(n\pi f_\alpha)}{(n\pi)^3}$ (2c)

in which m_α and m_β are the slopes of the liquidus at T_E (defined so that both are positive), f_α and f_β the relative phase amount, Γ_α and Γ_β the corresponding Gibbs–Thomson-coefficient, and θ_α and θ_β the triple point angels. $\Delta C_E := \Delta C_\alpha^E + \Delta C_\beta^E$ is the difference of the maximal solubilities of the α phase and β phase at T_E.

Using the minimum undercooling principle, Jackson and Hunt obtained the following relation between ΔT_E, λ_E and V

$$\lambda_E^2 V = \frac{K_2}{K_1} \frac{1}{P(f_\alpha)} \quad (3)$$

$$\lambda_E \Delta T_E = 2K_2 \quad (4)$$

Eq. (3) has become the well known scaling law for eutectic growth.

Already 2 years after the classical work of Jackson and Hunt, Donaghey and Tiller published a paper where they could relax two simplifications made in the JH-model (DT-model) [2]. Donaghey and Tiller applied the correct approach for $\Delta C_{L,\alpha}$ and $\Delta C_{L,\beta}$, that's $\Delta C_{L,\alpha}(x) = (1-k_\alpha)C(x,0)$ and $\Delta C_{L,\beta}(x) = (1-k_\beta)(1-C(x,0))$, and so they explicitly allow $\Delta C_{L,\alpha}$ and $\Delta C_{L,\beta}$ to be functions of x.[2] In addition, they did not restrict their approach to $P_E \ll 1$. Donaghey and Tiller got the following infinite linear system of equations for the determination of the B_n coefficients:

$$(a_0 - 1)B_0 + \sum_{k=1}^{\infty} a_k B_k = b_0 - C_\infty a_0, \quad (5a)$$

$$a_n B_0 + \tfrac{1}{2}(a_0 + a_{2n} - F_n)B_n + \sum_{k=1,\,k\neq n}^{\infty} \tfrac{1}{2}(a_{n+k} + a_{n-k})B_k = b_n - C_\infty a_n \quad n \neq 0, \quad (5b)$$

with $a_0 := f_\alpha(1-k_\alpha) + f_\beta(1-k_\beta)$ and $b_0 := (1-k_\beta) f_\beta$ (5c)

plus $a_n := (k_\beta - k_\alpha)\frac{\sin(n\pi f_\alpha)}{n\pi}$ and

$$b_n := (k_\beta - 1)\frac{\sin(n\pi f_\alpha)}{n\pi} \quad n \neq 0. \quad (5d)$$

As already mentioned in [3], Donaghey and Tiller had a small error in their result. Thus, the above given coefficients of the infinite linear system of equations are slightly different from those given in [2]. Note that Eqs. (5a) and (5b) are symmetrical with respect to a phase exchange, as with the change of α into β and vice versa, C_∞ have to be replaced by $(1-C_\infty)$. Donaghey and Tiller did not investigate whether the infinite system of equations posses finite subsystems of dimension m for which the solution converges with increasing m. They also did not give a general relation between ΔT_E, V, and λ_E.

Trivedi et al. [3] investigated the solution of the infinite linear system of equations, Eqs. (5a)–(5d), for two types of phase diagrams (TMK-model). These are (1) the metastable phase diagram is cigar-shaped so that below the eutectic temperature the solidus and the liquidus are parallel; and (2) the distribution coefficient is an arbitrary constant, but $k_\alpha = k_\beta$. They showed that for these two cases a solution of the system of equations Eqs. (5a)–(5d) exists and that a relation between ΔT_E, V and λ_E analogue to Eq. (2a) can be derived. They found that the scaling law, Eq. (3), formally still holds, even though for large Peclet numbers $\lambda_E^2 V$ turned out to be not a constant.

In the present paper, we give the ΔT_E-V-λ_E relation for the general case $k_\alpha \neq k_\beta$ and any Peclet number, together with the general scaling law for eutectic growth. For extremely different parameter sets, we found solutions for the infinite linear system of equations, Eqs. (5a)–(5d). Especially for a highly unsymmetrical phase diagram we discuss the solutions and compare them with the results from the TMK-model for symmetrical phase diagrams.

2. Theory

By using the Fourier series approach, Eqs. (1a) and (1b), the average composition in the liquid at the interface in front of the α phase and the β phase can be obtained without knowing the B_n coefficients explicitly:

$$\bar{C}_{L,\alpha} = C_\infty + B_0 + \frac{P_E \Delta C_0}{f_\alpha} P(f_\alpha, P_E, k_\alpha, k_\beta, C_\infty) \quad (6a)$$

$$\bar{C}_{L,\beta} = C_\infty + B_0 - \frac{P_E \Delta C_0}{f_\beta} P(f_\alpha, P_E, k_\alpha, k_\beta, C_\infty) \quad (6b)$$

where the generalised P-function is introduced

$$P(f_\alpha, P_E, k_\alpha, k_\alpha, C_\infty) := \frac{1}{P_E \Delta C_0} \sum_{n=1}^{\infty} \left[B_n \frac{\sin(n\pi f_\alpha)}{n\pi} \right]. \quad (6c)$$

[2] With temperature depending distribution coefficient k_α and k_β have to be taken at the temperature of the eutectic front $T* = T_E - \Delta T_E$.

The prefactor in the definition of the P-function is chosen so that the results from the present study can be directly compared with the JH- and the TMK-model. $\Delta C_0 := (1 - k_\alpha) + (1 - k_\beta)$ can differ from the difference of the maximal solubilities of the α phase and β phase at T_E as the front might be significantly undercooled. The dependence of the P-function from f_α, P_E, k_α, k_β and C_∞ can be deduced from the coefficients of the infinite linear system of equations, Eqs. (5a)–(5d). Note that from Eq. (5a) it can be derived that

$$B_0 = B_0' - \frac{P_E(k_\beta - k_\alpha)}{a_0 - 1} P(f_\alpha, P_E, k_\alpha, k_\beta, C_\infty). \tag{7}$$

with $B_0' := (b_0 - C_\infty a_0)/(a_0 - 1)$. Thus, $\bar{C}_{L,\alpha}$ and $\bar{C}_{L,\beta}$, are linear functions of the P-function. For $k_\alpha = k_\beta = k$ Eq. (7) becomes Eq. (15) in [3] and $B_0' = B_0$. For this case the P-function defined in Eq. (6c) turns into Eq. (16) in [3].

By an analogue procedure as the one described by Jackson and Hunt in [1] the relation between average undercooling at the front, ΔT_E, interlamellar spacing, λ_E and growth velocity, V, can be obtained without knowing the B_n coefficients explicitly. This generalised relation is

$$\overline{\Delta T_E} = \tilde{K}_1 V \lambda_E P(f_\alpha, P_E, k_\alpha, k_\beta, C_\infty) + \frac{K_2}{\lambda_E}. \tag{8}$$

The constants $\tilde{K}_1$ is now defined as $\tilde{K}_1 := K_1(\Delta C_0/\Delta C_E)$. K_2 is defined in Eq. (2b). Note that this ΔT_E-V-λ_E relation is equivalent to Eq. (2a) with the exception that the P-function is defined without knowing the B_n coefficients.

Applying the minimum undercooling principle, we get from Eq. (8) (again without knowing the B_n coefficients), the generalised scaling law for eutectic growth

$$\lambda_E^2 V = \frac{K_2}{K_1'} \frac{1}{P + \lambda_E \partial P/\partial \lambda_E}. \tag{9}$$

The undercooling at the eutectic interface is obtained as

$$\lambda_E \Delta T_E = K_2 \left(1 + \frac{P}{P + \lambda_E \partial P/\partial \lambda_E}\right). \tag{10}$$

With $\lambda_E \partial P/\partial \lambda_E = P_E \partial P/\partial P_E$, it is obvious that the right hand side of Eqs. (9) and (10) depends on f_α, P_E, k_α, k_β and

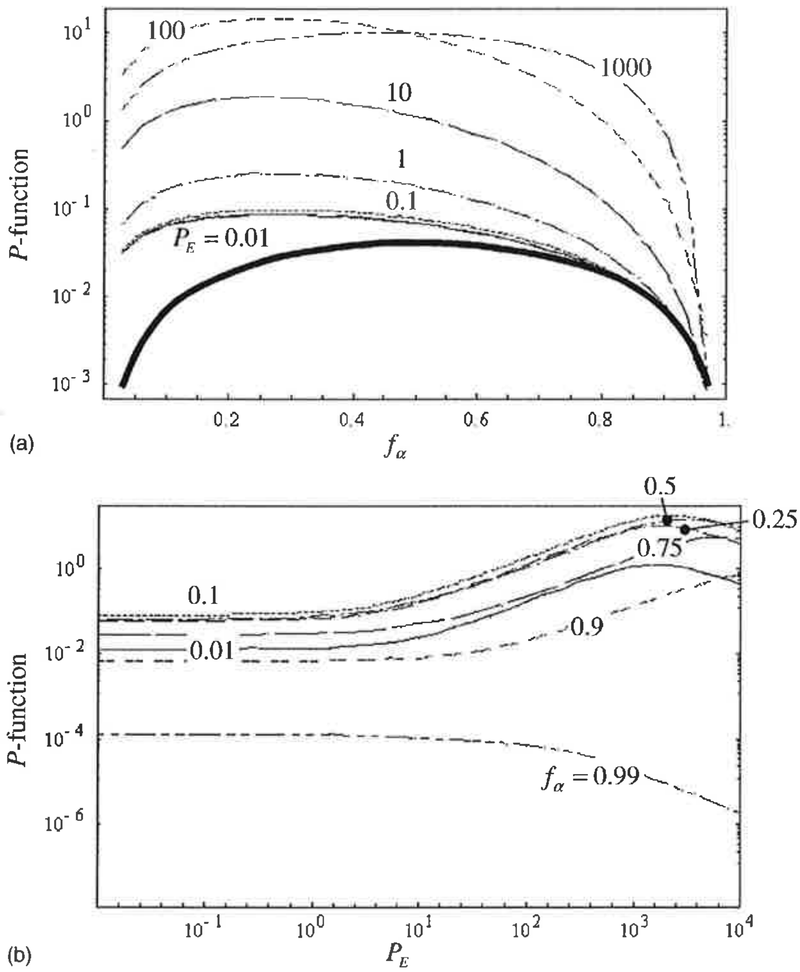

Fig. 2. The variation in the P-function: (a) with f_α for different values of P_E, and (b) with P_E for different values of f_α. The distribution coefficients were chosen extremely different with $k_\alpha = 10^{-6}$ and $k_\beta = 0.999999$. C_∞ was set to 0.5. The bold black curve in (a) represents the prediction of the Jackson–Hunt model.

C_∞. Eqs. (9) and (10) have been derived earlier by Trivedi et al. for the two considered types of phase diagrams mentioned above (see Eqs. (12) and (14) in [3]). However, we have not yet specified the unknown B_n coefficients, whereas in [3] these coefficients were explicitly determined.

To solve the infinite linear system of equations, Eqs. (5a)–(5d), we have used MATHEMATICA (version 4.1) as numerical tool. For $0 < f_\alpha < 1$, $10^{-2} < P_E < 10^4$, $C_\infty = 0.1$ and 0.5 as well as $k_\alpha = k_\beta = 10^{-6}$ and 0.999999 and $k_\alpha = 10^{-6}$ and $k_\beta = 0.999999$ and vice versa, we have tested the convergence of the P-function calculated from the solutions of finite subsystems of dimension m. For all investigated parameter combinations the P-function converged fast: by solving a subsystem with a dimension of only $m = 20$ the accuracy for the P-function is better than 0.1%.

At the end of the theory section, we would like to mentioned that the solute conservation equation, $f_\alpha \bar{C}_{L,\alpha} + f_\beta \bar{C}_{L,\beta} = C_\infty$, cannot be used to estimate f_α as it has already been used in the theory. This is true for the JH-, the TMK- and the presented general approach. As suggested by Kassner and Misbah [4] f_α should be implicitly estimated from the condition of an isothermal interface, $\Delta T_\alpha = \Delta T_\beta$ which gives

$$(|m_\alpha| + |m_\beta|)[C_\infty + B_0 - C_E] + P_E P(f_\alpha, P_E, k_\alpha, k_\beta, C_\infty) \times \left(\frac{|m_\alpha|}{f_\alpha} - \frac{|m_\beta|}{f_\beta}\right) = \Gamma_\beta \left(\frac{\sin\theta_\beta}{f_\beta(\lambda_E/2)}\right) - \Gamma_\alpha \left(\frac{\sin\theta_\alpha}{f_\alpha(\lambda_E/2)}\right). \quad (11)$$

3. Results and discussion

For $k_\alpha = 0.9999999$ and $k_\beta = 10^{-6}$ the variation in the P-function with f_α for different values of P_E is shown in Fig. 2a and the variation in the P-function with P_E for different values of f_α is shown in Fig. 2b The corresponding figures for $k_\alpha = k_\beta \to 0$ can be found in Fig. 4a and b in [3] and for $k_\alpha = k_\beta \to 1$ in Fig. 3a and b in [3]. For a symmetrical case ($k_\alpha = k_\beta = k$) and small P_E the variation in the P-function with f_α is independent of k and equal to the

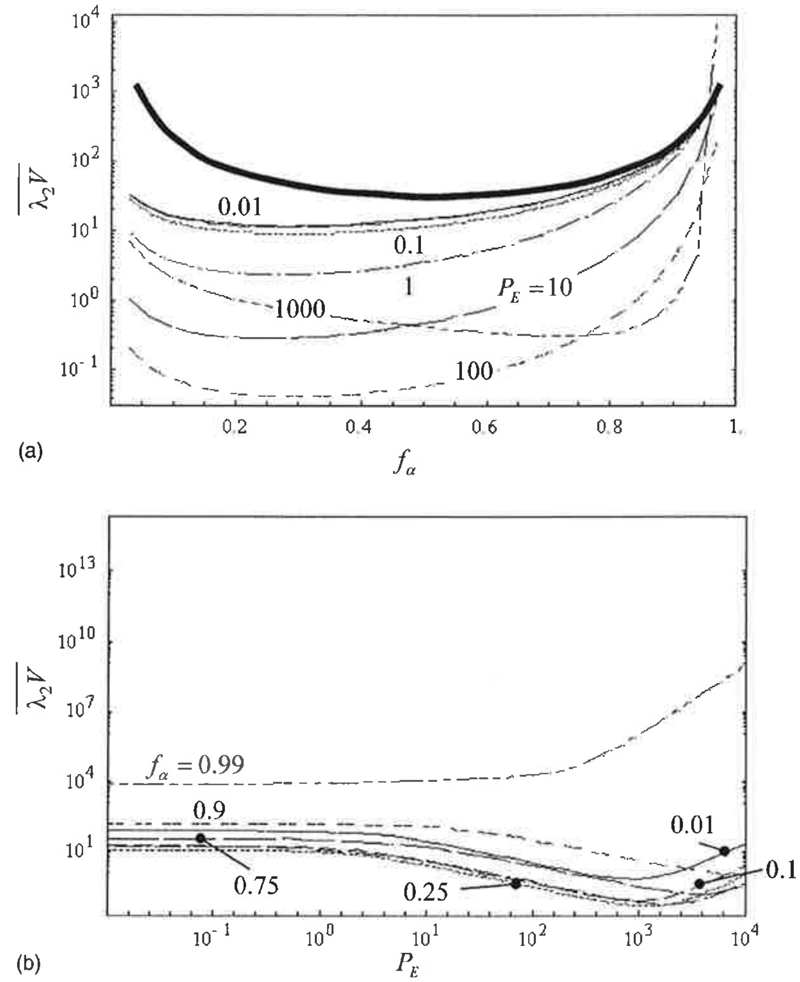

Fig. 3. The variation in $\overline{\lambda_E^2 V}$: (a) with f_α for different values of P_E, and (b) with P_E for different values of f_α. The distribution coefficients were chosen extremely different with $k_\alpha = 10^{-6}$ and $k_\beta = 0.999999$. C_∞ was set to 0.5. The bold black curve in (a) represents the prediction of the Jackson–Hunt model.

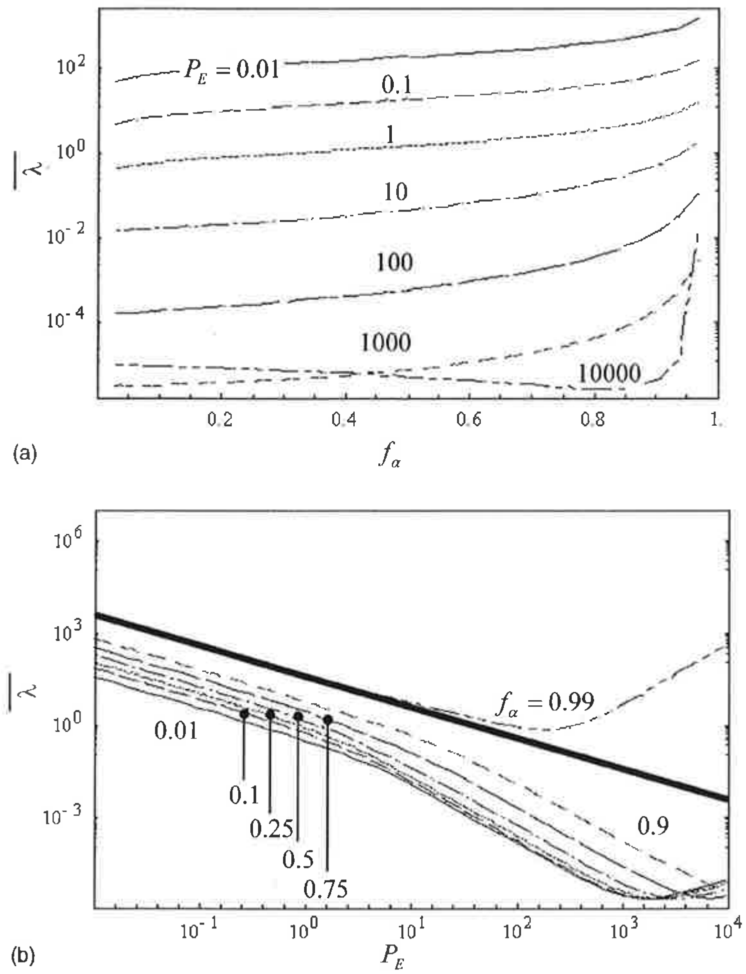

Fig. 4. The variation in $\bar{\lambda}_E$: (a) with f_α for different values of P_E, and (b) with P_E for different values of f_α. The distribution coefficients were chosen extremely different with $k_\alpha = 10^{-6}$ and $k_\beta = 0.999999$. C_∞ was set to 0.5. The bold black curve in (b) represents the prediction of the Jackson–Hunt model for $f_\alpha = 0.99$.

P-function of the Jackson–Hunt approach [3]. This curve is also shown in Fig. 2a (bold black). The comparison shows that for $k_\alpha \neq k_\beta$ the *P*-function becomes unsymmetrical with respect to f_α even for small P_E. Therefore, it is generally not described by the JH-model. For the symmetrical case with small *k* the *P*-function increases with increasing P_E (Fig. 4b in [3]), for large *k* the *P*-function decreases with increasing P_E (Fig. 3b in [3]). As shown in Fig. 2, in the unsymmetrical case the *P*-function increases with increasing P_E for most value of f_α, only for $f_\alpha = 0.99$ it decreases. From that finding it is obvious that in the unsymmetrical case, the smaller of the two distribution coefficients dominates the eutectic growth process. In the present case ($k_\alpha = 0.9999999$, $k_\beta = 10^{-6}$) the α-lamellae grow without any significant solute redistribution. On the other hand, the solute redistribution ahead of the β-lamellae is large and further growth of these lamellae can only occur if diffusion reduces the high solute concentration in front of the lamella. Therefore, diffusion of the element with the smaller *k* is more important than diffusion of the other species.

These two findings, that for $k_\alpha \neq k_\beta$ the prediction of the JH-model may become wrong even for small P_E and that the smaller of the two *k*'s dominates the growth process, can also be seen from the variation in $\overline{\lambda_E^2 V}$ with f_α for different values of P_E as shown in Fig. 3a and the variation in $\overline{\lambda_E^2 V}$ with P_E for different values of f_α as shown in Fig. 3b. The corresponding variation in $\bar{\lambda}_E$ and $\overline{\Delta T}_E$ is resented in Fig. 4a and b and Fig. 5a and b. The dimensionless correspondence of $\lambda_E^2 V$ and the dimensionless spacing $\bar{\lambda}_E$ and dimensionless undercooling $\overline{\Delta T}_E$ are defined by

$$\overline{\lambda_E^2 V} := \lambda_E^2 V \frac{K_1'}{K_2} = \frac{1}{P + \lambda_E \partial P / \partial \lambda_E} \tag{12}$$

$$\bar{\lambda}_E := \lambda_E \frac{2DK_1'}{K_2} = \frac{1}{P_E} \frac{1}{P + \lambda_E \partial P / \partial \lambda_E} \tag{13}$$

$$\overline{\Delta T}_E := \Delta T_E \frac{2DK_1'}{f_\alpha f_\beta} = \frac{2P_E}{f_\alpha f_\beta} \left(2P + \lambda_E \frac{\partial P}{\partial \lambda_E} \right). \tag{14}$$

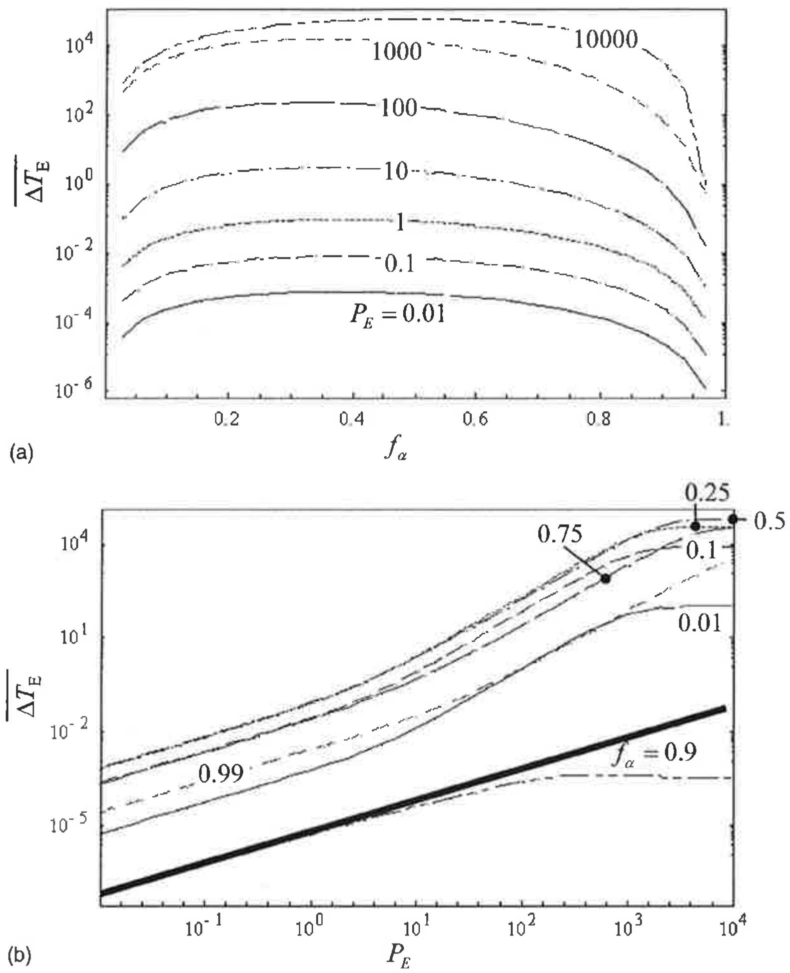

Fig. 5. The variation in $\overline{\Delta T_E}$: (a) with f_α for different values of PE, and (b) with PE for different values of f_α. The distribution coefficients were chosen extremely different with $k_\alpha = 10^{-6}$ and $k_\beta = 0.999999$. C_∞ was set to 0.5. The bold black curve in (b) represents the prediction of the Jackson–Hunt model for $f_\alpha = 0.9$.

The motivation for these definitions are from Eqs. (9) and (10) and Eqs. (20) and (21) in [3]. In order to estimate the right hand side of Eqs. (12)–(14), we have first calculated $P + \lambda_E \partial P/\partial \lambda_E = P + P_E \partial P/\partial P_E$ by using the approximation $P + P_E \Delta P/\Delta P_E$. Unfortunately, in many cases this approximation tends to zero as soon as $\Delta P/\Delta P_E$ becomes negative. As a result the quantity $(P + P_E \Delta P/\Delta P_E)^{-1}$ often reveals a singularity at $P_E > 1$, even for the symmetrical TMK case. With this approach we were not able to reproduce Fig. 8 from [3]. We believe that numerical inaccuracies are the reason for this problem. We decided to use the following approach. From the definition of the P-function, Eq. (6c), we got

$$\frac{\partial P}{\partial P_E} = -\frac{P}{P_E} + \frac{1}{P_E \Delta C_0} \sum_{n=1}^{\infty} \left[\frac{\partial B_n}{\partial P_E} \frac{\sin(n\pi f_\alpha)}{n\pi} \right] \qquad (15)$$

so that the following expression is obtained

$$P + \lambda_E \frac{\partial P}{\partial \lambda_E} = \frac{1}{\Delta C_0} \sum_{n=1}^{\infty} \left[\frac{\partial B_n}{\partial P_E} \frac{\sin(n\pi f_\alpha)}{n\pi} \right]. \qquad (16)$$

This equation was then used by replacing the derivative by the ratio of differences. So we have solved the system of equation, Eqs. (5a)–(5d), twice for two adjacent Peclet numbers and apply Eq. (16).

As already predicted by the symmetrical approach of the TMK-model, the eutectic scaling law $\lambda_E^2 V =$ const. does not hold at large P_E. Opposite to the behaviour of the P-function, $\lambda_E^2 V$ (or more exactly $\overline{\lambda_E^2 V}$) decreases with increasing P_E for $k_\alpha = k_\beta \to 0$ until a certain limit slightly above $P_E = 10^3$. For $k_\alpha = k_\beta \to 1$ $\lambda_E^2 V$ increased with increasing P_E. Here, no upper limit was found. In the present unsymmetrical case, the extreme small value of k_β leads to a decreasing characteristic of the $\overline{\lambda_E^2 V}$–P_E curves even for large amounts of f_α. A similar statement is true for the variation of $\bar{\lambda}_E$ with P_E (Fig. 4b). As the solidification interval is inverse proportional to the distribution coefficient, small k's result in large solidification intervals ΔT_S. Trivedi et al. [3] found that for $k_\alpha = k_\beta = k$ the eutectic undercooling approaches for large P_E towards the corresponding solidification interval. For the presented unsymmetrical case it becomes obvious from Fig. 5, that for large P_E, ΔT_E approaches the solidification interval of the element with the smaller k. In this regime the eutectic growth should become a one-phase growth only and the corresponding phase amount should approach 1.

4. Conclusions

The outcome of this work can be summarised by the following conclusions:

- To get a solution of the general planar eutectic problem ($k_\alpha \neq k_\beta$, any P_E) an infinite system of equations has to be solved.
- For all investigated parameter combinations the solution of finite subsystems converged rapidly.
- The TMK eutectic scaling laws are valid even without knowing the explicit solution of the infinite system of equations.
- Solutions for $k_\alpha \neq k_\beta$ are unsymmetrical (even if $P_E \ll 1$). They are dominated by the solute with the smaller k.
- f_α should be estimated from the implicit equation $\Delta T_\alpha = \Delta T_\beta$ rather than from the solute conservation equation.

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