Theoretical and numerical investigation of diffusive instabilities in multi-component alloys

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ABSTRACT

Diffusive instabilities of the Mullins-Sekerka type are one of the principal mechanisms through which microstructures form during solidification. In this study, we perform a linear stability analysis for the perturbation of a planar interface, where we derive analytical expressions to characterize the dispersion behavior in multi-component alloys under directional and isothermal solidification conditions. Subsequently, we confirm our calculations using phase-field simulations for different choices of the inter-diffusivity matrices. Thereafter, we highlight the characteristics of the dispersion curves upon change of the diffusivity matrix and the velocity. Finally, we also depict conditions for absolute stability of a planar interface under directional solidification conditions.

1. Introduction

Morphological instability of a solid-liquid interface to small perturbations is the basis for the most commonly observed solidification microstructure of dendrites. Experimentally, a planar solidification front during solidification is usually perturbed, either by random thermal fluctuations or due to interactions with insoluble impurities [1]. An unstable solidification front is characterized by amplification of such interfacial perturbations which ultimately develop into cellular or dendritic structures. Any random infinitesimal perturbation can be thought of as a linear combination of a multitude of wavelengths with different amplitudes (which are small during early stages). Thus, the stability of a solid-liquid interface to the amplification of these perturbations can be understood by investigating the growth behavior of the individual modes. Mullins and Sekerka in their classical work [2] present a linear stability analysis of an interface perturbed by any generic wavelength and provide expressions for their growth rates for a binary alloy. This allows the determination of the maximally (fastest) growing wavelength that can be approximately related to the length scales in the cellular or dendritic microstructures. Cells and dendrites, being the most commonly observed solidification structures, have been investigated theoretically [3–10], experimentally [11–18], as well as through simulations [19–24]. The theory in [2] (also confirmed by phase-field simulations in [25]) reveals that the instability length scale in a binary alloy is a function of the equilibrium compositions in the solid and the liquid, the Gibbs-Thomson coefficient, the growth velocity and the solute diffusivity.

For multicomponent systems, a linear stability analysis of a directionally solidifying ternary alloy is first reported by Coates et al. [26]. The study assumed no diffusional interaction amongst solutes, with the dispersion behavior (the amplification rates for different wavelengths of perturbations) calculated assuming a steady state behavior in the perturbed state. The correctness of this assumption is investigated by Coriell et al. [27] by solving for the time dependent problem which leads to the validation of the steady-state assumption in [26]. The effect of coupled solute diffusivities on the stability of the system to infinitesimal perturbations is studied by Hunziker [28].

In this paper, we have three principal aims. First we derive analytical expressions for the growth rates of the perturbations as a function of imposed wavelengths by performing a linear stability analysis for directional as well as isothermal solidification conditions, in a generic multi-component alloy. Here, while the problem of directional solidification is well posed in that there exists a steady-state condition (constant velocity) for the planar interface which is to be perturbed, the problem of isothermal solidification has a planar growth solution which only admits a relation where the displacement of the interface scales with the square root of time. We show that an equivalent perturbation analysis can be performed for both conditions, notwithstanding this difference in the initial states which are being perturbed. Secondly, we utilize a phase-field model based on a grand-canonical density formulation and validate our analytical calculations for both sets of solidification conditions by performing simulations for a varied set of diffusivity matrices. Thirdly, we comment on the nature of the dispersion curves in terms of the maximal growth rates and

maximally growing wavelengths and their variation with the change in the diffusion length scales. Here, we also highlight the conditions for absolute stability which is applicable for directional solidification conditions. We begin with the theoretical analysis of the growth of perturbations and thereafter describe the phase-field model, followed by the results.

2. Theory

In this section, we derive an analytical linear stability analysis of the diffusive instability of a planar interface under two conditions. In the first, a) the planar interface is driven in a temperature gradient of defined magnitude (G) with a velocity (V) and secondly, b) for solidification in a uniform undercooled melt. While in a) the velocity of the interface is imposed, in b) the system chooses a growth coefficient that linearly relates the square of the displacement of the interface with time, depending upon the alloy composition and diffusivity matrices.

2.1. Directional solidification

2.1.1. Steady state

We begin with steady-state (planar front) solidification of a K component alloy. The $K-1$ independent components have no diffusional interaction (i.e., the diffusivity matrix is diagonal; the general case is treated later) in the liquid, while there is no diffusion in the solid. The governing equation in a frame attached to the interface growing at a velocity V writes as,

$$D_{ii}\frac{\partial^2 c_i}{\partial z^2} + V\frac{\partial c_i}{\partial z} = 0,\qquad(1)$$

where c_i denotes the concentration and D_{ii} the diffusivity of the i'th component in the liquid, with $i = 1, 2, 3, \ldots, K-1$. z is the direction normal to the solid-liquid interface (located at $z=0$). Consideration of uncoupled diffusion of solutes enables us to present the following discussion in terms of a generic component i, which stands for all the components in a system.

Eq. (1) has a solution whereby the solid grows with a composition that is the same as the liquid composition. This can be easily obtained by integrating the Eq. (1) twice and respectively applying the boundary conditions at the interface,

$$Vc_{i,eq}^l(1-k_i) = -D_{ii}\frac{\partial c_i}{\partial z}\bigg|_{z=0} = -D_{ii}G_{c,i},\qquad(2)$$

which is the Stefan boundary condition at a solid-liquid interface moving with velocity V, with $G_{c,i}$ as the compositional gradient in c_i at the planar interface and k_i as the equilibrium partition coefficient corresponding to the selected tie-line; the other boundary condition being that of the equilibrium compositions at the interface,

$$c_i = c_{i,eq}^l, \quad \text{at } z = 0.\qquad(3)$$

The resultant form of the equation (after the second integration) admits a solution only when the far-field liquid composition is the same as the solid composition. Thereby, the composition of the liquid $c_{i,eq}^l$ as well as the temperature of the interface can be determined uniquely, given the starting alloy composition (far-field liquid composition). This completes the solution to Eq. (1) as given by,

$$c_i = c_{i,eq}^l + \frac{G_{c,i}D_{ii}}{V}\left[1 - \exp\left(\frac{-Vz}{D_{ii}}\right)\right].\qquad(4)$$

2.1.2. Linear stability analysis

The steady-state solidification described above is now modified by introducing a sinusoidal perturbation given by,

$$z = \Phi = \delta(t)\sin\omega x,\qquad(5)$$

with x being one of the directions parallel to the unperturbed interface (normal to z). Despite δ being a function of time (t), a stability criterion derivable from the steady state solution will not differ appreciably from that obtained by solving the time dependent problem [26,27] (quasi-stationary approximation [6]), which leads to the following governing differential equation describing a system with interfacial perturbations,

$$D_{ii}\frac{\partial^2 \tilde{c}_i}{\partial z^2} + D_{ii}\frac{\partial^2 \tilde{c}_i}{\partial x^2} + V\frac{\partial \tilde{c}_i}{\partial z} = 0,\qquad(6)$$

where the modified composition field of any generic component i, under interfacial perturbation is denoted by $\tilde{c}_i$. The form of the solution to Eq. (6) is obtained by adding a term to the steady-state solution given by Eq. (4), which represents a sinusoidal variation in the composition fields in response to the interfacial perturbation of a similar character. It must be taken into account that such an effect diminishes in magnitude with distance from the interface, leading to the following expression,

$$\tilde{c}_i = c_i + E_i\sin\omega x\exp(-k_\omega^{(i)}z) = c_{i,eq}^l + \frac{G_{c,i}D_{ii}}{V}\left[1 - \exp\left(\frac{-Vz}{D_{ii}}\right)\right]$$
$$+ E_i\sin\omega x\exp(-k_\omega^{(i)}z),\qquad(7)$$

where $k_\omega^{(i)}$ and E_i are constants. The constant $k_\omega^{(i)}$ is determined by the requirement that the composition profile given by Eq. (7) satisfies the governing Eq. (6), resulting in a quadratic equation in $k_\omega^{(i)}$, which yields,

$$k_\omega^{(i)} = \frac{V}{2D_{ii}} + \sqrt{\left(\frac{V}{2D_{ii}}\right)^2 + \omega^2}.\qquad(8)$$

The compositions in the liquid at the perturbed interface are no longer given by the equilibrium tie-lines considered during steady-state growth because of the Gibbs-Thomson correction. The composition deviations conform to the interfacial curvature, which is approximated by the second derivative of z with respect to x from Eq. (5) and can be seen to be of the same form as the perturbation itself. Thus, the composition in the liquid at the perturbed interface is given by,

$$c_{i,\Phi} = c_{i,eq}^l + b_i\delta\sin\omega x,\qquad(9)$$

where b_i is a constant. Evaluating the solution to the perturbed problem given by Eq. (7) at the perturbed interface (see Eq. (5)), we retrieve,

$$c_{i,\Phi} \approx c_{i,eq}^l + (G_{c,i}\delta + E_i)\sin\omega x,\qquad(10)$$

where we have retained terms proportional to the first order in the perturbation, Φ. Separately comparing the Fourier coefficients and the leading order constant from Eqs. (9) and (10), we derive,

$$E_i = \delta(b_i - G_{c,i}).\qquad(11)$$

Eq. (11) is only a reformulation of E_i in terms of b_i, which are still unknown. The b_i's (b_1, b_2, $\cdots$, b_{k-1}) are related to each other through the fact that each of the composition fields ($\tilde{c}_i$) satisfies the Stefan condition at the perturbed interface, moving at a velocity ($v(x)$). This implies that the same amplification factor ($\dot{\delta}/\delta$) must be obtained by considering the diffusion field of any one of the components. The expression for the Stefan condition at the perturbed interface is given by,

$$v(x) = (V + \dot{\delta}\sin\omega x)c_{i,\Phi}(1-k_i) = -D_{ii}\frac{\partial \tilde{c}_i}{\partial z}\bigg|_{z=\delta\sin\omega x},\qquad(12)$$

where $\dot{\delta}$ is $d\delta/dt$. The above equation can be re-expressed as,

$$(V + \dot{\delta}\sin\omega x) = \frac{-D_{ii}}{c_{i,\Phi}(1-k_i)}\frac{\partial \tilde{c}_i}{\partial z}\bigg|_{z=\delta\sin\omega x}.\qquad(13)$$

From Eq. (9),