Self-consistent modeling of morphology evolution during unidirectional solidification

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Abstract

A self-consistent model is developed to describe the morphology evolution during unidirectional solidification, which shows that, for a given temperature gradient, the interface morphology will go planar → shallow cell → deep cell → dendrite → cell → planar with increasing growth velocity. By examining the interaction of adjacent cells/dendrites, a wide allowable range of primary spacing for given growth conditions is determined, which shows a good agreement with experimental results. Numerical results show that cellular/dendritic and dendritic/cellular transitions appear not at a unique velocity but over a range of velocities, the critical velocity for the transition being dependent on the primary spacing before the transition. © 2001 Elsevier Science Ltd. All rights reserved.

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1. Introduction

A variety of interesting patterns, e.g. planar, cellular and dendritic, can form during unidirectional solidification. Solidification interface morphology, as a typical non-equilibrium self-organizing mode, has been focused on widely by many metallurgists, physicists and mathematicians for several decades [1,2]. Since the constitutional supercooling criterion of morphological instability of a solidifying solid/liquid interface was put forward [3], many significant efforts have been directed to describe morphology evolution and to predict morphological characteristics such as perturbation wavelengths $\lambda_{s}$, tip radius $\rho$, primary spacing $\lambda_{1}$, etc. [4–13].

A rigorous self-consistent model of dendritic growth was first proposed by Ivantsov [10] for a isothermal and/or isoconcentrate interface. He found that there exist an infinite number of solutions, which connect solidification velocity $V$ and tip radius $R$ for a given undercooling. From then on, many attempts have been done to find a unique solution by using minimum undercooling conditions [11], the marginal stability principle [12] or the microscopy solvability approach [13]. Most theoretical and experimental researches were performed to find a definite scaling law, which can correlate growth conditions with the resulting microstructures [6–8,11]. However, with the further understanding of the non-linear effect on the morphology evolution of the solidification interface, it has been gradually recognized that there exists a wide allowable range of primary cellular/dendritic spacing under given growth conditions, and that the pattern formation and mode selection of the solidification system are dependent not only on the current governing parameters, but also on the sequence of events by which the system has been set into motion. Based on the above knowledge, Warren and Langer [14] carried out a linear stability analysis by perturbing the solidification front at the dendritic tips in an array. They found that a range of spacing values exists for stable dendritic arrays and the minimum stable spacing was determined at the same time.

Actually, morphology evolution is a self-consistent free boundary or moving boundary problem, where the solid/liquid interface must be obtained as part of the solution. However, most of the models earlier made some ad hoc assumption about the shapes of the cells/dendrites. At the same time, it is very difficult and complex to solve this self-consistent problem analytically, which involved numerous mathematical equations and complicated formula derivation, e.g. the microscopy solvability approach. With the rapid development of computer techniques, from the viewpoint of convenient and understandability, numerical simulation has become an increasingly more useful method, which translates the complicated governing differential
equations to simple algebraic equations by suitable discretization. Since 1984, McFadden et al. [4] and Ungar et al. [5] had implemented a finite-difference method and finite-element technique for tracking the families of steady cellular interface, respectively. Ungar et al. predicted a fairly complex bifurcation diagram very close to the planar interface stability threshold and found that deep finger-like cells are often terminated by a bubble closure such as that observed in some experiments. Recently, Hunt and Lu [15] developed a numerical finite-difference model to describe the deep cellular/dendritic array growth. Through investigating the stability of the cellular/dendritic array, a range of primary spacing was found to be stable under given growth conditions. Their model shows good agreement with many experimental results in different alloy systems. It should be indicated that, however, most of the earlier theoretical researches only describe some local processes during unidirectional solidification, and there is little analysis which can describe the complete morphology evolution from the constitution supercooling limit (CSL) to the absolute stability limit (ASL).

In this paper, a self-consistent model is developed to morphology evolution between CSL and ASL during unidirectional solidification.

2. Formulation of the model

With considering an axisymmetric interface shape and neglecting the effect of convection, it is assumed that the latent heat of fusion is sufficiently small and that the thermal conductivities of the liquid and solid are sufficiently large and close to each other that the temperature throughout the system is determined by imposing a moving linear temperature field:

\[ T = T_0 + Gz \]  

(1)

where \( T_0 \) is the liquidus temperature of the bulk alloy composition \( C_0 \), \( G \) the applied temperature gradient, \( z \) is the axial position in a coordinate system moving at the pulling velocity \( V_0 \) and this coordinate system is referred to as the gradient coordinate system. The solute diffusion equation with this coordinate system is:

\[ D_s \nabla^2 C_i + V_0 \frac{\partial C_i}{\partial z} = \frac{\partial C_i}{\partial t} \]  

(2)

where \( C \) is the solute composition, \( D \) is the solute diffusion coefficient, and subscript \( i \) is either liquid (liquid) or solid (solid).

The far field conditions are:

\[ C_L = C_0, \text{ at } z \to +\infty \]  

(3)

\[ \frac{\partial C_L}{\partial z} = 0, \text{ at } z \to -\infty \]  

(4)

At the interface:

\[ C_S = kC_L \]  

(5)

\[ V_s(1-k)C_L = D_s \frac{\partial C_S}{\partial n} - D_L \frac{\partial C_L}{\partial n} \]  

(6)

The temperature \( T_1 \) at the interface is given by the modified Gibbs–Thomson equation:

\[ T_1 = T_0 + m(C_L - C_0) - \Gamma \kappa - \frac{V_n}{\mu} \]  

(7)

where \( k \) is the distribution coefficient, \( m \) is the liquidus slope, \( \Gamma \) is the Gibbs–Thomson coefficient, \( \kappa \) is the interface curvature, and \( \mu \) is the interface kinetic coefficient.

The diffusion equation is solved by the finite-difference method through control volume integration discretization. A self-consistent interface can be obtained by cooperating the temperature field with the solute field along the solid/liquid interface through Eq. (7). In order to cover a wider range of morphology, two kinds of initial shapes are used, as in Fig. 1.

At the near low velocity planar limit or absolute stability planar limit, a finite amplitude sine or cosine curve is used, whilst for a deep cell or dendrite, a modified Scheil shape is

Fig. 1. Schematic illustration of a symmetric repeat unit (initial shape): (a) near low velocity planar or absolute stability planar and (b) deep cell or dendritic.

Fig. 2. Variation of the undercooling against the spacing, where 1,2,3 indicate the different growth conditions.
used and a liquid composition gradient condition is applied in the intercellular groove, i.e. \((\partial C/\partial z) = (Glm)\).

3. Numerical results and discussion

It should be indicated that, for a given solidification condition, two kinds of interface shapes can be obtained for different spacing during the calculation of a deep cell or dendrite, which distribute in two spacing ranges, as in Fig. 2. In the range with smaller spacing, the interface shows a cellular shape, and in the range with larger spacing, it shows a parabolic dendrite-like shape.

In order to determine which shape appears at a given condition, the array stability limit with different shapes must be considered first. It is shown, from experiments of cellular and dendritic growth, that there exist two kind of spacing competition adjustment mechanisms, i.e. over-growth and tip splitting (for cell) or growth of tertiary arm (for dendritic), the lower limit of the stable spacing range being determined by the overgrowth mechanisms and the upper limit by tip splitting or growth of a tertiary arm [16]. According to this, the interaction of adjacent cells/dendrites is examined using a method similar to the Hunt–Lu model [15]. With increment of the spacing, when a cell or dendrite with some spacing can transport the solute to the other cell or dendrite with smaller or larger spacing, this spacing is examined as the minimum stable spacing, i.e. the lower limit of the stable array spacing range. For cellular growth, perturbing the cellular shape with a cosine perturbation of the form \(\delta \cos (2\pi r/\lambda_p)\), where \(\lambda_p\) is the wavelength of the perturbation, \(r\) is the radial position of the interface point, and \(\delta = \min (R_{tip}, \lambda_1)/h\), \(n\) is typically 100. By analyzing the tip stability, an upper limit can be obtained. For dendritic growth, new dendrite forming must deal with the growth competition between the tertiary arm and the secondary arm, while the present model only simulates a smooth dendrite-like shape without side-branch development. This shows that still much work should be done to predict the upper limit of stable dendritic spacing.

Fig. 2 shows three kinds of variation in the tip undercooling for cellular/dendritic growth for different solidification conditions. For a given temperature gradient, numerical results predict that the variation in the tip undercooling will change as \(1 \rightarrow 2 \rightarrow 3 \rightarrow 2 \rightarrow 1\) with increasing growth velocity. For condition 1, the tip undercooling of the cell with spacing in the whole stable spacing range is lower than that of dendrite, so that cellular growth will appear then. For condition 3, in contrast to condition 1, dendritic growth will appear. For condition 2, the undercooling ranges of cellular and dendritic growth overlap each other, so the coexistence of cells and dendrites may appear according to the spacing distribution. Thus it can be seen that cellular/dendritic and dendritic/cellular transition is actually dependent on the primary spacing. The remarkably history-dependent selection of primary spacing will make the cellular/dendritic and dendritic/cellular transitions appear not at a unique velocity but in a velocity range. The critical velocity for transition is dependent on the primary spacing before transition.

Fig. 3 shows several typical interface shapes with different growth velocities. Fig. 3(a) shows a series of

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Fig. 3. Typical interface shapes: (a) shallow cell; (b) deep cell; and (c) dendrite.

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Fig. 4. Allowable range of dendritic spacing: (a) SCN = 1.0%Ace, \(G = 4.0\) K/mm; (b) Al = 0.53%Zn, \(G = 14.5\) K/mm; and (c) Cu = 26.6%Mn, \(2.2 \times 10^3\) K/mm.

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finite amplitude cells, which correspond to constitutional supercooling and absolute stability regimes, Fig. 3(b) shows a series of deep cell shapes obtained in low and high growth velocity ranges, Fig. 3(c) shows the more parabolic dendrite-like shapes obtained for moderate growth velocities. The numerical results indicate that, for a low temperature gradient, the interface morphology will go planar → shallow cell (Fig. 3(a)) → deep cell (Fig. 3(b)) → dendrite (Fig. 3(c)) → deep cell (Fig. 3(b)) → shallow cell (Fig. 3(a)) → planar with increasing solidification velocity. However, the deep cell obtained at high velocity is different from that at low velocity, and the ratio of cellular trunk to spacing in high growth velocity is much lower than that for low growth velocity. Fig. 4 compares the predicted spacing with the experimental results during different solidification conditions in three alloy systems, and shows very good agreement.

4. Summary

A self-consistent model is developed to describe the morphology evolution under unidirectional solidification. It is shown that, for a given temperature gradient, the interface morphology will go planar → shallow cell → deep cell → dendrite → cell → planar with increasing solidification velocity. By examining the interaction of the solute field between adjacent cells/dendrites and tip stability of a cell, a wide allowable range of primary spacing for given growth conditions is determined, which shows a good agreement with the experimental results. At the same time, the model predicts that cellular/dendritic and dendritic/cellular transition appear not at a unique velocity but in a velocity range, the critical velocity for the transitions being dependent on the primary spacing before transition.

Acknowledgements

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