Free Dendrite Growth of Fe–0.5mass%C Alloy –Three-dimensional Phase-field Simulation and LKT Model–

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The free dendrite growth of iron–0.5mass% carbon alloy has been investigated using a three-dimensional phase-field model with thin-interface-limit parameters. The dendrite growth velocity and the tip radius of curvature are determined at different degrees of undercooling, and then the stability parameter for the alloy is estimated. The dendrite growth velocities obtained by phase-field simulations are compared with those obtained using the LKT model with the stability parameter of 0.05. The two are in good agreement and the validity of the LKT model is discussed with regard to practical applications.

KEY WORDS: phase-field model; dendrite growth; iron–carbon alloy; LKT model.

1. Introduction

Understanding dendritic solidification is of great importance because the scale of a dendrite determines the segregation patterns, microstructures, and consequently the properties of the material. For this reason, this process has been extensively studied both theoretically and experimentally for several decades. In the proposed dendrite models, it is assumed that a dendrite has the shape of a parabolic revolution and an isothermal or non-isothermal interface. For independently predicting the growth velocity, V, and the tip radius of curvature, ρ, the maximum-velocity criterion was used until Langer and Mueller-Krumbhaar proposed the marginal-stability criterion. The marginal-stability criterion stipulates that the tip radius of a dendrite is equal to the marginal wavelength of the perturbation to a planer interface. They performed the linear-stability analysis on an Ivantsov dendrite neglecting the anisotropy of interface energy and determined the stability parameter, σ*, to be $\frac{2a\tau_0}{\rho^2V}$.

Their criterion was successful in explaining the $V-\rho$ relationship obtained in the experimental results of succinonitrile dendrite growth, subsequently, the studies on dendrite growth advanced to a highly theoretical stage.

In the late 1980s the solvability theory showed that the anisotropy of interface energy played an essential role in dendritic evolution, and that tip splitting takes place without the anisotropy of interface energy. In addition, it was shown that the stability parameter is proportional to the 7/4th power of anisotropy intensity. Since then, different types of computational approaches have been developed to simulate the complete time-dependent evolution of a free dendrite, and the values of stability parameters for pure materials were also numerically evaluated. The growth velocity and tip radius of a dendrite have been obtained from simulations, and the stability parameter is evaluated using

$$\sigma^* = \frac{2a\tau_0}{\rho^2V} \quad \cdots \cdots \cdots \cdots (1)$$

where $\alpha$ is the thermal diffusivity, $\rho$ is the equilibrium partition coefficient, and $\tau_0 = \sigma^0 T_m C_p / \Delta H$ is the capillary length, $\sigma^0$ is the interface energy, $T_m$ is the melting temperature, $C_p$ is the heat capacity per volume and $\Delta H$ is the latent heat of fusion.

The stability parameter for dendritic growth of alloys, $\sigma_{D,*}$, was also defined in a similar manner and is given by

$$\sigma_{D,*} = \frac{2D G}{\rho^2 k \Delta T_0} \quad \cdots \cdots \cdots \cdots (2)$$

where $D$ is the diffusion coefficient, $k$ is the equilibrium partition coefficient, and $\Delta T_0 = \Gamma \Delta T_0$ is the chemical capillarity length, where $\Gamma$ is the Gibbs–Thomson coefficient defined by $\sigma \cdot T_m / \Delta H$, and $\Delta T_0$ is the solidification range of an alloy. Kurz and Fisher proposed a model based on the marginal-stability criterion so as to analyze the constrained dendrite growth of alloys. They defined the stability parameter in a different manner:

$$\rho = 2\pi \sqrt{\frac{\Gamma}{mG_c - G}} \quad \text{or} \quad \sigma^* = \frac{\Gamma}{\rho^2 (mG_c - G)} \quad \cdots \cdots \cdots \cdots (3)$$

where $G_c$ is the solute concentration gradient at the dendrite tip, $G$ is the average temperature gradient, and the values of $G_c$ and $G$ are obtained using the Ivantsov solution. The model was extended to the LGK model and the LKT model which cover the dendrite from solutal to thermal-solutal growth. In particular, the high Péclet number correction was introduced in the LKT model making it possible.
to analyze dendrite growth under a wide range of rapid solidification conditions. In the LKT model, the stability parameter is defined by \( ^{(31)} \)

\[
\sigma^* = \frac{\Gamma}{\rho^2 (mG_G^c - G)}
\]

\[
G = \frac{\kappa_S G^S_S + \kappa_L G^L_L}{\kappa_S + \kappa_L}
\]

where \( \kappa \) is the thermal conductivity, \( \xi \) is the correction function for high Péclet numbers, and the subscripts S and L denote the solid and liquid phases, respectively. Using the LKT model, intensive studies were carried out on the morphological changes, phase-selections and solute trapping phenomena during rapid solidification processes, and a large number of useful results for industrial applications were obtained. In addition, the LKT model is often used to obtain the dendrite growth velocity data necessary for mesoscopic numerical simulations using a cellar-automaton algorithm. \( ^{(22,23)} \) In spite of the publicity of the LKT model, it was not quantitatively examined until the phase-field simulation for alloy solidification became popular. \( ^{(24)} \)

In this study, three-dimensional simulations of the free dendrite growth of an iron–carbon alloy are performed using a thin-interface-limit phase-field model. \( ^{(25)} \) The dendrite growth velocity and the tip radius of curvature are determined at different degrees of undercooling and the value of the stability parameter is estimated. Dendrite growth velocity and the tip radius of curvature are discussed from the viewpoint of practical applications of the LKT model.

2. Calculations

In the simulation, both the phase-field equation and the solute diffusion equation are numerically solved using an explicit finite difference scheme. The governing equations are

\[
\frac{1}{M} \frac{\partial \phi}{\partial t} = \varepsilon^2 \nabla^2 \phi - \frac{\partial f}{\partial \phi} \tag{5}
\]

\[
\frac{\partial c}{\partial t} = - \nabla \left( \frac{D}{f} \frac{\partial \phi}{\partial f} \nabla \phi \right) \tag{6}
\]

where \( f \) is the free energy density, subscript \( c \) denotes partial derivatives, \( D \) is the diffusion coefficient, and \( M \) and \( \varepsilon \) are phase-field parameters. The free energy density of an alloy is given by,

\[
f(\phi, c) = h(\phi) f^S(\phi) + (1 - h(\phi)) f^L(\phi) + Wg(\phi) \tag{7}
\]

where superscripts \( S \) and \( L \) indicate solid and liquid phases, respectively, \( f^S \) and \( f^L \) are free energies of liquid and solid phases, and \( W \) is the height of the imposed interfacial potential. The solid fraction function, \( h(\phi) = \phi^3(3 - 2\phi) \) is a monotonically increasing function from \( h(0) = 0 \) to \( h(1) = 1 \), and the interfacial potential, \( g(\phi) = \phi(1 - \phi) \), is a parabolic function with \( g(0) = 0 \) and \( g(1) = 0 \). By assuming a dilute solution, the terms in Eqs. (1) and (2) are rewritten into simplified forms. Note that the solute content at the interface region is given as the sum of fraction-weighted solute contents of solid and liquid phases and that the solid and liquid solute contents are obtained under the condition of equilibrium potentials in both solid and liquid phases. \( ^{(25)} \) The anisotropies of interface energy are directly related to that of the gradient energy coefficient, \( \varepsilon \), in phase-field models. The phase-field parameters of \( e_0 \) and \( W \) are related to the interface energy, \( \sigma_0 \), and interface width, \( 2\lambda \), and the parameter, \( M \), is related to the kinetic coefficient, \( \beta_0 (= 1/\mu_0) \), where \( \mu_0 \) is the linear kinetic coefficient. The details of the governing equations and the thin-interface-limit phase-field parameters can be found in the literature. \( ^{(15,26,27)} \)

The simulation is carried out in the same manner as in the last work, using an adaptive mesh algorithm. The program achieved sufficient execution efficiency using a computer with a Xeon CPU of 3.76 GHz. In the calculations, all variables are rewritten as dimensionless forms using the following units of capillary length, \( d_{\text{co}} \) and time, \( d_{\text{co}}^2/D \). The physical properties of Fe–0.5mass%C alloy used in the calculation are shown in Table 1. \( ^{(15,28-30)} \) The dimensionless mesh sizes in the calculations are from 0.88 to 0.18. Note that the reduced interface width method for solutal diffusion \( ^{(31)} \) is applied instead of the anti trapping current model \( ^{(25)} \) so as to eliminate abnormal interface effects in the thin-interface-limit model.

3. Results and Discussion

Before performing the three-dimensional simulations, the effect of thermal conduction on the dendrite growth of an alloy is examined in two-dimensional simulations, in which the growth velocity is calculated coupled with and without the heat conduction equation. The results agree with each other and the effect of thermal field is deemed negligible. Therefore three-dimensional simulations are carried out as for pure solutal dendrite growth.

Figure 1 shows the stability parameter obtained at different degrees of dimensionless undercooling. The stability parameter values are slightly scattered and the mean value is 0.21. From the results of two- and three-dimensional sim-

| Properties          | Unit  | Values |
|---------------------|-------|--------|
| Interface energy, \( \sigma \) | J/m²  | 0.024  |
| Melting temperature, \( T_m \) | K    | 1811   |
| Diffusivity of solid, \( D_s \) | m²/s  | 6×10⁻⁹ |
| Diffusivity of liquid, \( D_l \) | m²/s  | 2×10⁻⁴ |
| Molar volume, \( V_m \) | solvant | 7.7×10⁻⁴ |
| Equilibrium distribution coefficient, \( k \) | m³/mol | 0.17   |
| Liquidus slope, \( m_e \) | K/mol | 1774   |
| Kinetic coefficient, \( \beta \) | Ks/m  | 0.638  |
| Anisotropy, \( \nu \) |       | 0.02   |
ulations for iron, the stability parameter of iron is estimated to be 0.11 and 0.086, respectively. As the solute diffusion in the solid phase is negligible, solutal dendrite growth is regarded as a one-sided problem, and thus the value of the stability parameter, $\sigma_D^*$, is expected to be twice that for iron. The results in Fig. 1 fall in such a range, from 0.22 to 0.18.

Figures 2 and 3 show the relationship between the dimensionless dendrite growth velocity and dimensionless undercooling and the relationship between the dimensionless tip radius of curvature and dimensionless undercooling, respectively. In the figures, open circles and the solid line show the results of three-dimensional phase-field simulations and the prediction by the LKT model, respectively. Note that the stability parameter for the LKT model is set to be 0.05, because the stability parameter for a solutal dendrite should be twice that for a thermal dendrite. The growth velocities obtained by phase-field simulations are slightly smaller than that obtained using the LKT model, but the two results are in rather good agreement in spite of the difference in the stability parameters. The tip radius obtained using the LKT model is smaller than those obtained by phase-field simulations as expected from the difference in stability parameters.

In spite of the different stability parameters, the growth velocity and tip radius of an alloy dendrite obtained using the LKT model and by phase-field simulations are close to each other. This is simply explained by the fact that an alloy dendrite grows with an isoconcentration interface having a shape approximated to be a parabolic revolution. Figure 4 shows the dendrite shape on the $x$–$z$ plane simulated using the phase-field model. In the figure the thick solid line shows the interface, thin solid lines with numbers are isoconcentration lines normalized by the mean concentration, the circle is represents the tip radius of curvature evaluated in the phase-field simulation, and the dotted line shows the parabolic interface with the same tip radius of curvature assumed. As the tip radius of curvature predicted by the LKT model is slightly larger than that by the phase-field simulation as shown in Fig. 3, the LKT interface is close to the simulated interface shape in shape and size. In most cases of alloy solidification, kinetic undercooling is small and curvature undercooling is presumably reduced by solute diffusion. As the interface of a dendrite agrees with an iso-concentration contour and its shape is approximated to be parabolic, the LKT model gives a good approximation of the dendrite growth of alloys. On the contrary, the situation for the dendrite growth of pure materials is completely different. Figure 5 shows...
the interface shape and normalized isothermal lines in the $x$–$z$ plane obtained by phase-field simulations for iron. The dotted line shows the parabolic interface with the same tip radius of curvature by the phase-field simulation assumed and the interface shape still could be approximated to be a parabolic revolution. However, the interface is completely different from with the isothermal contour. Therefore the assumption of the isothermal interface in the LKT model is not satisfied. Even thought the assumptions in the LKT model deviate from the reality of a dendrite, the LKT model can be a useful tool for predicting the dendrite growth velocity of pure materials when the values of the stability parameter and the interface kinetic coefficient are carefully selected as described in the previous article.\(^{15}\)

4. Conclusions

Three-dimensional phase-field simulations of the free dendrite growth of Fe–0.5mass%C alloy have been successfully performed. Using the phase-field model, the growth velocity and the tip radius of curvature for the alloy dendrites are evaluated at different degrees of undercooling. The stability parameter is estimated to be about 0.2, and which is approximately twice the value for iron. The prediction using the LKT model with the stability value of 0.05 is compared with the results of phase-field simulations, and the growth velocity and the tip radius of curvature for both are close to each other. Therefore, it is concluded that the LKT model gives good predictions for the dendrite growth of alloys from the viewpoint of practical use, when its stability parameter is set to be twice the usual value of $1/4\pi^2$.

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