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Phase Field Modeling of Dendritic Growth and Coarsening

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1. Introduction

Phase field models are known to be very powerful in describing the complex pattern evolution of dendritic growth. It is a useful method for simulating microstructure evolution involving diffusion, coarsening of dendrites and the curvature and kinetic effects on the moving solid-liquid interface. Such models are efficient especially in numerical treatment because all the governing equations are written as unified equations in the whole space of the system without distinguishing the interface from the mother and the new phase, and direct tracking of the interface position is not needed during numerical calculation. In the last decade, the phase field method has been intensively studied as a model of solidification processes [1-5]. The dendritic coarsening behavior affects the distribution of length scales, microsegregation and other microstructural characteristics of the materials, all of which determine the physical and chemical properties of materials in terms of strength, ductility and corrosion resistance. Therefore, understanding coarsening and being able to study the morphology of the dendritic structure is of technological importance^[6-9]. Many properties of cast materials are intimately related to the dendritic morphology that is largely set by coarsening. Even if the effect of the dendritic microstructure is altered by subsequent heat treatment, they rarely fully disappear.

In this part, the dendritic growth of and the subsequent dendritic coarsening as well as the effect of undercooling on coarsening in Al-2mol%Si alloy during isothermal solidification are simulated using phase field model.

2. Phase field model

The phase field model used in this paper was developed by Kim et al. So it would be briefly mentioned here. Readers can refer to literatures [10,11] for details of the formulation. The model includes two variables: one is a phase field $\phi(x,y,t)$ and the other is a concentration field c(x,y,t). The variable $\phi(x,y,t)$ is an ordering parameter at the position (x,y) and the time t, $\phi=1$ means being solid and $\phi=0$ liquid. The solid-liquid interface is expressed by the steep layer of ϕ connecting the value 0 and 1. So the phase field model can be described as

$$\frac{\partial \phi}{\partial t} = M \Big(\varepsilon^2(\theta) \nabla^2 \phi - f_\phi \Big) \tag{1}$$

$$\frac{\partial c}{\partial t} = \nabla \bullet \left(\frac{D(\phi)}{f_{cc}} \nabla f_c \right)$$
(2)

Where,
$$f(c,\phi) = h(\phi) f^{S}(c_{S}) + (1 - h(\phi)) f^{L}(c_{L}) + Wg(\phi)$$
 (3)

$$D(\phi) = D_{\rm L} + h(\phi) (D_{\rm S} - D_{\rm L})$$
⁽⁴⁾

$$h(\phi) = \phi^{3} (6\phi^{2} - 15\phi + 10)$$
(5)
$$g(\phi) = \phi^{2} (1 - \phi^{2})$$
(6)

$$c = h(\phi)c_{\rm S} + (1 - h(\phi))c_{\rm L}$$
 (7)

$$\mu^{\mathrm{S}}((c_{\mathrm{S}}(x,t)) = \mu^{\mathrm{L}}(c_{\mathrm{L}}(x,t))$$
(8)

$$f^{\rm S} = c_{\rm S} f^{\rm S}_{\rm B}(T) + (1 - c_{\rm S}) f^{\rm S}_{\rm A}(T)$$
⁽⁹⁾

$$f^{\rm L} = c_{\rm L} f^{\rm L}_{\rm B}(T) + (1 - c_{\rm L}) f^{\rm L}_{\rm A}(T)$$
(10)

$$\varepsilon(\theta) = \varepsilon_0 \left\{ 1 + \nu \cos(k\theta) \right\} \tag{11}$$

$$M^{-1} = \frac{\varepsilon^2}{\sigma} \left[\frac{RT}{V_{\rm m}} \frac{1 - k^{\rm e}}{m^{\rm e}} \frac{1}{\mu} + \frac{\varepsilon}{D_{\rm L} \sqrt{2W}} \varsigma \left(c_{\rm S}^{\rm e}, c_{\rm L}^{\rm e} \right) \right]$$
(12)

$$\varsigma\left(c_{\rm S}^{\rm e}, c_{\rm L}^{\rm e}\right) = f_{cc}^{\rm S}\left(c_{\rm S}^{\rm e}\right) f_{cc}^{\rm L}\left(c_{\rm L}^{\rm e}\right) \left(c_{\rm L}^{\rm e} - c_{\rm S}^{\rm e}\right)^{2} \\
\times \int_{0}^{1} \frac{h(\phi) \left[1 - h(\phi)\right] \mathrm{d}\phi}{\left[1 - h(\phi)\right] \mathrm{d}\phi} \tag{13}$$

$$\mathcal{E} = \sqrt{\frac{6\lambda}{2.2}\sigma}$$
(14)

$$W = \frac{6.6\sigma}{\lambda} \tag{15}$$

Where *M* and ε are the phase field mobility and gradient energy coefficient, respectively. *f* is the free energy density of the system. The subscripts under *f* indicate the partial derivatives. $D(\phi)$ is the diffusivity of solute as a function of phase field. D_S and D_L are the diffusive coefficient in the solid and liquid respectively. $h(\phi)$ and *W* correspond to solid fraction and the height of double-well potential, respectively. c_L and c_S are the solute concentration in liquid and solid, respectively. ε_0 is the mean value of ε , θ is the angle between the normal to the interface and the x-axis, θ =arctan(ϕ_y/ϕ_x). v is the strength of anisotropy and k is the

mode number, k=4. m^{e} is equilibrium slope of the liquids, k^{e} is the equilibrium partition coefficient. The phase field parameters of ε and W are related to the interface energy σ and the interface width 2λ .

In addition, stochastic noise introduced into the phase field model causes fluctuations at the solid/liquid interface that leads to the development of a dendrite structure. Herein, noise is introduced by modifying the phase field equation

$$\frac{\partial \phi}{\partial t} \rightarrow \frac{\partial \phi}{\partial t} + 16g(\phi)\chi\omega \tag{16}$$

Where χ is a random number distributed uniformly between -1 and 1, a new number is generated for every point of the grid, at each time-step. ω is an amplitude of the fluctuations, here ω =0.01 in the calculation.

The Al-2mol%Si alloy is considered in this work. Isothermal computations are performed using the model described above. The grid sizes of the phase field and the concentration field are 1.0×10^{-8} m. The governing equations are discretized on uniform grids by using an explicit finite difference scheme. The thermo-physical data given as: σ =0.093 J·m⁻¹, T_m =922K, V_m =1.06×10⁻⁵ m³/mole, k^e =0.0807, D_L =3×10⁻⁹ m²·s⁻¹, D_S =1×10⁻¹² m²·s⁻¹, m^e =-939.0, v=0.03, ω =0.01.

3. Results and discussion

3.1 Simulation of single dendrite growth during isothermal solidification

Fig.1 shows the evolution of simulated single dendrite during isothermal solidification of Al-2mol%Si alloy at the temperature of 870K. Fig.2 illustrates the concentration profiles calculated during single dendrite isothermal solidification of Al-2mol%Si alloy at the temperature of 870K. It can be seen from Fig.1, the nucleus grows and becomes unstable and then the dendrite forms. In the early stage of solidification, the dendrite develops the main arms along the crystallographic orientations. The secondary and tertiary arms as well as the necking phenomenon can be observed. In the case of equiaxed dendrite growth, there exists a solute build-up ahead of the dendrite tip, as shown in Fig.2. The crystal shape is dictated mainly by the diffusion of solute. Nevertheless, a remaining anisotropy in properties leads to the growth of dendrite arms in specific crystallographic direction.



Fig. 1. Simulated single dendrite evolution during isothermal solidification of Al-2mol%Si alloy at the temperature of 870K. The snapshots (a), (b), (c) and (d) correspond to solidification time of 0.01ms, 0.04ms, 0.08ms and 0.14ms respectively.

Supercooling



Fig. 2. Concentration profiles calculated during single dendrite isothermal solidification of Al-2mol%Si alloy at the temperature of 870K. The snapshots (a), (b), (c) and (d) correspond to solidification time of 0.01ms, 0.04ms, 0.08ms and 0.14ms respectively.

The oriented growth of single dendrite from the bottom-left corner toward the top-right corner of the square domain was simulated in Al-2mol%Si alloy. Fig.3 shows the evolution of oriented growth of single dendrite during isothermal solidification in Al-2mol%Si alloy at temperature of 870K. Fig.4 illustrates the concentration profiles during isothermal solidification of Al-2mol%Si alloy at temperature of 870K. The snapshots (a), (b), (c), (d) and (e) in Fig.3 and Fig.4 correspond to solidification time of 0.04, 0.10, 0.14, 0.18, and 0.22ms respectively. Fig.5 shows the snapshot of the oriented dendrite at solidification time of 0.26ms. Fig.6 shows the curve of dendritic tip velocity versus time.



Fig. 3. Simulated oriented growth of single dendrite during isothermal solidification in Al-2mol%Si alloy at temperature of 870K. The snapshots (a), (b), (c), (d) and (e) correspond to solidification time of 0.04, 0.10, 0.14, 0.18 and 0.22ms respectively.



Fig. 4. Concentration profiles calculated oriented growth of single dendrite during isothermal solidification in Al-2mol%Si alloy at temperature of 870K. The snapshots (a), (b), (c), (d) and (e) correspond to solidification time of 0.04, 0.10, 0.14, 0.18, and 0.22ms respectively.

The simulation results show the morphology of dendrite with primary and secondary arms as well as tertiary arms. The process of dendrite growth and the competition between the dendrite arms are reproduced. At the early stage of solidification, the dendrite develops the main arms along the crystallographic orientations. Near the primary arm, the small secondary arms compete with each other and some overgrown secondary arms survive. The dendrite grows fast at the early stage as shown in Fig.6, in which the dendritic tip velocity increases steeply with the time. In the process of dendrite growth, the secondary arms are sometimes eliminated by their neighbors, and a number of them grow perpendicularly to the primary arm. The survived secondary arms grow until being screened by the tertiary arms, whereas the dendritic tip velocity changes in a small range due to the addition of noise.

Because of the concentration redistribution in the front of solid-liquid interface, the interdendritic liquid always has a different composition compared to that of the dendrite arms. The concentration of Si in dendrite arms is the lowest. The highest concentration corresponds to the interdendritic liquid, as shown in Fig.4.

In addition, an interesting phenomenon is found that the tertiary arms only grow at one side of some secondary arms which can be seen in Fig.5. The simulated result is in accordance with the result of Seong Gyoon KIM^[12].



Fig. 5. Snapshot of oriented growth dendrite at time of 0.26ms



Fig. 6. The curve of growth velocity versus time

3.2 Simulation of multi-dendrite growth during isothermal solidification

3.2.1 Simulation of multi-dendrite growth during isothermal solidification

Fig.7 shows the evolution of simulated multi-dendritic growth during isothermal solidification of Al-2mol%Si alloy at the temperature of 880K. Fig.8 illustrates the concentration profiles calculated during isothermal solidification of Al-2mol%Si alloy at the temperature of 880K. The dendrites grow freely and independently in the melt and finally impinge on one another for arbitrarily oriented crystals, which can be seen from Fig.7 (a) and (b). In the process of dendritic growth, the secondary arms are eliminated by their neighbors, and a number of them grow perpendicularly to the primary arm. However, the growth of some main arms is suppressed by nearby dendrite. As solidification proceeds, growing and coarsening of the primary arms occur, together with the branching and coarsening of the secondary arms, as shown in Fig.7 (c) to (e). Due to the concentration redistribution in the front of solid-liquid interface, as shown in Fig.8, the interdendritic liquid always has a different composition compared to that of the dendrite arms. The concentration of Si in dendrite arms is the lowest. The highest concentration corresponds to



Fig. 7. Simulated multi-dendrite evolution during isothermal solidification of Al-2mol%Si alloy at the temperature of 880K. The snapshots (a), (b), (c), (d) and (e) correspond to solidification time 0.04, 0.08, 0.12, 0.16 and 0.2ms respectively.

the interdendritic liquid. Once the diffusion fields of dendrite tips come into contact with those of the branches growing from the neighboring dendrites, the dendrites stop growing and being to ripen and thicken.



Fig. 8. Concentration profiles calculated during multi-dendrite isothermal solidification of Al-2mol%Si alloy at the temperature of 880K. The snapshots (a), (b), (c), (d) and (e) correspond to solidification time 0.04, 0.08, 0.12, 0.16 and 0.2ms respectively.

3.2.2 Effect of anisotropy strength on the dendrite morphology

Fig.9 shows the effect of anisotropy strength on the dendrite morphology during isothermal solidification of Al-2mol%Si alloy at 870K for solidification time of 0.04ms, 0.1ms and 0.2ms correspond to anisotropy strength 0, 0.01, 0.03 and 0.08 respectively. All the parameters except the strength of anisotropy (v) are fixed and v is increased gradually from zero. Fig.9 (a) shows the microstructure where v=0, namely perfect isotropic growth. In Fig.9 (b)-(d), the strength of anisotropy (v) is v=0.01, v=0.03 and v=0.08 respectively.

In Fig.9 (a), where v=0, namely perfect isotropic growth is considered, the simulated patterns similar to the viscous fingering obtained. Tip splitting is seen as the dendrite growth. This figure shows the growth of a dense-branching morphology. For v=0.01 in Fig.9 (b), the shape of the crystal growth has both the features of isotropic and the dendrite structure. It can be seen the dendrite and the viscous finger-like structure. For v=0.03 in Fig.9 (c), the results show one typically dendrite structure. For v=0.08 in Fig.9 (d), the results show another type of dendrite arms are gradually disappeared. In addition, it can be seen the "necking" during the dendritic growth process. From these simulations, the dendrites grow up faster along with the crystal-axis direction.



(a) For *v*=0, namely perfect isotropic growth. This figure shows the growth of a dense-branching morphology. The snapshots correspond to time of 0.04ms, 0.1ms and 0.2ms, respectively.

Supercooling



(b) For v=0.01, the shape of the crystal growth has both the feature of isotropic and of the dendrite structure. The snapshots correspond to time of 0.04ms, 0.1ms and 0.2ms, respectively.



(c) For v=0.03, the simulation patterns of dendrite growth show typical dendrite structure. The snapshots correspond to time of 0.04ms, 0.1ms and 0.2ms, respectively.



(d) For v=0.08, the simulations show another type of dendrite structure. The snapshots correspond to time of 0.04ms, 0.1ms and 0.2ms, respectively.

Fig. 9. Effect of anisotropy strength on the dendrite morphology of Al-2mol%Si alloy at 870K

Comparing these simulations, it is obvious that the dendrite structure is very sensitively dependent on the strength of anisotropy (v).

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3.3 Dendritic coarsening under different undercoolings

Dendritic coarsening for Al-2mol%Si alloy under different undercoolings of Δ T=37K, Δ T=47K, Δ T=57K and Δ T=67K during isothermal holding is carried out. Fig.10 illustrates the fraction of solid phase as a function of solidification time for different undercoolings. The value of fraction of solid phase with solidification time under different undercoolings is listed in Table1. Fig.11 shows the evolution of the dendritic microstructure during isothermal holding at time of 0.20ms, 0.30ms, 0.40ms, 0.58ms, 0.72ms and 1.60ms respectively at undercoolings of (a) Δ T=37K, (b) Δ T=47K, (c) Δ T=57K and (d) Δ T=67K.



Time /ms

Fig. 10. The fraction of solid phase as a function of solidification time under different undercoolings

Solidification time	Fraction of solid phase (%)			
(ms)	ΔT=37K	ΔT=47K	ΔT=57K	ΔT=67K
0.20	5.5	10.0	13.4	15.2
0.30	11.9	24.0	31.6	36.4
0.40	21.6	42.5	52.0	57.7
0.58	42.4	64.2	71.2	76.0
0.72	55.5	67.8	72.7	76.4
1.60	62.6	69.2	73.9	77.2

Table 1. The fraction of solid phase with solidification time under different undercoolings

The dendritic morphology and dendritic coarsening mechanisms as well as the solidification kinetics are varied with the undercooling. With the undercooling increasing, the fraction of

solid phase is also increased. The curve of solidification kinetics is similar for $\Delta T=37K$, $\Delta T=47K$, $\Delta T=57K$ and $\Delta T=67K$, but the final fraction of solid phase is quite different, and the coarsening time is also different. The coarsening time is 0.88ms, 0.70ms, 0.62ms and 0.58ms for undercooling $\Delta T=37K$, $\Delta T=47K$, $\Delta T=57K$ and $\Delta T=67K$ respectively, after that the fraction of solid phase keeps stable basically, and the stable fraction of solid phase is 62.6%, 69.2%, 73.9% and 77.2% respectively. The higher of the undercooling, the faster of the dendrite growth and the shorter of reaching coarsening time. When undercooling $\Delta T=37K$ and $\Delta T=47K$, the evolution of microstructure show typical dendritic morphology, as shown in Fig.11 (a) and (b). The mechanisms of isothermal dendritic coarsening are melting of samll dendrite arms (as indicated in the circled area in Fig.11 (a) and (b)) and smoothing of dendrites (as indicated in the rectangled area in Fig.11 (a) and (b)). It shows dendrite remelting from tips towards roots and coalescence between neighboring branches. Dendrite remelting is found to be dominant in the early stage of dendrite growth, whereas



(d) ∆T=67K

Fig. 11. The evolution of the dendritic microstructure for Al-2mol%Si alloy during isothermal holding at time of 0.20ms, 0.30ms, 0.40ms, 0.58ms, 0.72ms and 1.60ms respectively for undercooling of (a) Δ T=37K, (b) Δ T=47K, (c) Δ T=57K and (d) Δ T=67K

coalescence and smoothing of dendrites are dominant during isothermal holding. When undercooling ΔT =57K and ΔT =67K, the evolution of microstructure shows viscous dendritic morphology, as shown in Fig.11 (c) and (d). The mechanisms of isothermal dendritic coarsening are coalescence of dendrites with the entrapment of liquid droplets (as indicated in the parallelogram area in Fig.11 (c) and (d)) and smoothing of dendrites (as indicated in the elliptical area in Fig.11 (c) and (d)) as well as the rounding of interdendritic liquid droplets (as indicated in the rhombus area in Fig.11 (c) and (d)).

With the undercooling increasing, the interdendritic liquid is reducing. When solidification time is 1.60ms, the interdendritic liquid takes up 37.4% of the domain for ΔT =37K, and 22.8% of the domain for ΔT =67K. In addition, the pattern of the interdendritic liquid is also different with the undercooling. When the undercooling ΔT =37K, the liquid phases in the dendritic structure show platelike structure. With the undercooling increase, the liquid phases in the dendritic structure show rodlike structure. The simulated results are in accordance with Wang's simulation results^[13].

4. Conclusions

- 1. The simulation of single dendrite and multi-dendrite growth for Al-2mol%Si alloy during isothermal solidification are carried out by phase field method. The primary and secondary arms as well as the necking phenomenon can be observed. For the oriented growth of single dendrite from the bottom-left corner toward the top-right corner of the square domain The survived secondary arms grow until being screened by the tertiary arms. An interesting phenomenon is found that the tertiary arms only grow at one side of some secondary arms.
- 2. For multi-dendrite simulation, the dendrites grow freely and independently in the melt and finally impinge on one another. As solidification proceeds, growing and coarsening of the primary arms occurs, together with the branching and coarsening of the secondary arms. Due to the concentration redistribution in the front of solid-liquid interface, the interdendritic liquid always has a different composition compared to that of the dendrite trunks. When the diffusion fields of dendrite tips come into contact with those of the branches growing from the neighboring dendrites, the dendrites stop growing and being to ripen and thicken.
- 3. For Al-2mol%Si alloy, when the undercooling is $\Delta T=37K$ and $\Delta T=47K$, the evolution of microstructure shows typical dendritic morphology, and the mechanisms of isothermal dendritic coarsening are melting of samll dendrite arms, coalescence of dendrites and smoothing of dendrites. When undercooling is $\Delta T=57K$ and $\Delta T=67K$, the evolution of microstructure shows viscous dendritic morphology, and the mechanisms of isothermal dendritic coarsening are coalescence of dendrites with the entrapment of liquid droplets and smoothing of dendrites as well as the rounding of interdendritic liquid droplets. The solidification kinetics is similar for different undercoolings, but the coarsening time and the final fraction of solid phase is quite different. The higher of the undercooling, the faster of the dendrite growth and the shorter of reaching coarsening time.

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Supercooling Edited by Prof. Peter Wilson

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Supercooled liquids are found in the atmosphere, in cold hardy organisms, in metallurgy, and in many industrial systems today. Stabilizing the metastable, supercooled state, or encouraging the associated process of nucleation have both been the subject of scientific interest for several hundred years. This book is an invaluable starting point for researchers interested in the supercooling of water and aqueous solutions in biology and industry. The book also deals with modeling and the formation subsequent dendritic growth of supercooled solutions, as well as glass transitions and interface stability.

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