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Anisotropic growth of multigrain in equiaxial solidification simulated with the phase field method*

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The phase field method has been mainly used to simulate the growth of a single crystal in the past. But polycrystalline materials predominate in engineering. In this work, a phase field model for multigrain solidification is developed, which takes into account the random crystallographic orientations of crystallites and preserves the rotational invariance of the free energy. The morphological evolution of equiaxial multigrain solidification is predicted and the effect of composition on transformation kinetics is studied. The numerical results indicate that due to the soft impingement of grains the Avrami exponent varies with the initial melt composition and the solidification fraction.

Keywords: multigrain, solidification, transformation kinetics, phase field method
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1. Introduction

In the last decade, the phase field method has been widely used to investigate the solidification process. Most of the works focused on the calculation of the growth of a single crystal from the melt.[1−3] However, understanding of the microstructural evolution of a polycrystalline material is essential for practical applications. In practical casting process, many crystallites with different crystallographic orientations independently grow out of the solution and subsequently interact through diffusive processes. In order to simulate such a phenomenon, crystallographic orientation must be included in the model.

Morin *et al*[4] constructed the free energy density having n wells, corresponding to n crystallographic orientations. Chen and Yang[5], and Steinbach *et al*[6] assigned n order parameters to the n allowed orientations. In this kind of models, large number of phase fields are needed to properly address multigrain growth problem, and even so, these models cannot naturally accommodate the formation of new, randomly oriented grains. In all approaches mentioned above, the free energy density depends on the orientation of the crystal with respect to a fixed reference frame, which is unphysical. The first model of polycrystalline solidification that both incorporates crystallographic orientations and has a rotationally invariant free energy is due to Kobayashi *et al.*[7] Various modifications of this model have been applied to describe the impingement of solidifying particles and grain boundary problems.[8−10] Granasy *et al*[11−13] adapted this model and incorporated the nucleation process. They calculated the properties of the critical fluctuations (nuclei) using the phase field theory and then placed such critical fluctuations randomly into the simulation window.

In this paper, we develop a phase field model for polycrystalline solidification based on the work of Kobayashi *et al*[7] and Granasy *et al.*[12,13] A simple Poisson seeding method[14] is used to handle nucleation. The anisotropic growth of multigrain in equiaxial solidification of a binary alloy is simulated. The results are discussed in terms of the formal theory of transformation kinetics (the JMAK theory).

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2. Methods

2.1. Formulation of the model

Following the procedure of Granasy *et al*,[12,13] we postulate the form for free energy functional as

$$F = \int \left[f(\phi, c) + \frac{\varepsilon^2}{2} |\nabla\phi|^2 + f_{\text{ori}} \right] \mathrm{d}v, \qquad (1)$$

where f is the free energy density that depends on phase field variable ϕ and concentration c. The second term in the integrand is a gradient correction to f. The orientational contribution f_{ori} is the excess energy due to inhomogeneities in crystal orientation in space, $f_{\text{ori}} = M[1 - P(\phi)]\,|\nabla\theta|$. In order to describe multigrain solidification in the presence of anisotropy, a non-conserved orientational field θ (a normalized orientational angle) is introduced, which is random in the liquid, and has a definite value between 0 and 1 in the crystal that determines crystal orientation in the reference frame. M is a scaling parameter. $P(\phi)$ is chosen to have the form $P(\phi) = \phi^3(6\phi^2 - 15\phi + 10)$, which equals 0 in the solid and 1 in the liquid. As a result, the excess energy acts in the solid only[9] and is proportional to $|\nabla\theta|$. The governing equations for the phase field and orientation field can be written as

$$\dot{\phi} = -M_\phi \frac{\delta F}{\delta \phi}, \qquad (2)$$

$$\dot{\theta} = -M_\theta \frac{\delta F}{\delta \theta}. \qquad (3)$$

The evolution of the concentration variable c is given by the conservation law and the irreversible thermodynamics linear law as

$$\dot{c} = \nabla \cdot \left(M_c \nabla \frac{\delta F}{\delta c} \right). \qquad (4)$$

In the above equations, M_ϕ, M_θ, M_c are corresponding mobilities. Evaluating the functional derivatives yields the following governing differential equations:

$$\frac{\partial \phi}{\partial t} = M_\phi[\nabla \cdot (\varepsilon^2 \nabla\phi) - (1-c)H^{\mathrm{A}} - cH^{\mathrm{B}} + M\,|\nabla\theta| \cdot P'(\phi)], \qquad (5)$$

$$\frac{\partial c}{\partial t} = -\nabla \cdot [-D_c \nabla c + M_c (H^{\mathrm{A}} - H^{\mathrm{B}}) \nabla\phi], \qquad (6)$$

$$\frac{\partial \theta}{\partial t} = M_\theta M \nabla \cdot \left[[1 - P(\phi)] \frac{\nabla\theta}{|\nabla\theta|} \right], \qquad (7)$$

where H^{A} and H^{B} are defined as

$$H^{\mathrm{A}} = P'(\phi) L^{\mathrm{A}} \left(\frac{1}{T} - \frac{1}{T_{\mathrm{m}}^{\mathrm{A}}} \right) + W^{\mathrm{A}} g'(\phi), \qquad (8)$$

$$H^{\mathrm{B}} = P'(\phi) L^{\mathrm{B}} \left(\frac{1}{T} - \frac{1}{T_{\mathrm{m}}^{\mathrm{B}}} \right) + W^{\mathrm{B}} g'(\phi). \qquad (9)$$

In the above equations, $L^{\mathrm{A,B}}$ and $T_{\mathrm{m}}^{\mathrm{A,B}}$ are the latent heat per unit volume and the melting temperature of the pure components A and B, respectively. $W^{\mathrm{A,B}}$ is the well height. The solute diffusivity is defined as $D_c = D_{\mathrm{l}} P(\phi) + D_{\mathrm{s}}(1 - P(\phi))$, where D_{l} and D_{s} denote the diffusivities in the liquid and in the solid, respectively. M_c depends upon c and ϕ as $M_c = \frac{v_{\mathrm{m}}}{R} c(1-c) D_c$, where v_{m} is the molar volume and R is the gas constant.

In order to incorporate interfacial free energy anisotropy in the model, we express the gradient coefficient ε as $\varepsilon = \bar{\varepsilon}\,[1 + \gamma \cos k(\sigma - \vartheta)]$, in which $\bar{\varepsilon}$ is the mean value of ε, σ is the angle between the normal to the interface and the x-axis, $\sigma = \arctan\left[\frac{\partial\phi/\partial y}{\partial\phi/\partial x}\right]$, $\vartheta = 2\pi\theta/k$, which is the angle between the crystallographic axis and the x-axis. k is the rotational symmetry of the crystal and γ is the magnitude of anisotropy.

2.2. Modelling of nucleation

Crystal nucleation can be modelled by including white noise into the governing equations. But this method requires sampling at a very high frequency in order to observe nucleation events, which is often prohibitively time consuming. In addition, the proper amplitude of the noise is difficult to determine which both can trigger nucleation and does not differ from the real critical fluctuations too much. To avoid the difficulties associated with triggering nucleation by noise, in this work, nucleation is modelled by randomly placing supercritical particles in the simulation domain.[14] Nuclei are introduced into individual cells randomly, but maintaining the desired nucleation rate. Nucleation and growth are treated as two separate stages. After nucleation, growth ensues without any new nucleus introduced.

3. Results and discussion

Isothermal computations are performed using the model described above. The familiar Ni–Cu alloy is considered in this work because its physical parameters are available. The material parameters used are as follows (A–Ni, B–Cu): T_{A}=1728K, T_{B}=1358K, $L_{\mathrm{A}} = 2350\mathrm{J/cm^3}$, L_{B}=1728J/cm^3, $D_{\mathrm{l}} = 10^{-5}\mathrm{cm^2/s}$, $D_{\mathrm{s}} = 10^{-9}\mathrm{cm^2/s}$, surface tension $\sigma_{\mathrm{A}} = 3.7 \times 10^{-5}\mathrm{J/cm^2}$, $\sigma_{\mathrm{B}} = 2.9 \times 10^{-5}\mathrm{J/cm^2}$, interface thickness $\delta = 5 \times 10^{-6}$cm, mesh size $\Delta = 3.5 \times 10^{-6}$cm, magnitude of anisotropy γ=0.05, magnitude of noise

α=0.3. The computational domain size is 1500×1500 in terms of mesh grid number.

3.1. Morphological evolution of multigrain solidification

Figure 1 shows the microstructural development of multigrain solidification. At early stage, the crystallites with different orientations grow independently, and the solute rejected by the solidified crystallites diffuses unimpeded. With growth of the crystallites, some of them begin to interact through their diffusion fields, a phenomenon termed "soft impingement". The growth of the soft impinged dendrite trunks and arms stagnates, but the others unblocked continue growing. The solute rejected piles up in the interdendritic zone, which hinders the solidification of the isolated liquid there. It should be noted that duo to the fourfold symmetry, orientations of crystallites that differ by 90° multiples are equivalent.

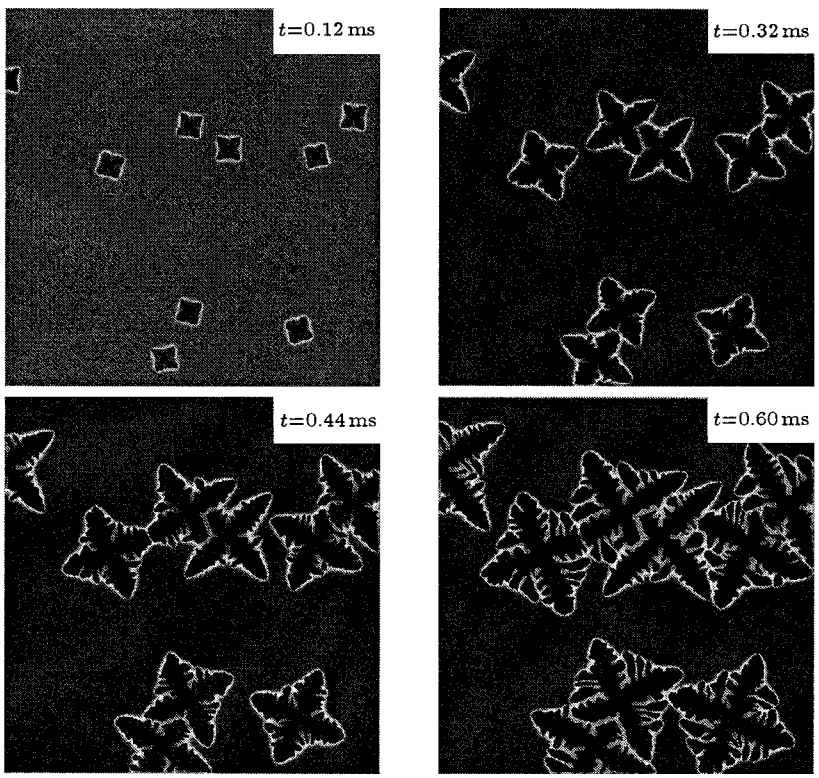


Fig.1. Morphological evolution of multigrain solidification.

3.2. Discussion

The multigrain solidification processes with different compositions are simulated (Fig.2). The melt temperature is T_0=1543K for all cases. The compositions are c_0=0.4946 at.%, c_0=0.4900 at.% and c_0=0.4875 at.% for simulations (a) , (b) and (c), respectively. Same numbers of nuclei are planted in the simulation window at the start for the three simulations. Because a smaller composition corresponds to a larger undercooling for Ni–Cu alloy, so that with decreasing the composition, more nuclei can retain and grow up. For simulation Fig.2(a), most nuclei disappear and the residual nuclei have enough space to grow, as a result the crystallites develop into full dendritic morphology. For simulation Fig.2(b), more nuclei are retained than Fig.2(a). The dendrites are not well developed because of the lack of space. In Fig.2(c), even more nuclei are retained and the shape of the grains is compact.

According to the JMAK theory for a nucleation-growth process, the transformed fraction follows the relation

$$X(t) = 1 - \exp\{-(t/t_0)^p\}, \tag{10}$$

in which $X(t)$ is the transformed fraction, t is transformation time and t_0 is a constant parameter. The "Avrami exponent" p is representative of the phase transformation mechanism and can be evaluated from

the slope of the plot ln[-ln(1-X)] vs ln$t$,

$$p \ln t = \ln[-\ln(1.0 - X(t))] + K_0. \qquad (11)$$

The Avrami exponents as functions of the normalized solidification fraction $X(t)/X_{\max}$ are presented in Fig.2(d–f), where $X_{\max}$ is the final solidification fraction. For diffusion controlled growth with zero nucleation rate, the Avrami exponent p is expected to be 1.5.[15] For the simulation Fig.2(a), the mean value of p is about 1.8 (Fig.2(d)), very close to 1.5. For simulation Fig.2(b), the exponent is about 2.2 due to the intensive interaction between the particles. For simulation Fig.2(c), the corresponding Avrami exponent decreases with increasing the transformed fraction (Fig.2(f)). Because of the large initial supersaturation, the composition of the solid formed at early stage is close to that of the liquid, so that the growth at early stage is interface controlled, as opposed to diffusion controlled. This results in a delay of the onset of diffusion controlled growth, which causes the value of p to decrease with time. This effect is prominent only in case of large initial solute supersaturation and abundant nuclei, where this transient period is comparably long with respect to the total solidification time. Comparing Fig.2(d), Fig.2(e) and Fig.2(f) indicates that the Avrami exponents increase with decreasing composition.

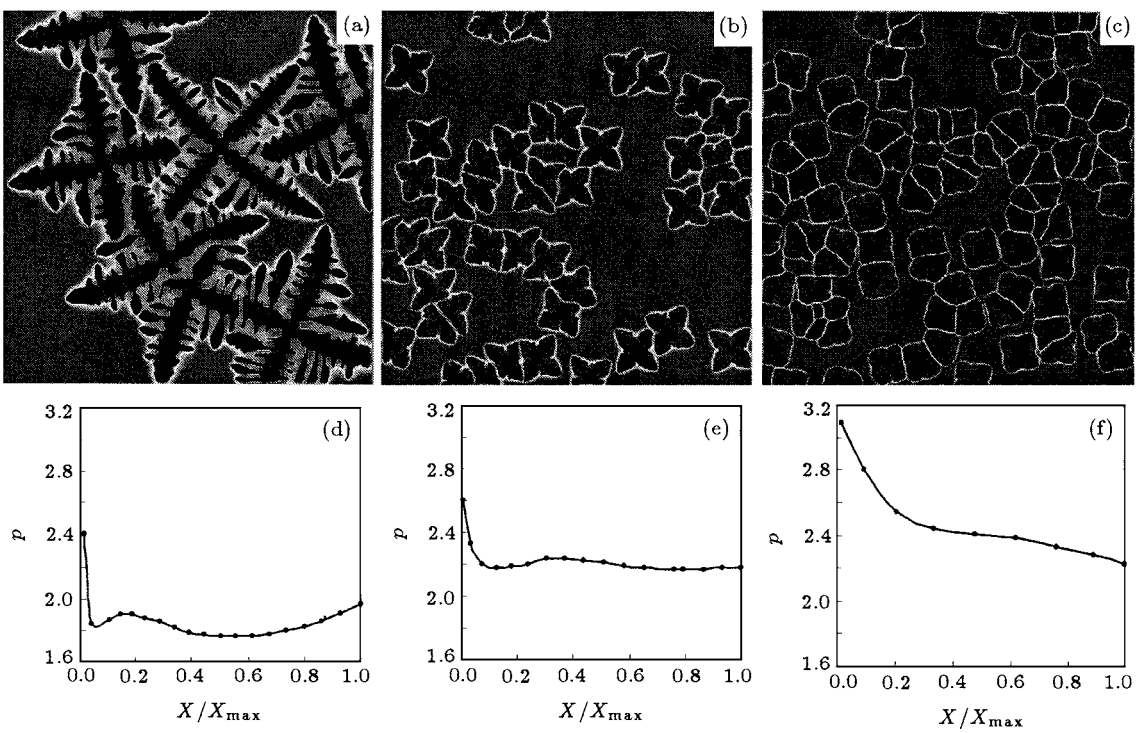


Fig.2. Multigrain solidification with different compositions. (a)–(c) Concentration distribution for c_0=0.4946 at.%,c_0=0.4900 at.% and c_0=0.4875 at.% respectively. (d)–(f) The Avrami exponents versus normalized transformed fraction corresponding to (a)–(c) . $X_{\max}$ is the final solidification fraction. The melt temperature T_0=1543K.

4. Conclusions

A phase field model that restores the rotational invariance of the free energy is presented and used to simulate the anisotropic growth of multigrain in solidification of a binary alloy. It is indicated that the phase field model is able to describe the complex morphologies in multigrain solidification. The soft impingement phenomenon is observed in the simulation. Due to the interaction between crystal grains, the Avrami exponent varies with the initial melt composition and the solidification fraction.

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