Simulation on the instability of a solid–liquid interface from a molar flux with a diffuse interface layer

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Abstract

A simulation method based on a diffuse-interface-flux model is proposed for the pattern formation during crystal growth. A numerical scheme was extended for the general treatment of solidification from simple analytical conditions. A diffuse interface with width δ is assumed between the solid and liquid phases. The molar flux between solid and liquid is balanced at the diffuse interface layer as a field of diffusion. The shape of the interface proceeds under the phase transition of the interface layer to solid or liquid. To ascertain the validity of the method, uniaxial solidifications of dilute binary alloy were simulated to observe some morphological transitions from planner to cellular interface. The wavelengths of cellular interface agree well with those from the Mullins–Sekerka model. © 2002 Elsevier Science B.V. All rights reserved.

1. Introduction

Since pattern-formation at the solid–liquid interface is caused from energy dispersion under non-equilibrium conditions, flux balance at the interface is the key factor to understand the mechanism on solidification. Under the controlled uniaxial solidification, when a one-sided model holds [1], molar flux balance is more essential than heat flux balance. Because constitutional undercooling causes the planner-to-cellular transition of the interface [2], behavior of the molar flux might be the mechanism in this morphological transition [3].

To clarify the planner–cellular transition at the solid–liquid interface of binary alloy, the author has investigated the balance of two molar fluxes, one is caused from constitutional undercooling and the other is from interfacial diffusion within the diffuse interface layer [3]. This analytical model, named diffuse-interface-flux model, can quantitatively explain the interfacial instability, which well agrees with that from the Mullins–Sekerka model; such as the relation among wavelength, solute concentration and growth rate without concerning Gibbs–Thomson effects explicitly. This result suggests that the diffuse-interface-flux model has a potential to clarify the free boundary problems in a more direct way.

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In this study, we propose a new simulation method based on the diffuse-interface-flux model. Numerical scheme and boundary condition are extended from the simple analytical treatment for the calculation of the general morphological studies. To ascertain the validity of the method we simulate the morphological instability of a solid–liquid interface in a binary alloy.

2. Numerical method

2.1. Numerical scheme

The evolutional equation is calculated with explicit solution from the diffusion equation by the finite difference method except at the interface. Simulation size is $15\,\mu m$ in the direction of temperature gradient, x-axis, and $5\,\mu m$ in the direction of the vertical y-axis. At the initial condition, planner interface is set parallel to the y-axis. Additionally, the solid phase with concentration kc_0 occupies $\frac{1}{3}$ of the whole system and the liquid phase with concentration c_0 occupies $\frac{2}{3}$. Size of regular grid in both direction, Δx or Δy, is $\Delta u = 0.1\,\mu m$. Like phase-field model, each grid belongs to either solid, liquid or interface. The width of the interface layer, δ, is always fixed constant as one (or two) grid size, $\delta = \Delta u$ (or $2\Delta u$). Although δ should be several nm as the diffuse interface, it remains large, since a thin diffuse interface layer requires large computational power. Unit time, $\Delta t = 10^{-7}\,s$, is determined to realize convergence and stability of the calculations in the explicit solution of the diffusion equation. Periodic boundary condition is set at y direction and the concentration at the edge of the liquid is set constant, c_0.

2.2. Physical model of simulation

Simulation algorithm on uniaxial solidification based on the diffuse-interface-flux model is expressed in the following steps. At first, the initial condition is given. In the next, after the time evolution of the solute concentration, the phase of each grid is determined. Then, the whole system is moved into the x direction under the constant

growth rate, adding a new liquid layer with concentration c_0 at the front of the liquid. These steps are repeated until a certain number of times, N_{step}. Concentration of each grid is calculated from the continuity equation of molar fluxes. Within the same phase, the molar flux is merely a diffusive transport. Molar flux within the liquid (solid, interface) is $\mathbf{j}_{ll} = -D_l\nabla c_l$ ($\mathbf{j}_{ss} = -D_s\nabla c_s$, $\mathbf{j}_{ii} = -D_i\nabla c_i$), where D_l (D_s, D_i) is the diffusion coefficient in the liquid (solid, interface), and c_l (c_s, c_i) is the concentration of solute in the phase.

Between the liquid and the interface, two molar fluxes exist; $\mathbf{j}_e$ caused from the phase equilibrium and $\mathbf{j}_i$ caused from the free energy difference under constitutional undercooling [3]:

$$\mathbf{j}_e = -D_i\,\frac{c_l - c_i/k}{\Delta u}. \tag{1}$$

$$\mathbf{j}_i = -\frac{c_i D_i \Delta H_f}{R T_i T_f}(m_l G_c - G)\mathbf{n}, \tag{2}$$

where R is the gas constant, H_f is the latent heat of fusion per mole, T_f is the melting point, T_i is the temperature at the solid–liquid interface, $\mathbf{n}$ is the normal unit vector at the interface, m_l is the liquidus slope, $G_c (= \nabla c_l = \Delta c_{lx}/\Delta x + \Delta c_{ly}/\Delta y)$ is the interfacial concentration gradient in liquid and $G (= \Delta T_i = \Delta T_x/\Delta x + \Delta T_y/\Delta y)$ is the interfacial temperature gradient.

The molar flux between solid and interface is merely a diffusive transport,

$$\mathbf{j}_{si} = -D_i\,\frac{c_s - c_i}{\Delta u}. \tag{3}$$

Phase is determined from the evaluation function, $f_{ph}(T, c)$, in regard of molar depression of freezing point. However, Gibbs–Thomson effect is not explicitly evaluated in the function itself. Effect of interfacial energy is implicitly involved within the solute balance at the interface layer:

$$f_{ph}(T, c) = T_f + m_s c_i - T, \tag{4}$$

where m_s is the solidus slope. To consider the effect of latent heat and to prevent oscillatory phenomena during phase transition, the degree of undercooling, ΔT, is also set. In case $f_{ph}(T, c) > \Delta T$, phase is solid, and in case $f_{ph}(T, c) < 0$, phase is liquid, and otherwise, $0 \leqq f_{ph}(T, c) \leqq \Delta T$, the phase will not be transformed to remain the same

as before. In order to consider the effect of thermal fluctuation, the solute concentration of liquid slightly and randomly varies with $\Delta c < c_0/1000$ at a middle layer.

2.3. Specific parameter values

In the present calculation, we simulate two-dimensional uniaxial solidification on a Cu–Ag eutectic system with dilute Ag concentration. Specific parameter values of the simulation on Cu–Ag system are the following: melting point, $T_f = 1358$ K, latent heat of fusion per mole, $\Delta H_f = 2.09 \times 10^5$ J, $m_l = -30$ K/mol, $k = 0.25$, $D_l = 10^{-9}$ m^2/s, $D_i = 10^{-9}$ m^2/s, $D_s = 10^{-13}$ m^2/s and $\Delta T = 1$ K. Since the layer of diffuse interface is much thicker than the real one, the present interfacial energy per mol, $\gamma_m = RT_f\delta$, which is obtained from the density functional theory [4], is almost 50 times overestimated. To simulate planer-to-cellular transition at the interface, several parameters, initial concentration of Ag; $c_0 = 0.001 \sim 0.2$ mol, growth rate of solidification; $V = 0.0001 \sim 0.001$ m/s, and temperature gradient; $G = 2000 \sim 20\,000$ K/m, are given. N_{step} is 10^6 steps, which means simulation time is equivalent to 0.1 s.

3. Numerical results and discussions

3.1. Profile of solute concentration

During the uniaxial solidification, except at the initial stage, the position and the concentration of the interface become in the steady state. Because of the excess solute rejected from the solid, concentrations of the liquid and solid are piled up to be increasing into c_0 and c_0/k, respectively. As shown in Fig. 1, since the simulated characteristic diffusion length in the liquid, $d_D = 3.97$ μm, is equivalent to that obtained from analytical theory, $D/kV = 4.00$ μm [2], solute profile shows reasonable results in the present calculation.
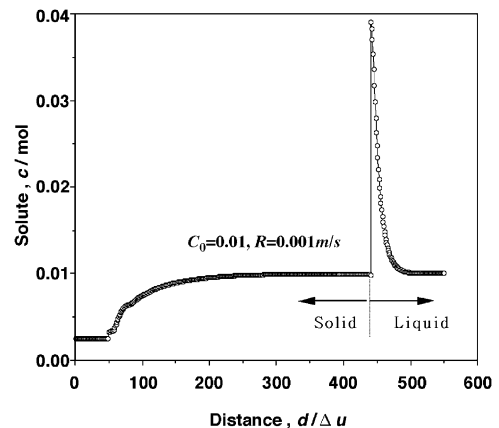

Fig. 1. Distribution profile of solute concentration along the temperature gradient axis.

3.2. Solute distribution at the interface

Like experimental observations during planner to cellular transition, solute distribution at the interface is expected to be localized. Fig. 2 shows solute distribution of the interface layer at the instability criterion under the constant growth rate $V = 0.001$ m/s, and the temperature gradient $G = 2000$ K/m. When $c_0 = 0.01$, interfacial concentration was kept constant. However, it became periodically fluctuating with wavelength $\lambda = 2.7$ μm (or 1.9 μm), when $c_0 = 0.1$ (or 0.2). The interfacial instabilities were not observed without the concentrational fluctuation in the liquid. Since the both wavelengths well agree with the analytical results from the Mullins–Sekerka model ($\lambda = 2.8$ μm (or 2.0 μm) at $c_0 = 0.1$ (or 0.2)) [5], the present method indicates the ability to simulate the instability of solid–liquid interface. Other morphological phenomena of phase transition are expected to be understood in the same manner.

3.3. Oscillation of concentration under the interfacial instability

As shown in Fig. 3, fluctuation of the interfacial concentration occurs suddenly during the uniaxial crystal growth, when the interfacial concentration reaches the instability criterion. Since the
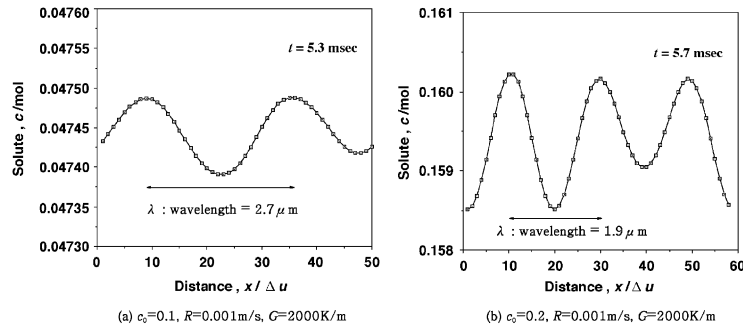
(a) c_0=0.1, R=0.001m/s, G=2000K/m   (b) c_0=0.2, R=0.001m/s, G=2000K/m

Fig. 2. Distribution of solute concentration at the solid–liquid interface layer.
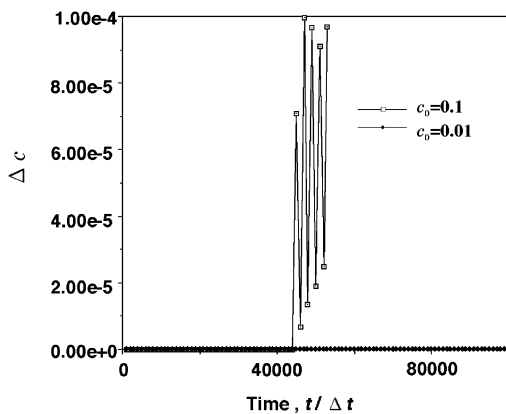

Fig. 3. Time evolution of the amplitude of solute concentration at the solid–liquid interface layer.

fluctuation increases drastically, it limits the ability of the present calculation within the initial transition of the instability. This oscillation does not affect the results of the wavelength and the diffusion length of the simulation. The increase of the amplitude on the solute concentration might be caused by the frequent molar flux flows between the diffuse interface layer and the liquid. This numerical problem might be solved by treating the interface as a continuous layer, such as the phase-field parameter, ϕ, or by regarding the effect of latent heat at the interface more precisely. Since the time fluctuation is accompanied with the spatial instability, the oscillation might be one of

the important features during the interfacial instability.

4. Conclusions

Based on the diffuse-interface-flux model, a new simulation method is proposed to investigate the interfacial instability during the uniaxial solidification of binary alloy. The following results were obtained;

(1) Analytical diffuse-interface-flux model can well be translated into the simulation method to study more general phenomena in solidification.
(2) Criterion and the wavelength of interfacial instability are well simulated from the present numerical scheme.
(3) Interfacial concentration shows abrupt increase and frequent oscillation at the instability criterion.

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