Phase-field Model for Solidification of Ternary Alloys

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The phase-field model for dilute ternary alloys is proposed and the relationships between phase-field parameters and material properties are derived at a thin interface limit. One-dimensional and two-dimensional numerical simulations show that the model reproduces the equilibrium conditions including Gibbs–Thomson effect. We apply the model for ternary alloys to micro-segregation analysis in Fe–C–P alloy and the results are compared with analytical models. The results show good agreement with those of Clyne–Kurz equation.

For two-dimensional computations, isothermal dendritic growth is simulated for Fe–C and Fe–C–P alloys. The change in phosphorous concentration affects significantly the interface velocity and the dendrite shape even when the phosphorous concentration is much lower than that of carbon because of the small diffusivity of phosphorous.

KEY WORDS: phase-field model; thin interface limit; ternary alloy; micro-segregation; isothermal dendrite.

1. Introduction

The phase-field model is becoming a powerful tool which can describe the complex interface pattern evolution. This method treats the interface as a finite area where the phase changes from solid to liquid gradually. Hence all the governing equations can be written in unified forms without distinguishing the interface and it is a great advantage in numerical calculation.

The phase-field models have been originally developed for solidification of pure materials1–9) and then extended to that of binary alloys.10–20) The first model for alloy solidification was due to Wheeler et al. (WBM model).15,16) They have defined the free energy density for binary alloys by assuming the interface as a mixture of solid and liquid phases with a same composition. The diffusion equation is derived in order to describe solute diffusion in matrix and solute redistribution at interface. In the WBM model, phase-field parameters are derived at a sharp interface limit. Hence the fine calculation mesh is required for quantitative calculation to reduce the error in the driving force for solidification.

Karma and Rappel21) have determined the phase-field parameters for pure materials using a thin interface limit condition, in which temperature varies in the interface region. Since the error in driving force disappears, the model enables us to use large calculation mesh size and an arbitrary value of kinetic coefficient. The thin interface limit condition has been applied to the WBM model, too.22) It has been also shown that the WBM model can reproduce the solute trapping phenomena at the high interface velocity.22–24) A problem in the WBM model is that the parameters vary depending on the calculation mesh size. In the free energy curves of the solid and liquid in the interface region, there exists an extra potential profile relating with the difference between free energy curves and the common tangent line. This extra potential in the WBM model may be negligible compared with the imposed double-well potential at either sharp interface limit or in an alloy with a very small difference of solid and liquid equilibrium compositions where the height of the extra potential itself is small. For most alloys, however, the extra potential is comparable with the imposed double-well potential even in a rather dilute alloy. Hence the extra potential can bring an obstacle preventing computational efficiency.25)

On the other hands, Tiaden et al. have proposed the model in which the interface is assumed as a mixture of liquid and solid at different concentrations.23) Kim et al. have applied the idea to their thin interface limit model in a thermodynamically consistent way.25) The model is equivalent with the WBM model but has a different definition of the free energy density in the interface. Since the extra potential in the WBM model disappears in the model because the free energy function corresponds to the common tangent line itself, it enables us to make calculation with large mesh size. Furthermore, it is possible to use vanishing kinetic coefficient by adjusting the parameters with the any mesh size. So far, however, the application of the model is limited to binary alloys though most of the important industrial materials are ternary or multi-component alloys.

In this study, we extend the phase-field model by Kim et al. to ternary alloys applying a dilute solution approximation for the free energy density. The relationship between
phase-field parameters and material properties are determined at a thin interface limit. We examine whether the model reproduces the equilibrium state given by the phase diagram in the 1-D calculations. Then we estimate the Gibbs–Thomson coefficient at different temperatures and compositions in the 2-D calculations. Finally, we show the application examples of the ternary phase-field model of micro-segregation and isothermal dendrite analysis in Fe–C–P alloy.

2. Phase-field Model for Ternary Alloys

2.1. Governing Equations

We use a dilute solution approximation to define the free energy. In a dilute ternary alloy with concentrations \(c_1\) and \(c_2\), chemical potentials of the elements \(\mu_1\), \(\mu_2\), and \(\mu_3\) are expressed,

\[
\mu_1^{S(c_1)} = \mu_1^{OS} + \frac{RT}{V_m} \ln(\gamma_1 c_{1S})
\]

\[
\mu_2^{S(c_2)} = \mu_2^{OS} + \frac{RT}{V_m} \ln(\gamma_2 c_{2S})
\]

\[
\mu_3^{S(c_1, c_2)} = \mu_3^{OS} + \frac{RT}{V_m} \ln(1-(c_{1S}+c_{2S}))
\]

where the subscripts of 1 and 2 represent the solute species and subscript 3 represents the solvent. The superscripts \(L\), \(S\) and \(O\) represent liquid, solid phases and the standard state, respectively. \(\gamma\) is the activity coefficient which will be eliminated in the process of deriving the governing equations. The equilibrium conditions are,

\[
\mu_1^{OS} + \frac{RT}{V_m} \ln(\gamma_1 c_{1S}) = \mu_1^{OL} + \frac{RT}{V_m} \ln(\gamma_1 c_{1L})
\]

\[
\mu_2^{OS} + \frac{RT}{V_m} \ln(\gamma_2 c_{2S}) = \mu_2^{OL} + \frac{RT}{V_m} \ln(\gamma_2 c_{2L})
\]

\[
\mu_3^{OS} + \frac{RT}{V_m} \ln(1-(c_{1S}+c_{2S})) = \mu_3^{OL} + \frac{RT}{V_m} \ln(1-(c_{1L}+c_{2L}))
\]

where subscript \(e\) represents the equilibrium value.

The free energy density is the sum of the free energy for liquid and solid bulk phases and an imposed double-well potential, which can be written as

\[
f = h(\phi)(\phi^5(c_{1S} + c_{2S}) + (1-h(\phi))\phi^4(c_{1L} + c_{2L}) + Wg(\phi)
\]

\[
= h(\phi)[c_{1S}\mu_1^{OS} + c_{2S}\mu_2^{OS} + (1-c_{1S} + c_{2S})\mu_3^{OS}]
\]

\[
+ (1-h(\phi))[c_{1L}\mu_1^{OL} + c_{2L}\mu_2^{OL} + (1-c_{1L} + c_{2L})\mu_3^{OL}]
\]

\[
+ Wg(\phi)
\]

where \(\phi\) is phase-field \((\phi=0\) in liquid and \(\phi=1\) in solid, respectively), \(t\) is time, the subscript \(i\) denote the partial derivative, and \(M\) and \(\varepsilon\) are phase-field parameters which will be determined later.

The solute concentration, \(c_j\), in a \(n\)-component alloy is given by,

\[
\sum_{j} c_j = 1
\]

\[
\frac{\partial c_j}{\partial t} = -\nabla J_{c_j}
\]

where subscript \(j\) means alloy component, \(\mu_j\) is chemical potential, \(V_j\) is molar volume, \(n\) denotes the solvent and \(L_{jk}\) is a positive constant associated with diffusivity of the solute. We input the term \(V_j/\bar{V}_n\) in the right hand of the equation is equal to zero for an interstitial diffusion. The flux of \(j\) component depends only on the chemical potential gradient of the \(j\) component itself because the cross-term in the concentration field equation is negligible in case of dilute solution. Hence the concentration field equation for a dilute ternary alloy is given by,

\[
\frac{\partial c_j}{\partial t} = \nabla \left( D_j(\phi) \frac{\nabla f_{c_j}}{f_{c_j}} \right) \tag{4}
\]

\[
D_j(\phi) = D_j^0 (\phi > 0.9), \quad D_j(\phi) = D_j^0 (\phi \leq 0.9)
\]

where subscripts under \(f\) mean the first or the second partial derivatives by \(c_j\), \(D^0\) and \(D^1\) are solute diffusivity in the liquid and solid phases, respectively. The functions \(f_{c_j}\) and \(f_{c_j}\) are given by differentiating Eq. (2) by \(\phi\) and \(c_j\) respectively. Present model is composed of Eqs. (3) and (4) together with the mass conservation condition and the equal chemical potential condition

\[
f_j^L(x) = f_j^S(x), \quad j=1, 2 \tag{5}
\]

\[
c_j = h(\phi)c_j^0 + (1-h(\phi))c_j^f \tag{6}
\]

The explicit forms of the terms \(f_{c_j}\), \(f_{c_j}\) and \(f_{c_j}\) are dependent on the definition of the free energy density for the interface region. Following the similar way with Ref. 26), they can be expressed explicitly in terms of compositions at the interface \(c_j^L, c_j^S\) and \(\phi\). For a dilute ternary alloy, the resulting phase-field and solute-field equations are given by

Phase-field equation:

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

\[
+ \frac{RT}{V_m} \ln \left( \frac{1-(c_{1S}+c_{2S})}{(1-(c_{1L}+c_{2L}))} \right) H(\phi) - Wg'(\phi)
\]

\[
\left. \right) \tag{7}
\]

where \(\phi\) is phase-field \((\phi=0\) in liquid and \(\phi=1\) in solid, respectively), \(t\) is time, the subscript \(i\) denote the partial derivative, and \(M\) and \(\varepsilon\) are phase-field parameters which will be determined later.
Concentration field equations:

\[
\frac{\partial c_1}{\partial t} = \nabla \left[D_1(\phi) \left[1-h(\phi)\right] \frac{c_{1L}(1-c_{1L}+c_{2L})}{1-c_{2L}} \right] + \frac{h(\phi)}{1-c_{1S}} \left(1-c_{1S}+c_{2S}\right) \nabla \ln \frac{c_{1L}}{1-(c_{1L}+c_{2L})}
\]

\[
\frac{\partial c_2}{\partial t} = \nabla \left[D_2(\phi) \left[1-h(\phi)\right] \frac{c_{2L}(1-c_{1L}+c_{2L})}{1-c_{1L}} \right] + \frac{h(\phi)}{1-c_{1S}} \left(1-c_{1S}+c_{2S}\right) \nabla \ln \frac{c_{2L}}{1-(c_{1L}+c_{2L})}
\]

(8)

2.2. Phase-field Parameters

In the phase-field equation, there are three unknown parameters of \( M, \varepsilon \) and \( W \). The \( W \) and \( \varepsilon \) must be matched with the interface energy \( \sigma \) and the interface thickness \( 2\lambda \). 1-D equilibrium solution \( \phi_0(x) \) of the phase-field Eq. (7) is

\[
\phi_0(x) = \frac{1}{2} \left[ 1 - \tanh \left( \frac{\sqrt{W} x}{\sqrt{2 \varepsilon}} \right) \right] \quad \text{..............(9)}
\]

When the interface region is defined as \( 0.1 < \phi < 0.9 \), interface thickness and interface energy are obtained from Eq. (10) and Eq. (11), respectively.

\[
2\lambda = \int_{0}^{0.9} \frac{dx}{d\phi_0} = \frac{\varepsilon}{\sqrt{W}} \quad \text{..............(10)}
\]

\[
\sigma = \varepsilon \int_{-\lambda}^{\lambda} \left( \frac{d\phi_0}{dx} \right)^2 dx = \frac{\varepsilon \sqrt{W}}{3 \lambda} \quad \text{..............(11)}
\]

The phase-field mobility \( M \) is related to the interface kinetic coefficient, that is, the ratio of interface velocity to the driving force of solidification. To find the relationship at the thin interface limit condition, we rewrite the governing equations in 1-D steady state

\[
-v_n \frac{d\phi}{dx} = \varepsilon \frac{d^2\phi}{dx^2} + h(\phi) \left[f_{c_1}(c_{1L}, c_{12}) - f^S_{c_1}(c_{1S}, c_{2S})\right] - f_{c_2}(c_{1L}, c_{12}) - f^S_{c_2}(c_{1S}, c_{2S}) - \nabla \cdot \left( \nabla \phi \right)
\]

(12)

\[
v_n \frac{dc_j}{dx} = \frac{d}{d\phi} \left( D_j(\phi) \frac{d\phi}{dx} f_{c_j} \right) \quad \text{..............(13)}
\]

where \( v_n \) is the interface velocity. In order to find the interface velocity at the thin interface limit condition where the diffusion length \( D/v_n \) is much larger than the interface width \( 2\lambda \), we can straightforwardly follow the way described in Ref. 26. By integrating Eq. (13) assuming the solute diffusion in solid is negligible, we can get the chemical potential profiles in the interface region,

\[
f_{c_j}(x) = f^S_{c_j}(c_{1S}(x)) = f^S_{c_j}(c_{2S})
\]

\[
-v_n \int_{x_j}^{x} \frac{f_{c_j}}{D_j(\phi)} \left[ c_{j}(x) - c_{j}^0 \right] dx \quad \text{.............(14)}
\]

By substituting the Eq. (14) into the Eq. (12) and by integrating Eq. (12) twice, we can get the following equation to the first order in \( 2\lambda \) and \( v_n/D \),

\[
\alpha_v = \frac{RT}{\varepsilon^2} \left[ c_{1S}(1-k_1^e)+c_{2S}(1-k_2^e) \right] - \left[(1-k_1^c)c_{1S}^e+(1-k_2^c)c_{2S}^e\right] \quad \text{.............(15)}
\]

where the \( c_j^e \) represents the interface concentration at liquid or solid side and \( \alpha \) is defined by

\[
\alpha = \frac{\sigma}{M\varepsilon} = \frac{\varepsilon}{\sqrt{2W}} \sum \frac{1}{D_i}(\phi_j, c_j^5) \quad \text{.............(16)}
\]

\[
\zeta_j = \frac{RT}{\varepsilon^2} (c_j^5 - c_j^0)^2
\]

\[
\times \int_{h(\phi_0)}^{-1} \frac{\left[1-h(\phi_0)\right]}{\phi_0(1-\phi_0)} d\phi_0 \quad \text{.............(17)}
\]

and where \( k_j^e \) is equilibrium partition coefficient. If we apply a vanishing kinetic coefficient, that is \( \alpha = 0 \), the phase-field mobility is expressed in

\[
\frac{1}{M} = \frac{\varepsilon^3}{\sigma \sqrt{2W}} \left( \frac{1}{D_i} \zeta_i(c_{1S}, c_{1S}) + \frac{1}{D_{2i}} \zeta_2(c_{2S}, c_{2S}) \right)
\]

3. Numerical Calculations

3.1. Thermodynamics Data

To obtain the phase diagram for a ternary alloy, we assume the equilibrium partition coefficients in the ternary alloy system are the same as those of binary alloys. Hence we can get the relationship between \( c_{1S}^e \) and \( c_{2S}^e \), and also \( c_{1S}^5 \) and \( c_{2S}^5 \), under the ideal dilute solution approximation condition. Then, for determining the thermal dependence of the liquidus and solidus line, we assume the \( c_{1S}^e \) of the ternary alloy at \( c_{1S}^e = 0 \) is equal to that of the corresponding

![Fig. 1. Phase diagram of Fe–C–P alloy and calculated equilibrium concentrations.](Image)
3.2. Calculation Procedure

To calculate the governing equations, there are seven unknown values. The three of them are phase-field parameter. Both of W and $\varepsilon$ are determined by solving the Eqs. (10) and (11) simultaneously. Since the Phase-filed mobility is a function of temperature, it should be calculated with the temperature during the computation. The value of solute concentration in liquid $c_{\text{L}}$ and solid $c_{\text{S}}$ are also required. They are determined from Eqs. (5) and (6) depending on $\phi$ and $c_j$ at each point and at every time step.

3.3. Micro Segregation Analysis

The governing equations, Eqs. (7) and (8), are numerically solved using a finite difference scheme. In the calculations the system temperature is uniform and continuously decreased with a constant cooling rate from the initial temperature of 1779 K, which is slightly lower than the liquidus temperature of the Fe–0.5mol%C–0.02mol%P alloy. The constant values of cooling rate $T$ and primary dendrite arm spacing $\lambda$ were determined using the experimental data on carbon steels$^{29)}$ and the following equations,

$$\tilde{T} = GR \quad \lambda = AG^{1/2}R^{-1/4}$$

where $G$ is a temperature gradient and $R$ is an interface velocity. With the experimental value of $G$ as 3400 K/m and the value of $A$ determined from the experimental data, $\tilde{T}$ and $\lambda$ are estimated as 4.6 K/s and 100 $\mu$m respectively. We also calculate the case of $T=18$ K/s for comparison. The calculation area is divided into 200 meshes and a small solid area of ten meshes is set as the initial solid. The interface width was prepared to be six times of the mesh size.

3.4. Isothermal Dendrite Growth

For the simulation of isothermal dendrite growth the phase-field equation is modified so as to include the anisotropy of interface energy.$^{4)}$ The anisotropy is introduced by putting the coefficient in the phase-field parameter as followings,

$$\varepsilon' = \varepsilon \{ 1 + v \cos(k\theta) \}$$

where $v$ is the magnitude of anisotropy, $k$ is the mode number and $\theta$ is the angle between the direction of the phase-field gradient and the reference axis of the system. We use $v=0.03$ and $k=4$ in the calculation. During the calculation a stochastic noise is imposed at the interface in order to simulate the random fluctuations, which give the well-developed secondary arms. We impose the noise on the liquid composition in the vicinity of the interface. The noise level is set to vary within 1% of liquid composition at each grid, with the way conserving the masses of all the species. The calculation area of 500×1000 meshes is prepared and a small triangle solid of 20×20 meshes is initially put at the corner of the area. The mesh size is $1.0 \times 10^{-5}$ m and the system temperature is 1780 K.

4. Result and Discussion

4.1. Equilibrium States

In order to test the model, we examined the equilibrium solute concentrations in 1-D calculation at different temperatures. The symbols in the Fig. 1 are the calculated equilibrium liquid and solid concentrations for Fe–0.5mol%C–0.02mol%P alloy at different temperatures. The differences between the calculated solute concentrations and the values evaluated from the phase diagram are within the error of 0.1%. In 2-D simulations we can examine whether the model reproduces the Gibb–Thomson condition properly. In the calculations no anisotropy and fluctuation were imposed. We put a small initial solid area at the center of the calculation area and continued the calculation until the equilibrium state would be attained. The final shape of the solid becomes a circle and the equilibrium concentrations at the corner of the area. The mesh size is $1.0 \times 10^{-5}$ m and the system temperature is 1780 K.

4.2. Micro Segregation

The results of micro segregation calculation are shown in this section. Figure 3 shows the change in solid fraction with time for the case with cooling rate of 4.6 and 18 K/s. The solid fraction increases rapidly in the early stage of the solidification and its increasing rate decreases gradually. It is due to the decrease of the phase-field mobility with the temperature. In a usual micro-segregation analysis the solid fraction is assumed to be proportional to time or the square root of time. In the present phase-field calculations, the
movement of the interface is self-consistently determined by the thermodynamic driving force and no assumption on growth rate is made. The results show that the simple assumption of linear or parabolic growth is not applicable.

We compare the results of phase-field calculation with the predictions by the Scheil’s equation and by the Clyne–Kurz’s equation. The parameter $a$ in the latter are 0.487 for carbon and 0.150 for phosphorus. The calculated profiles for carbon are in good agreement with that by Clyne–Kurz’s equation as shown in Fig. 4. It is because the solute diffusion in liquid is fast enough. The calculated profiles for phosphorous in Fig. 5 agree better with the Clyne–Kurz’s equation than Scheil’s equation when the solid fraction is above 0.5. The difference in early stage of solidification would be the effect of the initial transient because of small diffusivity.

Figures 6 and 7 show the concentration profiles of carbon and phosphorus, respectively, when the 95% of the area is solidified. The straight lines in the figures represent the $c_s/k_{eq}=1$ and $1/k_{eq}$, respectively. In case of carbon, the concentration gradient disappears because of the back diffusion effect. On the other hand, large concentration gradient of phosphorous still exists in solid though the initial transient effect disappears. Figure 7 also shows that the concentration gradient becomes small in case of small cooling rate.

These results show that the Clyne–Kurz’s equation can estimate the same composition in the liquid for both carbon and phosphorous as that of phase-field calculation. Both analytical models, however, can not predict the composition profile in solid. These results show that the phase-field model can simulate not only concentration in liquid at the late stages of solidification but also a concentration profile in solid.

4.3. Isothermal Dendrite Growth

We analyze isothermal dendrite growth in two-dimensional system. The dendrite shapes of Fe–0.5mol%C binary alloy, Fe–0.5mol%C–0.001mol%P, Fe–0.5mol%C–0.01 mol%P and Fe–0.5mol%C–0.02mol%P ternary alloys at 1780 K are shown in Fig. 8. The secondary arms develop well and the arm spacing becomes narrow with a small ad-
ondary arm spacing significantly changes with a small addition of phosphorous. The secondary arm spacing significantly changes even when it does not change the interface velocity much.

5. Conclusions

The phase field model for ternary alloy systems was derived using a dilute solution approximation, and the phase field parameters with the thin interface limit condition are determined for the Fe–C–P alloy. At equilibrium condition in one-dimensional numerical simulation, the calculation results show good agreement with the compositions given by the phase-diagram. In case of a curved interface in 2-D simulations, local equilibrium conditions with the Gibbs–Thomson effect are reproduced.

Using the model, the micro-segregation behavior in Fe–0.5mol%C–0.02mol%P ternary alloy are analyzed and compared with the analytical models. The simulated growth rate is neither linearly, nor parabolically dependent on time as usually assumed. The liquid concentration profiles show good agreement with those of Clyne–Kurz equation.

For two-dimensional computations, isothermal dendrite growth is simulated for Fe–C and Fe–C–P alloys. The change in phosphorous concentration affects the interface velocity and the dendrite shape even when the amount of the phosphorous is much smaller than that of carbon because of the small diffusivity of phosphorous. The secondary arm spacing significantly changes with a small addition of phosphorus even when it does not change the interface velocity much. The results seem to be quite reasonable and show the wide potentiality of phase field model to the applications.

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