

Phase field simulation for non-isothermal solidification of multicomponent alloys coupled with thermodynamics database

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Received 10 July 2012; accepted 16 April 2013

Abstract: In order to quantitatively model the real solidification process of industrial multicomponent alloys, a non-isothermal phase field model was studied for multicomponent alloy fully coupled with thermodynamic and diffusion mobility database, which can accurately predict the phase equilibrium, solute diffusion coefficients, specific heat capacity and latent heat release in the whole system. The results show that these parameters are not constants and their values depend on local concentration and temperature. Quantitative simulation of solidification in multicomponent alloys is almost impossible without such parameters available. In this model, the interfacial region is assumed to be a mixture of solid and liquid with the same chemical potentials, but with different composition. The anti-trapping current is also considered in the model. And this model was successfully applied to industrial Al–Cu–Mg alloy for the free equiaxed dendrite solidification process.

Key words: phase-field; multicomponent alloys; coupling; thermodynamics; non-isothermal solidification; simulation

1 Introduction

The phase-field model becomes a powerful tool which can describe the complex interface pattern evolutions [1–5]. It describes the microstructure using a set of conserved and nonconserved field variables that are continuous across the interface regions. Phase field models of solidification have been originally developed for pure materials [6,7] and then extended to alloys [8–11]. Since industrial alloys are almost multicomponent alloys and contain more than two main solutes, phase field models are then extended to multicomponent systems [12–16]. But most of the phase field models for alloys solidification are under the isothermal assumption and do not consider the latent heat release [17,18]. Recently, the non-isothermal phase field models for alloy solidification [19–22] are also developed, but none of them is fully coupled with thermodynamics database, especially for the thermal transportation equations.

In multicomponent alloys, phase equilibrium, solute diffusion behaviors and latent heat release are no longer

the same as that in binary alloys because of solute interactions. A convenient way to solve this problem is coupling the solidification model with thermodynamic and diffusion mobility database [23,24]. Thus, phase equilibrium, solute diffusion transportation and latent heat release can be accurately predicted. There has been a great effort in the past years concerned with the assessment of thermodynamic and diffusion mobility database in multicomponent alloys [25–27].

In this work, we develop a non-isothermal phase field model for multicomponent alloy fully coupled with thermodynamic and diffusion mobility database to quantitatively model the real solidification process of industrial multicomponent alloys. Furthermore, industrial Al–Cu–Mg alloy is taken as an example to study the free equiaxed dendrite solidification process by this model.

2 Non-isothermal phase-field model for multicomponent alloys

2.1 Governing equations

The total free energy of the system is defined as

$$F(\phi, C_1, C_2, \dots, C_{n-1}) =$$

$$\int_{\Omega} [f(\phi, C_1, C_2, \dots, C_{n-1}) + \frac{1}{2} \varepsilon^2 |\nabla \phi|^2] d\Omega \quad (1)$$

where F is the total free energy of the system, f is the free energy density, Ω is the volume integration on space, ε is the gradient energy coefficient and ϕ is the phase field ranging from zero in liquid to one in solid.

In this work, the free energy density is defined as [28]

$$f(\phi, C_1, C_2, \dots, C_{n-1}) = h_p(\phi) f^S(C_1^S, C_2^S, \dots, C_{n-1}^S) + [1 - h_p(\phi)] f^L(C_1^L, C_2^L, \dots, C_{n-1}^L) + w g(\phi) \quad (2)$$

where

$$C_i = h_p(\phi) C_i^S + [1 - h_p(\phi)] C_i^L \quad (3)$$

where S is the solid phase, L is the liquid phase and h_p is a mixing function for phase; the equilibrium compositions C_i^S and C_i^L in a given point are not independent of each other, but restricted by the same chemical potential condition; where $h_p(\phi)$ and $g(\phi)$ are given as

$$h_p(\phi) = \phi^3 (6\phi^2 - 15\phi + 10) \quad (4)$$

$$g(\phi) = \phi^2 (1 - \phi)^2 \quad (5)$$

Then the governing equations for the phase-field can be expressed as [29]

$$\frac{\partial \phi}{\partial t} = -M \frac{\delta F}{\delta \phi} = M (\varepsilon^2 \nabla^2 \phi - f_{\phi}) \quad (6)$$

$$f_{\phi} = h'_p(\phi) \left[f^S - f^L + \sum_{i=1}^{n-1} (C_i^S - C_i^L) \mu_i \right] + w g'(\phi) \quad (7)$$

where M is the phase-field mobility and μ_i is the chemical potential of the i th solute.

The time evolution of diffusion field under mass conservation condition can be given by

$$\frac{\partial C_i}{\partial t} = \nabla \sum_{k=1}^{n-1} M_{ik} \nabla \frac{\delta F}{\delta C_k} = \nabla \sum_{k=1}^{n-1} M_{ik} \nabla \mu_k \quad (8)$$

where M_{ik} is the diffusion mobility. If the diffusion mobility follows a mixture rule assumed as

$$M_{ik} = h_d(\phi) M_{ik}^S + [1 - h_d(\phi)] M_{ik}^L \quad (9)$$

where h_d is a mixing function for diffusion, the Eq. (8) for diffusion field can be re-written as

$$\frac{\partial C_i}{\partial t} = \nabla \sum_{k=1}^{n-1} \left\{ h_d(\phi) \sum_{j=1}^{n-1} M_{ik}^S f_{C_k C_j}^S \nabla C_j^S + [1 - h_d(\phi)] \sum_{j=1}^{n-1} M_{ik}^L f_{C_k C_j}^L \nabla C_j^L \right\} \quad (10)$$

$$D_{ij}^p = \sum_{k=1}^{n-1} M_{ik}^p f_{C_k C_j}^p \quad (11)$$

where D_{ij}^p is the diffusion coefficient in solid ($p = S$) or liquid ($p = L$) phase. Under the assumption that $D_S \ll D_L$, we can re-write the diffusion equation as

$$\frac{\partial C_i}{\partial t} = \nabla \left\{ [1 - h_d(\phi)] \sum_{j=1}^{n-1} D_{ij}^L \nabla C_j^L \right\} \quad (12)$$

In order to eliminate chemical potential jump [30] effect, an antitrapping term should be introduced into the diffusion equation:

$$\frac{\partial C_i}{\partial t} = \nabla \left\{ [1 - h_d(\phi)] \sum_{j=1}^{n-1} D_{ij}^L \nabla C_j^L \right\} + \nabla \left[\frac{\varepsilon}{\sqrt{2w}} (C_i^L - C_i^S) \frac{\partial \phi}{\partial t} \frac{\nabla \phi}{|\nabla \phi|} \right] \quad (13)$$

where

$$h_d(\phi) = \phi \quad (14)$$

Using the energy conservation law, the thermal diffusion equation can be described as

$$c_p \frac{\partial T}{\partial t} = \nabla k \nabla T + \Delta H \cdot h'_p(\phi) \frac{\partial \phi}{\partial t} \quad (15)$$

where T is temperature, k is thermal conductivity, ΔH is the latent heat, c_p is specific heat capacity and can also be described by the mixture rule:

$$c_p = c_p^S + [1 - h_p(\phi)] c_p^L \quad (16)$$

Now, the evolution of phase field, solute concentration field and temperature field are governed by Eq. (6), Eq. (13) and Eq. (15), respectively. The phase field control and solute field control equation using explicit difference algorithm is named as Euler method. And the temperature control equation using alternating direction implicit method is also referred to the ADI method. In this work, the phase equilibrium between solid and liquid in Eq. (3) and (7), chemical potentials in Eq. (7), solute diffusion coefficients in Eq. (12) and (13), latent heat in Eq. (15) and specific heat capacity in Eq. (16) can be gotten by fully coupling with the thermodynamics and diffusion mobility database, which will be shown in Section 3.

2.2 Phase-field parameters

In the phase-field equation, there are three parameters: phase-field mobility M , gradient energy coefficient ε and height of the parabolic potential w . w and ε can be obtained from the interfacial energy σ and the interface width 2λ using the one-dimension equilibrium solution [29]:

$$\sigma = \frac{\varepsilon \sqrt{w}}{3\sqrt{2}} \quad (17)$$

$$2\lambda = \frac{\varepsilon}{\sqrt{2w}} \ln \frac{\phi_b(1-\phi_a)}{\phi_a(1-\phi_b)} \quad (18)$$

where the interface 2λ is defined as the width over which ϕ changes from ϕ_a to ϕ_b .

The phase-field mobility M is correlated with the interface kinetics and its equation can be given as [29]

$$\frac{1}{m} = \frac{1}{M} \frac{\sqrt{w}}{3\sqrt{2\varepsilon}} - A \frac{\varepsilon}{\sqrt{2w}} \xi \quad (19)$$

If a infinite interface mobility is applied [31], which is $m \rightarrow \infty$, the phase-field mobility can be expressed as [31]

$$M = \frac{w}{3\varepsilon^2 A \xi} \quad (20)$$

where

$$\xi = \sum_{i=1}^{n-1} (C_i^{L,E} - C_i^{S,E}) \sum_{j=1}^{n-1} f_{C_i, C_j}^{L,E} \sum_{k=1}^{n-1} d_{jk}^L (C_k^{L,E} - C_k^{S,E}) \quad (21)$$

$$A = \int_0^1 \frac{h_p(\phi)}{\phi} d\phi \quad (22)$$

where E is the equilibrium state.

3 Thermodynamics and diffusion description in multicomponent alloys

3.1 Thermodynamics

In the case of multicomponent alloys, integral Gibbs energy for each phase depends on its constitution, temperature and pressure, and this can be described by a thermodynamic model [32] as

$$G = G^0 + G_{\text{mix}}^{\text{ideal}} + G_{\text{mix}}^{\text{xs}} \quad (23)$$

where G^0 is the contribution of pure components of phase to the Gibbs energy, $G_{\text{mix}}^{\text{ideal}}$ is the ideal mixing contribution and $G_{\text{mix}}^{\text{xs}}$ is the contribution due to non-ideal interaction between components, also known as the Gibbs excess energy of mixing. When Gibbs energy in Eq. (23) is obtained, the other thermodynamics related parameters, such as phase equilibrium concentration, chemical potential, latent heat and specific heat capacity, can all be calculated out accurately, for example:

$$\Delta H = H^L - H^S = G^L - G^S + T \left(\frac{\partial G^S}{\partial T} - \frac{\partial G^L}{\partial T} \right) \quad (24)$$

$$c_p^S = \frac{\partial H^S}{\partial T} \quad \text{and} \quad c_p^L = \frac{\partial H^L}{\partial T} \quad (25)$$

There are a large number of thermodynamic models for various substances in different state [33]. For example, for FCC, BCC, HCP solid solution phases and liquid phase, the Gibbs energy G , for ternary alloy can be written as [34]

$$G = \sum_{i=1}^3 C_i G_i^0 + RT \sum_{i=1}^3 C_i \ln C_i + G^{\text{ex}} \quad (26)$$

$$G^{\text{ex}} = \sum_{i=1}^2 \sum_{j=i+1}^3 C_i C_j \sum_{n=0}^m [L_{ij,n} (C_i - C_j)^n] + C_1 C_2 C_3 L^{\text{ter}} \quad (27)$$

Such work has been proved powerful, since there are several available software systems which are capable of estimating the phase equilibrium as a function of pressure, temperature and the composition of alloying elements, such as Thermo-Calc [35], PANDAT [36], and MTDATA [37]. In this work, the properties of thermodynamics are all obtained by Thermo-Calc through TQ interface. The thermodynamic description of Al–Cu–Mg system will be shown in Section 4.

3.2 Diffusion coefficients

For the case that the elements are substitutional, the diffusion coefficient defined in the volume-fixed frame of reference, D_{ij}^V , can be expressed as [38]

$$D_{ij}^V = \sum_{k=1}^n (\delta_{ki} - C_i) C_k \Omega_k \frac{\partial \mu_k}{\partial C_j} \quad (28)$$

where C_i is the mole fraction of component i , μ_k is the chemical potential of component k , and δ_{ki} is the Kronecker delta. It can be observed that the $\partial \mu_k / \partial C_j$ is a purely thermodynamic parameter which corresponding to the thermodynamic factor and can thus be evaluated from the thermodynamic description of the system. The parameter Ω_k is the mobility of specie k in a given phase and will be discussed later.

The chemical diffusion coefficients, by eliminating the concentration element n , can be deduced as

$$D_{ij}^n = D_{ij}^V - D_{in}^V \quad (29)$$

where n is the solvent and D_{ij}^V is given in equation (28). The diffusivity D_{ij}^n is the most convenient one for practical calculations. Determination of the diffusion mobility parameter in Eq. (28) requires the use of experimental diffusion data. Tracer or self diffusivity D_i^* , is generally determined from diffusion studies using isotopes and directly related to the mobility Ω_k by means of the Einstein relation as

$$D_i^* = RT \Omega_i \quad (30)$$

Using the diffusion coefficient expression in Eq. (29), the concentration of element n is eliminated and an $(n-1) \times (n-1)$ chemical diffusion matrix is obtained. Then the concentration dependent solute diffusion coefficients in Eq. (13) can also be accurately obtained.

It is worth noting here that the thermodynamic factors in Eq. (28) are also obtained by Thermo-Calc through the TQ interface.

4 Application to Al–Cu–Mg alloy

The thermodynamic description of Al–Cu–Mg system now can be used from BUHLER et al [39]. Using the parameters for the thermodynamic description of Al-rich ternary Al–Cu–Mg alloys, G^{ex} for liquid phase can be evaluated as

$$G_{\text{Liquid}}^{\text{ex}} = x_{\text{Al}}x_{\text{Cu}}[(-66622 + 8.1T) + (46800 - 90.8T + 10T \ln T)(x_{\text{Al}} - x_{\text{Cu}}) - 2812(x_{\text{Al}} - x_{\text{Cu}})^2] + x_{\text{Al}}x_{\text{Mg}}[(-12000 + 8.566T) + (1894 - 3T)(x_{\text{Al}} - x_{\text{Mg}}) + 2000(x_{\text{Al}} - x_{\text{Mg}})^2] + x_{\text{Cu}}x_{\text{Mg}}[(-36984 + 4.7561T) - 8191.29(x_{\text{Cu}} - x_{\text{Mg}})] \quad (31)$$

For Al-FCC primary solution, G^{ex} is given by

$$G_{\text{FCC}}^{\text{ex}} = x_{\text{Al}}x_{\text{Cu}}[(-53520 + 2T) + (38590 - 2T)(x_{\text{Al}} - x_{\text{Cu}}) + 1170(x_{\text{Al}} - x_{\text{Cu}})^2] + x_{\text{Al}}x_{\text{Mg}}[(4971 - 3.5T) + (900 + 0.423T)(x_{\text{Al}} - x_{\text{Mg}}) + 950(x_{\text{Al}} - x_{\text{Mg}})^2] + x_{\text{Cu}}x_{\text{Mg}}(-22279.28 + 5.868T) \quad (32)$$

Tracer diffusion is the diffusion of a tracer in the matrix of an alloy. The impurity or self diffusivity D_i^* , in ternary Al–Cu–Mg is given in Table 1. The other parameters used in the simulation are given in Table 2.

Table 1 Diffusion data used in simulation

Parameter	$D_i^*/(\text{m}^2 \cdot \text{s}^{-1})$
Impurity diffusion coefficient of Cu in liquid Al [40]	$1.06 \times 10^{-7} \exp(\frac{-24000}{RT})$
Impurity diffusion coefficient of Mg in liquid Al [40]	$9.9 \times 10^{-5} \exp(\frac{-71600}{RT})$
Self diffusion coefficient of liquid Al [41]	$1.16 \times 10^{-7} \exp(\frac{-21330}{RT})$

Table 2 Parameters used in simulation

Parameter	Value
Interface energy, $\sigma/(\text{J} \cdot \text{m}^{-2})$	0.093
Initial alloy composition (mole fraction, %) $C_{\text{Cu}}=1.1$, $C_{\text{Mg}}=1.8$	
Initial temperature/K	900
Initial cooling rate/ $(\text{K} \cdot \text{s}^{-1})$	1.0×10^6
Molar volume, $V_{\text{m}}/(\text{m}^3 \cdot \text{mol}^{-1})$	1.06×10^{-5}
Grid size, $\Delta x/\text{m}$	1.0×10^{-8}
Thermal conductivity/ $(\text{W} \cdot \text{m}^{-1} \cdot \text{K}^{-1})$	190

Figures 1(a) and (b) show the temperature field and phase field in the whole system, respectively. The solidification time is 4.5×10^{-5} s. Figures 1(c) and (d) show the concentration field of Cu and Mg, respectively, both in solid and liquid. They show that the temperature field has less gradient than concentration fields since the thermal conductivity is almost 10^3 times larger than the solutes diffusion coefficients.

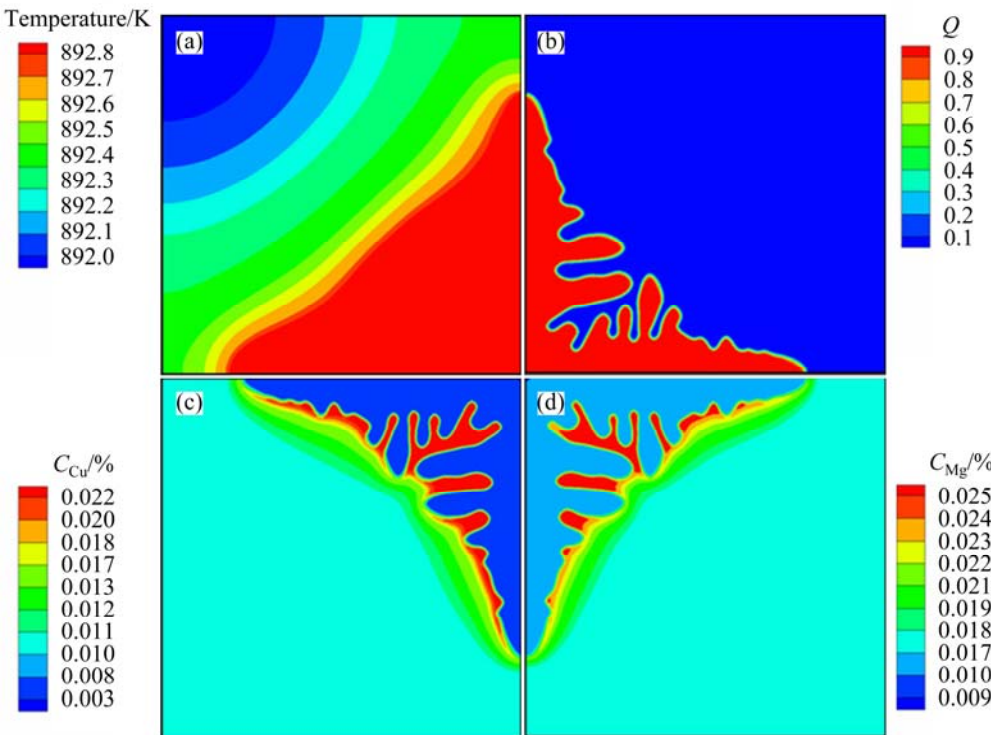


Fig. 1 Simulation results of Al–1.1% Cu–1.8% Mg dendritic solidification process ($t=4.5 \times 10^{-5}$ s): (a) Temperature field; (b) Phase field; (c) C_{Cu} ; (d) C_{Mg}

Figure 2 shows the distribution of diffusion coefficients in the whole system. It shows that the diffusion coefficients are all highly concentration dependent, both diagonal (D_{CuCu} and D_{MgMg}) and off-diagonal (D_{CuMg} and D_{MgCu}). It shows that the off-diagonal diffusion coefficients (D_{CuMg} and D_{MgCu}) are about 10% of diagonal ones (D_{CuCu} and D_{MgMg}).

Figure 3 shows the latent heat and specific heat capacity distribution in the whole simulation area. It shows that the latent heat and specific heat capacity are both concentration and temperature dependent. They both change a lot in the whole system. It is almost impossible to get these parameters accurately if the model is not coupled with thermodynamics database. Quantitative simulation of solidification in multicomponent alloys is also impossible without such parameters available.

5 Conclusions

1) A non-isothermal phase field model was developed for multicomponent alloy fully coupled with thermodynamic and diffusion mobility database, which can accurately predict the phase equilibrium, solute diffusion coefficients, specific heat capacity and latent heat release in the whole system.

2) These parameters are not constants and their values are dependent on local concentration and temperature. Quantitative simulation of solidification in multicomponent alloys is almost impossible without such parameters available.

3) In the model, the interfacial region is assumed to be a mixture of solid and liquid with the same chemical potentials, but the different compositions.

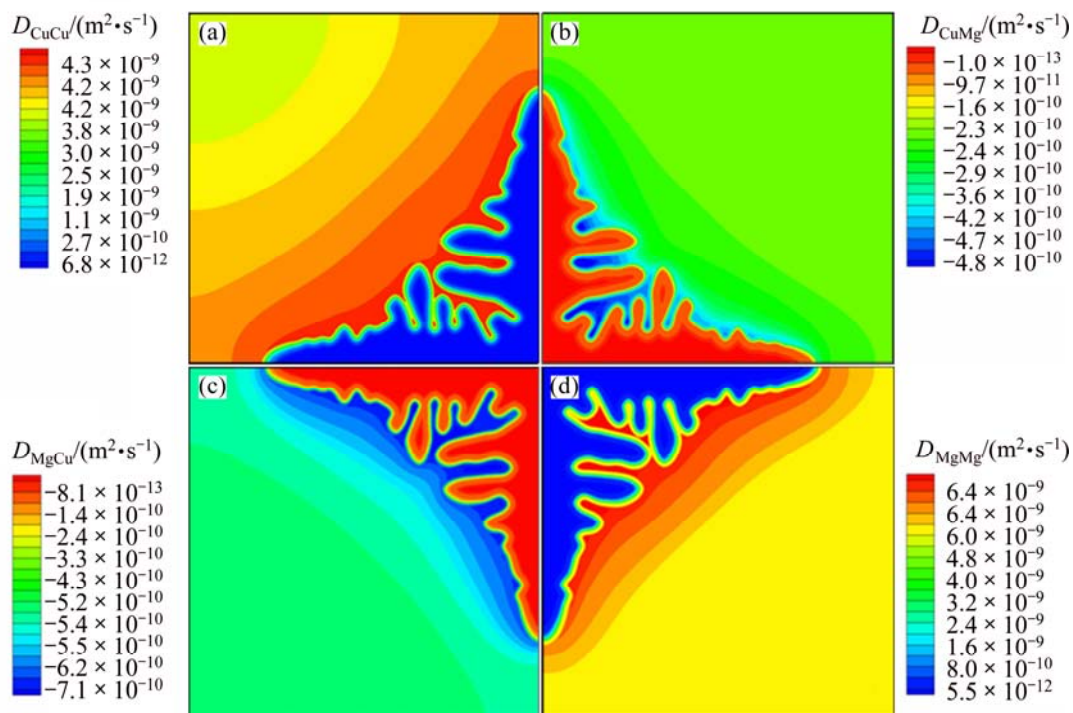


Fig. 2 Diffusion coefficients distribution at solidification time 4.5×10^{-5} s: (a) D_{CuCu} ; (b) D_{CuMg} ; (c) D_{MgCu} ; (d) D_{MgMg}

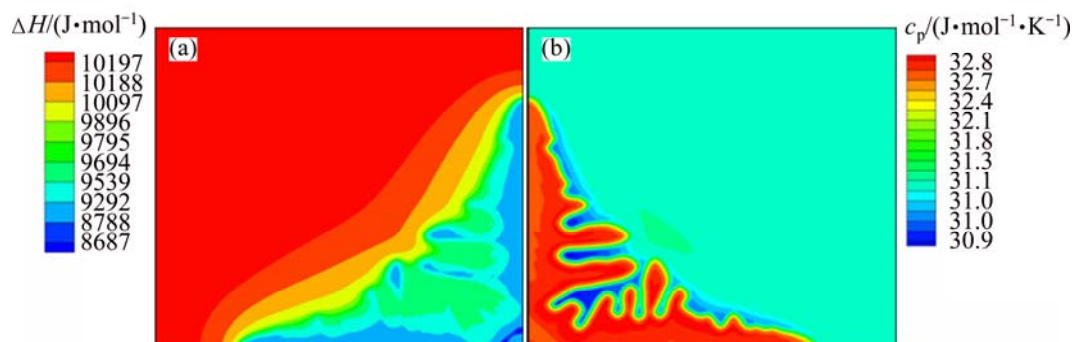


Fig. 3 Latent heat and specific heat capacity distribution at solidification time 4.5×10^{-5} s: (a) Latent heat; (b) Specific heat capacity

4) The anti-trapping current is also considered in the model. For example, industrial Al–Cu–Mg alloy for the free equiaxed dendrite solidification process can be studied by the above model.

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多元合金耦合热力学数据非等温凝固的相场模拟

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摘 要: 研究一种多元合金的非等温相场模型, 定量地模拟工业多元合金的真实凝固过程, 结合热力学和扩散迁移率的数据来预测整个系统中的相平衡、溶质扩散系数、比热容和放出的潜热。结果表明: 这些参数不是常数, 它们的值与成分和温度有关。没有这些参数, 定量模拟多元合金的凝固几乎是不可能。在这个模型中, 界面区域假设是由有同样化学势的固相和液相混合而成的, 但组成不同。这个模型中同样考虑反溶质截流。一并将模型应用到工业 Al–Cu–Mg 合金的各向枝晶自由长大的凝固过程。

关键词: 相场; 多元合金; 耦合; 热力学; 非等温凝固; 模拟

(Edited by Chao WANG)