

Phase-field model of isothermal solidification with multiple grain growth*

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This paper develops a new phase-field model for equiaxed dendrite growth of multiple grains in multicomponent alloys based on the Ginzberg–Landau theory and phase-field model of a single grain. Taking Al–Cu and Al–Cu–Mg alloys for example, it couples the concentration field and simulates the dendrite growth process of multiple grains during isothermal solidification. The result of the simulation shows dendrite competitive growth of multiple grains, and is reapplied to the process of dendrite growth in practical solidification.

Keywords: phase-field, multiple grains, multicomponent alloys, equiaxed dendrites

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1. Introduction

With the ability to model the kinetics and the pattern formation for solidification, a phase field model has been studied by many scientists. The phase field method has successfully been applied to predict microstructure evolution in metallic alloys such as dendritic solidification.^[1,2] Dendrites are intricate patterns that make up the microstructure of many important commercial alloys.^[3,4] Currently available models,^[5–9] however, are restricted to a single grain. Many commercial materials are multicomponent and multiple grains, whose properties are determined by the microstructure that develops during solidification and subsequent processing. In order to apply the method quantitatively to these phenomena occurring in technical alloys, the method needs to be able to treat multicomponent multi grain systems. In this work, a new phase field model is developed to study the solidification of multiple grains, based on the Ginzberg–Landau theory and phase-field model of a single grain. Phase-field simulations on multiple grain growth for Al–Cu and Al–Cu–Mg alloys are carried out, and reapplied to the process of dendrite growth in practical solidification.

2. Phase-field model

2.1. Governing equations for binary alloys

The phase field theory is an advanced computational approach which generally describes the evolution of so-called ‘phase fields’ ϕ . In this model, the field describes the different phases of grains. The phase field, $\phi(x, t)$, characterizes the physical state of the system at each position and time: $\phi = 1$ for the solid, $\phi = 0$ for the liquid, and $0 < \phi < 1$ at the interface. The free energy density of a solid–liquid mixture may be written in the form

$$f(\phi, c) = h(\phi)f^s + [1 - h(\phi)]f^l + wg(\phi), \quad (1)$$

where f^s and f^l are the free energy density of the solid and liquid phase, respectively,

$$f^s = cf_B^s + (1 - c)f_A^s, \quad (2)$$

$$f^l = cf_B^l + (1 - c)f_A^l, \quad (3)$$

where the subscripts A and B represent the elements, the superscripts l and s represent liquid and solid phases, and where the solid fraction, $h(\phi)$, is given by $\phi^3(10 - 15\phi + 6\phi^2)$, w is the height of the parabolic

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potential, and $g(\phi)$ is the imposed parabolic potential, which is given by $\phi^2(1 - \phi^2)$.

The governing equations for the phase-field and the concentration-field are expressed as follows:^[10]

$$\frac{\partial \phi}{\partial t} = M[\varepsilon^2(\theta_i)\nabla^2\phi - f_\phi], \quad (4)$$

$$\frac{\partial c}{\partial t} = \nabla \left(\frac{D(\phi)}{f_{cc}} \nabla f_c \right), \quad (5)$$

$$\varepsilon(\theta_i) = \varepsilon_0(1 + \nu \cos(k\theta_i)), \quad (6)$$

where t is the time, M and ε_0 are phase-field parameters to be determined later, and $D(\phi)$ is the solute diffusivity, ν is the magnitude of anisotropy, k is the mode number and θ_i is the angle between the direction of the orientations of grains and the reference axis of the system, the subscripts i are the number of grains. The functions f_ϕ , f_c and f_{cc} are obtained by differentiating Eq.(1) with respect to ϕ and c respectively.

2.2. Governing equations for multicomponent alloys

Take ternary alloys for example, the Gibbs free energy for the liquid and solid phases are expressed respectively as

$$G_1^{\text{reg}}(c_{1l}, c_{2l}, c_{3l}) = \sum_{j=1}^3 (c_{jl}\mu_{jl}^0 + RTc_{jl} \ln c_{jl}) + G_1^{\text{ex}}(c_{1l}, c_{2l}, c_{3l}), \quad (7)$$

$$G_s^{\text{reg}}(c_{1s}, c_{2s}, c_{3s}) = \sum_{j=1}^3 (c_{js}\mu_{js}^0 + RTc_{js} \ln c_{js}) + G_s^{\text{ex}}(c_{1s}, c_{2s}, c_{3s}), \quad (8)$$

where the subscripts 1, 2 and 3 represent the elements, and the subscripts l and s denote liquid and solid phases. The notations μ_{jl}^0 and μ_{js}^0 represent the standard state of chemical potential. The excess free energy for the mixture, G^{ex} , is given later.

The free energy density can be written as

$$f = h(\phi)G_s^{\text{reg}} + (1 - h(\phi))G_l^{\text{reg}} + wg(\phi). \quad (9)$$

The governing equations for the phase-field of binary alloys and the equations of ternary alloys are expressed in the same way. The governing equation for the concentration-field is expressed as follows:

$$\frac{\partial c_j}{\partial t} = \nabla \left(\frac{D_j(\phi)}{f_{c_j c_j}} \nabla f_{c_j} \right), \quad (10)$$

where the subscripts j represent the solute elements.

2.3. Phase-field parameters

In the phase-field equation, there are three parameters: the gradient energy coefficient ε_0 ; the height of the parabolic potential w ; the mobility M . The parameters w and ε_0 can be obtained from the interface energy σ and the interface width 2λ . Using the one-dimensional equilibrium solution, we can obtain the following relationships:

$$\varepsilon_0 = \sqrt{\frac{6\lambda\sigma}{2.2}}, \quad (11)$$

$$w = \frac{6.6\sigma}{\lambda}. \quad (12)$$

The equation of the phase-field mobility M of binary alloys is given as^[6,7]

$$M^{-1} = \frac{\varepsilon^3}{\sigma\sqrt{2W}} \left(\frac{1}{D} \zeta(c_1^e, c_s^e) \right), \quad (13)$$

$$\begin{aligned} \zeta &= \frac{RT}{V_m} (c_1^e - c_s^e)^2 \\ &\times \int_0^1 \frac{h(\phi)[1 - h(\phi)]}{[1 - h(\phi)]c_1^e(1 - c_1^e) + h(\phi)c_s^e(1 - c_s^e)} \\ &\times \frac{d\phi}{\phi(1 - \phi)}. \end{aligned} \quad (14)$$

The equation of ternary alloys is given as^[8,9]

$$\begin{aligned} M^{-1} &= \frac{\varepsilon^3}{\sigma\sqrt{2W}} \left(\frac{1}{D_{1l}} \zeta_1(c_{1l}^e, c_{1s}^e) + \frac{1}{D_{2l}} \zeta_2(c_{2l}^e, c_{2s}^e) \right), \end{aligned} \quad (15)$$

$$\begin{aligned} \zeta_j &= \frac{RT}{V_m} (c_{jl}^e - c_{js}^e)^2 \\ &\times \int_0^1 \frac{h(\phi)[1 - h(\phi)]}{[1 - h(\phi)]c_{jl}^e(1 - c_{jl}^e) + h(\phi)c_{js}^e(1 - c_{js}^e)} \\ &\times \frac{d\phi}{\phi(1 - \phi)}. \end{aligned} \quad (16)$$

2.4. Physical properties of Al-Cu alloy and Al-Cu-Mg alloy

Using the parameters for the thermodynamic description of Al-rich ternary Al-Cu-Mg alloy,^[11] G^{ex} for the liquid phase can be evaluated as

$$\begin{aligned}
G_1^{\text{ex}} = & c_{\text{Al}}c_{\text{Cu}}[(66622 + 8.1T) + (46800 - 90.8T + 10T \ln T)(c_{\text{Al}} - c_{\text{Cu}}) - 2812(c_{\text{Al}} - c_{\text{Cu}})^2] \\
& + c_{\text{Al}}c_{\text{Mg}}[(-12000 + 8.566T) + (1894 - 3T)(c_{\text{Al}} - c_{\text{Mg}}) + 2000(c_{\text{Al}} - c_{\text{Mg}})^2] \\
& + c_{\text{Cu}}c_{\text{Mg}}[(-36984 + 4.7561T) - 8191.29(c_{\text{Cu}} - c_{\text{Mg}})].
\end{aligned} \quad (17)$$

For the fcc Al primary solution, G^{ex} is given by

$$\begin{aligned}
G_s^{\text{ex}} = & c_{\text{Al}}c_{\text{Cu}}[(-53520 + 2T) + (38590 - 2T)(c_{\text{Al}} - c_{\text{Cu}}) + 1170(c_{\text{Al}} - c_{\text{Cu}})^2] + c_{\text{Al}}c_{\text{Mg}}[(4971 - 3.5T) \\
& + (900 + 0.423T)(c_{\text{Al}} - c_{\text{Mg}}) + 950(c_{\text{Al}} - c_{\text{Mg}})^2] + c_{\text{Cu}}c_{\text{Mg}}(-22279.28 + 5.868T),
\end{aligned} \quad (18)$$

where T is the temperature of the system.

The physical properties of the Al–Cu alloy used in this calculation are listed in Table 1. The physical properties of the Al–Cu–Mg alloy are listed in Table 2.

Table 1. Physical parameters of the Al–Cu alloy.

parameter	Al–2%Cu(at)
energy of interface of Al $\sigma/(\text{J} \cdot \text{m}^{-1})$	0.093
melting point of Al (T_m/K)	933.3
diffusion coefficient in solid $(\text{m}^2 \cdot \text{s}^{-1})$	3.0×10^{-9}
diffusion coefficient in liquid $(\text{m}^2 \cdot \text{s}^{-1})$	3.0×10^{-13}

Table 2. Physical parameters of the Al–Cu–Mg alloy.

parameter	Al–Cu–Mg alloy
energy of interface of Al $\sigma/(\text{J} \cdot \text{m}^{-2})$	0.093
melting point of Al T_m/K	933.3
diffusion coefficient of Cu in solid $(\text{m}^2 \cdot \text{s}^{-1})$	$4.44 \times 10^{-5} \exp(-16104/T)$
diffusion coefficient of Cu in liquid $(\text{m}^2 \cdot \text{s}^{-1})$	$1.06 \times 10^{-7} \exp(-2887/T)$
diffusion coefficient of Mg in solid $(\text{m}^2 \cdot \text{s}^{-1})$	$0.37 \times 10^{-4} \exp(-14854/T)$
diffusion coefficient of Mg in liquid $(\text{m}^2 \cdot \text{s}^{-1})$	$9.90 \times 10^{-5} \exp(-8610/T)$
molar volume (m^3/mol)	1.06×10^{-5}

2.5. Nucleation

Under equiaxed dendrite growth conditions nucleation is a very important parameter for microstructure formation as continuous growth is not extremely pure as in the case of technical alloys, formation of the primary phases is dominated by heterogeneous nucleation. In this model, the system makes use of heterogeneous nucleation, puts several seeds randomly in the area of simulation before the calculation of system begins. The density of seeds in the simulation must be less than the biggest nucleation density calculated by the Gauss formula,^[12] written as

$$\frac{dn}{d(\Delta T)} = \frac{n_{\text{max}}}{\sqrt{2\pi}\Delta T_\sigma} \exp\left(-\frac{(\Delta T - \Delta T_{\text{max}})^2}{2\Delta T_\sigma^2}\right), \quad (19)$$

where ΔT_{max} is the biggest nucleation undercooling, ΔT_σ is standard modification undercooling, n_{max} is the biggest nucleation density.

3. Numerical calculations and results

The calculation area of 1200×1200 meshes is prepared and several seeds (small solid triangles) are initially put at the random districts of the area. The Zero-Neumann boundary conditions for ϕ and c are imposed at the boundaries of the computational domain. The temperature condition is constant, whereas the boundary is fixed at 895 K.

3.1. Application to the Al–Cu alloy and simulation results

Al–Cu alloys and Al–Cu–Mg alloys are frequently used aluminium alloys for industrial production.^[11] The multiple dendrite shapes of the Al–2at%Cu binary alloy and distribution of concentration profile are shown in Fig.1. In Fig.1.(a), growth of the upper primary stalk of grain B, which became bent, is affected

by grain A. Because grain A grew to the area before grain B, solidified and increased the temperature of the area, it induced grain B to grow to the lower temperature area and become bent. The upper left stalk of grain A became bent too because of the effect of

other grains. Through Fig.1(b), it can be seen that the primary stalk has a low concentration, but the regions between the secondary arms and grains have the highest concentration. These features are commonly observed in real equiaxed dendrites.

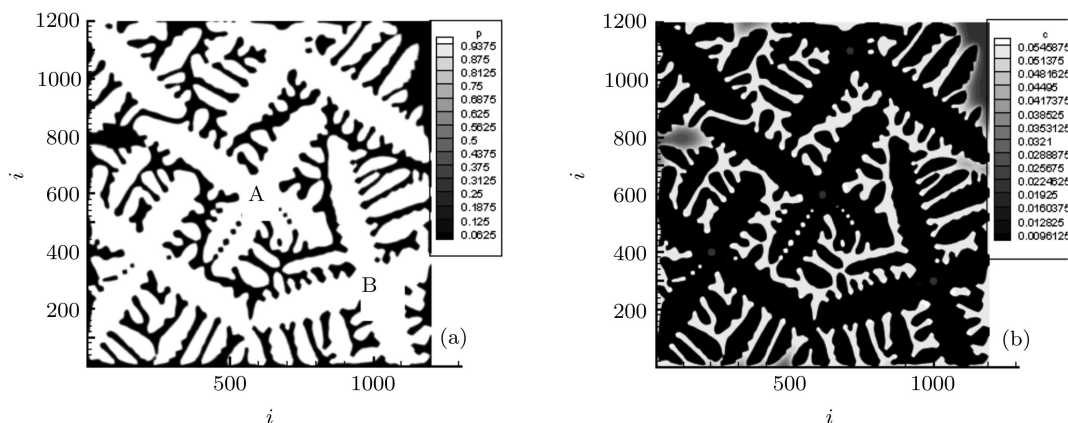


Fig.1. Simulations of equiaxed dendrite solidification of an Al-Cu alloy. (a) Multiple grain dendritic pattern, (b) concentration field profile for dendrite growth process.

The microstructural form of Al-2at.%Cu alloy (result of the experiment) are shown in Fig.2. Comparing the simulated microstructure and experimental microstructure of the equiaxed dendrite in Al-2at.%Cu, we can see that there are some differences. This is because the simulated result was calculated in 2-dimensional conditions, and the picture of the microstructure form of the Al-2at.%Cu alloy shown in Fig.2 was a slice of a 3-dimensional solid. The grains of simulation shown in Fig.1 are the slices on axis, and the grains in the terminal stage of solidification in the Al-Cu alloy shown in Fig.2 are not all like this. Another reason is that the result of the simulation is isothermal solidification, the other is not. It shows the concentration distribution of Cu in the area of grid number $i = 600$, in Fig.3, the regions between the grains have the highest concentration, while the grain itself has a low concentration.

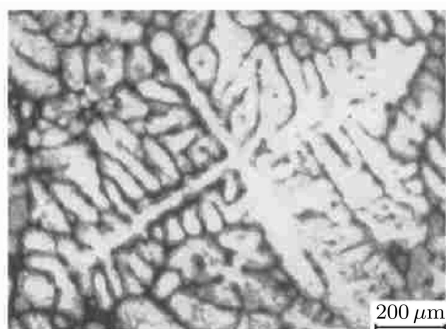


Fig.2. Microstructures formed in the terminal stage of solidification in the Al-Cu alloy.^[12]

These features show good agreement with the composition distribution given by the real equiaxed dendrite growth.

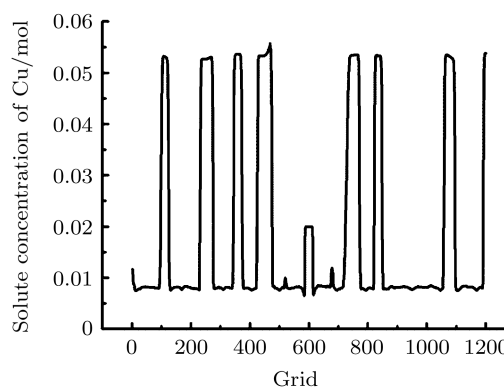


Fig.3. Concentration profile of Cu in simulation areas.

3.2. Application to the Al-Cu-Mg alloy and simulation results

The results of simulation of equiaxed dendrite solidification of Al-2at.%Cu-3.5at.%Mg alloy are shown in Fig.4. Figure 4(a) shows five equiaxed dendrite competitive growth and Fig.4(b) shows ten grains competitive growth. The grains of Fig.4(a) are coarser than the grains in Fig.4(b). It should be noted that the predicted dendritic morphology is affected by density of seeds, the higher seed density it is, the more refined and globular grains become. A conclusion can be drawn after comparing Fig.1(a) with Fig.4(a), that

the composition change (adding Mg to Al–Cu alloy in order to get Al–Cu–Mg alloy) does not significantly change the shape of grains. Figure 5 shows the solid phase rate of the simulation area in Figs.4(a) and 4(d). In this paper, the definition of solid phase rate is the fraction of the solid phase in the simulation region. It can be seen that, under the higher seed density, the velocity of the solid becomes faster and the grains are more refined. This result shows good agreement with real solidification, in many industrial alloy solid-

ification processes, an amount of inoculants is used to change the micro-structural morphology and enhance the properties of the materials. Figures 4(b) and 4(c) show the concentration distribution of solute Mg and Cu, respectively. It can be noticed that the solute Mg in grains is more well-proportioned than Cu. Figures 4(e) and 4(f) are concentration profiles of Fig.4(b) and Fig.4(c) respectively. They show the solute distribution of the area in the simulation (Figs.4(b) and 4(c)) where the grid number $i = 400$. The trough of the

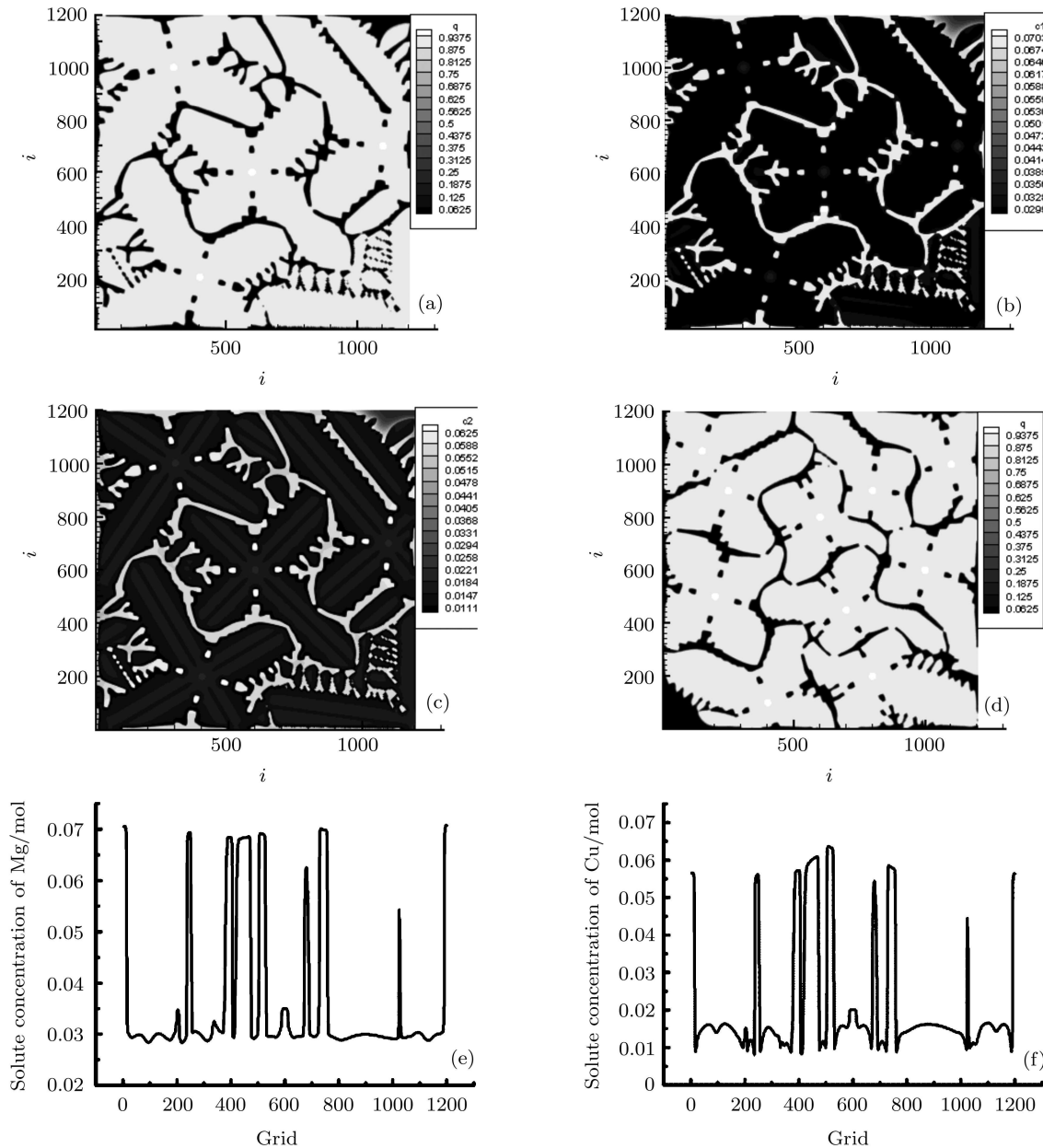


Fig.4. Results of simulation for Al–Cu–Mg alloy. (a) Morphology of dendrite growth of five grains; (b) distribution of concentration Mg; (c) distribution of concentration Cu; (d) morphology of dendrite growth of ten grains; (e) concentration profile of Mg in simulation areas; (f) concentration profile of Cu in simulation areas.

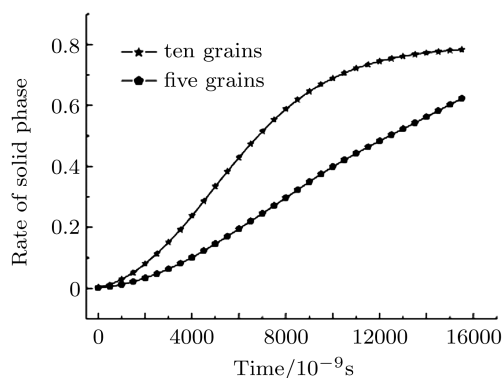


Fig.5. The solid phase rate due to different magnitudes of original grains in simulation range.

curve represents low solute concentration in grains, the wave crest represents high concentration in liquid (in front of the interface). Obviously, the curve of solute Cu in grains is more undulate than that of solute Mg, that means the solute Mg in grains is more well-proportioned than Cu in grains, this is because that the diffusion coefficient of solute Mg in solid grains is bigger than that of solute Cu. Considering Fig.3, we can learn that solute Mg changes the concentration distribution of solute Cu in equiaxed dendrite and increases the concentration of Cu in grains. The concentration distribution of solute Cu in grains without Mg in the alloy is more well-proportioned than that with

Mg in the alloy. That means solute Mg will increase the microsegregation of solute Cu in grains.

4. Conclusion

A phase-field model for multiple grain growth of multicomponent alloys has been developed in this research. The model has been used for the simulation of Al-Cu and Al-Cu-Mg alloys, and the predicted microstructures agree with experimental observations. It has been shown that the phase-field method can be a valuable tool for simulating equiaxed solidification processes in technical alloys. Qualitative effects like the grain refining mechanisms of inoculants and the effect on the grain size and dendritic morphology in Al-Cu-Mg alloy, the effect of the relation between alloy composition and microstructure (Al-Cu alloy vs. Al-Cu-Mg alloy) have been shown. While obtaining quantitative results is hard, the major problem lies in the lack of physical parameters such as the critical nucleation undercoolings or seed density curves for different phases and so on. This information cannot be obtained without intensive calibration using solidification experiments, which are performed under well-determined conditions. Thus, there is a lot of work to be done in this region.

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