

Phase-field Model for Diffusional Phase Transformations in Elastically Inhomogeneous Polycrystals

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Abstract. A phase-field model is described for predicting the diffusional phase transformation process in elastically inhomogeneous polycrystals. The elastic interactions are incorporated by solving the mechanical equilibrium equation using the Fourier-spectral iterative-perturbation scheme taking into account elastic modulus inhomogeneity. A number of examples are presented, including grain boundary segregation, precipitation of second-phase particles in a polycrystal, and interaction between segregation at a grain boundary and coherent precipitates inside grains. It is shown that the local pressure distribution due to coherent precipitates leads to highly inhomogeneous solute distribution along grain boundaries.

Introduction

Most materials in engineering applications are polycrystalline, containing grains of different crystallographic orientations separated by grain boundaries. During heat treatment, grain growth through grain boundary migration and phase transformations and microstructure evolution within the grain structure take place simultaneously. On one hand, the presence of grain boundaries results in inhomogeneous distribution of solute atoms and/or new phase particles, which has important implications to the materials' mechanical properties [1-4]. On the other hand, the existence of phase microstructures such as precipitate particles may inhibit grain boundary migration and thus reduce the rate of grain growth. Therefore, the evolutions of grain and phase microstructures are inherently coupled. The main objective of this article is to report a phase-field model for predicting the thermodynamics and kinetics of solute segregation, phase transformations, and microstructure evolution in polycrystals containing elastically anisotropic grains.

Model Description

Free Energy Model & Kinetic Equations. Phase-field method has been employed to model microstructure evolution for many different materials processes [5-10]. In a phase-field model, the total free energy F of an inhomogeneous microstructure is described as a function of a set of field variables which are continuous across the interface. In the model, we use conserved field variables $X(\vec{r})$ and non-conserved field $\eta_p(\vec{r})$ to describe the compositional and structural distributions of a solid solution. The total free energy F of the system is given by the following volume integral [5]

$$F = \int_V \left\{ f(X(\vec{r}), \eta_p(\vec{r})) + \frac{\kappa_c}{2} (\nabla X)^2 + \frac{\kappa_o}{2} \sum_p (\nabla \eta_p)^2 + E_{el} \right\} dV, \quad (1)$$

where f is the local free energy density, κ_c and κ_o are gradient energy coefficients, and E_{el} is the local elastic energy density.

The temporal evolution of the compositional fields and non-conserved order parameters are governed by the Cahn-Hilliard equation (Eq. 2) [11] and Allen-Cahn relaxation equation (Eq. 3) [12], respectively,

$$\frac{\partial X(\vec{r}, t)}{\partial t} = \nabla \cdot M \nabla \left(\frac{\delta F}{\delta X(\vec{r}, t)} \right), \quad (2)$$

$$\frac{\partial \eta_p(\vec{r}, t)}{\partial t} = -L \left(\frac{\delta F}{\delta \eta_p(\vec{r}, t)} \right), \quad (3)$$

where M is the interdiffusion mobility, L is the kinetic coefficient related to interfacial mobility, t is time, $\left(\frac{\delta F}{\delta X} \right)$ is the variation of the free energy function with respect to composition, and $\left(\frac{\delta F}{\delta \eta_p} \right)$ is the variation of the free energy function with respect to the order parameter fields.

Inhomogeneous Elasticity Model. There are generally two types of elastic inhomogeneity contributions in polycrystals. Firstly, the elastic constants at the grain boundaries are expected to be different from those within the grains. Secondly, each grain with different orientation will have different elastic constants with respect to a reference orientation on a global coordinate system since a crystalline grain is always elastically anisotropic. To take into account both inhomogeneity, we describe the elastic modulus of a polycrystal in the global reference coordinate system in terms of the field variables describing a grain structure [13]:

$$C_{ijkl}(\vec{r}) = \sum_g \eta_g^2(\vec{r}) a_{im}^g a_{jn}^g a_{ko}^g a_{lp}^g C_{mnop}, \quad (4)$$

where the non-conserved field variable $\eta_g(\vec{r})$ represents the orientation of the grains in a polycrystalline system, a_{ij}^g are the components of an axis transformation matrix representing the rotation from the coordinate system defined on a given grain g to the global reference coordinate system, and C_{mnop} on the right-hand side is the elastic modulus in the coordinate system defined on a given grain.

Since the mechanical equilibrium is generally established much faster than the phase transformation processes, we solve the mechanical equilibrium equation to obtain the local elastic field in solids,

$$\nabla_j \sigma_{ij} = \nabla_j C_{ijkl}(\vec{r}) \cdot [\varepsilon_{kl}(\vec{r}) - \varepsilon_{kl}^o(\vec{r})] = 0, \quad (5)$$

where σ_{ij} is the local elastic stress, $C_{ijkl}(\vec{r})$ is the position-dependent elastic modulus tensor, $\varepsilon_{ij}(\vec{r})$ is the local total elastic strain, and $\varepsilon_{ij}^o(\vec{r})$ is the local stress-free strain (or eigenstrain) due to phase transformation, thermal mismatch, etc. For example, the stress-free strain due to compositional inhomogeneity, $\varepsilon_{ij}^o(\vec{r})$, is given by

$$\varepsilon_{ij}^o(\vec{r}) = \delta_{ij} K_m (X(\vec{r}) - X_o), \quad (6)$$

where δ_{ij} is the Kronecker delta function, K_m is the composition expansion coefficient of lattice parameter, $X(\vec{r})$ is the composition field, and X_o is the overall composition of the parent phase.

We employed the iterative-perturbation scheme to solve the elasticity equations in systems with inhomogeneous elastic moduli [14-15]. The elastic modulus can be separated into two parts. One is a constant isotropic part, and the other is a position-dependent perturbation, i.e.

$$C_{ijkl}(\vec{r}) = C_{ijkl}^{iso} + \left[\sum_g \eta_g^2(\vec{r}) a_{im}^g a_{jn}^g a_{ko}^g a_{lp}^g C_{mnop} - C_{ijkl}^{iso} \right], \quad (7)$$

where C_{ijkl}^{iso} is the isotropic elastic stiffness tensor, and remaining part within the square bracket is considered as perturbation. The total strain $\varepsilon_{ij}(\vec{r})$ can be separated into a sum of homogeneous strain ($\bar{\varepsilon}_{kl}$) and heterogeneous strain ($\delta\varepsilon_{kl}(\vec{r})$). The homogeneous strain can be calculated by minimizing the elastic energy with respect to the homogeneous strain [16] while the heterogeneous strain can be expressed by the elastic displacement ($u_i(\vec{r})$) as the following equation [17]:

$$\varepsilon_{kl}(\vec{r}) = \bar{\varepsilon}_{kl} + \delta\varepsilon_{kl} = \bar{\varepsilon}_{kl} + \frac{1}{2} \left(\frac{\partial u_k}{\partial r_l} + \frac{\partial u_l}{\partial r_k} \right). \quad (8)$$

Plugging Eq. 7 and Eq. 8 into Eq. 5, we have

$$C_{ijkl}^{iso} \frac{\partial^2 u_k}{\partial r_j \partial r_l} = \nabla_j \left\{ \left(\sum_g \eta_g^2(\vec{r}) a_{io}^g a_{jp}^g a_{kq}^g a_{lr}^g C_{opqr} \right) (\varepsilon_{kl}^o(\vec{r}) - \bar{\varepsilon}_{kl}) \right\} - \frac{\partial}{\partial r_j} \left[\left(\sum_g \eta_g^2(\vec{r}) a_{io}^g a_{jp}^g a_{kq}^g a_{lr}^g C_{opqr} - C_{ijkl}^{iso} \right) \frac{\partial u_k}{\partial r_l} \right]. \quad (9)$$

By iteratively solving this equation in the Fourier space, we can obtain the elastic displacement ($u_i(\vec{r})$), and calculate the heterogeneous strain field.

Simulation Results & Discussion

Generation of Static Grain Structures. Our present simulations on diffusional phase transformations were conducted on a static grain structure for simplicity. To investigate the diffusion phenomena near the grain boundary, we generated a simple two-dimensional grain structure containing two or four grains separated by grain boundaries on 256×256 square lattices through the phase-field grain growth model [18] under the periodic boundary condition using non-conserved field variables $\eta_g(\vec{r})$ as shown in Fig.1 (a) and (b). In addition, each grain is assigned by an angle that represents the orientation of the grain. For example, the left-hand side grain (Grain I) is ascribed a misorientation angle $= 0^\circ$ with respect to a fixed reference, while an angle $= \theta$ ($\neq 0^\circ$) is ascribed for the right-hand side grain (Grain II) in Fig. 1 (a).

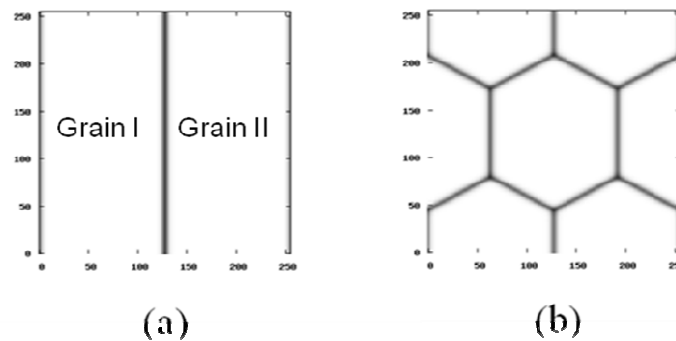


Fig. 1 Grain structures generated by phase-field simulations for (a) bi-crystal and (b) 4 grain system (4 order arameters).

Coherent precipitates inside grains and grain boundary segregation. We considered diffusion of species at or near the grain boundaries and within the bulk. In order to describe the grain structure-dependence of the local free energy density on the static grain structure, we propose a local free energy density (f) based on the regular solution free energy model:

$$f = X(\vec{r}) \cdot \mu(\vec{r}) + (1 - X(\vec{r})) \cdot \mu_h + [X(\vec{r}) \cdot \ln X(\vec{r}) + (1 - X(\vec{r})) \cdot \ln(1 - X(\vec{r}))] + A(\vec{r}) \cdot X(\vec{r}) \cdot (1 - X(\vec{r})) \quad (10)$$

where $\mu(\vec{r})$ is the position-dependent chemical potential of solute, μ_h is the chemical potential of the host crystal, and $A(\vec{r})$ is the position-dependent regular solution parameter. The position-dependent chemical potential is defined as

$$\mu(\vec{r}) = \mu_{bulk} + (\mu_{gb} - \mu_{bulk}) \cdot \left[1 - \sum_g \eta_g^2(\vec{r}) \right] / \left[1 - (\sum_g \eta_g^2(\vec{r}))_{\min} \right], \quad (11)$$

where μ_{bulk} is the chemical potential of solutes within grains, and μ_{gb} is the chemical potential of solutes at the grain boundary. The chemical potential is a function of the order parameters describing the grain structure. The summation of squares of order parameters is equal to unity within a grain, and is less than unity at the grain boundary. The behavior of solute segregation can be controlled by adjusting the difference between μ_{bulk} and μ_{gb} . In a similar way, we define the position-dependent regular solution parameter as

$$A(\vec{r}) = A_{bulk} + (A_{gb} - A_{bulk}) \cdot \left[1 - \sum_g \eta_g^2(\vec{r}) \right] / \left[1 - (\sum_g \eta_g^2(\vec{r}))_{\min} \right], \quad (12)$$

where A_{bulk} is the regular solution parameter within grains, and A_{gb} is the regular solution parameter at the grain boundary.

The temporal and spatial evolution of the composition field ($X(\vec{r})$) can be obtained by solving the Cahn-Hilliard nonlinear diffusion equation (Eq. 2). Substituting the total free energy F (Eq. 1) with the assumption that M is a constant, and plugging the local elastic energy (E_{el}) [17]

$$E_{el} = \frac{1}{2} C_{ijkl}(\vec{r}) (\bar{\varepsilon}_{ij} + \delta\varepsilon_{ij}(\vec{r}) - \varepsilon_{ij}^o(\vec{r})) (\bar{\varepsilon}_{kl} + \delta\varepsilon_{kl}(\vec{r}) - \varepsilon_{kl}^o(\vec{r})), \quad (13)$$

into Eq. 2, we obtain

$$\frac{\partial X}{\partial t} = M \nabla^2 \left(\frac{\partial f}{\partial X} - \kappa_c \nabla^2 X - C_{ijkl}(\vec{r}) (\bar{\varepsilon}_{ij} + \delta\varepsilon_{ij}(\vec{r}) - \varepsilon_{ij}^o(\vec{r})) \frac{\partial \varepsilon_{kl}^o}{\partial X} \right). \quad (14)$$

The diffusion equation (Eq. 14) was solved using the semi-implicit Fourier-spectral method [19-20], i.e. the temporal evolution of the compositional distribution was obtained by solving

$$\tilde{X}(\vec{k}, t + \Delta t) = \frac{\tilde{X}(\vec{k}, t) - \Delta t (k)^2 \left[\left(\frac{\partial f}{\partial X} \right)_{\vec{k}} - \left(C_{ijkl}(\vec{r}) (\bar{\varepsilon}_{ij} + \delta\varepsilon_{ij}(\vec{r}) - \varepsilon_{ij}^o(\vec{r})) \frac{\partial \varepsilon_{kl}^o}{\partial X} \right)_{\vec{k}} \right]}{1 + \Delta t \kappa_c (k)^4}, \quad (15)$$

where \vec{k} is the wave vector in Fourier space, k is the magnitude of \vec{k} , \tilde{X} is the Fourier transform of X , Δt is the time step for integration, and the subscript \vec{k} means the Fourier transform.

We first introduced two circular precipitates into the grain structure in Fig. 1(a). One was embedded in Grain I, while the other was embedded in Grain II. The initially sharp interfaces were relaxed by solving the Cahn-Hilliard equation without elasticity for hundred time steps. The chemical potentials of solutes in the bulk and the grain boundary were chosen to be $\mu_{bulk}^* = 0.5$ and $\mu_{gb}^* = 0.25$, respectively, to induce the segregation of solutes to a grain boundary. In addition, we used $A_{bulk}^* = 3.0$ and $A_{gb}^* = 3.0$ as the regular solution parameters to make the free energy density a double-well type potential. To obtain the morphologies of precipitates under inhomogeneous elasticity, the diffusion equation containing inhomogeneous elasticity contribution (Eq. 15) was numerically solved for 10000 time steps. As shown in Fig. 2(a) and (b), the morphology of the precipitate is cubic with rounded corners. The precipitate embedded in Grain II is rotated by 60° with respect to the precipitate embedded in Grain I in Fig. 2(a). From the morphologies of the precipitates, we can confirm that the inhomogeneous, anisotropic elasticity is properly incorporated by our proposed model, and the model successfully describes the orientation of the precipitates in the differently oriented grains. We assigned different misorientation angles such as 45° for Grain II as shown in Fig. 2 (b) and applied the inhomogeneous elasticity model to the case of multi-grain system as shown in Fig. 2 (c). In all cases, we obtained precipitates with correct orientations.

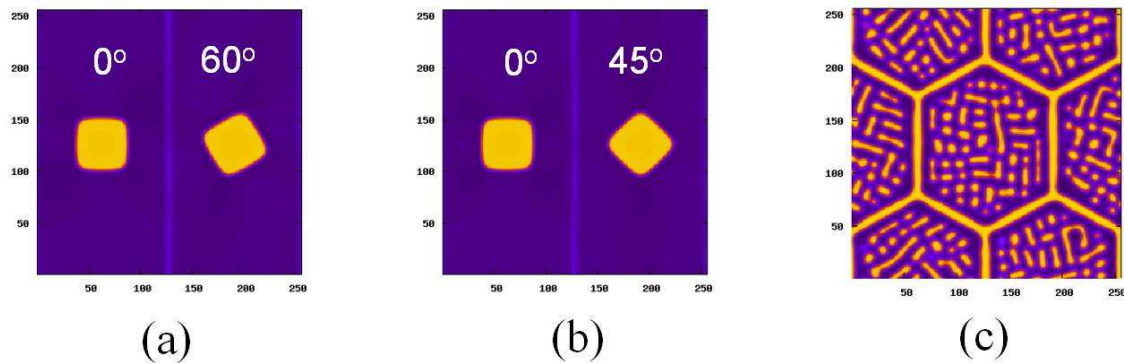


Fig. 2 Coherent precipitates inside grains in different crystallographic orientations in the case of (a) bi-crystal (60° grain II), (b) bi-crystal (45° grain II), and (c) 4 grains system.

The presence of the precipitates inside the grains generates the elastic strain field. In addition, the elastic field directly affects the diffusion process according to the equation [21]:

$$J = cM(F + X_i \Delta \Omega \nabla P), \quad (16)$$

where J is the flux, c is the total concentration, M is the diffusion mobility, F is the driving force for diffusion except the local pressure effect, X_i is the mole fraction of species i , $\Delta \Omega$ is the pure dilation during the atomic jump, and P is the local pressure defined by $P = \frac{\sigma_{xx} + \sigma_{yy}}{2}$ in two dimensions.

Consequently, the diffusion kinetics can be significantly affected by the local pressure fields. We monitored the effects of the local pressure on the solute segregation at grain boundaries. The composition and local pressure profiles along the grain boundary are shown in Fig. 3. The solute segregation at the regions with locally maximum compressive local pressure is suppressed. On the other hand, the segregation composition at the locally minimum compressive region has a local maximum value. Since we assume dilatational eigenstrain for solute atoms, the solute atoms do not favor regions with compressive local pressure since these regions provide smaller space for accommodation of the atoms. The solute segregation behavior under local pressure is similar to that near a dislocation [22].

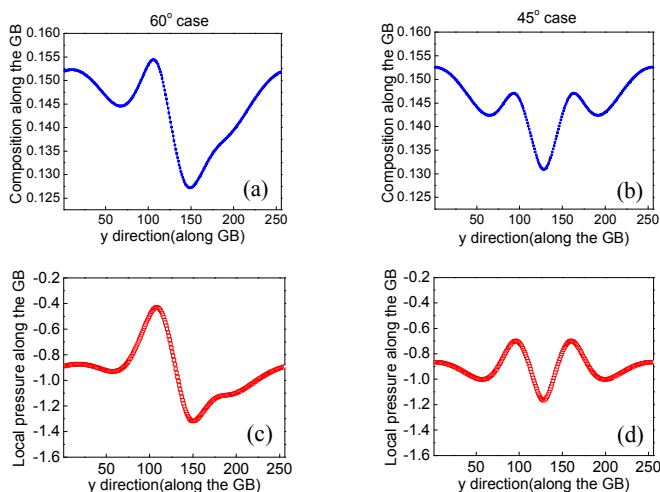


Fig. 3 Grain boundary segregation composition ((a): 60° case and (b): 45° case) and local pressure ((c): 60° case and (d): 45° case) distributions along the grain boundary

Summary

We successfully implemented the inhomogeneous elasticity model for predicting the diffusional phase transformation in polycrystals in phase-field simulations. The model is able to predict both the morphology of coherent precipitates within grains and grain boundary segregation. However, stress relaxation due

to the atomic rearrangement near grain boundary region, e.g., through grain boundary migration at high temperatures, is not considered in this work since the present work is focused on phase transformations in a static grain structure. Three-dimensional simulations of precipitate microstructure evolution in both static and evolving polycrystalline Ni-alloys are currently underway.

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References

- [1] M.P. Seah: J. Phys. F: Metal Physics Vol.10 (1980), p. 1043.
- [2] T. Kinoshita, S. Munekawa and S.-I. Tanaka: Acta mater. Vol. 45 (1997), p. 801.
- [3] L. Heatherly and E. P. George: Acta mater. Vol. 49 (2001), p. 289.
- [4] T. Kinoshita and S. Munekawa: Acta mater. Vol. 45 (1997), p. 2001.
- [5] L. Q. Chen: Annu. Rev. Mater. Res. Vol. 32 (2002), p. 113
- [6] W. J. Boettinger, J. A. Warren, C. Beckermann and A. Karma: Annu. Rev. Mater. Res. Vol. 32 (2002), p. 163
- [7] L. Granasy, T. Pusztai, T. Borzsonyi, G. Toth, G. Tegze, J. A. Warren and J. F. Douglas: J. Mater. Res. Vol. 21 (2006), p. 309
- [8] H. Emmerich: Adv. Phys. Vol. 57 (2008) p. 1
- [9] N. Moelans, B. Blanpain and P. Wollants: Comput. Coupling Phase Diagr. Thermochem. Vol. 32 (2008), p. 268
- [10] I. Steinbach: Modelling Simul. Mater. Sci. Eng. Vol. 17 (2009), p. 073001
- [11] J. W. Cahn: Acta Metall. Vol. 9 (1961), p. 795
- [12] S. M. Allen and J. W. Cahn: Acta Metall. Vol. 27 (1979), p. 1085
- [13] S. Bhattacharyya, T. W. Heo, K. Chang and L. Q. Chen: submitted to Commun. Comput. Phys. (2010)
- [14] S. Y. Hu and L. Q. Chen: Acta mater. Vol. 49 (2001), p. 1879
- [15] P. Yu, S. Y. Hu, L. Q. Chen and Q. Du: J. Comput. Phys. Vol. 208 (2005), p. 34
- [16] D. Y. Li and L. Q. Chen: Acta Mater. Vol. 45 (1997), p. 2435
- [17] A. G. Khachaturyan: *Theory of Structural Transformations in Solids* (John-Wiley and Sons, New York 1983).
- [18] L. Q. Chen and W. Yang: Phys. Rev. B Vol. 50 (1994), p. 15752
- [19] L. Q. Chen and J. Shen: Comput. Phys. Commun. Vol. 108 (1998), p. 147
- [20] J. Zhu, L. Q. Chen, J. Shen and V. Tikare: Phys. Rev. E Vol. 60 (1999), p. 3564
- [21] R. W. Balluffi, S. M. Allen and W. C. Carter: Kinetics of Materials (John-Wiley and Sons, New Jersey, 2005).
- [22] S. Y. Hu and L. Q. Chen: Acta mater. Vol. 49 (2001), p. 463

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