Equilibrium Morphology of Misfit Particles in Elastically Stressed Solids under Chemo-Mechanical Equilibrium Conditions

Xujun Zhao[1] , Stéphane P.A. Bordas[2] and Jianmin Qu[3a)]

[1]*Mathematics and Computer Science Division, Argonne National Laboratory, Lemont, IL 60439, USA*
[2]*Research Unit in Engineering, Computational Mechanics Group, University of Luxembourg, Luxembourg*
[3]*Department of Mechanical Engineering, Tufts University, Medford, MA 02155, USA*

Abstract

This paper studies the effects of chemical, elastic and interfacial energies on the equilibrium morphology of misfit particles due to phase separation in binary alloys under chemo-mechanical equilibrium conditions. A continuum framework that governs the chemo-mechanical equilibrium of the system is first developed using a variational approach by treating the phase interface as a sharp interface endowed with interfacial excess energy. An extended finite element method (XFEM) in conjunction with the level set method is then developed to simulate the behaviors of the coupled chemo-mechanical system. The coupled chemo-mechanics model together with the numerical techniques developed here provides an efficient simulation tool to predict the equilibrium morphologies of precipitates in phase separate alloys.

Keywords: precipitates, chemo-mechanics, interfacial energy, XFEM.

Introduction

When a solid solution is in a non-equilibrium state, which may be caused by external factors such as heat treatment or other perturbations, its internal microstructure will gradually evolve into other states that are more thermodynamically stable. During this process, atoms tend to rearrange their spatial positions to reduce the total system free energy. Consequently, second phase particles may form, grow, and gradually change their size, shape, morphology, and even chemical composition. Since the thermomechanical properties of such phase separated alloys, to a large extent, depend on the microstructure of the separated phases, the ability to control the morphological evolution of the phases enables the design and processing of high performance alloys. Therefore, it is of paramount importance to understand the morphological development of phase separations resulting from diffusional phase transformations. Furthermore, effective and efficient simulation tools are needed to predict the stable morphology of the separated phases.

This paper resents a two-phase continuum model for a generally anisotropic elastic system with misfit particles incorporating the chemical free energy, elastic strain energy and interfacial energy, which allows precipitate particles to change both their sizes and shapes under the conditions that the system satisfies chemical equilibrium, mechanical equilibrium and mass conservation. We employ a coherent sharp interface model to implicitly describe the geometry of the precipitate-matrix interface using the level set method. The interface is endowed with an interfacial excess energy, which includes two parts: the interfacial chemical free energy and the interfacial elastic energy. The interfacial elastic energy is described using the Gurtin-Murdoch interface elasticity model [1]. The chemo-mechanically coupled equilibrium equations and the interfacial equilibrium conditions are derived by minimizing the total system free energy using a variational approach, and the kinetics of phase transformations is derived using a time derivative approach. These equations are then solved via a coupled nonlinear extended finite element method (XFEM), which allows the phase interface to be completely

[a)] Corresponding author. Email: jianmin.qu@tufts.edu

independent of the underlying meshes, so that remeshing is avoided when the interface evolves. The driving force for the interface to evolve in the direction of reducing the total system energy is also derived and connected to the level set evolution equation.

Problem Formulation

We consider a binary solid A_cB_{1-c} containing species A and species B, where c is the molar fraction (concentration) of species A. We assume that the solid in consideration can be represented by the network model of Larche and Cahn [2]. Specifically, we assume that the lattice sites of species B form a network within which species A can move (diffuse). This allows the definition of a displacement and hence a strain of the solid. We further assume that the solid can segregate into two phases, the matrix phase occupying the volume V_m and the precipitate phase occupying the volume V_p. The total volume of the solid is thus $V = V_m + V_p$. The precipitate-matrix interface denoted by Γ is assumed coherent.

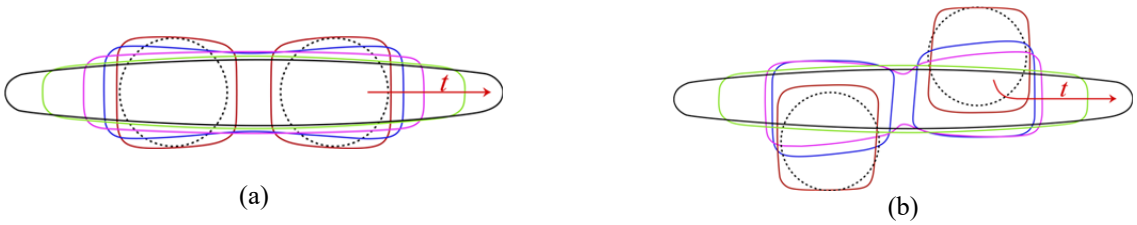
In the bulk, the equations that govern the morphological evolution are derived by considering the total strain energy, $F_{elastic} = \int_V w(c,\boldsymbol{\varepsilon})dV = \frac{1}{2}\int_V (\boldsymbol{\varepsilon} - \boldsymbol{\varepsilon}^c) : \mathbf{C} : (\boldsymbol{\varepsilon} - \boldsymbol{\varepsilon}^c)dV$ and the total chemical free energy, $F_{chem} = \int_V f(c)dV$, where $f(c) = cf_A + (1-c)f_B + \Omega c(1-c) + k_B T\left[c\log c + (1-c)\log(1-c)\right]$.

On the interface, the excess free energy, $F_{surf} = \int_\Gamma \left[\gamma(c_s, \boldsymbol{\varepsilon}^s) + f_s(c_s)\right]dS$, will need to be considered, which includes both the elastic part γ and the chemical part f_s.

The equilibrium state of the system is achieved by minimizing $F = F_{chem} + F_{elastic} + F_{surf}$, or setting $\delta F = 0$. To keep track of the evolution of the interface, we take advantage of the level set approach. By enforcing that at each time step of the morphological evolution, the total material derivative of F must be non-positive, we obtain the normal velocity of the interface $v_n = \mathbf{v}\cdot\mathbf{n} = \mathbf{n}\cdot[\![\Sigma]\!]\cdot\mathbf{n} + (\gamma + \pi_s)\kappa - \mathbf{n}\cdot\nabla_s\cdot\left(\boldsymbol{\sigma}^s\cdot\nabla_s\mathbf{u}\right)$.

Numerical Examples

The interface velocity is a function of chemical concentration, elastic deformation and interfacial energy/stress, which are obtained from $\delta F = 0$, and the mass conservation condition. These equations are fully coupled and need to be solved simultaneously. In this work, we developed a hybrid nonlinear extended finite element method (XFEM) that combines with the level set method to compute the coupled chemo-mechanical fields. Showing in Fig. 1a is an example of initially two separate collinearly aligned particles that eventually merged to form an elongated needle-like particle. A similar example is shown in Fig, 1b where the two separate particles are not aligned initially. The precipitate particles considered here are $\gamma - \gamma'$ in Ni-alloys. Cubic elastic anisotropy is included.



(a) (b)

Fig. 1 Morphological evolution of two-particle systems with dilatational eigenstrain when (a) the particles are aligned initially, (b) the particles are not aligned initially. The dotted lines represent the initial shapes of particles and the arrows point to the evolution directions of interfaces when time step increases.

Conclusions

This work studied the effects of elastic and interfacial energies on equilibrium morphologies of misfit particles due to phase separation in binary alloys under chemo-mechanical equilibrium conditions. First, a continuum framework that governs the chemo-mechanical equilibriums of both the bulk and interface is developed using a variational approach by treating the phase interface as a sharp interface endowed with interfacial excess energy. Second, an extended finite element method (XFEM) in conjunction with the level set method is developed to simulate the behavior of the coupled chemo-mechanical system.

By using the XFEM developed here, numerical examples have been carried out on Ni-Al alloys to investigate the effects of chemical energy, elastic strain energy, and the interfacial energy on the morphological evolution of precipitate particles. The simulated results show that the morphological evolution of a phase separate binary alloy is governed by the competition among three energetic driving forces, bulk chemical energy, bulk elastic strain energy and interfacial excess energy. To study the roles that these energies play, two dimensionless parameter α and β are introduced, which represent the ratio between the bulk chemical free energy density and the bulk elastic strain energy density, and the ratio between the bulk elastic strain energy and the interfacial excess energy, respectively. Under a given α, in material systems with larger β, the interfacial excess energy is relatively low. Therefore, misfit particles tend to grow in size and may eventually coalesce. The relatively low interfacial excess energy density (which is assume isotropic) also leads to square-like or rod-like precipitate shapes due to the system's tendency to reduce its elastic strain energy by growing the misfit particle along elastic soft directions. Vice versa, for systems with smaller β, interfacial excess energy dominates. Thus, misfit particles are usually spherical. Smaller ones tend to shrink in size and may eventually disappear. Multiple misfit particles may choose to stay separated because the high interface energy suppresses particle growth, and there is very little driving force for the particle to move toward each other without changing their volumes. In general, misfit particles tend to align themselves along an elastically soft direction. Under given β, material systems with larger α have higher chemical energy, and lower elastic strain energy, which favors separation. Thus, misfit particles tend to grow. On the other hand, the same misfit particles in systems with smaller α may shrink and eventually disappear.

The simulated behavior for the Ni-alloys agrees well with experimental observations, demonstrating that the coupled chemo-mechanics model together with the numerical techniques developed here provides an efficient simulation tool to predict the equilibrium morphologies of precipitates in phase separate alloys.

References

[1] Gurtin, M.E. and A.I. Murdoch. *Archive for Rational Mechanics and Analysis*, 1975. 57(4): p. 291-323.

[2] Larche, F. and J.W. Cahn. *Acta Metallurgica*, 1973. 21(8): p. 1051-1063.