Computational Materials Science 112 (2016) 128–138

Contents lists available at ScienceDirect

Computational Materials Science

journal homepage: www.elsevier.com/locate/commatsci

Editor's Choice

A nucleation algorithm for the coupled conserved–nonconserved phase field model

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ARTICLE INFO

Article history:
Received 8 July 2015
Received in revised form 5 October 2015
Accepted 6 October 2015
Available online 19 November 2015

Keywords:
Phase field modeling
Finite element method
Nucleation
Time adaptivity
Mesh adaptivity

ABSTRACT

This paper presents a refinement to the existing nucleation algorithm for a coupled conserved–nonconserved phase field model. In the new method, which offers greater ease of implementation as compared to the existing approach, only the nonconserved order parameter is modified to seed supercritical nuclei (thus termed order-parameter-only seeding). The order-parameter-only seeding method naturally satisfies the conservation law for the conserved order parameter. In addition, the implementation within a finite element framework is described. The evolution of a single nucleus is examined to ensure that the precipitate growth kinetics are not affected by the seeding method. We find that, after a brief initial transient period, order-parameter-only nucleation yields similar precipitate growth characteristics to that of the existing model. The kinetics of a phase transformation exhibiting concurrent nucleation and growth is analyzed in the form of the Avrami equation, and a statistical analysis is performed to determine if mesh and/or time adaptivity affects the simulation results. The statistical analysis indicates that the nucleation algorithm is amenable to adaptive meshing and adaptive time stepping.

1. Introduction

Nucleation and growth are key phenomena in phase transformations such as solidification and solid–solid precipitation. Nucleation behavior can have a major impact on the resulting microstructure and material properties. For example, precipitation hardening of alloys involves the formation of second-phase particles. The mechanical properties of such materials are dependent on precipitate size and spatial distribution; these characteristics are controlled in part by the nucleation behavior [1]. Thus, modeling of nucleation and growth is of major technological and scientific importance for materials development and design [2,3]. One means of modeling microstructural evolution is the phase field approach, which has been successfully employed to simulate phase transformations such as spinodal decomposition [4–7], coarsening [8–12], solidification [13–15], and thin film growth [6,16–19]. Comprehensive descriptions and reviews of phase field modeling are found in Refs. [20–24].

In a phase field model, the microstructure is described by one or more continuous conserved or nonconserved field variables, termed order parameters. An order parameter is generally denoted as $\phi(\mathbf{r}, t)$ and indicates the phase at $\mathbf{r}$, where $\mathbf{r}$ is position and t is time. Each phase is designated by a bulk value (e.g., $\phi = 1$ for the α phase and $\phi = 0$ for the β phase), and the value of ϕ changes smoothly between phases to yield a diffuse interface that has a finite width. The position of the interface between the phases is described by an intermediate value (e.g., $\phi = 0.5$). Thus, the phase field methodology eliminates the need to track interface positions explicitly. The free energy of the system is described as a functional of the order parameters, and the evolution of the system is driven by the reduction of the free energy.

Nucleation is commonly handled in one of two approaches within a phase field model [20], though other approaches exist as well. In the first method, a random noise term satisfying the fluctuation–dissipation theorem is added to the time evolution equation to model atomic-scale thermal fluctuations, giving rise to homogeneous nucleation [25]. However, the spatial and temporal resolutions required to accurately describe these fluctuations are computationally prohibitive with existing resources except for when modeling extremely small volumes. In practice, unphysically large random noise is often used in the early stage of a simulation to induce the formation of second-phase particles. After their formation, the noise is deactivated and growth of the particles ensues [25–27]. Similarly, heterogeneous nucleation on microstructural defects and walls has been modeled by the use of white- and colored-noise terms in the evolution equation [28].

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http://dx.doi.org/10.1016/j.commatsci.2015.10.009
0927-0256/

An alternative to the aforementioned noise-based methods is explicit introduction of nuclei into the phase field simulation. In this method, classical nucleation theory is applied to a phase field model described by coupled conserved–nonconserved order parameters [29]. The local nucleation probability for each discretized volume in the simulation is calculated as a function of the local nucleation rate, and nucleation occurs stochastically as a function of the local nucleation probability. If nucleation occurs, a supercritical nucleus is introduced into the simulation by changing the value of the local composition field. To satisfy mass conservation, a depletion region around the nucleus must be created in the composition field. The algorithm was initially implemented with a depletion region described by a discontinuous concentration profile [29]. In subsequent work, a depletion region profile derived from the Zener gradient approximation [30] was demonstrated. This method was further modified by introducing a diffusion smoothing step after nucleus introduction [31] to create a smooth composition gradient at the nucleus/matrix interface.

Several other phase field treatments of nucleation exist in addition to the two aforementioned approaches. Microscopic phase field formulations of heterogeneous nucleation on defects were developed, in which the energy of a defect, such as a dislocation, is added into the local free energy to induce nucleation on the defect [32,33]. In addition, several methods have been developed to numerically determine the critical nucleus shape. In one approach, the critical nucleus profile under solidification conditions was determined by finding the time-independent solutions of the phase field evolution equations for a given concentration [34]. To obtain the shapes of critical nuclei in solid–solid phase transformations, which could deviate from a spherical shape due to elastic effects, both the nudged elastic band method [35] and the minimax technique [36–38] have been utilized.

In this paper, a refinement to the existing nucleation algorithm for a coupled conserved–nonconserved phase field model is presented. In the new method, only the nonconserved order parameter is modified to seed supercritical nuclei (thus termed order-parameter-only, or OPO, seeding). The OPO seeding method is easier to implement than the original method [29,30,39] and naturally satisfies the conservation law for the conserved order parameter. The implementation within a finite element framework is described, and the evolution of a single nucleus is discussed. The kinetics of a phase transformation exhibiting concurrent nucleation and growth is analyzed in the form of the Avrami equation, and a statistical analysis is performed to determine if mesh and/or time adaptivity affects the simulation results. While the implementation was performed within a finite element framework, the method may be used with other numerical methods such as the finite difference method.

2. Methods

2.1. Phase field model

The proposed OPO seeding method relies on the coupling between a conserved order parameter and a nonconserved order parameter through the free energy. Although OPO seeding can be implemented into any coupled conserved–nonconserved phase field model, a model describing the zirconium/zirconium hydride system [40] is adopted to demonstrate the approach. The zirconium/zirconium hydride system is described using a conserved field variable, c, that represents the concentration of hydrogen, and a nonconserved structural order parameter, η, that distinguishes the different structures of the two phases, where $\eta = 0$ for zirconium and $\eta = 1.5$ for the hydride. To demonstrate the nucleation algorithm, the free energy functional from Ref. [40] is simplified by neglecting elastic energy and by retaining only one structural order parameter corresponding to one of the three equivalent orientation variants. The free energy is given as

$$F = \int_V \left\{ f_{chem}(c,\eta) + \frac{\kappa_c}{2}|\nabla c|^2 + \frac{\kappa_\eta}{2}|\nabla \eta|^2 \right\} dV, \tag{1}$$

where κ_c is the gradient energy coefficient for concentration, κ_η is the gradient energy coefficient for the structural order parameter, f_{chem} is the bulk chemical free energy density, and V is the volume. The expression for f_{chem} used in this work is

$$f_{chem}(c,\eta) = \frac{A_1}{2}(c-c_1)^2 + \frac{A_2}{2}(c-c_2)\eta^2 - \frac{A_3}{4}\eta^4 + \frac{A_4}{6}\eta^6, \tag{2}$$

where c_1, c_2, A_i $(i = 1,\ldots,4)$ are constants. The values of c_1 and c_2 are the equilibrium concentrations of hydrogen in the matrix and hydride, respectively, while the A_i's control the shape of the free energy surface [40].

The microstructural evolution of the system is governed by coupled conserved–nonconserved dynamics. The Cahn–Hilliard equation [7] governs the evolution of the concentration of hydrogen as

$$\frac{\partial c}{\partial t} = \nabla \cdot [M \nabla \mu], \tag{3}$$

where the chemical potential, μ, is

$$\mu = \frac{\partial f_{chem}}{\partial c} - \kappa_c \nabla^2 c, \tag{4}$$

t is the time, and M is the mobility of hydrogen. In addition, the Allen–Cahn equation [41] governs the evolution of the nonconserved structural order parameter as

$$\frac{\partial \eta}{\partial t} = -L\left[\frac{\partial f_{chem}}{\partial \eta} - \kappa_\eta \nabla^2 \eta\right], \tag{5}$$

where L is the kinetic coefficient. Both the mobility and the kinetic coefficient are assumed to be isotropic. Following Ref. [40], we use $L = M = 0.4$, $A_1 = 18.5$, $A_2 = -8.5$, $A_3 = 11.5$, $A_4 = 4.5$, $\kappa_c = \kappa_\eta = 1.5, c_1 = 0.006$, and $c_2 = 0.59$ in nondimensionalized units. Nondimensionalization of the governing equations is found in Appendix A.

2.2. Explicit nucleation algorithm

Simmons et al. pioneered the development of an algorithm that explicitly introduces nuclei in a phase field simulation based on classical nucleation theory [29]. Hereafter, this method will be referred to as the explicit nucleation algorithm. The concepts from that algorithm that are incorporated into our model are summarized in this section. The local nucleation rate for critical nuclei, $J^*(\mathbf{r},t)$, is calculated following classical nucleation theory [42] as [29]

$$J^*(\mathbf{r},t) = ZN\beta^* \exp\left(\frac{-\Delta G^*}{k_B T}\right) \exp\left(\frac{-\tau}{t}\right), \tag{6}$$

where Z is the Zeldovich correction factor, N is the number of atoms in the element, β^* is the frequency at which a critical nucleus becomes supercritical, T is the temperature, k_B is the Boltzmann constant, τ is the incubation time, and ΔG^* is the critical nucleus activation energy. For a given volume, the nucleation rate is then given by

$$J^*(\mathbf{r},t) = Zn\Delta v\beta^* \exp\left(\frac{-\Delta G^*}{k_B T}\right) \exp\left(\frac{-\tau}{t}\right), \tag{7}$$

where $N = n\Delta v$, n is the number density of atoms and Δv is the volume of the element. Following Ref. [29], Eq. (6) is simplified to

$$J^*(\mathbf{r},t) = k_1 \exp\left(\frac{-k_2}{\Delta c}\right), \tag{8}$$