Computational Materials Science 147 (2018) 353–362

Contents lists available at ScienceDirect

Computational Materials Science

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

How to apply the phase field method to model radiation damage

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ARTICLE INFO

Article history:
Received 30 October 2017
Received in revised form 31 January 2018
Accepted 3 February 2018

Keywords:
Radiation damage
Phase field method
Computational nuclear materials

ABSTRACT

The phase field (PF) method provides a valuable means of predicting radiation induced microstructure evolution of domains ranging roughly from 100 nm to 100 μm in size and for lengths of time ranging from microseconds to years, depending on the rate of diffusion. In this work we summarize how to apply the PF method to modeling radiation damage. We begin by summarizing the PF method, and discuss the numerical solution of the PF equations. We then summarize approaches for representing the free energy of defects within a material. Next, three PF approaches for representing extended defects are discussed: the Wheeler, Boettinger, and McFadden model, the Kim, Kim, Suzuki model, and the Grand Potential model. We use a simple 1D void growth problem to compare the predictions of these models, as well as their computational expense. We end by describing how defect generation, recombination and annihilation at sinks can be described by the PF method, as well as discussing modifications required to model nucleation.

1. Introduction

Materials under irradiation are bombarded with energetic particles, knocking atoms from their lattice sites and creating atoms of other elements due to fission and transmutation. Though this damage occurs at the atomic scale, it results in microstructure evolution that significantly impacts the macroscale behavior and properties of the material (see Fig. 1 for two examples of irradiation-induced microstructure evolution). Possible forms of microstructure evolution include void formation and swelling, dislocation loop formation and growth, irradiation-assisted creep, and constituent redistribution [1]. These changes directly impact material properties, including thermal conductivity, elastic modulus, radiation hardening, and embrittlement. It is essential that we understand and be able to predict the coevolution of the microstructure and properties of irradiated materials to ensure their safety and optimize their performance. Thus, modeling and simulation of irradiated materials is of great importance.

As irradiation damage is inherently an atomistic behavior, modeling and simulation of radiation damage has been done with atomistic simulation approaches for many years. These methods are critically important for understanding the impact of radiation, and they are summarized in other chapters in this collection. While these approaches are constantly being driven to larger length and time scales, they still cannot easily obtain the length and timescales needed to predict irradiation-induced microstructure evolution.

The earliest attempts at modeling microstructure evolution in irradiated materials employed mean field reaction-rate theory approaches [2–4]. In reaction-rate theory, chemical reaction rate equations are used to approximate the evolution of defect concentrations in irradiated materials. The behavior is assumed to be uniform throughout the material, resulting in spatially-independent equations that compute average defect concentrations. Defect generation, recombination, and annihilation at sinks are included with different terms in the rate equations that average these effects across the material. Initial work used small numbers of defects, e.g. monovacancies and monointerstitials to predict void swelling [2]. Later, this approach has been expanded to model many clusters in an approach that is often called cluster dynamics. Cluster dynamics is beginning to be expanded to be spatially resolved as well [5,6]. Rate theory and cluster dynamics are summarized in another chapter in this collection. While rate theory and cluster dynamics are valuable approaches for predicting the average behavior of the material, they do not provide information on the microstructure topology and therefore cannot be directly tied to property evolution. The computational cost of spatially resolved rate theory can be quite high, and therefore is primarily done in 1D for sizes up to 100 nm.

The phase field (PF) method is a flexible tool for modeling spatially-resolved microstructure evolution at diffusional time

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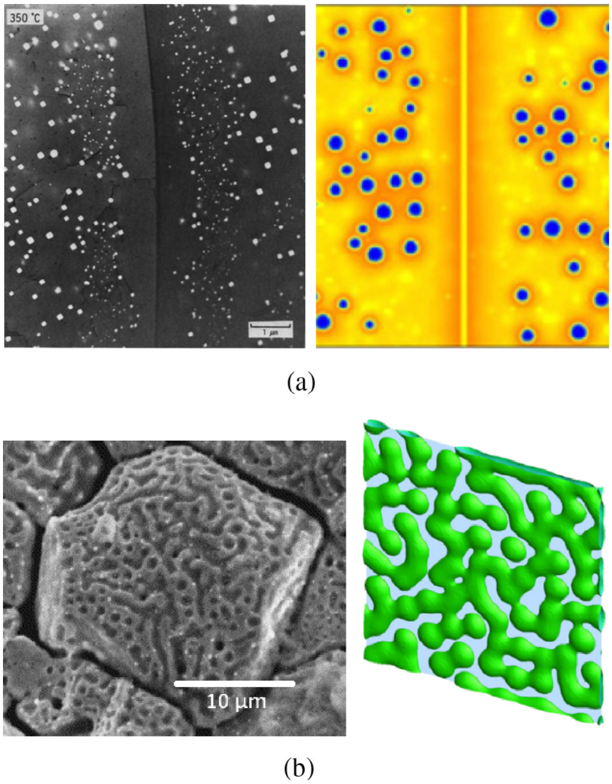
https://doi.org/10.1016/j.commatsci.2018.02.007



(a)

(b)

Fig. 1. Examples of irradiation induced microstructure evolution, with experimental microscopy and PF method simulations. Void formation near grain boundaries is shown in (a), with an experimental micrograph [14] on the left and an example of a PF simulation result [15] on the right. Fission gas bubble percolation on grain boundaries is shown in (b), with an experimental micrograph [16] on the left and a PF simulation of bubble percolation in a bicrystal [17] shown on the right.

scales and at length scales ranging from 100 nm to 100 µm [7–9]. In 2009, the PF method was first applied to model irradiated materials [10,11], and since then it has been used to model a large range of different irradiation-induced phenomena [12,13]. Two examples of PF simulations of radiation damage are shown in Fig. 1. The PF method provides a means of bridging from atomistic simulations to macroscale simulations of material behavior in a radiation environment. It can also employ reaction rates and sink strengths that have been developed for use in rate theory models.

The purpose of this work is to summarize how the PF method can be applied to model radiation damage. Rather then giving a general review of past work that applies the PF method to radiation damage, which has already been accomplished in previous reviews [12,13], here we provide an introduction and general tutorial on what is needed to develop a PF model of radiation damage.

We begin with a summary of how the PF method functions and all the information required to implement a quantitative model. We then include a brief summary of the numerical solution of the PF equations. Next, we discuss functionals that describe the impact of defect concentrations on free energy. The formation and growth of defects are treated similarly to the formation and growth of new phases, and we summarize three approaches to modeling phase transformation. We then discuss the addition of generation, recombination, and sink terms to the model. We end with a discussion on modeling nucleation in the PF method.

2. PF method summary

In the PF method, the microstructure of the material is not represented by a spatial discretization such as a mesh, but rather with variable values. Two types of variables are used, concentrations c_i that describe quantities such as defect or solute concentrations (related to the number and types of atoms present at a specific material point) that are conserved in closed systems, and order parameters ψ_j that describe distinct regions such as phases or grains (related to the structure of the atoms present at the material point) that are not conserved. The values of these variables spatially vary across the material domain, with small changes in value within a given microstructural feature and large changes in value across the interfaces between microstructural features. Thus, all interfaces in the material are diffuse with a finite width. The values of these variables evolve with time to describe the microstructure evolution. The variables evolve to minimize the free energy of the system, which is described by a function $F(c_i, \psi_j, T)$. The general PF approach is very versatile and has been used to model many types of microstructure evolution, as summarized in several review articles [7–9].

The free energy function used in PF models describes the total free energy stored in material domain Γ and has the general form

$$F = \int_\Gamma [f_{loc}(\mathbf{c}, \boldsymbol{\psi}, T) + f_{grad}(\nabla \mathbf{c}, \nabla \boldsymbol{\psi}) + f_{add}((\mathbf{c}, \boldsymbol{\psi})]\, dV, \tag{1}$$

where $\mathbf{c}$ and $\boldsymbol{\psi}$ are vectors containing all c_i and ψ_j used in the model, f_{loc} describes the local free energy density throughout the material and dictates the variable values that represent each microstructural feature. More information regarding f_{loc} for radiation damage models is given in Sections 4 and 5.

f_{grad} serves a numerical and a physical purpose. Numerically, it serves as an energy penalty for sharp interfaces and thus ensures that the interfaces are diffuse. Physically, it represents energy associated with inhomogeneities in chemistry or structure. Though its form does vary in some models, the most common form is

$$f_{grad} = \sum_i^N \frac{\kappa_i}{2} |\nabla c_i|^2 + \sum_j^M \frac{\kappa_j}{2} |\nabla \psi_j|^2, \tag{2}$$

where κ_i and κ_j are the gradient coefficients for the concentrations and order parameters, respectively. The values for the gradient coefficients determine both the width of the interface and the interface energy and can be determined by comparison to sharp interface models or by fitting.

f_{add} describes any additional sources of energy, such as the elastic energy or energy stored in dislocations during plastic deformation, and is not included in every model. The energy density due to elastic deformation is computed via

$$f_{add} = \frac{1}{2} \boldsymbol{\sigma}(\mathbf{c}, \boldsymbol{\psi}) \cdot (\boldsymbol{\epsilon}_{el} - \boldsymbol{\epsilon}_0(\mathbf{c}, \boldsymbol{\psi})), \tag{3}$$

where the elastic strain $\boldsymbol{\epsilon}_{el}$ is impacted by an eigenstrain $\boldsymbol{\epsilon}_0$ that can describe lattice mismatches between phases (which would be a function of order parameters ψ_j) or lattice strains caused by defects (which would be a function of concentrations c_i). The stress is typically computed from the elastic strain

$$\boldsymbol{\sigma} = \mathcal{C}(\mathbf{c}, \boldsymbol{\psi})(\boldsymbol{\epsilon}_{el} - \boldsymbol{\epsilon}_0(\mathbf{c}, \boldsymbol{\psi})), \tag{4}$$

where the elasticity tensor $\mathcal{C}$ is typically different in each phase, making it a function of order parameters ψ_j and could change with concentrations c_i.

The evolution of order parameters is defined by a modified Allen-Cahn equation

$$\dot{\psi}_j = -L_j \frac{\delta F}{\delta \psi_j}, \tag{5}$$

where L is the variable mobility and δ denotes a functional or variational derivative. When considering no sources of additional energy and substituting Eqs. (1) and (2) into (5), we obtain