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Modeling Microstructural Evolution Under Irradiation

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Abstract

Microstructural evolution of materials under irradiation is characterized by some unique features that are not typically present in other application environments. While much understanding has been achieved by experimental studies, the ability to model these microstructural evolution for complex materials states and environmental conditions not only enhances understanding, it also enables prediction of materials behavior under conditions that are difficult to duplicate experimentally. Furthermore, reliable models enable designing materials for improved engineering perform for their respective applications. Thus development and application of mesoscale microstructural model is important for advancing nuclear materials technologies. In the work, the two dominant microstructural models will be described. They are the Potts and phase-field models. Their application to nuclear materials will be reviewed and demonstrated on example microstructural evolution processes, and finally, the current trends in model development will be discussed. The current trend in microstructural models is the development of hybrid models that combine elements of these two models and others to simulate multiple physical phenomena that nuclear materials experience under irradiation.

1 Introduction

Microstructural evolution of nuclear fuels and claddings under irradiation is characterized by some unique features that are not typically present in other application environments. A very wide range of irradiation conditions, radiation with many different particle over a large range of energies, lead to formation of defects in crystalline materials. These defects can interact in many different ways with each other and existing defects such as grain boundaries to give rise to microstructural evolution not observed in other applications. These radiation-induced defects can alter the kinetics by enhancing diffusion. They can have more substantial effects such as void formation due to aggregation of vacancies. Under some conditions, radiation can create a large density of defects causing the crystalline material to transform to an amorphous state.

Swelling is another common consequence of radiation in many materials. The aggregation of vacancies to form voids is observed in many materials including some fuel rod claddings. The production of extra atoms by fission in nuclear fuel can strain the lattice leading to swelling, albeit to a lesser extent. Nuclear fuels can also swell due to precipitation of fission gas (primarily Xe and Kr) into nanosized intragranular bubbles and sub-micron intergranular bubbles. The creation of extra atoms by fission or transmutation of atoms alters the chemistry of the fuel (see the next section for a detailed discussion) and results in microstructural evolution that is unique to nuclear materials.

Another feature that is unique to some nuclear materials it the large temperature gradient present in these materials that lead to different microstructural evolution in the different regions and segregation of components. Restructuring of fast reactor fuels to form an axial pore along the central axis of the fuel pellet is an example of this. Another unique feature of nuclear fuels is the accumulation of radiation damage at the outer rim of the fuels, which in turn drives recrystallization in this region. Recrystallization is virtually non-existent in most ceramics making this phenomenon most interesting in itself, but with great import for fuel performance. Finally, radiation-induced segregation of components in two or more component materials occurs in many materials.

All these processes unique to irradiated materials drive a number of microstructural evolution processes, which are not observed in other materials. The ability to simulate these evolution processes at the microstructural scale would enhance our ability to predict their behavior and hence design them at the microstructural scale for optimal engineering performance. In this section, the application of microstructural evolution models to nuclear materials will be reviewed. While the models will be reviewed briefly, detailed descriptions of the models will be provided in a sister volume. While there are many models for simulating microstructural evolution including front tracking, level-set and cellular automata, two models have dominated the field, phase-field and the Potts model. These models will be reviewed briefly and their utility for understanding nuclear materials behavior and the ability to incorporate them into nuclear materials design will be discussed. Finally, the current trends in materials microstructural model development will be reviewed.

2 Mesoscale Microstructural Models

2.1 Potts kinetic Monte Carlo model

The Potts kinetic Monte Carlo (kMC) is a statistical-mechanical model that populates a lattice with an ensemble of discrete particles to represent and evolve the microstructure. The Potts kMC used for mesoscale simulations is distinct from other kinetic Monte Carlo models used for simulation of atomistic, chemical, neutronic and other materials simulations. In the following sections, the term kMC is used to refer to the Potts kMC model. The particles in the Potts kMC model represent a discrete quantity of material that is much larger than an atom, thus all atomistic information about the material system is aggregated into mesoscale model parameters. In kMC, the particles evolve in a variety of ways to simulate microstructural changes due short- and long-range diffusive processes. kMC methods have proven themselves to be versatile, robust and capable of simulating various microstructural evolution processes. They have the great advantage of being simple and intuitive, while still being a rigorous method that can incorporate all the thermodynamic, kinetic and topological characteristics to simulate complex processes. They are easy to code, readily extendable from 2D to 3D and can simulate the underlying physics of many materials evolution processes based on the statistical-mechanical nature of the model. These processes include curvature-driven grain growth[1,2], anisotropic grain growth[3], recrystallization[4], grain growth in the presence of a pinning phase[5,6], Ostwald ripening[7], and sintering[8,9,10].

Representation of microstructure: The microstructure consists of an ensemble of particles that occupy a regular lattice. Recently, some kMC models have started to use non-regular or non-lattice based methods. However, the vast majority of kMC models use regular lattices and for the purpose of this work, we shall confine our discussion to regular lattices. The particles can be considered to be a discrete amount of material that is much larger than an atom; thus all atomistic characteristics are aggregated into discrete extensive thermodynamic quantities such as mass and energy for each particle. For the simplest case of grain growth, the microstructure is represented by particles identified by an integer value signifying membership in a particular grain. This membership integer value is simply a distinct degenerate state identifying each particle as belonging to a certain grain with no other physical significance. In a two-phase system such as a porous material, the solid material can be assigned one set of spins and all the porosity a single distinct membership value. In a two-phase system consisting of two solid components such as a eutectic material, each phase can be represented by a set of membership integer values. Lattices can be 2D triangular or square, 3D cubic or face-centered cubic or other geometries. The most commonly used geometries are 2D square and 3D cubic lattices.

Energy and thermodynamics: Since kMC uses a discrete ensemble of particles to represent the microstructure, it follows that the total volumetric energy is the sum of energies of each particle. In addition, interfacial energies must also be included in the calculation of the total free energy of the system. Interfaces in kMC is defined by neighboring particle with unlike membership. Different types of interfaces with different energies can be defined. Neighboring particles belonging to two different grains would define a grain boundary or grain particles forming an interface with pore particles would define a pore surface. Each of these can be given its own energy value. The total interfacial energy of the system is then the length in 2D or area in 3D of each interface multiplied by its interfacial energy per unit length or area. Thus, the sum of all unlike neighbor interaction energies of all the particles is E_{int}.

$$E_{int} = \sum_{i=1}^{N} \sum_{j=1}^{n} J_{qi,qj}$$

where i is each particle, N is the total number of particles, j is the neighboring particle, n is the total number of neighbors being considered, qi is the spin of particle i and J is the interaction energy between particles i and j of spins qi and qj. The bulk energy of the system is the sum of the inherent energy of each particle and does not depend on its neighbors.

$$E_{vol} = \sum_{i=1}^{N} V_i$$

where V_i is the volumetric energy of particle i. Examples of volumetric energy may be the volumetric chemical free energy, elastic strain energy or the irradiation damage energy stored in the nuclear fuel. The interfacial and volumetric energies are very versatile and can be formulated to match virtually any energy that is characteristic of real materials. They can be functions of microstructure or materials, so that as the system evolves due to changing chemistry or accumulation of irradiation or other damage, the particle energies can reflect their current thermodynamic state. This ability to easily tailor particle energies makes kMC models highly versatile and widely applicable to many materials evolution processes.

Evolution and kinetics: kMC models have been shown to correctly simulate complex path-dependent evolutionary processes. They yield images with great detail of the microstructure and its evolution in response to a given set of starting conditions and applied conditions. The microstructure evolves in response to local conditions such as curvature, radiation damage, temperature, etc. These local conditions may also be changing with time and position. The basic mechanisms for change in a kMC model are the changes in membership of particles from one grain or phase to another or transport of the particles to a different lattice positions by exchanging places with neighboring particles. These types of changes can be used to simulate many types of transport mechanisms such as grain boundary motion, surface and bulk diffusion, dissolution and precipitation and other mechanisms. The spin change events frequencies are determined using the standard Metropolis algorithm with Boltzmann statistics. A spin change event is identified. The change in total energy of a spin change event is calculated using the equation of state described in the previous section. The probability of this change is

$$\begin{aligned} P &= 1 & \Delta E &\leq 0 \\ P &= \exp\frac{-\Delta E}{k_B T} & \Delta E &> 0 \end{aligned}$$

where k_B is the Boltzman constant, and T is the simulation temperature. The change is performed with this probability by choosing a random number R, uniformly distributed from 0 to 1. If $R < P$ then the event is accepted and a change is made. If not then no change is made, and the original spin is restored. kMC models reduce the total free energy of the system and when implemented correctly reduce free energy along the correct kinetic path of microstructural evolution. These characteristics makes the Potts model well suited for studying microstructural evolution in nuclear materials.

Application of kMC model to nuclear materials: The kMC model has been applied to a variety of processes that are relevant to the simulation of nuclear fuel, clads and other materials. Grain growth was the first and has been the most extensively studied microstructural evolution process. It has been shown that this model simulates curvature driven grain growth with the expected kinetics of $R^2 - R_o^2 = kt$, where R is the average grain radius and R_o is the average grain radius at time $t = 0$ and k is a kinetic constant related to grain boundary diffusivity. Grain growth in systems with immobile, mobile and evolving pinning phases are important processes for nuclear fuels.

Nuclear fuels are often porous at the start of their service and become more so as fission gases are produced during in-reactor service. This porosity can move and coarsen as the matrix coarsens as well. Solid fission products often segregate to grain boundaries and pin them to effectively reduce or stop grain growth. This processes has been extensively studied using the kMC model. Other fission products can segregate to grain boundaries and pin them so that grain growth is suppressed even at high temperatures. This two phase pinning model is described in detail elsewhere[6] and shown to incorporate all the processes necessary to simulate coarsening in a two-phase system such as that found in nuclear fuels. These processes include curvature driven grain growth mediated by pore drag on the grain boundaries, which leads to different grain boundary motion behaviors: slower grain boundary motion due to pore drag, grain boundary break away from very slow pores or complete stagnation of the grain boundary by pores.

In materials that have temperature gradients, particularly the large gradients seen in nuclear materials, microstructural evolution in different regions can vary greatly depending on the local temperature. Thermally activated processes such as grain boundary motion, and surface and bulk diffusion, occur at a faster rate at higher temperatures with the temperature-dependent rate or mobility $M(T)$ is given by the Arrhenius relationship $M = M_o \exp\left(\frac{-Q_A}{k_B T}\right)$ where Mo is a pre-exponential constant and Q_A is the activation energy for that process. In nuclear fuels and clads, most processes are indeed thermally activated, although a few are enhanced by radiation-induced defects as well. The thermally activated processes are treated in the kMC model by varying their mobility as a function of the local temperature. For example, consider grain growth. The grain boundary velocity v is proportional to the driving force (curvature) and mobility as $v = M\kappa$ where κ is the grain boundary curvature. This mobility term is introduced very easily in the kinetic equation as $P_M = M(T)P$ where P_M is the temperature dependent probability term and P is the original probability. It has been shown by Garcia et al.[11] that under continuously varying temperature gradients, the kinetics and topology of the grain growth are locally normal. The local grain size and grain size distribution are the same in the temperature gradient as they would be at the same temperature in an isothermal grain growth simulation. This method for simulation of temperature gradients is correct as long as the mechanism for microstructural evolution and the corresponding activation energy remain constant over the entire temperature range. In two phase systems, the Soret effect that segregates phases is important. This, too, can be simulated in the Potts model by introducing a heat of transport term, Q^* into the energy of the system. An example that combines all the processes described above has been combined in one simulation for illustrating the model capability. Coarsening in a two-phase system with a thermal gradient applied is shown in Figure 1. Curvature-driven grain growth with a mobile pinning phase, in this case gas, is evolving in a temperature gradient. The gas pockets coarsen by coalescence and move by surface diffusion. As one can see, coarsening is much faster at the high temperature region and the gas preferentially diffuses to the higher temperature region as this lowers the total free energy of the system.

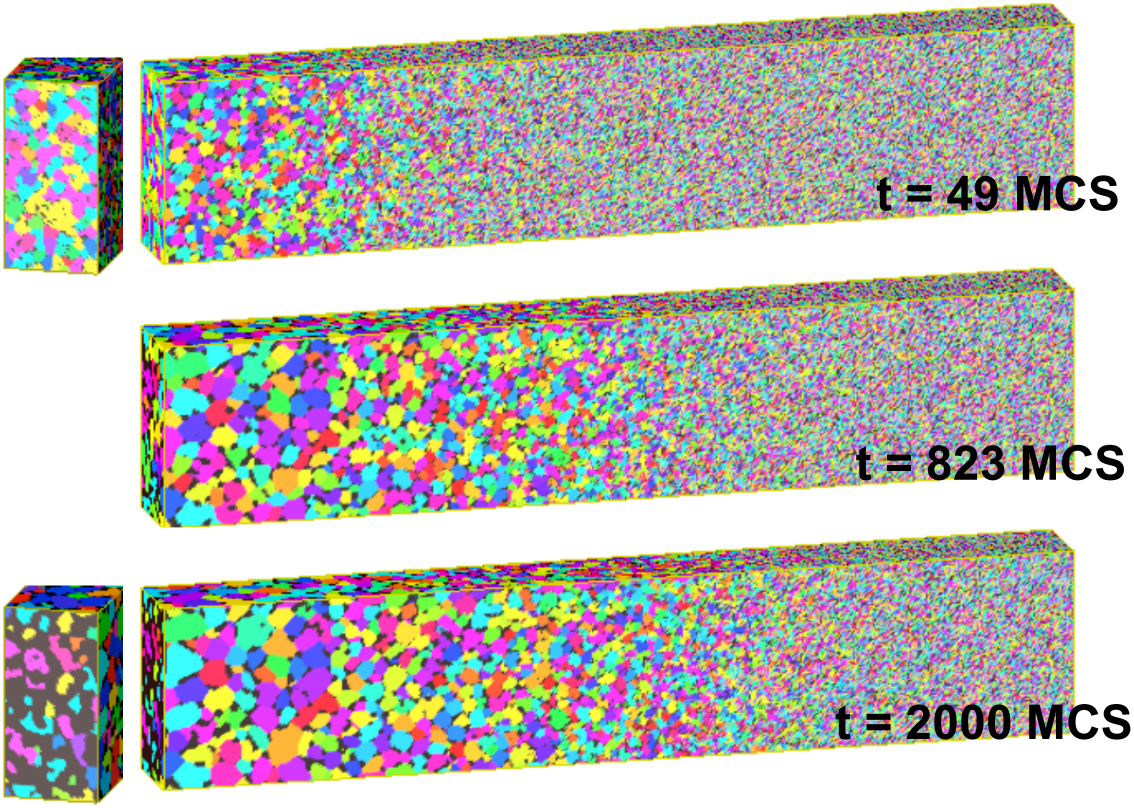

Figure 1. Coarsening of grains and pores in a thermal gradient. Grains and pores coarsen more quickly at the higher temperatures. Pores migrate to the high temperature end due to the Soret effect as shown by the difference in pore volume at the high-temperature edge between time t = 49 and 2,000 MCS.

Recrystallization is an important phenomenon observed in LWR and some other fuels. Rollet et al.[12] developed a recrystallization model and later used it to study abnormal growth[13]. Further evolution of this model was used to study dynamic recrystallization[14].

A preliminary application of kMC model to study recrystallization in the high burn-up rim region of LWR fuels was recently presented by Oh[15] and more developed one by Madison et al.[16] Another very important microstructural evolution process is swelling. This mechanism is very similar to densification during sintering of crystalline materials. A validated kMC model for sintering has been demonstrated by comparison to detailed experimental 3D images of microstructural evolution in Cu powder compacts[17]. While almost all the basic microstructural evolution processes at the mesoscale have been simulated to varying degrees using kMC methods, their application to nuclear fuel and cladding is in its infancy. In order to make significant advances, the models have to be adapted to the specific application conditions of nuclear fuels and coupled so that many different processes (i.e. fission gas generation, diffusion, bubble formation accompanied by swelling and recrystallization) can occur simultaneously. The adaptation of kMC models to the specific processes relevant to nuclear materials and development of coupled models is not trivial. However, the previous use of kMC models to correctly simulate various processes including coupled processes suggests that kMC is a powerful and useful method for modeling microstructural evolution in nuclear materials.

2.2 The phase-field model

In contrast to the Potts kMC model, the phase-field model is a deterministic, continuum, thermodynamic model that describes the microstructure and its evolution in terms of continuum phase-fields that are evolving. These phase-fields can be grain orientations, composition, phases or other microstructural and compositional descriptors. Similar to the analogous descriptor in the Potts model, all atomistic qualities of the system are not directly incorporated. While the phase-fields themselves are continuum quantities, they are digitized and evolved on some predefined grid – typically square in 2D and cubic in 3D. The size of the grid is such that the distance between solution points is much larger than typical interatomic distance but sufficiently small to resolve microstructural features such are grain boundaries. The phase-field method is a powerful mesoscale modeling method that is widely used to model the kinetics of microstructural evolution in materials and is described by Boettinger et al.[18] It has been used to study the evolution of gas bubbles and thermal conductivity in nuclear fuels,[19,20] melting of nuclear fuel[21], void ensembles under irradiation[22,23], precipitate morphology and evolution in alloys[24], nucleation and growth near a dislocation[25], domain structure evolution in ferroelectric materials[26], coarsening of precipitates[27] and solidification[2829]. The mathematical details of the method have been reviewed by Chen[27] and Boettinger et al.[18] and a brief description is presented here.

Microstructural representation: Microstructure is represented by continuum fields called phase-fields. These phase-fields have digital values at discrete grid points. The value of the phase-field can vary continuously over a range that may correspond to some physical descriptor such as composition or grain membership. Another feature of phase-fields is that they have diffuse interfaces meaning that features such as grain boundaries span several grid sites. For illustration, consider a representation of grains in a polycrystal as shown in Fig. 2. Each phase-field η_i in this figure represents member in that particular grain. Within each grain this phase-field is η_i = 1.0 and all others η's = 0.0. However at the grain boundaries they vary continuously with intermediate values to give a diffuse interface. Thus points in the grain boundaries can have partial membership in more than one grain.

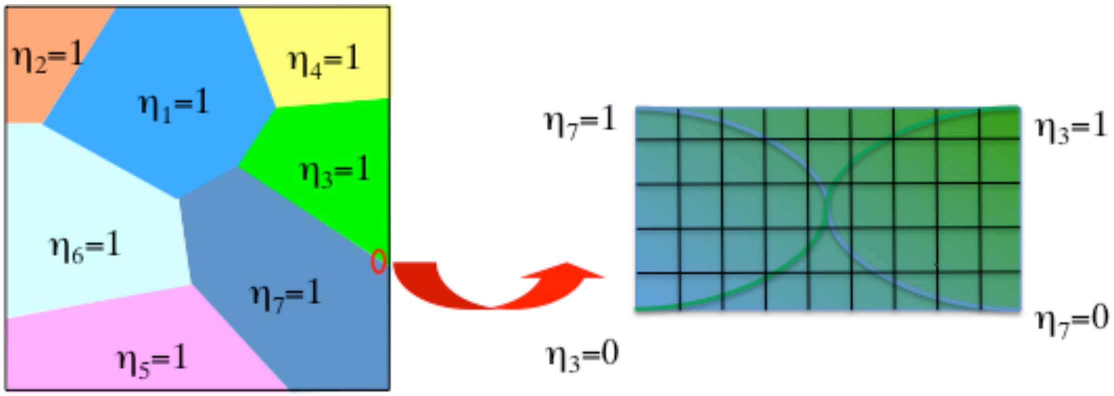

Figure 2. Grain structure is represented in the phase-field model by fields that represent member in different grains η. At grain boundaries the field values vary continuously as shown by the grain boundary between η_7 and η_5.

Thermodynamics and energy: The energy of the system is defined by a free energy functional, which is a function of the phase-fields and their gradients. For the case of grain growth a typical free energy functional that defines the total free energy of the system may be

$$F = \int \left[f_o + \sum_{i=1}^{p} \frac{\kappa_i}{2} (\nabla \eta_i(r))^2 \right] d^3 r$$

It is the integral over the simulation volume of the free energy f_o and all gradient terms at the interfaces. The free energy f_o is a function of all the local phase-fields, and κ is the phase-field gradient coefficient and defines the width of the diffuse interface. p is the total number of phase-fields or grains in this case. The free energy for this particular case is

$$f_o = -\frac{a}{2} \sum_{i=1}^{p} \eta_i^2 + \frac{b}{4} \left(\sum_{i=1}^{p} \eta_i^2 \right)^2 + \gamma \sum_{i=1}^{p} \sum_{j \neq i}^{p} \eta_i^2 \eta_j^2$$

Where a , b and γ are fitting constants. The volumetric free energy function is constructed so that when any one phase-field representing grain member is =1.0 and all others are = 0.0, the system has minimum volumetric free energy. Thus all the energy in the system is due to the interfaces. When calculated, the function defines the total grain boundary energy. The grain boundary energy is considered the only driving force for normal grain growth. For other simulations such as grain growth coupled to diffusion in a two-component system, the free energy can be defined to have volumetric terms that also drive microstructural evolution.

Kinetics and evolution: The evolution of the phase fields are simulated using the time-dependent Ginzburg-Landau equations

$$\frac{\partial \eta_i}{\partial t} = -M\left(\frac{\partial f_o}{\partial \eta_i} - \kappa \nabla^2 \eta_i\right)$$

Thus each phase field $\boldsymbol{\eta}_i$ at each grid location and time t is the variational derivative of the free energy F with respect to that phase field $\boldsymbol{\eta}_i$. M is a mobility term, which in this case is related to the grain boundary mobility. When the phase-field in conserved as in the overall composition of the system the Cahn-Hilliard equation applies

$$\frac{\partial C}{\partial t} = -M\nabla^2\left(\frac{\partial f_o}{\partial C} - \kappa \nabla^2 C\right)$$

Application of these equations result in simulations that minimize the total free energy of the system along paths that are defined by the exact free energy functional, mobilities and other simulations parameters in a deterministic manner.

Application of the phase-field model to nuclear materials: The model described above was used to simulate the precipitation of radiation-formed vacancies on to a void by Rokkum et al[23]. The void grows as the vacancies "deposit" on its surface. The model and simulation conditions are described in detail. The grain is assumed to be supersaturated with vacancies. When annealed at a higher temperature, these vacancies diffuse to the void and precipitate at its surface, thus causing the void to growth and depeleting the grain of vacancies. Figure 3 shows the microstructure of such a system where an intragranular void grows by precipitation of vacancies.

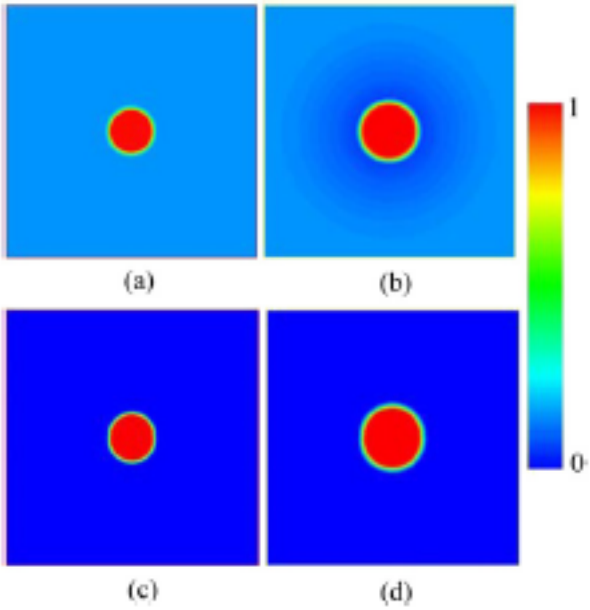

Figure 3. Phase-field simulation of void growing by precipitation of vacancies from a grain supersaturated with vacancies. As the void grows, the grain is depleted of vacancies as shown by the scale to the right showing vacancy concentration. (copyright permission is needed)

A key advantage of the phase-field method for nuclear fuel modeling is that various defects, fission gas bubbles, voids and microstructural features with moving boundaries can be easily treated. In addition, the thermodynamic driving force for microstructure evolution can be incorporated based on input from experimental databases and lower length scale models. These include bulk chemical free energy, interfacial/surface energy, elastic strain energy, and electrostatic energy. Traditionally, phase-field models have simulated quasi-equilibrium phenomena. However, the highly non-equilibrium nature of radiation effects in nuclear fuel can be captured by explicitly including the energetics of processes such as defect production by irradiation and fission gas re-solution as discussed previously.

Currently, it is possible to simultaneously treat the generation of point defects and fission gas atoms, nucleation of gas bubbles at vacancy clusters, gas re-solution due to the collision between gas bubbles and fission fragments or neutrons, interaction of point defects and gas atoms with line defects and interface sinks, elastic interaction between gas atoms and defects, and the inhomogeneity of elasticity and diffusivity[20]. However, simulating two, three or more of these simultaneously in a simulation remains challenging. Existing phase-field models are mainly two-dimensional. The model must be extended to three dimensions to realistically model nuclear fuel. Recently, a three-dimensional phase-field model has been applied[30] to study the microstructure evolution in Ni-based super alloys with construction of local free energy based on thermodynamic databases using the CALPHAD method[31]. Such an approach holds considerable promise for reliable modeling of fuel microstructure. Another major limitation of phase-field models at present is that they provide mainly qualitative information about nuclear fuel microstructural changes. Quantitative modeling requires accurate thermodynamic and kinetic input from lower scales, such as first principles calculations and classical MD simulations. The required input includes the size and spatial distribution of defect clusters in nuclear fuel, relationship between defect concentration and chemical free energy, solubility of fission products, point defect and defect cluster mobilities and binding energies, defect interaction energies, kinetics of gas resolution, and realistic models of grain structure. Quantitative modeling is essential for uncertainty quantification, verification and validation. Validated information from phase-field models can be passed on as constitutive equations to higher-level continuum models of heat conduction and oxygen diffusion in nuclear fuels. An additional challenge with phase-field models is that even when the thermodynamic data is available, incorporating into a simulation is often difficult due to a number of reasons including constructing complex free energy functionals with many different types of order parameters that are numerically stable and evolve correctly to simulate the desired behavior.

Continued application of mesoscale microstructural models to study microstructural evolution in nuclear materials will require further development of the models. The developments will be needed on several fronts. Models that can simulate additional materials processes with additional physics and chemistry, in response to different driving forces and conditions, with coupling of materials physics and processes, and bridging scales both to/from lower length and higher length scale simulations. Advances in numerical techniques and parallel processing would also be useful allowing larger simulations spanning a larger sample size. These might include adaptive meshing or developing techniques to parallelize inherently serial processes.

3 Current Trends and Future Development of Mesoscale Models.

Continued model development of the kMC model has taken the form of developing hybrid models that couple multiple materials physics that the kMC model cannot inherently treat. One of the early examples of this type of modeling was a sintering model that treated all the processes active during solid state sintering except densification by annihilation using kMC. Densification, however, was simulated by a unique method of addressing global shrinkage based on the local microstructural configuration[17]. Another example is for simulation of recrystallization. Application of the Potts model alone with a volumetric free energy modified to include the strain energy density of dislocations in the metal could not simulate the kinetics of recrystallization correctly. Rollett and Raabe[32] showed that a hybrid Potts cellular automaton model correctly simulated the kinetics of recrystallization.

In contrast, the phase-field model has remained essentially the same model. Its development has been in the form of introducing new phase fields to represent new physics. The numerical implementation of additional coupled fields has limited the advancement of phase-field models. Coupling phase fields which introduce additional physics is difficult as the development of free energy functional that are numerically stable and converge to the correct solution is difficult. The evolution of the phase-field model is in improved numerical techniques to enable larger simulations with more coupling of physics. Adaptation and use of advanced numerical solver that converge to solution with less computation has been a large effort to improve phase-field model performance. Development of adaptive meshes that reduce computation in stable non-evolving areas away from the interfaces has been the focus of model improvement. Another is reassignment of degenerate grain orientation locally to both reduce the number of phase fields required and to prevent the artifacts introduced by having a limited number of grain orientations.

An exciting development in mesoscale modeling has been the introduction of a hybrid model that couples phase-field with Potts model[33]. This type of hybrid combines the inherent efficiency and stability of the Potts model with the ability to treat continuously varying materials characteristics such as composition in the phase-field model to enable simulation of microstructural evolution in a new class of materials. One area of microstructural evolution problems has remained challenging. Models that can simulate microstructural evolution that is driven partially by mechanical stresses are the focus of much research. While, some solutions have been found under a limited set of conditions, the development of modeling techniques that would enable simulation microstructural evolution that is partially driving by mechanical stresses has not been achieved. The problems of interest to nuclear materials are swelling in nuclear fuels due to fission gas bubble formation, interactions between clad and pellet, hydride reorientation in cladding during long-term storage and many others.

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