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Design and analysis of forward and reverse models for predicting defect accumulation, defect energetics, and irradiation conditions

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ABSTRACT

The complexity of radiation effects in a material's microstructure makes developing predictive models a difficult task. In principle, a complete list of all possible reactions between defect species being considered can be used to elucidate damage evolution mechanisms and its associated impact on microstructure evolution. However, a central limitation is that many models use a limited and incomplete catalog of defect energetics and associated reactions. Even for a given model, estimating its input parameters remains a challenge, especially for complex material systems. Here, we present a computational analysis to identify the extent to which defect accumulation, energetics, and irradiation conditions can be determined via forward and reverse regression models constructed and trained from large data sets produced by cluster dynamics simulations. A global sensitivity analysis, via Sobol' indices, concisely characterizes parameter sensitivity and demonstrates how this can be connected to variability in defect evolution. Based on this analysis and depending on the definition of what constitutes the input and output spaces, forward and reverse regression models are constructed and allow for the direct calculation of defect accumulation, defect energetics, and irradiation conditions. This computational analysis, exercised on a simplified cluster dynamics model, demonstrates the ability to design predictive surrogate and reduced-order models, and provides guidelines for improving model predictions within the context of forward and reverse engineering of mathematical models for radiation effects in a materials' microstructure.

1. Introduction

Central to the problem of aging and degradation of metals and alloys subjected to radiation environments (e.g., neutrons, electrons, or heavy ions) is the large spectrum of possible microstructure evolution processes that can be simultaneously triggered. These processes range from the generation of point defects, such as vacancies and self-interstitial atoms (SIAs), as well as clusters of these defects, along with their migration, recombination, emission, and annihilation. Over time, the collective interactions between defects can lead to noticeable radiation effects in the materials' microstructure, e.g., void swelling in austenitic steels. In turn, these profound microstructure changes lead to macroscopic degradation of material properties; e.g., material strength, ductile-to-brittle transition temperature shift, etc. [1].

Beginning with the pioneering work of Brailsford et al. [2], a series of computational models have been proposed for predicting microstructure evolution under ion or neutron fluxes. Among others, both atomistic and object oriented kinetic Monte Carlo (akMC/okMC) frameworks [3–5], rate theory and more complex cluster dynamics (CD) methods [6–9] have been used with relative success to rationalize experimentally characterized irradiated microstructures. These computational tools necessitate parameterization from atomistic or density functional theory (DFT) calculations. Such results are used to establish a catalog of formation, migration, and binding energies associated with each defect species and reaction being considered.

Overall, the predictive capability of CD models depends on three aspects. First, the list of reactions being considered should be sufficiently exhaustive so as to not overlook key processes impacting microstructure evolution. For example, in the case of $\langle 0\,0\,1\rangle$ SIA loops in bcc-Fe alloys, their formation process resulting from the short-range interactions between $\frac{1}{2}\langle 1\,1\,1\rangle$ type loops could not be postulated *a priori* without guidance from atomistic simulations [8]. Second, the quantification of the intrinsic thermodynamic

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quantities associated with each defect species (e.g., monovacancy, divacancy, SIA clusters, etc.) necessarily needs to be accurate. However, interatomic potentials used in atomistic simulations can rarely be applied to complex engineering materials (e.g., alloys or strongly correlated materials). Further, DFT methods have intrinsic numerical limitations in representing paramagnetic material systems as well as cells sufficiently large to characterize defect clusters. Consequently, both kMC and CD type approaches are typically limited to either pure systems or idealized material proxies (i.e., focusing on the major alloying element). Third, despite recent progress [10], most models predicting radiation effects do not explicitly represent the materials microstructure (e.g., grain size, texture, dislocations, etc.). In consequence, the quantification of segregation of defects at sinks (i.e., dislocations or grain boundaries) and of their associated bias is obtained from a homogenization viewpoint (i.e. using analytic solutions obtained from auxiliary problems). However, in the case of dislocations, the spatial arrangements between line defects and the nature of the dislocation line (e.g., screw vs. edge) could largely influence the effective bias between vacancies and interstitials [11]. Such considerations are typically missing from kMC and CD computational efforts.

In parallel to continuing developments addressing the limitations of DFT and atomistic simulations, the aforementioned shortcomings can be addressed via the use of data analytics. Within the context of both kMC and CD frameworks, parametric sensitivity analyses have been performed to establish statistically representative correlations between the thermodynamics of defect recombination (input space) and grain boundary sink efficiencies (output space) [12–14] for instance. These types of prospective studies conveyed by forward modeling aim at establishing direct connections between an input space composed of intrinsic properties of various irradiation-induced defect species and a resulting output space composed of the defect clusters of various natures and sizes within the damaged microstructure.

This study largely expands upon these views and outlines a method for constructing forward and reverse reduced-order models linking damage state, intrinsic material defects properties, and irradiation conditions. Among other aspects, this study aims at demonstrating how the combined use of CD and statistical analysis can assist in identifying the essential intrinsic defect properties, as well as the necessary accuracy in their quantification, to develop and calibrate predictive CD type model in complex material systems. For the sake of demonstration, a simple CD model is used to perform a global sensitivity analysis with the intent to characterize parameter sensitivity (defect energetics and irradiation conditions) and to demonstrate how this can be connected to variability in defect evolution (defect concentration and size). A forward regression model allows for the calculation of defect cluster size and concentration for a given set of defect energies, dose rate, and temperature, whereas a reverse regression model enables the extraction of defect energetics for a given set of defect cluster size and concentration. Additionally, a so-called "forensic model" is established for the determination of irradiation conditions, e.g., temperature and dose rate, for a given set of defect energies, cluster size, and concentration.

The subsequent sections are organized to illustrate these points. Section 2 presents the modeling and statistical methodologies utilized to design and construct the forward and reverse reduced-order models. Specifically, Section 2.1 provides details of the simplified deterministic CD model while Sections 2.2 and 2.3 provide details on the parameter sensitivity analysis and the construction of the regression based surrogate models. Section 3 presents simulation results and notable observations obtained using this formulation. Finally, Section 4 draws conclusions regarding the quality of such an approach and discusses suggestions for future directions.

2. Methodology

The design and analysis of forward and reverse models for predicting defect accumulation, defect energetics, and irradiation conditions consists of three components: (i) a reasonably descriptive and physically motivated CD model providing a description of key processes governing defect evolution under specific irradiation conditions (dose rate and temperature); (ii) a sensitivity analysis approach in order to investigate the impact of the various defect behaviors on damage accumulation as a function of the irradiation conditions and; (iii) regression models trained against the representative simulations from the sensitivity analysis.

In this section, the physical model used to simulate damage accumulation in bulk α-Fe and the associated statistical techniques are presented.

2.1. Cluster dynamics model

The CD method utilizes the point defect balance equations and the mean field rate theory of defect transport to sinks to predict how the populations of point defects and clusters evolve during exposure to radiation [15]. The spatially dependent balance equation for a point defect cluster, n (i.e., n being the population of either vacancies or self-interstitials), is given as,

$$\frac{dC_n}{dt} = D_n \nabla^2 C_n + g_n + R_n(\vec{C}) - D_n k_n^2 C_n, \tag{1}$$

where D_n is the diffusion coefficient, C_n is the concentration, g_n is the rate of production in damage events, k_n^2 is the sink strength for removing the cluster from the mean field, and $R_n(\vec{C})$ represents the net rate of creation or annihilation of n through interactions with other clusters in the system. Defect clusters from monomer SIAs and vacancies, to dimers and trimers, up to agglomerations of points defects, such as prismatic dislocation loops and cavities large enough to be visible experimentally, are all governed by an equation of this form. The general expansion of the interaction term, $R_n(\vec{C})$, is given as,

$$R_n(\vec{C}) = \sum_{i+j\to n} k_{i,j}^{+} C_i C_j - \sum_{i+n\to m} k_{i,n}^{+} C_i C_n + \sum_{m-i\to n} k_{m,i}^{-} C_m - \sum_{n-i\to j} k_{n,i}^{-} C_n + \sum_{m-n\to i} k_{m,n}^{-} C_m, \tag{2}$$

where the first term describes the formation of n through the combination of two reactant clusters, the second term describes the loss of n through the combination of n with other clusters, the third term describes the formation of n through the dissociation of species i from a parent cluster, the fourth term describes the loss of n by the dissociation of i from it, and the fifth term occurs only if n is a species capable of dissociating from other clusters. The notation $a \pm b \to c$ indicates the set of all possible (mass conservative) reactions (+) or dissociations (−) which might occur between clusters. These CD models can range from fairly simple representations of Frenkel pair damage, similar to standard rate theory, to highly complex descriptions of cascade damage conditions that contain numerous source terms, mobile defect species, and dense reaction networks using large volumes of available atomistic and experimental data informing high-fidelity descriptions of a variety of defect clusters. A broad range of phenomena has been investigated using these methods, including noble gas bubbles [16–18], dislocation loop nucleation and growth [19–21], and precipitation [22–25], with the length scale of application ranging from bulk microstructure modeling [26,27] to spatially resolved descriptions of *in situ* experiments [7,28,29] or nanostructured polycrystals [10] to name a few.