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Computationally-efficient stochastic cluster dynamics method for modeling damage accumulation in irradiated materials

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ABSTRACT

An improved version of a recently developed stochastic cluster dynamics (SCD) method (Marian and Bulatov, 2012) [6] is introduced as an alternative to rate theory (RT) methods for solving coupled ordinary differential equation (ODE) systems for irradiation damage simulations. SCD circumvents by design the curse of dimensionality of the variable space that renders traditional ODE-based RT approaches inefficient when handling complex defect population comprised of multiple (more than two) defect species. Several improvements introduced here enable efficient and accurate simulations of irradiated materials up to realistic (high) damage doses characteristic of next-generation nuclear systems. The first improvement is a procedure for efficiently updating the defect reaction-network and event selection in the context of a dynamically expanding reaction-network. Next is a novel implementation of the τ-leaping method that speeds up SCD simulations by advancing the state of the reaction network in large time increments when appropriate. Lastly, a volume rescaling procedure is introduced to control the computational complexity of the expanding reaction-network through occasional reductions of the defect population while maintaining accurate statistics. The enhanced SCD method is then applied to model defect cluster accumulation in iron thin films subjected to triple ion-beam (Fe^{3+}, He^{+} and H^{+}) irradiations, for which standard RT or spatially-resolved kinetic Monte Carlo simulations are prohibitively expensive.

1. Introduction

The production and accumulation of defects in materials subjected to irradiation is a multiscale problem spanning multiple orders of magnitude in time and space. For the last several decades, the rate theory (RT) method for solving coupled ordinary differential equation (ODE) systems has been the workhorse for irradiation damage simulations [1–3], mostly owing to its much greater computational efficiency compared to more detailed methods such as molecular dynamics (MD) or kinetic Monte Carlo (kMC). RT involves solving a set of coupled ODEs such as:

$$\frac{dC_i}{dt} = \dot{\mathcal{F}}_i - \dot{\mathcal{L}}_i, \qquad (i = 1, \ldots, N) \tag{1}$$

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where each equation describes the time evolution of the average concentration of a particular type (species) of defect cluster denoted by index i. The terms on the right hand side are the loss rate $\dot{\mathcal{L}}_i$ of species i due to various kinetic processes, and the production rate $\dot{\mathcal{F}}_i$ of species i due to irradiation and reactions involving defect cluster species other than i. RT models achieve a high level of simulation efficiency at the cost of drastic simplifications in the underlying physical model, chief of which is the mean-field approximation that neglects spatial correlations and finite volume fluctuations. Another significant reduction in computational complexity is gained by limiting the number of species considered. In practice, the number of admissible defect species (and ODEs in the system) is truncated to achieve a satisfactory balance between accuracy and available computational resources. Large defect clusters not explicitly included in the set are accounted for only approximately (if at all) using a truncation model for the tail of the defect size distribution [4,5].[1] Once defined, the number of ODEs in the set must remain the same through the simulation. To allow simulations to realistically high irradiation doses, this number may need to be as high as 10^6 even in the simplest materials, e.g. pure iron. Furthermore, the number of distinct ODEs that need to be included in the set grows exponentially with increasing number of complex defect cluster types, e.g. simulations of V_mHe_n complexes of m vacancies and n helium atoms requires $(m \times n)$ equations to be included. This is yet another case of combinatorial explosion where the number of equations to be solved is far too large for practical numerical simulations. Consequently, current RT models have been limited to defect populations having no more than two and, in most cases, only one size dimension. This need to allocate an ODE for every possible defect cluster type even before the simulation starts is a serious limitation of the ODE-based RT method.

To overcome these limitations, Marian and Bulatov recently developed the stochastic cluster dynamics (SCD) method to model defect evolution in irradiated materials [6]. The SCD method is based on the stochastic simulation algorithm (SSA) proposed originally by Gillespie for simulations of chemical kinetics in well-stirred systems [7,8]. Whereas RT is formulated in terms of average species concentrations that can take arbitrary fractional values, SSA considers integer-valued species populations in a finite volume and interprets the ODEs defining the RT model as a set of stochastic master equations. The so-defined species population is then evolved stochastically, one reaction at a time, following a standard kMC algorithm. The SSA method has been widely used in the chemical engineering and biochemistry communities [9–13] but is still relatively unknown to computational materials scientists. SCD achieves additional efficiency through the use of dynamic data handling mechanisms where only defect clusters with nonzero populations are kept track of throughout the simulation time. This is a major advantage over RT in which every admissible defect cluster must be allocated a variable and an equation that persist through all stages of ODE integration. Importantly, the computational complexity of a SCD simulation is controlled by the value of the simulation volume and does not depend on the complexity (number of size dimensions) of admissible defect cluster types. Thus, SCD does not suffer from combinatorial explosion and can handle cluster populations with arbitrary number of size attributes. Several proof-of-principle studies have been carried out to demonstrate the applicability of the SCD method to simulations of irradiated materials [6,14].

Although SCD sidesteps combinatorial explosion, the method relies on a kMC algorithm to sample stochastic evolution trajectories from the master equation. Thus, SCD simulations face the usual computational challenges characteristic of kMC simulation methods, such as stiffness caused by a wide spectrum of event rates. Further applications of SCD to technologically relevant materials and irradiation conditions require improvements to make the method more robust and computationally efficient. In this paper, we present several enhancements to SCD, specifically (i) a dynamic reaction-network expansion mechanism to efficiently update the reaction channels and the total reaction rate, (ii) an implementation of the τ-leaping algorithm to accelerate SCD simulations by allowing several reaction events to be leaped over in one single time-step τ, and (iii) a volume scaling method in which the reaction volume is reduced adaptively in order to control the computational cost while preserving statistically significant defect populations. The τ-leaping method [15] was originally developed and used in SSA simulations with fixed variable spaces [13]. In SCD, where the size of the reaction network varies with time, an efficient algorithm for updating noncritical reactions and noncritical species and for computing the leap time is needed to reduce the overhead associated with τ-leaping. We apply the enhanced SCD method to simulations of defect populations in pure iron subjected to triple ion-beam irradiation. The predicted damage accumulation kinetics are verified by comparing them to the original SCD algorithm predictions. The same comparisons are used to quantify gains in computational performance over the original SCD simulations.

The paper is organized as follows. In Section 2, we overview the theory behind the SSA and the τ-leaping methods. In Section 3, we briefly overview our original SCD algorithm, our material model for iron and the types of reaction events considered in our radiation damage simulations. Improvements to the SCD method are described in Section 4 together with their algorithmic details. In Section 5, we present the numerical verification of the new improved SCD algorithm and compare its computational performance to the original algorithm. Finally, Section 6 summarizes our findings.

[1] Existing truncation schemes are ad hoc and unlikely to correctly capture the statistic of extreme values in the defect size distribution believed to be important for understanding material degradation under irradiation.