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On the transfer of cascades from primary damage codes to rate equation cluster dynamics and its relation to experiments

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ABSTRACT

Transferring displacement cascades from primary damage codes to rate equation cluster dynamics (RECD) is not straightforward, due to the inability of RECD to treat spatial correlations explicitly. A method, called "sphere homogenization kinetic Monte Carlo" (SHKMC), has been proposed to produce a effective source term from a cascade database. This paper reviews the method and a few applications. SHKMC is based on a modified kinetic Monte Carlo algorithm to keep track of the homogenization process of defects within cascades. The crucial parameter is the homogenization distance, which is not an intrinsic parameter of cascades but which is given by RECD simulations. SHKMC leads to a time-varying source term, even under constant irradiation flux. RECD with such a source term is able to reproduce reference kinetic Monte Carlo calculations of microstructure evolution under cascade conditions. It is also possible to provide a spatially-dependent source term for the simulation of ion irradiations. As an example, irradiation of iron with 10 MeV Fe ions is discussed. Analysis of the source term shows that the fraction of mono-defects is close to the fraction of freely-migrating defects determined experimentally and that it significantly varies with depth.

1. Introduction

The simulation of nucleation and growth of interstitial and vacancy clusters under irradiation can be performed with various methods, such as kinetic Monte Carlo (KMC) [1–4], phase field [5] and rate equation cluster dynamics (RECD) [6–11]. Among these methods, RECD is in general the most efficient owing to its mean field formalism. Ordinary differential equations on cluster concentrations are solved as a function of time. The combination of efficient time solvers [12] and numerical approximations such as grouping methods [6] or the Fokker-Planck equation [13] make this method unrivalled to reach doses as high as 100 dpa (displacements per atom) in a few minutes of wall-clock time [14,10,15]. For systems with clusters containing three types of elements or more, deterministic solving becomes numerically difficult and stochastic approaches can be used [16,17,11].

Efficiency of the RECD approach is obtained at the cost of substantial efforts to determine the parameters of the model. Among other things, cluster free energies [18–20] and reaction rates between clusters (so-called sink strengths) [21,22] are quantities that can be calculated from atomistic simulations. It has been shown that once it is properly parametrized, RECD gives results in good agreement with reference atomic KMC calculations under thermal aging [18,19,23]. Under irradiation, when isolated defects are produced, RECD results are also consistent with simulations by object kinetic Monte Carlo (OKMC) [24–26] or event kinetic Monte Carlo (EKMC) [27]. Under ion and neutron irradiations, an additional difficulty arises. Defects are produced inside displacement cascades, where their positions are spatially correlated. Spatial correlations cannot be taken into account directly in the formalism of RECD: due to its fundamental mean field hypothesis, it is assumed that defects are homogeneously distributed in the material. Therefore, simulations of cascade damage annealing [28,29] and irradiation [25] lead to marked differences between RECD and O/EKMC calculations (called hereafter KMC for simplicity).

To circumvent this problem, several methods have been proposed. All of them need a cascade database produced by a primary damage code, such as molecular dynamics (MD) or binary collision approximation (BCA) codes. The simplest method, based on the annealing of cascades, is also the oldest one. Originally it was motivated by the determination of the fraction of freely migrating defects (FMDs), *ie* defects which escape the cascade region and can diffuse over long distances to contribute, for example, to void and loop growth [30–32]. Then it was used to justify the concept of "production bias" in theories of void swelling [33,34]. It seems

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that it became popular to provide source terms for RECD only recently [35,36]. It consists in annealing each cascade with KMC (MD cannot simulate such large times), counting the remaining clusters after annealing and averaging the number of clusters of each type over the KMC runs to provide effective quantities of clusters created in cascades. Such quantities serve as effective source terms in RECD. During annealing, defects diffuse in the matrix, so they are more randomly distributed at the end of the simulation. It can be expected that if the annealing is sufficiently long, defects are nearly homogeneously distributed and the transfer to RECD is valid.

However, it is not possible to define a proper annealing time in all cases. It is quite common that cluster diffusion coefficients span several orders of magnitude. As an example, in α-iron, taking vacancy and interstitial migration energies equal to $E_v^m = 0.67$ eV and $E_i^m = 0.34$ eV respectively and the same diffusion prefactor, at 300 K the ratio of interstitial to vacancy diffusion coefficients is around 3.5×10^5. Therefore, in this case, interstitials will homogenize much more rapidly than vacancies. If the annealing time is limited to the point when interstitials can be considered homogeneously distributed, vacancies will have hardly diffused and they will still be spatially correlated. Thus the transfer to RECD is not expected to be completely satisfactory. Incidentally, another difficulty in the annealing method is to estimate when a defect can be considered as homogenized. As a first approximation, it is the case when the defect reaches the boundaries of the KMC simulation box, if the box dimension l is around $l = d^{-1/3}$, where d is the density of cascades [35]. Since this density varies with time, it appears difficult to find the correct value for l.

The second method, which is the topic of this article, is called the "sphere homogenization" KMC (SHKMC) method [37]. It was specifically designed to alleviate the problems inherent to the annealing method. It also relies on the annealing of defects, in the sense that defects are allowed to diffuse in a KMC box. However, a precise diffusion distance is given to estimate when defects are considered as homogenized. Such defects are handled in a specific way, to avoid them to perturb slower defects which are still not homogenized. The choice of the simulation time appears much less critical than in the annealing method; in general, it should be high enough for the slowest defects to homogenize, regardless of the fastest species. Using this method, it was shown that RECD simulations are in very good agreement with KMC simulations for irradiations with 20 keV primary knock-on atoms (PKAs) and 60 keV helium ions in α-iron.

Finally, a third method was recently proposed to incorporate cascade damage in RECD [38,11]. It is somewhat different from the two previously discussed methods, since it relies on a specific version of cluster dynamics, called "spatially resolved stochastic cluster dynamics" (SRSCD). In this kind of model, a system is divided into cells where classical mean field RECD equations are solved. Cells are coupled together by the diffusion of mobile clusters. This method bears some resemblance to spatial 1D-discretization which is performed in RECD to simulate depth-dependent microstructures [39,9,10], but here the discretization is performed along the three spatial dimensions. Due to the large number of equations to solve, a stochastic approach, similar to the one proposed in Ref. [16], is used. It was shown that provided the cell size is similar to the cascade size (around 10 nm for 20 keV PKAs), cascade effects can be simulated by directly injecting the number of defects due to a cascade into the appropriate cells.

In this article we review the SHKMC method presented in Ref. [37]. Simulation techniques are briefly reminded in Section 2. The SHKMC method is described in Section 3 and a few applications in α-iron are shown and discussed in Section 4. In Section 4.1 the validity of the method is assessed for the irradiation with 20 keV PKAs. The sensitivity of RECD results to the number of cascades in the cascade database is discussed in Section 4.2. Finally, SHKMC is used as a standalone technique to provide an estimation of the FMD fraction and to investigate its variation with depth for a heavy ion irradiation (Section 4.3).

2. Simulation techniques

Our goal is to simulate cluster distributions as a function of time under cascade damage conditions. For this purpose we first need a cascade database, produced by a primary damage code. A reference cluster distribution can then be obtained by performing a KMC simulation where cascades are introduced sequentially up to the targeted physical time. RECD calculations depend on a source term, which can be extracted directly from the cascade database (no-annealing case), or after using annealing or SHKMC codes on this database. Annealing and SHKMC were in fact performed with the same KMC code as the one used for the reference calculation, where a dedicated module for damage annealing and homogenization was introduced. In this section the three different codes are briefly described and the parameters are listed. The agreement of RECD with KMC is discussed in the case where isolated defects are produced, to show the consistency of KMC and RECD formalisms.

2.1. Primary damage calculations (iradina)

Cascades were produced with the *iradina* code [40]. *Iradina* uses a Monte Carlo approach to compute the transport of ions in a target and the associated damage. As SRIM [41] and MARLOWE [42] codes, it relies on the BCA approximation, which means that collision cascades are approximated by a sequence of binary collisions. The computed damage is less precise than with MD: among other things, shock waves [43], thermal spike [44] and the resulting intracascade clustering are not simulated, so only mono-interstitials and mono-vacancies are predicted by BCA codes. Point defect clusters are produced only by the diffusion and clustering of mono-interstitials and mono-vacancies. Source terms produced with *iradina* will therefore be different from source terms produced with MD and in order to compare simulation results with experiments, MD should be preferred. However, our goal here is more to emphasize methodological aspects than to discuss physical results. In the following, *iradina* could be replaced with MD without any difficulty.

The choice of *iradina* is explained by several reasons. We want a code which produces a large number of cascades in a limited amount of time, notably to investigate the sensitivity of RECD calculations to the number of cascades in the database (Section 4.2). This cannot be achieved easily with MD, so BCA codes are the natural candidates. SRIM is probably the most popular BCA code in the radiation damage community, but unfortunately the position of interstitials are not provided in output. *Iradina* has been shown to give results in excellent agreement with SRIM in the so-called "Full Cascade" mode, with a much higher efficiency [40]. In addition, as it is open-source, we modified it slightly to provide in output the position of interstitials and vacancies. We also added the possibility to introduce the incoming ion in the middle of the simulation box, in order to simulate the damage created by PKAs in a bulk material (Section 4.1).

In all calculations, the displacement threshold energy was $E_d = 40$ eV, which is the recommended value for iron [45]. Interstitials and vacancies which are closer to each other than $r_{iv} = 3.3a$, where a is the lattice parameter, are assumed to spontaneously recombine [46], so they were removed from the cascade description before the cascade database was transferred to KMC. The