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Relevancy of displacement cascades features to the long term point defect cluster growth

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ABSTRACT

Displacement cascades in iron have been generated by means of the MARLOWE binary collision approximation (BCA) code with primary knock-on atom (PKA) energies ranging from 5 to 100 keV. They serve as input for modelling long term evolution by means of the LAKIMOCA Object Kinetic Monte Carlo code. It is found that the size distributions of the fractions of vacancy and interstitial clustered in the long term are not significantly dependent on the PKA energy in this range. Since the subcascade formation, morphology and spatial extension, as well as the spatial correlations between primary point defect positions do depend on the PKA energy, it is concluded that the size distributions of clustered point defects fractions in the long term do not depend on these cascade features. In contrast, the size distributions of clustered point defect fractions in displacement cascades are found to be independent of the PKA energy while their spatial correlations strongly influence the cluster size distributions in the long term. The use of a mean field approximation in cluster growth kinetics predictions is thereby invalidated. Irradiation dose and dose-rate are also found to be determinant factors governing the long term evolution.

1. Introduction

Neutrons or ions impinging on a material are well-known to produce damage in it by knocking on atoms (primary knock-on atoms, or PKAs) which then lose the acquired energy, to a large extent by displacing other atoms, in a sequence known as a displacement cascade. The cascade development is characterised by an initial ballistic phase, a subsequent thermal spike and finally a relaxation stage. The overall process lasts in the picoseconds range. Cascade processes can be simulated at the atomic level by means of molecular dynamics (MD) [1] or its binary collision approximation (BCA) [2]. At the end of the cascade, a debris is left, formed by point defects and clusters thereof, concentrated in a volume of only a few hundreds nm^3 [3,4] and distributed in a way that reflects the complex mechanisms whereby they were produced. This is the so-called primary damage. The rearrangement of these defects by diffusion and mutual reactions over tens of years of continuous irradiation produces microstructural changes that directly affect the mechanical properties and even the integrity of the material. The problem of modelling all of these processes in a predictive way is thus of primary importance for the safety of existing nuclear power plants and a large effort in this direction, based on a multiscale approach, is being made [5]. In this framework, very different time- and length-scales need to be bridged, by reducing at each step the number of variables involved. For example, atomic scale systems are characterised by a number of degrees of freedom of the order of the Avogadro number (10^{23}), but can be described according to statistical mechanics by only a few macroscopic variables, namely temperature, pressure and concentration (when different chemical species are present). In the present case, the problem addressed is the following: in a displacement cascade regime, the primary damage state is defined by the point defect spatial distributions produced after each cascade. These distributions can be statistically analysed starting from the coordinates of the defects and characterised by functions describing the spatial correlations between vacancies and self-interstitial atoms (SIAs), such as pair correlation functions, cascade volumes, elongations, etc. The primary damage has been thereby shown to be different from a purely random point defect distribution [4,6], consistently with the fact that it is the consequence of physical processes governed by given interatomic interactions and by the crystal lattice. The evolution of the initial defect distributions, according to the migration properties of point defects and their clusters and to the possible mutual reactions, leads eventually to the

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formation of a microstructure, characterised by another distribution of point defects, describable in terms of a cluster size distribution. This final distribution is regarded here as the transformation of the initial one via a 'transfer function', determined by the defect diffusion properties and mutual reactions included in the model. The objective is to identify the reduced number of parameters, defining the primary damage state produced by a cascade, that mainly determine the cluster formation and growth in the long term. An extensive comparison between different primary damage databases and the different microstructure evolutions corresponding to each of them, modelled by applying an object kinetic Monte Carlo (OKMC) method [7], revealed that dramatic differences in the initial primary damage state, particularly in terms of defect clustered fractions, are largely smoothed away in the final microstructure [8–10]. The features of the 'transfer function' appear therefore to be more important in determining the final state than the initial distributions themselves. Nonetheless, if totally random initial point defect distributions are used as input, the final microstructure is found to be significantly different [8], thereby suggesting that some of the features of the defect distributions after a cascade must be of relevance in determining the microstructure evolution [10]. Here we present an attempt at distinguishing some of these features from others. Note that the scope of this work is limited to exploring the effect of the spatial inhomogeneities intrinsic to cascade damage production on the microstructure evolution. A detailed account of possible spectral effects, related to specifics modes of irradiation (electrons, protons, neutrons of different origin, ions, ...) is beyond the scope of the present work.

Section II provides a brief summary of the simulation models and of the distributions employed to characterise the primary damage state. The relationship between cascade debris features and size distributions of clustered point defects in the long term is described and discussed in Section III. Section IV considers the variations introduced in the results by dose and dose-rate changes. A discussion is given in Section V and the main conclusions are drawn in Section IV.

2. The models

In this work a set of displacement cascade debris in iron produced using the BCA was employed for the study of the corresponding microstructural evolution. BCA cascades and their evolution on the long term have been compared with the same features assessed from a fairly large existing MD cascade database [5,11]. Also in the present work, when necessary MD cascades produced by Stoller et al. [10,11] using the code Moldy [12] and the Finnis–Sinclair many-body potential for iron [13], were used. The primary damage has been characterized by treating separately the SIA and vacancy populations, using component analysis [14] coupled to a fuzzy partition technique [15] for the identification of subcascades. The latter method allows overlapping subcascades to be recognised, by finding the partition of each displacement cascade debris configuration which minimizes the overlap, without assuming a priori any subcascade prototype or number (for technical reasons, though, the algorithm cannot partition one cascade into more than 16 subcascades). Component analysis allows an ellipsoid to be associated with each subcascade and therefore to compute the corresponding volume and an aspect ratio. The latter is defined as $\alpha/\gamma-1$, where α and γ are, respectively, the major and the minor components of the correlation matrix between point defect positions, i.e. the longest and shortest semiaxes of the associated ellipsoid; it thus measures the offset from sphericity. The reader is referred to Refs. [4,14,15] for a more detailed description of the method.

A further characterisation of the cascade debris was provided by the pair distribution function which, in its integral form, may be written as

$$G(r) = \frac{1}{N(N-1)} \sum_{i=1}^{N} \sum_{j>i}^{N} \delta(r - r_{ij}) \quad \text{where} \quad G(r) = \frac{4\pi r^2}{\Omega} g(r) \tag{1}$$

$g(r)$ being the standard pair correlation function where angular correlations are disregarded and Ω being the volume. In a structureless infinite homogeneous medium, $G(r)$ increases with r^2 while $g(r)$ is unity. $G(r)$ is normalized in such a way that

$$IG(R) = \int_0^R G(r)\mathrm{d}r \tag{2}$$

is unity when $R \geq r_{ij}^{\max}$, the largest separation distance between point defects in a cascade. The integral (2) will be used here to analyze pair separation distances between point defects in cascades over all distances within the simulation box.

The shortcoming of the BCA is that only the ballistic phase is properly modelled and further approximations are needed to allow for the effect of the thermal spike and the relaxation phase on the final defect distribution. The great advantage, however, is that it is several orders of magnitude faster than full MD and allows a much more statistically significant sampling of cascade debris to be accumulated, within limited computer time. Series of 500 cascades for each PKA energy, in the range from 5 to 100 keV, could thus be produced using the code Marlowe [16]. A PKA is introduced by providing a randomly chosen atom, located at a lattice site, with a momentum in a direction also selected at random, corresponding to a preset, well-defined initial kinetic energy. The ballistic phase of the cascade is then traced as a time ordered sequence of binary collisions of a moving projectile atom with a target atom assumed at rest, interacting by means of a repulsive pair potential used to compute the scattering integrals. We chose for this the so-called universal potential [17], also popular in full MD codes for modelling the interaction between 'close' encounters. The annealing phase of the cascade is then modelled by assuming a recombination radius between created SIA and neighbouring vacancies. This recombination radius was adjusted so as to predict the same number of Frenkel pairs (FP) as full MD [4]. The validity and limits of this method are extensively discussed in the literature [4,8–10,18–24].

Each series of cascades was then used as input to model the microstructure evolution, whose dependence on PKA energy could thereby be evaluated. The microstructure evolution has been simulated using the LAKIMOCA code. The method and the code are described in detail elsewhere [7]. The OKMC is compared to the rate equation approach in [25]. Briefly, in the OKMC, defects and defect clusters are treated as objects with specific positions in a simulation volume. The probabilities for thermally activated transition mechanisms are calculated based on Arrhenius frequencies. After a certain event is chosen to occur, time is increased according to a residence time algorithm [26]. The basic aspects of the parameterization used are described in [7]. Three parameter sets were used, which only differ by the SIAs and SIA clusters mobility models. In set I, all SIA clusters (size $m \geqslant 2$) migrate in 1D, with a migration energy $E_m = 0.04$ eV and a prefactor decreasing with the size m according to the law $\nu_0 m^{-s}$ ($\nu_0 = 6 \times 10^{12}\ \mathrm{s}^{-1}$, $s = 0.51$, following Ref. [27]). In set II, small clusters ($m \leqslant 5$) migrate in 3D with $E_m = 0.4$ eV, as broadly suggested by recent *ab initio* calculations [28], while larger clusters maintain 1D motion with $E_m = 0.04$ eV. For large clusters the prefactor decreases with $s = 0.51$ and for small ones it decreases with $s = 10$. Finally, set III treats small clusters ($m \leqslant 5$) in the same way as set II, but assumes that larger clusters are completely immobile (see Table 1). For