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Modeling microstructure evolution of f.c.c. metals under irradiation in the presence of He

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Abstract

We use kinetic Monte Carlo simulations with input parameters obtained from molecular dynamics calculations to study microstructure evolution in irradiated f.c.c. metals in the presence of He. This model is able to reproduce the characteristic swelling curve of a f.c.c. metal and the dependence of this curve with dose rate. We provide an atomistic description of the evolution of defects produced by irradiation and particularly of void nucleation. Stable, nanometer size He–vacancy complexes, created at the early stages of damage accumulation can grow under irradiation forming voids. These voids are responsible for macroscopic changes in the material, such as volume changes (void swelling). These simulations show how the initial clustering of vacancies in the cascade core influences the evolution of the microstructure and the nucleation of bubbles and voids.

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1. Introduction

Irradiation of metals can alter significantly their mechanical properties. In the presence of gases, such as those appearing through transmutation in fission or fusion irradiation conditions or through alpha-decay in radioactive materials, an enhanced production of bubbles and voids may occur. This can cause significant dimensional changes in the material [1]. Despite the many years of research regarding the formation of bubbles and voids under irradiation there are still many unknowns, specially regarding the initial stages of nucleation of such defects.

In order to have a volume change it is necessary that interstitials produced by the irradiation diffuse to sinks such as dislocations or grain boundaries and eventually to surfaces leaving vacancies in the bulk that nucleate and grow three-dimensional structures or voids. This different behavior between vacancies and interstitials has been traditionally explained by the preferential trapping of interstitials at dislocations due to their stronger elastic interaction with the dislocation stress field, the so-called dislocation bias model [2]. This model although very successful in explaining swelling rates at high doses, is not able to explain the behavior of f.c.c. metals at very low irradiation doses [3]. In light of the new observations coming from molecular dynamics simulations, a new theory called production bias emerged recently to explain swelling in f.c.c. metals [4]. Molecular dynamics simulations have shown that in f.c.c. metals, the damage resulting from high energy recoils consist of vacancies and interstitials that are not isolated, but form clusters [5–7]. The same behavior has been observed in Cu [5–7], Au [8] and Pb [9], and at a lower extent in Ni [10]. This clustering occurs within the first few picoseconds of the cascade collapse. Moreover, self-interstitial clusters can diffuse very rapidly in a one-dimensional motion along the $\langle 1\,1\,0\rangle$ direction, according to exhaustive molecular

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dynamics simulations [11]. The production bias model states that the asymmetry created by the different clustering of self-interstitial atoms and vacancies is responsible for the microstructure evolution at low doses and the dose and temperature dependence of swelling [12].

We have used a kinetic Monte Carlo (kMC) model to simulate the production and evolution of defects in f.c.c. metals under irradiation, with input parameters obtained from molecular dynamics simulations. The advantage of kMC is that it treats space correlations between defects. The dislocation bias can be included in the model through the capture radius between defects. The production bias is inherent to the model since input from molecular dynamics simulations regarding damage due to irradiation is included in these simulations, that is, the initial clustering between vacancies and between self-interstitials in the cascade is obtained directly from the molecular dynamics calculations. One-dimensional diffusion of self-interstitial clusters is also included in the model. In this paper we describe the general input used in this kMC simulations and the results obtained for f.c.c. materials for irradiation at high concentration of He and very low dose rates.

2. Model for He-defect interactions in kMC

We use a kMC model with input data obtained molecular dynamics simulations. The methodology of the kMC used in these calculations has been explained elsewhere [13] and will not be detailed here. The source term or defects produced by the irradiation is introduced in the kMC model from a database obtained with molecular dynamics simulations. Molecular dynamics is a powerful technique to describe multiple interactions, such as those that take place during the deposition of energy in the lattice by an energetic recoil, and it has been used extensively during the last two decades [5–7]. In the particular set of simulations presented here a database for cascades in Au was used as input data for damage in an f.c.c. material [8]. Single vacancies and divacancies are considered mobile as well as all self-interstitial clusters with sizes smaller than 40. As we showed earlier [13], using a similar model for copper with input data from molecular dynamics simulations we can compare the results from kMC to experimental measurements.

In order to introduce the effect of an impurity such as helium in the defect evolution, the energetics of reaction of He atoms with defects must be known. Adams and Wolfer [14] have used molecular dynamics simulations to obtain the binding energy of He–vacancy (He–V) complexes in nickel for clusters with up to 20 vacancies and 10 helium atoms. We have used the results of these calculations for the dissociation energies of He–V complexes in the kMC model extrapolating for large numbers of He in a cluster as well as large number of vacancies. We consider in the calculations that the migration of the He_1V_1 (1 helium atom and 1 vacancy) complex is the same as the migration of a single vacancy. All other HeV complexes are assumed to be immobile.

All reactions are considered to be diffusion limited, that is, there is no barrier for reaction between two defects; as soon as two defects are within their capture radius the reaction will occur. The interaction radius between defects is: $r = r_{\mathrm{sph}} + \delta$, where δ is the jump distance and

$$r_{\mathrm{sph}} = \sqrt[3]{\frac{3N\Omega}{4\pi}} \tag{1}$$

with Ω the atomic value and N the number of defects in the cluster. Since the stress field of self-interstitials is larger than the one of single-vacancies a larger capture radius is considered for interstitials (r_{I}) interacting with loops (vacancy or interstitial clusters) that for the case of vacancies, r_{v}: $r_{\mathrm{I}} = 1.15 \times r_{\mathrm{v}}$, where r_{v} has been defined above. This therefore includes a bias for the interaction of interstitials with the other defects in the material [13]. We include a sink to both vacancies and interstitials within the simulation box. The capture radius is also biased with a value of $r_{\mathrm{I}}^{\mathrm{s}} = 1.4 \times r_{\mathrm{v}}^{\mathrm{s}}$, where $r_{\mathrm{I}}^{\mathrm{s}}$ is the capture radius of a self-interstitial to the sink and $r_{\mathrm{v}}^{\mathrm{s}}$ is the capture radius of a vacancy to the sink. This sink mimics the presence of a dislocation, and corresponds to a dislocation density of 2×10^{10} dislocations/cm^2.

With the introduction of a new element, helium, in the calculations we need to define the different reactions that can take place. Fig. 1 shows a diagram of the different reactions defined in the kMC model between He, vacancies (V) and self-interstitials (I). As we mentioned above, damage produced by a cascade in f.c.c. metals can result in clusters of vacancies, which can be loops or stacking fault tetrahedral (SFT), depending on the metal. Foreman and Singh [15] have shown that the formation of a void from the interaction of He atoms with the cascade damage is very unlikely. Therefore we will

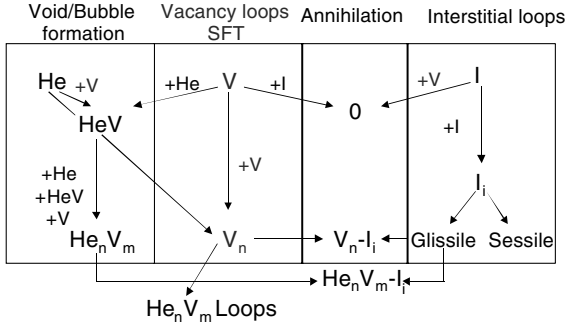

Fig. 1. Diagram of reactions between helium atoms, vacancies (V) and self-interstitials (I) used in the kMC model.

consider in this model that the interaction of the He atoms with a vacancy cluster will not contribute to the population of voids or bubbles. The nucleation of a void or a bubble will start with the interaction between He atoms and single vacancies. We distinguish between the loops or SFTs with He atoms and the void/bubbles, as shown in the diagram of Fig. 1. One of the drawbacks of kMC models that include such level of detail as the one we present here is that the highest doses reached are small (below 1 dpa) due to the computational demands of this calculation.

3. Results

The results presented here are in terms of displacement per atom (dpa) for the dose, where dpa is the standard unit of damage as defined in the ASTM standards [16]. All the values for temperatures are given in terms of the melting point, in order to generalize the results, since the aim of this paper is to obtain general trends in microstructure evolution.

For these particular simulations the helium concentration is of one helium atom per cascade, or a total of 1000 appm of He per dpa, which corresponds to conditions close to those of helium implantation. Fission and fusion irradiation conditions correspond to values of 1 and 10 appm/dpa respectively, lower than those considered in these simulations. We have studied the evolution of vacancy clusters, SIA clusters and bubbles/voids as a function of dose for different irradiation temperatures and dose rates. Fig. 2 shows the concentration of clusters of vacancies, Fig. 2(a), of interstitials, Fig. 2(b) and of bubbles/voids, Fig. 2(c), as a function of dose for five different temperatures and for a dose rate of 10^{-8} dpa/s. As can be observed in the figure, the total number of vacancy clusters decreases with increasing temperature. At a temperature of $0.24T_m$ small vacancy clusters start to dissociate and when the temperature reaches $0.37T_m$ none of the vacancy clusters survive. The concentration of interstitial clusters does also decrease for increasing temperatures, in this case due to recombination with mobile vacancies and fast migration to sinks. It is important to notice that the instantaneous concentration of vacancy clusters is much larger than that of SIA clusters because of the fast migration and biased diffusion of the latter. The evolution of HeV clusters is shown in Fig. 2(c). He atoms stabilize small vacancy clusters, therefore the decrease in HeV cluster concentration occurs at higher temperature than for the case of vacancy clusters, at $0.26T_m$. For the lowest temperatures studied, $0.21T_m$ the vast majority of the clusters contain one He atom, therefore, the ratio of He to vacancy content (He/V) is small. As the temperature increases the He/V ratio increases; for $0.29T_m$ clusters with as many as 5 He atoms are found. As an example,

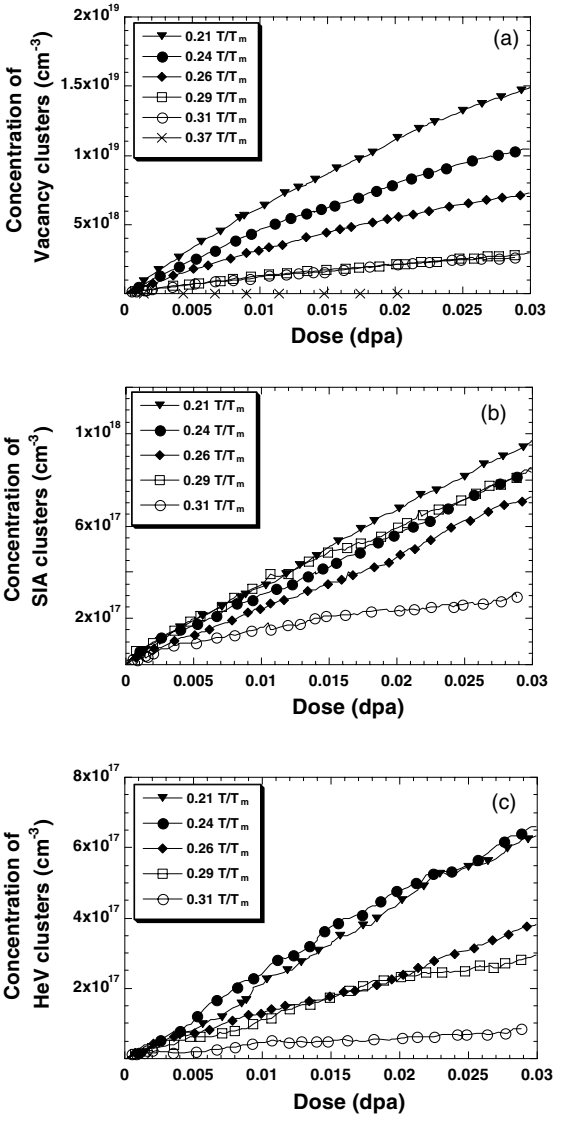

Fig. 2. Cluster concentration for (a) vacancies, (b) interstitials and (c) bubbles/voids as a function of dose for 5 different temperatures and a dose rate of 10^{-8} dpa/s.

Fig. 3 shows the concentration of HeV clusters as a function of number of vacancies in the clusters as well as number of helium atoms for a total dose of 0.03 dpa and a temperature of $0.29T_m$. Those HeV clusters with high He content could be considered as bubble embryos and are stable at this temperature. At higher temperatures, $0.31T_m$ small HeV complexes are unstable and nucleation of bubbles or voids becomes difficult, reducing the number of clusters of this type with dose.

The effect of dose rate on HeV complex nucleation and early growth can be seen in Figs. 4(a) and (b). Here we present the results for the same temperature and

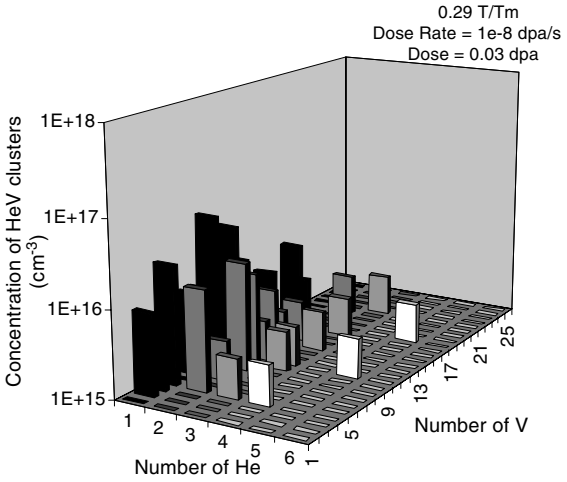

Fig. 3. Concentration of HeV clusters as a function of cluster size (number of vacancies in cluster) and helium concentration for 0.03 dpa, $T = 0.29T_m$ and dose rate of 10^{-8} dpa/s.

dose, $0.24T_m$ and 0.012 dpa respectively, and two different dose rates, 10^{-6} dpa/s in Fig. 4(a) and 10^{-8} dpa/s in Fig. 4(b). Note that 10^{-6} dpa/s is a dose rate typical of neutron irradiation in a fast reactor environment. Clearly, a much larger number of HeV complexes are present for the lowest dose rate. More significant is the fact that the distribution shifts towards larger, more stable clusters with larger He/V ratios that exceed the critical size for growth. At the higher dose rate the distribution consists almost exclusively of clusters with one He atom and multiple vacancies, but at lower dose rates it shifts towards more He atoms per vacancy. The results clearly show that at the lower dose rates HeV complexes nucleate and start to grow very efficiently, and it seems to indicate that the onset of void growth is at extremely low doses for low dose rates. We must point out the absence of impurities in these calculations, which might alter the evolution of the void nucleation and growth.

Due to the atomistic nature of these simulations we are able to obtain not only the concentration of clusters as a function of dose but also the total number of vacancies in each type of vacancy complex (loops, bubbles or voids). One important parameter that we can extract from these simulations is the total change in volume of the material due to void formation, $\Delta V/V$ for a particular dose ϕ as

$$\Delta V/V(\phi) = \sum_N C_v^{\mathrm{void}(N)}(\phi) \times \Omega_r(N),$$

where $C_v^{\mathrm{void}(N)}$ is the concentration of vacancies on voids or bubbles of size N and $\Omega_r(N)$ is the relaxation volume for a vacancy in a cluster of size N. In this calculation the relaxation volume per vacancy that we have considered is of 0.2 Ω, which is the limiting value for voids

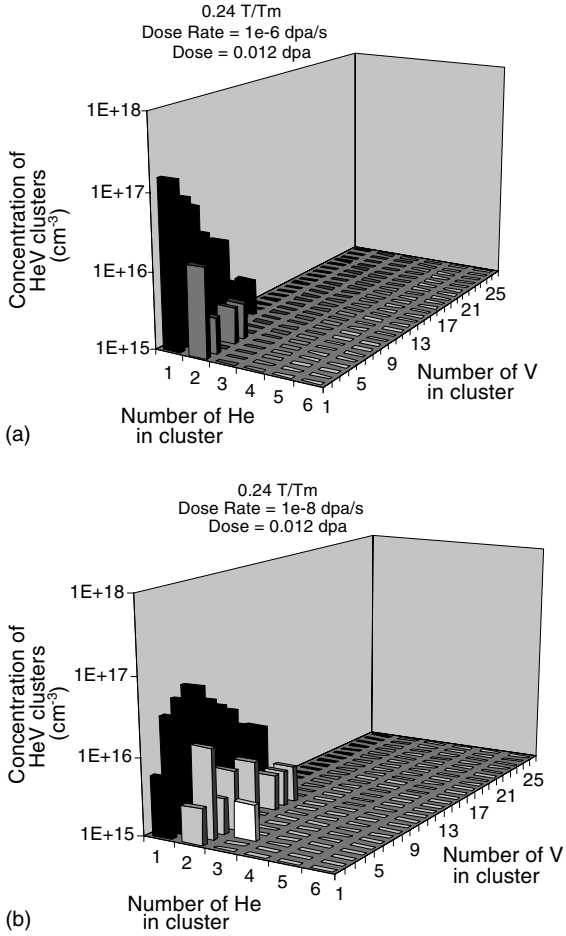

Fig. 4. Concentration of HeV clusters as a function of cluster size (number of vacancies in cluster) and helium concentration for $T = 0.24T_m$ and 0.012 dpa and two different dose rates (a) 10^{-6} dpa/s and (b) 10^{-8} dpa/s.

with more than 9 vacancies obtained by Shimomura [17] using molecular dynamics simulations. Using this approach the void swelling for the case of a dose rate of 10^{-8} dpa/s and different temperatures is presented in Fig. 5. Notice that the void swelling increases with temperature between 0.21 and $0.24T_m$ but there is a sharp decrease for 0.26 and higher temperatures and there is no swelling at temperatures of $0.37T_m$. Fig. 6 shows the temperature dependence of void swelling for a fixed dose value, 0.03 dpa, and for three different dose rates. The model is able to reproduce the characteristic swelling curve of FCC metals [18,19], with a swelling peak that in this case is centered around $0.24T_m$ for a dose rate of 10^{-8} dpa/s. In the case of copper, for example, under fission neutron irradiation, with a damage rate of 2×10^{-7} dpa/s and a gas concentration of ~0.3 appm He and ~15 appm H, the experimentally measured swelling peak is around $0.44T_m$ [18] at approximately 1 dpa. We

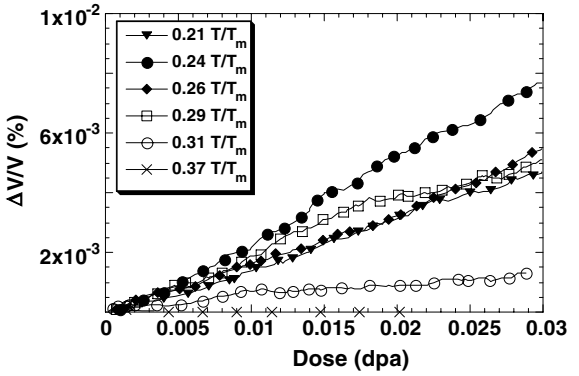

Fig. 5. Void swelling as a function of dose for a dose rate of 10^{-8} dpa/s and different temperatures as a function of dose.

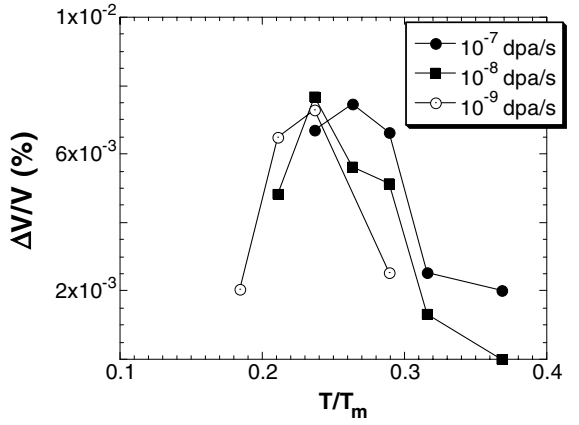

Fig. 6. Temperature dependence of void swelling for a total dose of 0.03 dpa and three different dose rates.

should point out that the higher helium concentration in the simulations presented here shifts the swelling peak towards lower temperatures [18] therefore we can not directly compare the results of these simulations to the experimental measurements.

The reason for this behavior is in the stability of vacancy clusters, HeV complexes and the bias diffusion of interstitials to sinks but also in the initial conditions of the damage, or source term. Most of the vacancies are in clusters within the first picoseconds of the cascade evolution. As a result at low temperatures these clusters will be stable and the concentration of single vacancies will be low. As we explained above the interaction of He atoms with vacancy clusters forming loops or SFT will not be able to induce a transformation of these clusters into three-dimensional voids. Therefore, the only mechanism of void formation is through nucleation by addition of single vacancies to He atoms. When the concentration of single vacancies is low, like at low temperatures, the concentration of nuclei for voids will also be low, as well as the number of vacancies arriving to these nuclei. At higher temperatures, when small vacancy clusters can dissolve the supersaturation of vacancies increases. These vacancies are then able to reach both sinks and HeV nuclei and therefore increase the number of vacancies in bubbles and voids. Only when the temperature is so high that small HeV nuclei are not stable the amount of vacancies contributing to voids and bubbles will decrease. These three regimes explain the characteristic swelling curve observed in f.c.c. metals, with a maximum swelling peak at a particular temperature.

The swelling peak depends strongly on the dose rate, as shown also in Fig. 6. For low dose rates the maximum of the swelling peak shifts towards lower temperatures. This is also observed experimentally for the case of f.c.c. metals [20]. Lower dose rate corresponds to longer times between cascades. For the same temperature, at a lower dose rate the damage produced by the cascade has longer time to evolve. At low dose rates nuclei for void formation will have time to dissolve before more vacancies arrive from the next cascade than at higher dose rates. Therefore, at the same temperature, the nuclei for void formation will be smaller for higher dose rates than for lower dose rates, since the nuclei will have to be stable for a shorter time, in order to achieve the critical size for void and bubble growth. As a consequence, the maximum temperature for void swelling will decrease towards lower temperatures as the dose rate decreases. For those temperatures where none of the nuclei dissolve as the time between cascades increases there is also more time for vacancies reaching HeV nuclei and the number of vacancies in bubbles or voids will be larger.

4. Conclusions

In summary, we have implemented a kinetic Monte Carlo model with input data from molecular dynamics simulations that account for the microstructure evolution of f.c.c. metals under irradiation in the presence of helium. This model is able to reproduce the characteristic swelling curves of f.c.c. metals as a function of temperature and dose rate. The temperature at which the maximum swelling occurs shifts towards lower values as the dose rate decreases, in agreement with experimental observations [20]. Due to its atomistic nature this model can be used to understand void nucleation as well as void growth. One of the most important input parameters in this simulation is the initial configuration of the damage after irradiation, or the source term. The fact that vacancies are forming clusters already in the first few picoseconds of the collision cascade is a critical factor in the later evolution of defects, and the dose and temperature dependence of void swelling.

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