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Displacement cascades and defect annealing in tungsten, Part III: The sensitivity of cascade annealing in tungsten to the values of kinetic parameters

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ABSTRACT

A study has been performed using object kinetic Monte Carlo (OKMC) simulations to investigate various aspects of cascade aging in bulk tungsten (W) and to determine its sensitivity to the kinetic parameters. The primary focus is on how the kinetic parameters affect the intracascade recombination of defects. Results indicate that, due to the disparate mobilities of SIA and vacancy clusters, annealing is dominated by SIA migration even at 2050 K. It was found that for 100 keV cascades initiated at 300 K, recombination is dominated by the annihilation of large defect clusters, while for all the other primary knock-on atom (PKA) energies and temperatures, recombination is primarily due to the migration and rotation of small SIA clusters, while the large SIA clusters escape the simulation cell. The annealing efficiency exhibits an inverse U-shaped curve behavior with increasing temperature, especially at large PKA energies, caused by the asymmetry in SIA and vacancy clustering assisted by the large differences in their mobilities. This behavior is unaffected by the dimensionality of SIA migration, and it persists over a broad range of relative mobilities of SIAs and vacancies.

1. Introduction

Defect production under neutron irradiation is primarily by the formation of displacement cascades, in which both mobile and immobile defects are produced locally in excess of the thermal equilibrium concentration. Long range migration of these defects and their interactions with other types of defects results in major microstructural changes [1]. Individual displacement cascades develop within a very short time-scale, and they are nearly impossible to analyze experimentally. Molecular dynamics (MD) is the most widely used simulation tool to study atomic-level details of displacement cascades, and it has been used extensively for this purpose [2–10]. Due to the requirement of femtosecond time steps, the simulation of the longer-time evolution of displacement damage is beyond the scope of MD. However, it can be studied using kinetic Monte Carlo (KMC) methods. Various groups have studied single cascade aging using the KMC method in Fe [11–15], Ni [16], Cu [17–20], V [21], SiC [22] and W [23], but there has not been a detailed study of single cascade aging to understand how the relative mobility of defects, which depends on defect migration barrier and cluster size, and temperature, affects the intracascade recombination of defects.

In a recent study on single cascade aging in W as a function of PKA energy and temperature using the KMC code KSOME (Kinetic Simulations Of Microstructure Evolution) [23] it was found that annealing is dominated by very fast self-interstitial atom (SIA) diffusion, and nearly all of the recombination and coalescence events occur in the very first few nanoseconds of the simulation. In addition, especially at higher PKA energies, a smaller fraction of defects are lost to intracascade recombination at 2050 K compared to that at 1025 K. This results in an annealing efficiency (fraction of defects lost to intracascade recombination) exhibiting an inverse U-shape curve as a function of temperature. It increases from 300 to 1025 K due to increased SIA rotation, but decreases from 1025 to 2050 K due to a decrease in the number of mobile SIA defect clusters and also the fraction of SIA clusters that can rotate due to the increased SIA clustering at 2050 K compared to 1025 K. Indeed, the escape rate of SIAs from the simulation box depends more sensitively on their cluster size distribution than on the temperature. In general, depending on the PKA energy and the temperature at

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which the cascades were initiated, 85–95% of SIAs will eventually escape and contribute to long-range defect migration.

The quantitative reliability of a model depends on the accuracy with which the physical mechanisms, such as point defect migration and their interactions with other defects, are understood at the atomic level, as well as how the parameters governing them are incorporated into the model. OKMC models require a large number of parameters *a priori*. This set of parameters is never complete, and there is always uncertainty in their quantitative value depending on how they are obtained. Moreover, many quantities cannot be measured experimentally and must be computed or estimated from other models. Therefore it is important to understand the variability of results as a function of these parameters. Such code parameterization problems for OKMC models have been well described by Becquart et al. in Ref. [24]. The present study is intended to elucidate the variability of observed cascade annealing behavior in W as a function of kinetic parameters, and to provide a comprehensive picture of the initial phase of long-term defect evolution.

2. Simulation details

This study was carried out using a database of cascades obtained from MD simulations [25] with a wide range of PKA energies at 300, 1025 and 2050 K. The values of migration energies and diffusion prefactors, capture radii and binding energies of defects used in the annealing simulations are taken from the *ab initio* calculations of Becquart et al. [26] and references therein. For brevity, this set of kinetic parameters is referred to as the "standard kinetic parameters" (see Table 1). A detailed discussion of the simulation methodology used in this study can be found in Ref. [23].

The annealing of a single cascade in any material depends on: (1) cascade morphology, which depends on the PKA energy, the lattice temperature, and the direction at which the cascade is initiated, (2) migration energies and relative mobilities of various types of defect clusters, which depend on temperature and cluster size, (3) dimensionality of SIA cluster diffusion (whether 1D or 3D or mixed 1D/3D motion), which depends on the temperature and SIA cluster size, and (4) the interactions between various types of defects, which depend on their type and size. In the present case the types of defect clusters are SIA and vacancy.

In order to understand the effects of the above factors on annealing, tests were carried out under the following conditions:

1. Only mono-SIAs are allowed to change the direction of their 1D motion,
2. All SIAs perform 3D diffusion (limiting case),
3. The relative mobilities of SIAs and vacancies are varied by
   a. Reducing the migration rate of SIA clusters by 100 times (by reducing its prefactor),
   b. Modifying the vacancy and SIA migration energy barriers to 1.3 eV and 0.05 eV, respectively
   c. Modifying the decay of SIA migration rate with SIA cluster size,
4. Cascades generated at 300, 1025, and 2050 K are each annealed at temperatures other than the temperature at which they are initiated. They were annealed at 300, 600, 900, 1200, 1500, 1800, and 2100 K.

Table 1
Standard kinetic parameters. $\nu_0 = 6 \times 10^{12}$ s^{-1}, $q = 1000$, $s = 0.5$ and n = defect cluster size.

Defect	Attempt frequency (s^{-1})	Migration energy (eV)	Dimensionality of migration
SIA	ν_0	0.013	1D along ⟨1 1 1⟩ E^{rot} = 0.38 eV
SIA cluster	$\nu_0\, n^{-S}$	0.013	1D
Vacancy	ν_0	1.66	3D
Vacancy cluster	$\nu_0\,(q^{-1})^{n-1}$	1.66	3D

The relationships between various factors affecting cascade annealing are non-linear and non-intuitive; therefore, it is expected that the variations in the above tests will reveal some important aspects of cascade annealing. Tests 1 and 2 are done to study the effect of dimensionality of SIA migration, which depends on cascade annealing temperature and SIA cluster size. Tests 1 and 2 also represent the two limiting cases of dimensionality of SIA migration corresponding to minimum and maximum recombination, respectively. Test 3 is done to examine the effect of relative mobilities of SIAs and vacancies, especially on intracascade recombination. Test 4 is done to determine the combined effect of the cluster size distribution and the relative mobilities of SIA and vacancy clusters on recombination. In this study no attempt has been made to examine the effect of capture radii on cascade annealing. Also, although the spatial distributions of defects in cascades are not explored explicitly, a key aspect of it is the spatial separation of SIAs at the periphery and vacancies at the cascade core.

3. Results and discussion

3.1. Effect of dimensionality of SIA migration

It is well accepted from MD simulations that SIA clusters produced in a cascade can develop into perfect dislocation loops and perform 1D migration with high mobility [12,27–35]. 1D diffusion of SIA loops in Fe and Fe-alloys has also been demonstrated experimentally under electron irradiation [36–40]. However, the presence of impurities can significantly influence SIA loop mobility [37–40]. MD simulations [32,41] of SIA clusters diffusing among oversized Cu impurities in an α-Fe matrix have shown that the diffusion prefactor decreased and the frequency of rotation from ⟨1 1 0⟩ to ⟨1 1 1⟩ dumbbell configuration increased by decreasing the energy difference [42] between those two configurations. Also the 1D confinement of SIA clusters through repulsive interaction with an oversized solute atom and SIA clusters was proposed [43] where escape of from these confined segments occurs only by changing their directions of motion. Depending on the confinement distance, SIA diffusion can effectively transition from 1D to 3D diffusion.

To illustrate the effect of dimensionality of SIA diffusion on cascade annealing, two limiting cases were considered: (1) each cluster diffuses in its own randomly chosen 1D direction and (2) clusters of all sizes diffuse in 3D. The "standard kinetic parameters" are defined such that SIA clusters larger than size five are constrained to 1D diffusion. A change in the dimensionality of SIA diffusion due to the presence of impurities represents an intermediate type of the two limiting cases above.

3.1.1. Effect of 1D migration of SIA clusters

Fig. 1 shows the effect of PKA energy on the fraction of surviving SIAs at 300, 1025 and 2050 K when all SIA clusters except mono-SIAs diffuse in 1D (bottom panels) compared to the standard parameters (top panels). All plots exhibit a general evolutionary behavior in which there is a rapid drop in the surviving fraction of SIAs at the beginning (Stage 1), and later on the surviving fraction decreases with an almost constant slope (Stage 2). Stage 1 is associated with intracascade recombination, while Stage 2 is due to SIAs escaping the simulation cell. At 300 K as seen in Fig. 1(a) and (d) there is no difference in either the fraction of SIAs lost to