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Computer simulations of the effects of temperature change on defect accumulation in copper during neutron irradiation

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Abstract

Kinetic Monte Carlo (kMC) computer simulations were used to study the effects of temperature change during low-dose 14-MeV fusion neutron irradiations on defect accumulation in copper. Instantaneous temperature changes from 300 to 473 K, from 300 to 673 K and from 373 to 573 K were simulated. All the high temperatures are above the Stage-V annealing temperature for copper, approximately 400 K. The total simulated dose was 0.01 dpa for all temperature-change simulations, at a dose rate of 10^{-6} dpa/s. In general, the concentrations of both self-interstitial atom (SIA) clusters and vacancy clusters decrease when the temperature is changed from low to high. The relative decrease in concentration of defect clusters when the temperature is increased is determined by the level of accumulation of vacancy clusters in the initial low-temperature part of the irradiation as well as by the specific temperatures involved. If the vacancy clusters are small and their concentration is high, the decrease of cluster density is prominent when the temperature increases, and the higher the second temperature, the easier it is for defect clusters to dissolve.

1. Introduction

The temperature of the first wall and divertor of fusion devices is expected to cycle due to the cyclic nature of the plasma burn. The effects of temperature change at low-dose irradiation have been studied by ion irradiation and by neutron irradiation with the Japan materials testing reactor (JMTR) [1–4]. Some temperature-change irradiation experiments at a relatively high dose (~5 dpa) are in progress in the high flux isotope reactor (HFIR) on a number of metals and alloys [5]. The experimental results show that the concentration of interstitial-type dislocation loops decreases when the temperature is changed from 473 or 573 K to 673 or 773 K [1,3,4].

The analyses of Fe–Cr–Ni alloy experiments based on standard rate theory indicate that a vacancy-rich state appears after the irradiation temperature is increased from 473 to 673 K, as a result of the dissolution of small vacancy clusters that were initially formed at the lower temperature. These mobile vacancies react with the newly formed interstitials produced at the high temperature [6]. However, in the rate theory analyses, the cascade effects on the formation of defect clusters are not considered because the rate theory is based on the mean field approximation, and it cannot accurately represent the effects of the discrete, stochastically produced damage regions containing point defects and defect clusters produced by individual cascades. Both SIA and vacancy clusters are directly formed in cascades at both temperatures, and these clusters will influence the microstructure evolution during the two-temperature irradiation.

Kinetic Monte Carlo (kMC) computer simulations of defect accumulation under irradiation are used in the present work to simulate the two-temperature irradiation experiments. Defect configurations from molecular dynamics (MD) simulations of cascades were used to provide realistic defect cluster input to the kMC simulations.

In the present study, single temperature changes from 300 to 473 K, from 373 to 573 K and from 300 to 673 K

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Table 1
Migration energies and dissociation energies for defect clusters in copper. V-n and I-n refer to vacancy clusters and SIA clusters of size n, respectively

Migration energies (eV)			Dissociation energies (eV)		
V-1	0.7	Experiment [8]	V-2	0.9	
V-2	0.7		V-3	0.9	
V-3	0.7		V-4	0.9	Calculation [11]
V-4	0.7		V-5	0.98	Calculation [11]
I-1	0.13	Calculation [9]	V-6	1.10	Calculation [11]
I-2	0.11	Calculation [9]	V-7	1.21	Calculation [11]
I-3	0.20	Calculation [9]	V-8	1.35	Calculation [11]
I-4–10 glide	0.10	Calculation [10]	V-9	1.45	Calculation [11]
			V > 9	1.56	Calculation [11]
			I-3	1.16	Calculation [9]
			I-4	1.16	Calculation [9]
			I > 4	2.62	Estimation [7]

were simulated in copper to investigate the effects of both the initial low-temperature dose and the range of temperature changes on cluster accumulation. Also, several requested temperature changes from 300 to 473 K were simulated to study the effect of the number and duration of irradiation cycles.

2. Simulation procedure

KMC-annealing simulations were carried out with the ALSOME code, which was developed to simulate the migration and interactions of defects by kMC methods as described in [7]. Individual atoms are not followed, but each defect is associated with a fcc lattice site and migrates by making ⟨1 1 0⟩ hops to the nearest neighbor lattice sites. To simulate the subcascades of high-energy cascades produced by 14-MeV fusion neutrons, results of MD simulations for a collection of 5 to 25 keV displacement cascades in copper were used as input for the kMC simulations. The basic set of cascades consisted of 14% 25-keV cascades, 43% 10-keV cascades and 43% 5-keV cascades. The simulations were carried out in a three-dimensional computational cell consisting of a cube of fcc lattice sites 150 lattice parameters on edge, containing 13 500 000 lattice sites. Random periodic boundary conditions were applied, in that a mobile defect jumping out of the cell through one cube face reenters the cell at a random position on the opposite cube face. The mobile defects are assumed to be absorbed at a sink, such as a grain boundary or a dislocation line, after traversing the cubic cell 7 times, representing a mean free path of about 270 nm in the unirradiated material.

SIA clusters and vacancy clusters containing up to 10 and 4 defects, respectively, are assumed to be mobile. Larger clusters are immobile and are subject to thermal instability. The values of migration energies and dissociation energies used in this work for Cu are listed in Table 1. In each case, the total irradiation dose was 0.01 dpa, with a dose rate of 10^{-6} dpa/s. Although it is now recognized as an important aspect of the kinetics of cascade defects, one-dimensional (1-D) migration of small SIA clusters in the form of glissile loops was not included in the present work. Because 1-D migrating SIA clusters are less likely to interact with another defect, one would expect fewer SIA clusters, more SIA absorbed at sinks, and perhaps more vacancy clusters if 1-D migration were included. On the other hand, the effects of the temperature changes during irradiation should be qualitatively the same with or without 1-D migration.

3. Formation of defect clusters during temperature change from low to high

3.1. Effects of irradiation dose at low temperature

Fig. 1 shows the densities of SIA clusters and vacancy clusters as a function of irradiation dose for several temperature histories. Results for continuous irradiation at 300 and 473 K are shown, along with two cases of two-temperature irradiation starting at 300 K, then instantaneously jumping to 473 K after doses of 0.0025 and 0.005 dpa, respectively. In addition, Fig. 1 contains the density of defect clusters after irradiation to 0.005 dpa at 300 K, then annealing at 473 K with no additional irradiation.

The density of vacancy clusters (Fig. 1(b)) increases with dose at 300 K, but decreases rapidly due to the dissolution of thermally unstable vacancy clusters when the temperature changes to 473 K. The thermal stability of the vacancy clusters is size dependent, so the smaller ones dissolve first. Under further irradiation at 473 K, the vacancy cluster density slowly decreases to an apparently constant level by 0.01 dpa. The curve showing the vacancy cluster density for continuous irradiation at

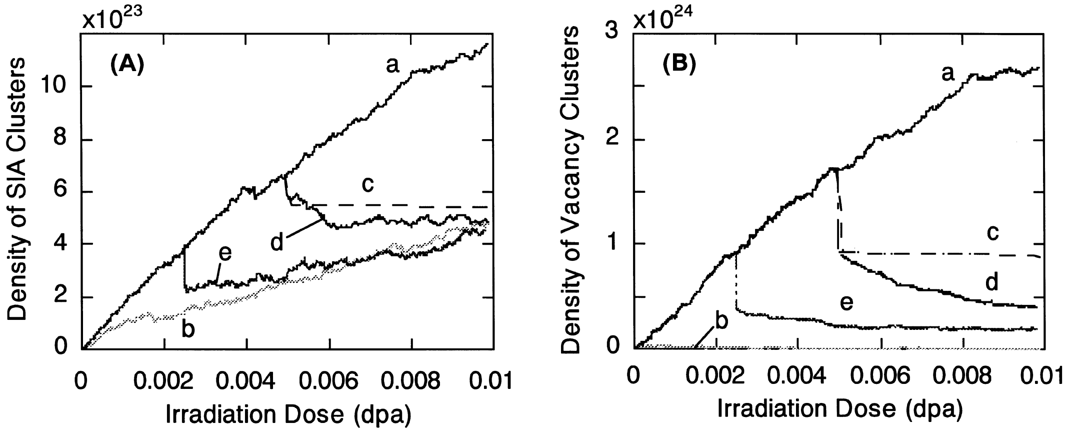

Fig. 1. The accumulation of SIA clusters (A) and vacancy clusters (B) in the irradiations with several temperature histories: (a) irradiation at 300 K; (b) irradiation at 473 K; (c) irradiation at 300 K to 0.005 dpa and annealing at 473 K; (d) initial irradiation at 300 K to 0.005 dpa and second irradiation at 473 K; (e) initial irradiation at 300 K to 0.0025 dpa and second irradiation at 473 K.

473 K is barely visible in Fig. 1(b), being 500 times lower than that at 300 K at these dose levels. The density of SIA clusters (Fig. 1(a)) increases with dose at 300 K and drops quickly when the temperature changes to 473 K due to the interaction of SIA clusters with the mobile vacancies liberated from the unstable small vacancy clusters at that temperature.

When the temperature is changed from 300 to 473 K during a two-stage 300 K/473 K irradiation, the densities of both SIA and vacancy clusters decrease (e.g., the d curves for the temperature change at 0.005 dpa). The initial decrease is caused simply by the temperature change which results in the instability of small vacancy clusters (compare with the c curves for annealing only), but under continued irradiation at the higher temperature, there are further decreases in both SIA and vacancy cluster density. This is primarily because SIA from the continuing irradiation at 473 K interact with small but relatively stable vacancy clusters, reducing them in size until they become unstable and dissolve. Excess vacancies released from these dissolving clusters reduce the number of SIA clusters in the system as well as coarsening the vacancy cluster distribution.

The d and e curves in Fig. 1 show the results for 300 K/473 K irradiations with the temperature changes occurring at 0.005 and 0.0025 dpa, respectively. Initially in both cases, the densities of SIA and vacancy clusters increase with increasing dose at 300 K. In the case where the temperature increases to 473 K after 0.0025 dpa (Fig. 1(e)), the density of SIA clusters drops quickly and stays low until about 0.005 dpa, where it increases similarly to the irradiation that was performed in its entirety at 473 K (Fig. 1(b)). The vacancy cluster density also drops and stays constant, but at a level much higher than that for case b, reflecting the continuing presence of large vacancy clusters produced during the initial portion of the irradiation at 300 K. The situation for the case where the temperature was increased at 0.005 dpa (Fig. 1(d)) is much the same as for that at 0.0025 dpa (Fig. 1(e)), except that for case d, it takes a higher dose of additional irradiation at 473 K to eliminate the greater number of vacancy clusters produced at 300 K. It appears that for the SIA clusters, curve d will coincide with curves b and e at about 0.01 dpa. Also, it is clear that the total number of all residual vacancy clusters is larger (and the number of large residual vacancy clusters is larger) when the dose of irradiation is higher at 300 K before the temperature change. Fig. 2 shows the variation in average SIA and vacancy cluster sizes as a function of dose for the 300 K/473 K irradiation. Both SIA and vacancy clusters show constant average cluster size during the 300 K portion of the irradiation, followed by coarsening during the subsequent 473 K part of the irradiation.

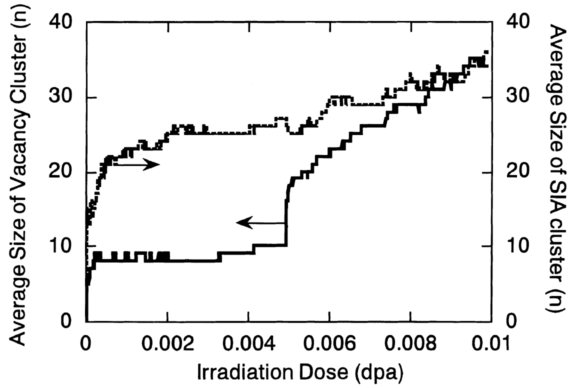

Fig. 2. The average sizes of vacancy clusters and SIA clusters for 300K/473K temperature-change simulations.

3.2. Effects of temperature change range

For all the temperature change simulations considered in this work, the lower temperature is below Stage V and the higher temperature is above Stage V.

Qualitatively, the immediate effects of changing the temperature are similar for all temperature pairs examined, but the magnitudes of the immediate effects as well as behavior under continued irradiation depend on the particular temperatures used. Fig. 3 shows the densities of SIA and vacancy clusters produced in 300 K/473 K, 300 K/673 K and 373 K/573 K irradiations. The densities of defect clusters immediately after the temperature change for the 300 K/673 K irradiation are significantly lower than those for the 300 K/473 K irradiation, because at 673 K, larger vacancy clusters dissolve than at 473 K, causing greater loss of SIA clusters. Also, there are fewer vacancy clusters, but their average size is larger. When the initial temperature is higher (373 K/573 K), the increase in temperature has a smaller effect on the cluster densities because 373 K is near the Stage V temperature, and some small clusters have already started dissolving from the beginning of the irradiation. It is interesting to note that at 0.01 dpa, the concentrations of SIA clusters are about the same for all the temperature histories examined here. However, since the average sizes of residual defect clusters at 0.01 dpa depend on the temperature history, one would expect that the evolution of the defect population under continuing irradiation beyond 0.01 dpa at different temperatures would result in different microstructures.

3.3. Effects of the number of temperature cycles

The effects of the cyclic variation of temperature during irradiation on the formation of defect clusters are

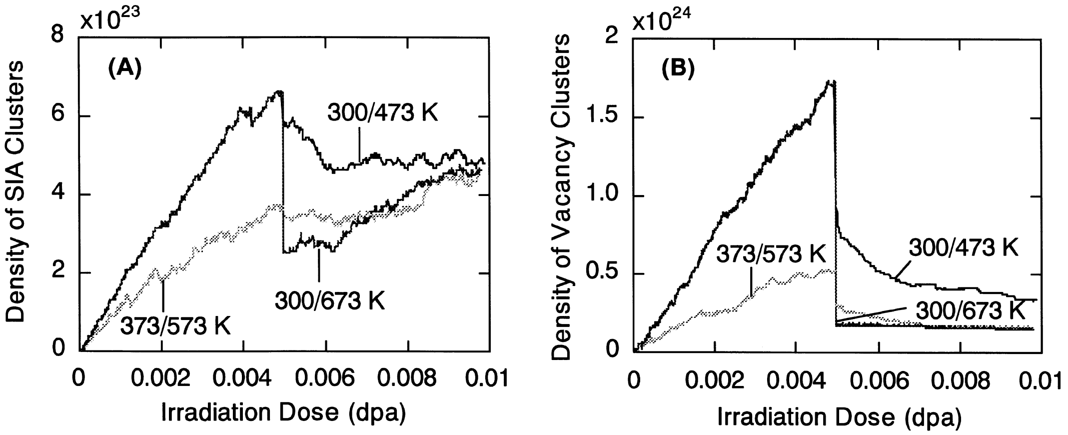

Fig. 3. Effects of the range of temperature-change irradiations on accumulation of SIA clusters (A) and vacancy clusters (B).

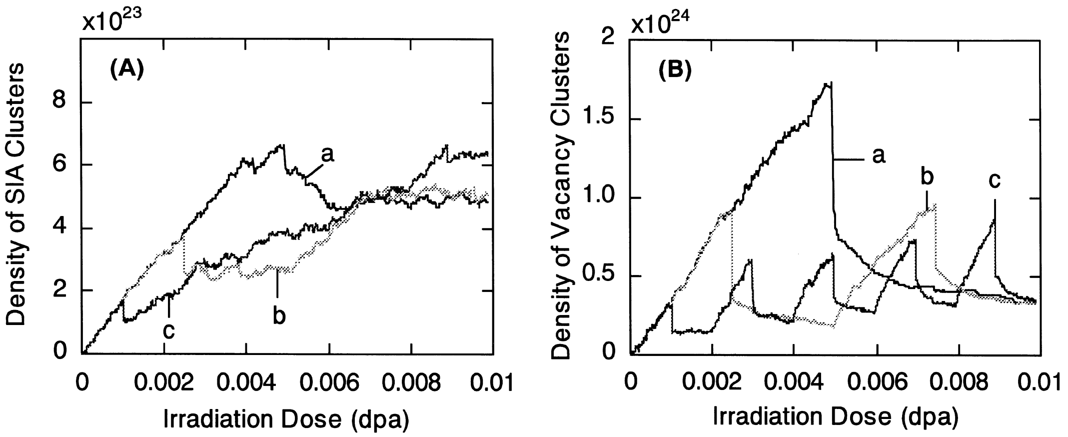

Fig. 4. Effect of number of temperature-cycled irradiations on accumulation of SIA clusters (A) and vacancy clusters (B): (a) temperatures changed one time from 300 to 473 K; (b) temperatures changed two times from 300 to 473 K; (c) temperatures changed five times from 300 to 473 K.

shown in Fig. 4. Alternating temperature runs were done for one, two and five low/high-temperature cycles of equal duration. For all three cases, the total dose is 0.01 dpa and the total times at low temperature and high temperature are equal. As shown in Fig. 4, when there are five short-temperature cycles, the density of SIA clusters increases almost linearly with the dose, showing very small decreases in SIA cluster density during each high-temperature portion. The frequent anneals at 473 K prevent the build up of large vacancy clusters, since the dose at the lower temperature is too small to build up a population of large vacancy clusters while most of the small ones dissolve during the next anneal. It is interesting to note that the density of vacancy clusters at 0.01 dpa is the same for all three numbers of cycles. However, the average vacancy cluster sizes are not the same in each case, which will undoubtedly affect the subsequent evolution under continued irradiation.

4. Conclusions

KMC simulations of the effects of temperature change during irradiation on defect accumulation in copper show that the densities of both SIA and vacancy clusters decrease significantly when the temperature increases from low to high. The decrease of cluster densities is caused by thermal annealing of the defect population introduced at the low temperature as well as by interaction with defects produced in the continuing irradiation at high temperature. The formation of vacancy and SIA clusters in cascades during the low-temperature irradiation and the cluster-size-dependent thermal instability of the vacancy clusters are responsible for the variations in microstructure caused by the temperature changes studied here. Sessile SIA clusters that are thermally stable (at these temperatures) anchor the SIA cluster population, but they can be reduced to mobile size by interaction with vacancies. The vacancy clusters provide a source of mobile defects that depends on temperature, and can likewise be reduced in size and stability by interactions with SIA. Thus, the evolution of the microstructure at any time depends on the interplay of the currently mobile fractions of vacancies and SIAs with the vacancy clusters having sizes at the edge of stability, and the SIA clusters having sizes at the edge of mobility. Microstructure evolution under conditions of changing temperature is sensitive to the magnitude and frequency of the temperature changes in relation to the damage rate and recoil spectrum.

The effect of 1-D migration of SIA defects can change the SIA reaction kinetics significantly [12]. The relationship between these effects and the results of temperature changes reported here will be investigated in future simulations.

Acknowledgements

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