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Simulation of the kinetics of defect accumulation in copper under neutron irradiation

H.L. Heinisch [a,*,1], B.N. Singh [b]

[a] *Pacific Northwest National Laboratory, P.O. Box 999, MS P8-15, Richland, WA 99352, USA*
[b] *Materials Department, Risø National Laboratory, DK-4000, Roskilde, Denmark*

Abstract

Stochastic annealing simulations were originally developed for and successfully used to describe the short-term annealing stage of defect evolution within individual displacement cascades. Applied to a sufficiently large volume with periodic boundaries, stochastic annealing simulations can also provide a description of the evolution of defects produced during continuous irradiation. The irradiation is simulated by successive introduction of collections of defects representing the primary damage state of individual cascades placed in the simulation volume randomly in time and space. Cascade energies and the rate of their occurrence are chosen to approximate the damage due to the neutron flux of the 14 MeV neutron source Rotating Target Neutron Source-II (RTNS-II). The cascades are chosen from a library of cascades generated in molecular dynamics (MD) simulations for recoil energies from 5 to 25 keV. The numbers of each type of defect, defect cluster size distributions, as well as the positions of the defects within the crystal, are monitored as a function of time. Simulated defect cluster densities as a function of dose up to 0.1 dpa at room temperature are compared to experimental results. The simulated cluster densities are within about a factor of two of the experimental results over several orders of magnitude of dose. The effects on damage accumulation due to dose, dose rate, cascade overlap and interstitial cluster mobilities are demonstrated with examples.

1. Introduction

Under cascade-producing irradiation, defect production and subsequent defect accumulation are strongly influenced by features of the initial cascade damage event. The temporal and spatial inhomogeneities of cascade damage, as well as details of damage that manifest themselves on the atomic scale, make it necessary to develop atomic-scale models as the basis for understanding the effects of cascade production on microstructure evolution and mechanical property changes in irradiated materials. The ability to correlate and extrapolate irradiation test data on fusion materials hinges on understanding radiation damage at this scale, as well as understanding its influence on behavior at the macroscopic scale.

Advances in atomistic descriptions of cascade events by molecular dynamics (MD) simulations in the past decade, coupled with experimental information, have resulted in the identification of cluster formation directly in cascades, and the subsequent behavior of those clusters, as key elements affecting the evolution of the microstructure. Stochastic annealing simulation provides a bridge between the atomistic and macroscopic scales that enables the direct effects of cascade production on the microstructure to be investigated. The work reported on here is the next step beyond the application of stochastic annealing simulations to the local short-term annealing stage of individual cascades, which has been reported elsewhere [1,2]. We report here on the simulation of the kinetics of damage accumulation under low doses of cascade-producing irradiation in copper at room temperature.

* Corresponding author. Tel.: +1-509 376 3278; fax: +1-509 376 0418; e-mail: hl_heinisch@pnl.gov

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2. The simulations

The computer simulations were performed using the stochastic annealing code ALSOME, which is described in detail in earlier publications [1]. For defect accumulation simulations, the same kinetic model is used as for individual cascades, along with the same defect parameter values. In particular, small clusters of self-interstitial atoms (SIAs) containing up to 10 SIA are assumed to be highly mobile, with clusters from sizes 4 to 10 gliding in thermally activated, one-dimensional random motion as discussed in Ref. [1]. The volume for the accumulation simulations is significantly larger than for individual cascades, and new cascades are introduced into the volume as the annealing progresses to simulate the conditions of an ongoing irradiation. To make the computation tractable with respect to computer time, a relatively small cubic volume 54 nm on edge was chosen, and periodic boundaries were applied. With one cascade in this volume, the smallest possible dose is about 10^{-5} dpa (displacements per atom) and the minimum cluster density is about 10^{21} m^{-3}. These values correspond to the lowest-fluence data obtained in 14 MeV neutron irradiations using Rotating Target Neutron Source-II (the RTNS-II, used for fusion materials research in the 1980s).

Room temperature irradiation of copper by 14 MeV neutrons was simulated with a flux of 5, 10 and 25 keV cascades generated in MD simulations by Diaz de la Rubia and Guinan [3] and by Foreman et al. [4]. Fusion neutrons produce recoil atoms with an average energy of about 300 keV, but cascades produced by those high-energy recoils are formed as a series of subcascades having average energies of 20–30 keV each [5]. More than 95% of the damage in Cu irradiated by 14 MeV neutrons is produced in cascades with energies above the threshold for producing subcascades [5], so low-energy events with high-defect survival efficiency make a negligible contribution to the damage produced by 14 MeV neutrons. Annealing simulation studies of closely spaced subcascades [2] show that little interaction of the defect populations of adjacent subcascades takes place as they anneal simultaneously. This is primarily because the SIAs are spread about the periphery of each subcascade, and they are significantly more mobile than the vacancies, which are concentrated near the center of each subcascade. Thus, the flux of 5–25 keV MD cascades introduced into the simulation volume should be a reasonable first approximation to the individual subcascades formed in irradiations by 14 MeV neutrons. A dose of slightly more than 0.1 dpa was obtained by introducing approximately 12,000 cascades into the simulation volume. This large set of cascades was produced by repeated use of the 13 available MD cascades.

Although periodic boundaries were used, the existence of a finite grain size (or a density of other intrinsic sinks) was simulated by imposing a limit on the total distance a mobile defect could travel (including periodic continuations of the simulation volume). Upon reaching this limit, the defect is removed from the simulation and counted as being absorbed. In the present simulations, defects that migrate the equivalent of 380 nm on average are considered to have reached a grain boundary or other intrinsic sink, and they are removed. Mobile clusters may grow or shrink (and glissile SIA clusters may change direction) by interaction with other defects they encounter as they traverse the crystal to an eventual sink.

3. Results

Doses up to 0.1 dpa were simulated at various damage rates, and the damage accumulation was observed as a function of dose. Fig. 1 shows the defect cluster density as a function of dose for a simulated irradiation at a dose rate of 10^{-10} dpa/s. The vacancy and SIA cluster densities are shown separately, as well as their total. Only clusters containing more than 10 point defects are counted in the cluster densities for either type. At room temperature there are not likely to be any SIA clusters of size 10 or less, because of the high mobility of these small clusters. In sympathy with this limit for SIA clusters, and also because it may be unlikely that vacancy clusters with 10 or fewer vacancies can be consistently observed by TEM, the limiting size for reported vacancy clusters was also set arbitrarily at 10. No other attempt to scale the simulation results to reflect defect cluster visibility has been made yet. Also in Fig. 1, the simulation results are compared to experimentally observed cluster densities in copper irradiated at room temperature in RTNS-II. The experimental data are compiled from three different experiments [6–8]. The RTNS-II test specimens were irradiated at dose rates varying from about 10^{-11} to 10^{-9} dpa/s.

The apparent scatter in the simulation results in Fig. 1 and others in this report at doses less than a few times 10^{-4} dpa is an artifact of the simulation method. It results from the size of the periodic volume. To get smooth curves down to doses of 10^{-5} dpa would require a significantly larger periodic volume. At less than about 3×10^{-4} dpa there are more vacancy clusters than SIA clusters, since the highly mobile SIA clusters, moving rapidly to sinks, are slower to start building a concentration of immobile clusters. At doses above 10^{-3} dpa there are more SIA clusters than vacancy clusters, and the ratio of their numbers stays approximately constant throughout the remainder of the irradiation. The simulated total density of clusters is in very good agreement with the experimentally reported cluster density, being within about a factor of 2 throughout the range of doses.

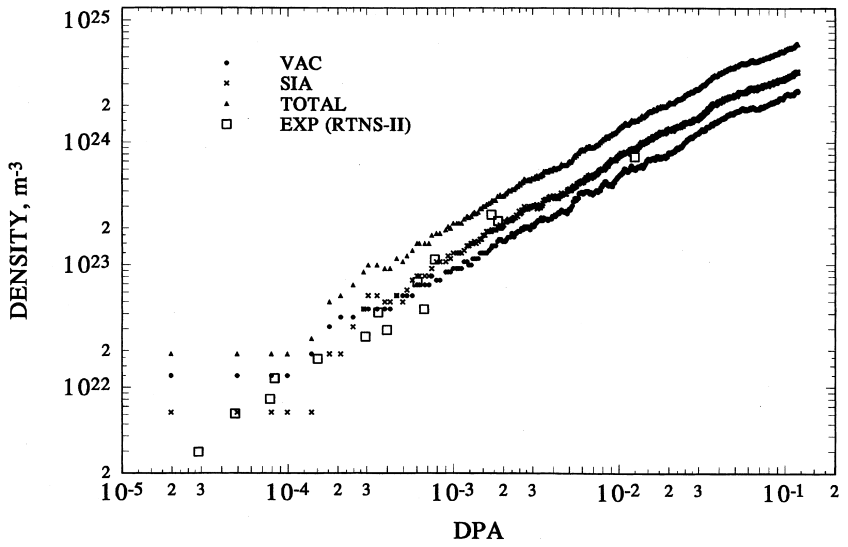

Fig. 1. Vacancy, SIA and total cluster densities as a function of accumulated dose for copper irradiated at 300 K under cascade-producing irradiation (as by 14 MeV neutrons) at 10^{-10} dpa/s. Simulation results are compared to experimental data from TEM observations of copper irradiated in RTNS-II [6–8].

Fig. 2 shows results of the same simulation carried out at damage rates well above those of the RTNS-II experiments, but spanning the range of existing neutron irradiation facilities. As expected, at higher dose rates more clusters are formed as more new defects arrive in the volume before those already there have an opportunity to escape. The effects of damage rate are further shown in Figs. 3 and 4, where the surviving defect fractions and the numbers of defect clusters are shown respectively as a function of dose rate. The surviving defect fractions are the total numbers of point defects of each type (including those in clusters) remaining in the system relative to the calculated dpa value for the irradiation dose (0.05 dpa for these examples). The damage rate does not affect the SIA population as strongly as it affects the vacancies because the SIAs migrate

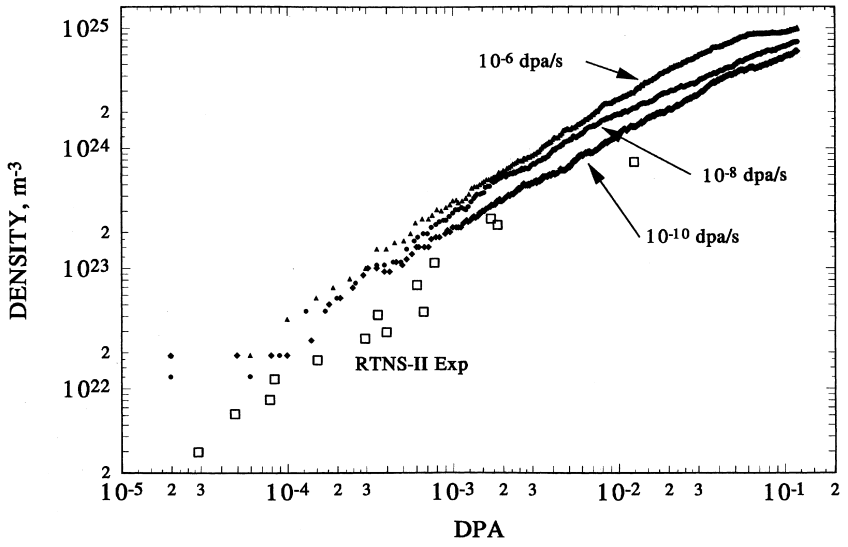

Fig. 2. Total defect cluster densities as a function of accumulated dose for copper irradiated at 300 K under cascade-producing irradiation (as by 14 MeV neutrons) at three dose rates. Simulation results are compared to experimental data from TEM observations of copper irradiated in RTNS-II [6–8].

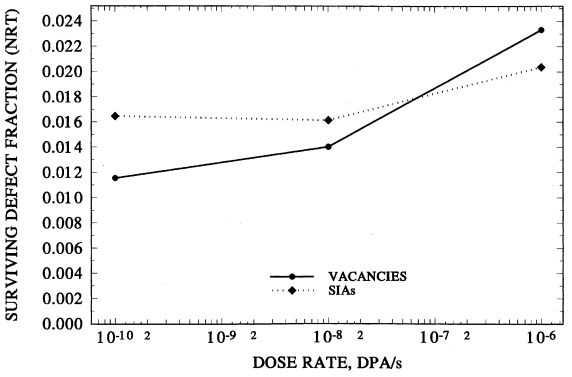

Fig. 3. Surviving defect fractions as a function of dose rate for cascade-producing irradiation (as by 14 MeV neutrons) at 300 K. The fractions are relative to the calculated NRT dpa value for an irradiation to 0.05 dpa.

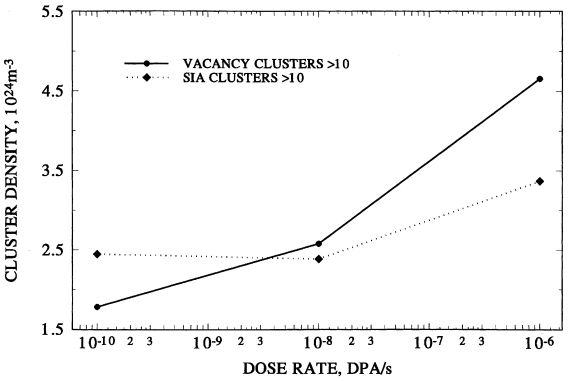

Fig. 4. Vacancy and SIA cluster densities as a function of dose rate for cascade-producing irradiation (as by 14 MeV neutrons) at 300 K. The densities are for an irradiation to 0.05 dpa.

significantly faster than the vacancies at room temperature, so they are less likely to be affected by the defects of the 'next cascade'. It is interesting to note that the surviving defect fractions are in the range of only 1–2% of the calculated dpa. This corresponds to the survival of about 5–10% of the defects that are actually introduced into the volume during the simulation as the remaining defects of the primary damage state (i.e. after MD simulations terminate at about 10 ps).

Various MD results indicate that small SIA clusters are formed directly in cascades and migrate rapidly by gliding in one-dimensional motion, and this migration mechanism was incorporated into the ALSOME code in earlier simulations of the annealing stage of individual cascades [1,9]. However, the details of SIA migration by glide and the effect of cluster size on this phenomenon are still not well known. The present computer simulations of measurable defect accumulation offer an opportunity to investigate the sensitivity of the model's results to variation in the definition of gliding SIA clusters. For example, the simplest case is the effect of changing the maximum size at which clusters are glissile. Fig. 5 shows the results of repeating the simulation referred to in Fig. 1, but changing the maximum size of glissile clusters from 10 to 19 SIA. With larger glissile clusters, considerably fewer defect clusters are produced or survive during the irradiation, and the dose dependence of the cluster density exhibits a smaller slope. More than an order of magnitude fewer SIA clusters are produced when the glissile cluster size is increased to 19. Thus, the damage accumulation model does show a significant dependence on the size of glissile SIA clusters, indicating that this issue requires careful further study at the atomic scale.

Under continuing irradiation at room temperature, saturation of the defect cluster density might be expected for two reasons: (1) as the inter-defect spacing becomes smaller, it becomes more likely that defects in the next cascade will react with the existing clusters at the expense of forming new clusters and (2) with increasing dose, it becomes more likely that the next cascade will occur on top of existing defects, causing the overlapped damaged regions to heal and resulting in fewer net defects than if there were no overlaps. The effects of cascade overlap were included explicitly in the simulations by recombining any existing defects within the volume of an overlapping cascade. The results shown in Figs. 1–5 include the effects of cascade overlap, which appear to be stronger at higher dose rates. Some simulations were done without the cascade overlap feature to assess its effects. The results at 10^{-6} dpa/s with and without cascade overlap are compared in Fig. 6. Up to a dose of 0.1 dpa there seems to be only a small tendency toward saturation caused simply by high-cluster density, in contrast to including the cascade overlap.

4. Discussion and conclusions

Stochastic simulations of the kinetics of damage accumulation in copper under continuous, low-dose neutron irradiation have produced results in good agreement with experimentally determined defect cluster densities using a simple stochastic hopping model and defect migration properties obtained largely from theoretical calculations [1,9]. The results were not scaled or fitted to the experimental data. By applying 'visibility criteria' to the simulation results, even better agreement between the simulation and experimental results could be expected. However, until more simulations are performed on low-dose experiments under other conditions for which good experimental data exist, it may be premature to expect that the simulation model is sufficiently descriptive of the defect accumulation phenomena. On the other hand, the present direct comparison of results

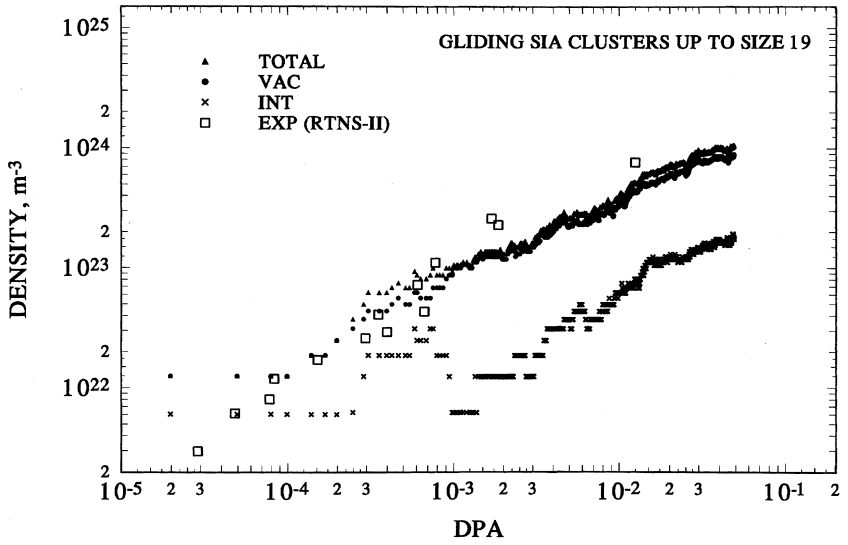

Fig. 5. Vacancy, SIA and total cluster densities using a model for gliding SIA clusters up to size 19 SIA as a function of accumulated dose for copper irradiated at 300 K under cascade-producing irradiation (as by 14 MeV neutrons) at 10^{-10} dpa/s. Simulation results are compared to experimental data from TEM observations of copper irradiated in RTNS-II [6–8].

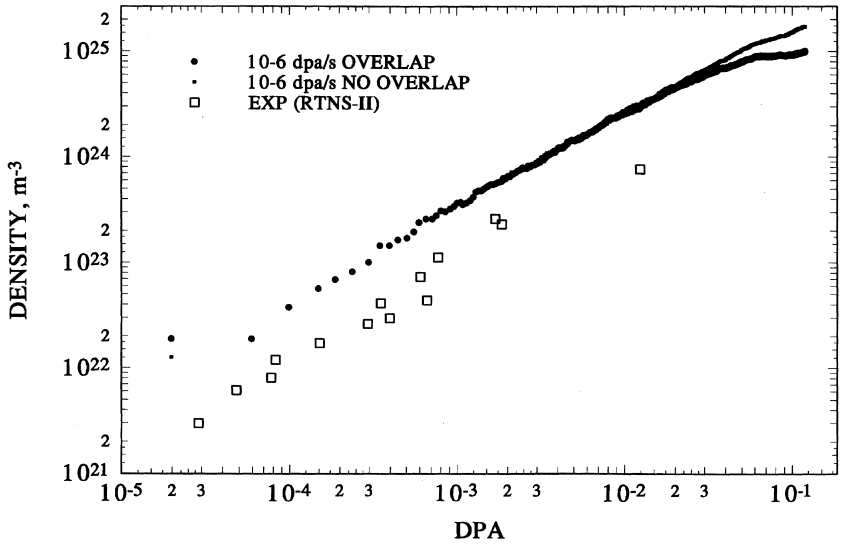

Fig. 6. Effects of cascade overlap on defect accumulation at a dose rate of 10^{-6} dpa.

of simulations and experiments provides a good opportunity to explore the sensitivity of the model and perhaps of the phenomena themselves, to the variation of many important defect and environmental parameters. As a test of sensitivity to the recoil energy spectrum, initial simulations of electron irradiations under similar dose rates as reported here have shown no defect accumulation whatever, which is consistent with experiments. Simulations of room temperature electron irradiations at a dose rate of 1 dpa/s revealed a distribution of vacancy clusters that form after the irradiation ceases at 0.01 dpa. This example illustrates the validity of the annealing simulation model over an extreme variation of parameters, but more exploration of various reactor-relevant conditions is required.

Simulations with even more realistic models, experimentally validated, will give us the opportunity to probe behavior we cannot see directly by experiments: the fraction of invisible clusters, the surviving defect fractions, the expected concentrations of vacancy and SIA

clusters and loops, and, at higher temperatures, the level of vacancy supersaturation. We can explore void nucleation models and the roles of impurities. All of this information – or at least much better approximations to the real situation than is presently available or possible – will be valuable input to the global models that describe the effects of radiation damage at the macroscopic scale.

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