

THE EFFECTS OF SELF-INTERSTITIAL CLUSTERS ON CASCADE DEFECT EVOLUTION BEYOND THE PRIMARY DAMAGE STATE - H. L. Heinisch (Pacific Northwest National Laboratory¹)

OBJECTIVE

The objective of this work is to determine the energy and temperature dependence of defect production and microstructure evolution for the development of fission-fusion correlations.

SUMMARY

The intracascade evolution of the defect distributions of cascades in copper is investigated using stochastic annealing simulations applied to cascades generated with molecular dynamics (MD). The temperature and energy dependencies of annihilation, clustering and free defect production are determined for individual cascades. The annealing simulation results illustrate the strong influence on intracascade evolution of the defect configuration existing in the primary damage state. Another factor significantly affecting the evolution of the defect distribution is the rapid one-dimensional diffusion of small, glissile interstitial loops produced directly in cascades. This phenomenon introduces a cascade energy dependence of defect evolution that is apparent only beyond the primary damage state, amplifying the need for further study of the annealing phase of cascade evolution and for performing many more MD cascade simulations at higher energies.

PROGRESS AND STATUS

Using MD, cascade simulations can follow the evolution of the cascade region through the quenching of the thermal spike (about 10 ps) and perhaps somewhat beyond (about 100 ps), depending on the energy. Once the cascade energy is dissipated during the quenching stage, little happens on the picosecond time scale. However, the cascade, consisting of a high concentration of defects in a small volume, is in a metastable state that continues to evolve by thermally activated diffusion, during which intracascade annealing results in recombination and clustering, as well as escape of some defects from the cascade region. Thus, simulating the annealing stage of cascade evolution offers the possibility of determining definitively the defect distributions individual cascades contribute to the global picture.

The annealing simulation code used for these studies and its input parameters are described in detail elsewhere [1]. The essential input parameters for the annealing simulations are the relative jump probabilities of the mobile defects at the temperature simulated and the critical reaction distances. Another element of the present annealing model is the dissolution of clusters. The relative frequency of single defect emission from a cluster as a function of cluster size is calculated using cluster binding energies from computer simulations [1].

The self-interstitial atom defects (SIA) are assumed to have two forms, depending on size. MD simulations of cascades in copper clearly show that SIA clusters exist in the primary damage state, i.e., that SIAs form clusters directly in cascades during the first 10 ps, and that the clusters collapse to glissile loops that move one-dimensionally. The present annealing model assumes that SIA clusters of size 1-3 are mobile in 3

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dimensions and that SIA clusters of size 4-9 are mobile in 1 dimension. Clusters larger than size 9 are considered immobile. The activation energy for 1-dimensional glide is taken as 0.1 eV, about the same as for the motion of a single SIA, independent of size. This model is based on Trinkaus, Singh and Foreman's interpretations of MD simulations [2], which is probably correct in concept, but not quantitatively definitive. There still does not exist enough information, either from MD simulations or experiments to sufficiently determine the characteristics of the formation of glissile SIA loops in cascades and their kinetics during the annealing stage. Stochastic annealing simulations, however, provide an excellent tool for studying the sensitivity of the effects of glissile loop properties on the annealing stage.

Figure 1 shows the fraction of surviving defect pairs and the fractions of escaping vacancies and SIAs after the annealing stage of two 25 keV cascades simulated with MD in copper as a function of temperature. The average fractions are determined relative to the number of defects remaining

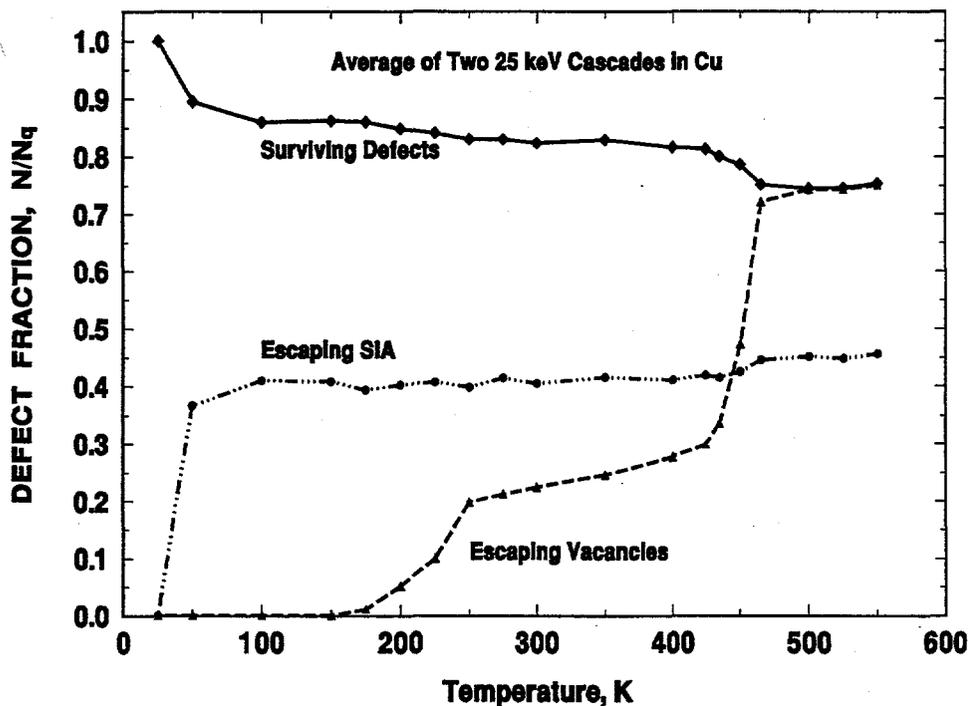


Figure 1. The fractions of point defects surviving recombination and of vacancies and SIAs escaping from 25 keV cascades in copper during the annealing stage, as a function of cascade annealing temperature. Each data point is the average of 100 anneals of each of two 25 keV cascades in copper generated in MD simulations.

in the cascades after quenching (i.e. at the end of the MD simulations) for 100 separate anneals of each of the two cascades. During the annealing stage, about 20% of the initial defects recombine and about 40% of the SIAs escape the cascade, with weak dependence on the temperature. The other 40% of the initial SIAs remain in the cascade region as immobile clusters. Approximately 60% of the SIAs that escape are in the form of small glissile loops moving in 1-dimensional random walks. Almost all the recombination

occurs as a result of the diffusion of the SIAs. An increasing fraction of the original mobile vacancies escapes the cascade at temperatures from 175-250 K. From 250-400 K small clusters also start to dissolve. Above 450 K all vacancy clusters are unstable and readily dissolve, resulting in slightly more recombination and escape of all remaining vacancies.

The situation depicted in Fig. 1 changes significantly when the size limit for glissile loops is changed in the annealing model. The same information in Figure 1 is plotted in Figure 2 for four values of the size limit for glissile SIA loops. The curve labeled $G=9$ refers to the escaping SIAs in the "standard" model which has glissile loops of size 4-9, as depicted in Figure 1. The curve labeled $G=\text{none}$ is when no glissile loops are permitted and only SIA clusters of size 1-3 are mobile, moving in three dimensions. The fraction of SIAs that escape the cascade region varies from 10-80%, depending on the size of SIA clusters that are assumed to be glissile loops in the annealing model. It is obvious that few clusters consisting of more than 20 SIAs are formed in these cascades, even during the annealing stage. Because of this narrow size range, the upper limit for glissile loop formation has a large impact.

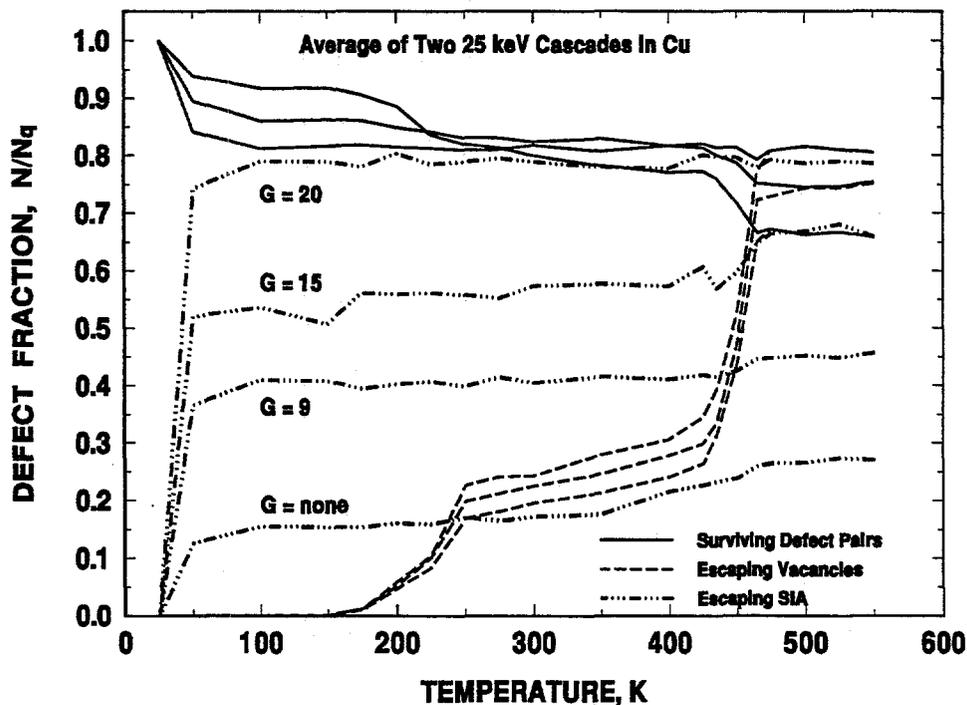


Figure 2. The fractions of surviving and escaping defects as a function of temperature for 25 keV cascades as in Fig. 1. The effects of changing the size limit for glissile SIA loops on the annealing behavior is illustrated.

DISCUSSION

Figure 2 reveals the importance of knowing the characteristics of SIA loop formation and kinetics in cascades. It is also of interest to note from Fig. 2 that the fractions of surviving defects and escaping

vacancies vary much less with the glissile SIA loop size limit than does the fraction of escaping SIAs, implying that the glissile SIA loops have few interactions with the vacancy population, and that recombination is due almost entirely to the diffusion of the small SIA clusters. This vividly illustrates the significant difference between the 1-dimensional random walk of glissile SIA loops and the 3-dimensional random walk of the small SIA clusters.

Sensitivity to the glissile SIA loop size limit during the annealing stage also implies a possible energy dependence in the global field of defects that has gone unrecognized. The probability of producing large SIA clusters is smaller for lower energy cascades. Even in the energy range of 5-25 keV the ratio of mobile to immobile SIAs that are formed may vary significantly. This energy sensitivity also affects the rates of SIA survival, which depend on the relative numbers of SIAs in glissile loops. It is likely that SIA loops up to size 20 and larger are glissile, and also likely that the migration energies of glissile loops increase with size. Additional MD simulations can shed considerable light on this key issue.

Determining glissile loop characteristics may now be the most important issue for annealing simulations, but it is not the only important issue. Virtually all the activation energies and pre-factors for the diffusion and emission processes need to be evaluated using consistent simulation approaches, and they need to be checked against experimental information where possible. Critical reaction distances need to be evaluated carefully as well. Atomic-scale defect studies need to be carried out to verify the use of homogeneous random hops in the annealing simulations, or to determine what correlated motions of defects are required to accurately model the annealing stage. Finally, but very important, is the need to establish a data base containing many more higher energy cascades than presently exist for each material of interest. Energies should range up to, and perhaps beyond, the threshold for subcascade formation, which is approximately 25 keV for copper [3]. Although the damage energy and the number of initially displaced atoms in cascades of the same energy have fairly narrow distributions, the distribution of sizes and shapes, which can affect defect interactions during the annealing stage, is quite broad [3]. The two 25 keV cascades used in this work so far are both somewhat compact, so some conclusions drawn from the results here may not be representative of the average behavior. In general, the higher the cascade energy, the less likely any single cascade, or small sample of cascades, can be considered to be typical of the entire distribution.

FUTURE WORK

The results discussed here were included in a presentation at the Symposium on Microstructure Evolution During Irradiation, 1996 Fall MRS Meeting, Dec. 2-6, 1996, Boston, and will appear in the proceedings. These results were also discussed at the workshop on Defect Production, Accumulation and Materials Performance in Irradiation Environment, Davos, Switzerland, October 2-8, 1996. Proceedings will be published in the Journal of Nuclear Materials. Annealing simulations are in progress on a library of cascades in copper generated by Alan Foreman et al. at Harwell. We are also working on annealing studies of cascades in iron and other materials in collaboration with the University of Liverpool.

REFERENCES

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