

EFFECTS OF IN-CASCADE DEFECT CLUSTERING ON NEAR-TERM DEFECT EVOLUTION - H.L. Heinisch (Pacific Northwest National Laboratory)*

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 effects of in-cascade defect clustering on the nature of the subsequent defect population are being studied using stochastic annealing simulations applied to cascades generated in molecular dynamics (MD) simulations. The results of the simulations illustrate the strong influence of the defect configuration existing in the primary damage state on subsequent defect evolution. The large differences in mobility and stability of vacancy and interstitial defects and the rapid one-dimensional diffusion of small, glissile interstitial loops produced directly in cascades have been shown to be significant factors affecting the evolution of the defect distribution. In recent work, the effects of initial cluster sizes appear to be extremely important.

PROGRESS AND STATUS

Introduction

The production of defect clusters directly in cascades in copper is well established, both in atomic-scale MD computer simulations [1,2] and experimentally in results of diffuse x-ray scattering experiments [3]. The consequences of intracascade clustering of self-interstitial atoms (SIAs) and vacancies, and of the thermal stability of these clusters, have been specifically addressed in the recently proposed "Production Bias Model" (PBM) [4]. The PBM has been further developed to account for the very important consequences of one-dimensional transport of small, glissile SIA clusters [5,6] and of the continuous transformation of larger sessile SIA clusters into glissile ones due to their interactions with single vacancies[7]. The PBM or any other such model can predict the global behavior of the system over macroscopic sizes and times. However, it has the limitation of not being able to deal explicitly with the problem of temporal and spatial fluctuations in the defect cluster production and size distributions occurring continuously under cascade damage conditions. At present, these cascade phenomena are dealt with by assuming some global average values for the relevant parameters. Clearly, accurate and reliable predictions from the PBM or other such models are possible only if physically realistic values for their input parameters can be obtained either experimentally (unlikely) or from atomic-scale models that include explicit temporal and spatial information. The determination of parameters for PBM from results of these models is, therefore, of crucial importance.

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Even after the longest practical MD simulation, the defect clusters of a single cascade remain spatially localized in a "metastable" state far from the spatial and temporal uniformity required for the global picture to which rate theory may be applied. Atomic scale stochastic annealing simulation provides the necessary link between the localized, short-term, atomistic view of individual MD cascades and the spatially averaged global view required for the application of rate theory. In the present work, stochastic annealing simulations are being done on individual, isolated cascades, as well as on simulated irradiations represented by continuous introduction of new cascades into an annealing volume with periodic boundaries. Preliminary results of the effects of clustering are presented here.

Clusters of Self-interstitial Atoms

Self-interstitial atoms form small clusters in cascades, with size up to $n = 14$ SIAs observed in x-ray scattering experiments [3]. In MD simulations, SIA clusters up to about size $n = 15$ are observed [1], and small ($n = 4-6$) clusters have been observed to form initially sessile loop configurations that unfault to glissile loops on (110) planes within picoseconds after the cascade occurs [2]. In earlier annealing simulations performed in the present program [8], it was assumed that SIA clusters up to $n = 9$ spontaneously become glissile loops, while larger clusters become sessile loops requiring greater energy to unfault. Little is known about the kinetics and energetics of SIA loops in copper, but MD-based studies are in progress elsewhere. The consequences of changing the assumptions of the earlier annealing simulations are being determined to study the effects of defect cluster properties.

In earlier work [8], cascades in copper generated with MD at 25 keV [1] were annealed as individual, isolated cascades as a function of temperature. Even at temperatures above $0.3 T_m$ (melting temperature), where all vacancy clusters dissociate, the SIA clusters with $n > 9$, which were assumed sessile, remained essentially intact in the cascade region, unaffected by the dissipation of the local vacancy concentration. In recent annealing simulations using lower energy cascades of 5-10 keV [2], which have no clusters/loops larger than $n = 9$, all SIA clusters escape the cascade region, except for a very few cases where clusters coalesce to form sessile clusters. Thus, with the present model (SIA loops up to $n = 9$ being glissile), there is a significant energy dependence in the annealing stage of cascades that is manifested at relatively high cascade energy. If the maximum allowable size of glissile SIA loops is increased in the model, all SIA loops escape the cascade region at all energies, as expected. Clearly, more details of loop formation are needed.

Defect evolution during irradiation is being studied by performing annealing simulations on fluxes of recoils represented by cascades of various energies placed randomly in space and time into a large, periodic volume. Dose rates and total doses are well below where cascade overlap would be expected. A very low sink density is represented by absorption of mobile defects that have traveled at least several microns, which is simulated by their wrapping around the periodic boundaries a limiting number of times. Using 5-25 keV cascades in the approximate ratios representing a fast reactor irradiation, and with the present $n = 9$ glissile SIA loop limit, the net vacancy population initially dominates at 400 K. However, during the simulation the population of large sessile SIA clusters eventually builds to become quite dominant. Reduction of large sessile loops to glissile size by absorption of vacancies does not occur under the conditions studied. To achieve a

vacancy super saturation, it is probably necessary to let SIA clusters much larger than $n=9$ be glissile or easily become glissile.

Vacancy Clusters

Vacancy clustering is observed in MD simulations [1,2] usually as somewhat organized agglomerations of vacancies. The very stable stacking fault tetrahedra (SFT) have not been observed to form during the time that can be followed by MD simulations. At temperatures well above $0.3 T_m$ the amount of vacancy clustering in cascades is not particularly important, since clusters are quite unstable at those temperatures. At lower temperatures, the vacancy cluster distribution in cascades can have an effect on the fraction of vacancies that escape from the cascades. Figure 1 shows the fractions of escaping vacancies as a function of temperature for cascades of various energies. The 25 keV cascades, containing some larger, more stable vacancy clusters, display a different temperature dependence than the lower energy cascades, which contain only a few small clusters that easily dissociate at lower temperatures. Thus, the clustering behavior of vacancies in cascades also results in a recoil energy dependence. However, when the potential formation of more stable vacancy configurations (e.g. SFT) is taken into account, the effect may be somewhat different from that displayed in Fig. 1.

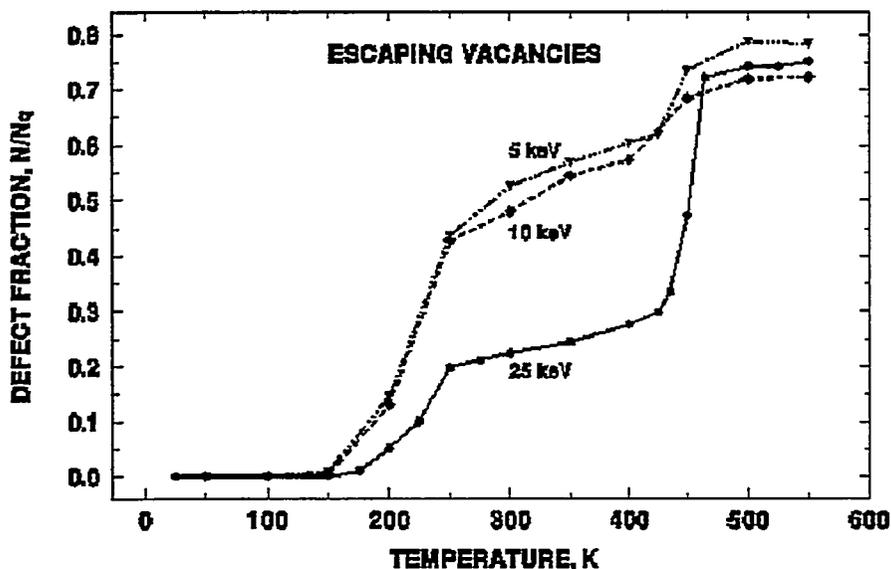


Figure 1. The fractions of vacancies escaping from the cascades during annealing as a function of annealing temperature. The fractions are relative to the initial number of vacancies at the end of the MD simulation in each cascade.

CONCLUSIONS

The preliminary results in this study indicate that defect evolution beyond the primary damage state modeled by MD simulations is sensitive to the characteristics of cluster formation in cascades. To make models more realistic, it is necessary to know more details about the production, migration and interaction of both SIA and vacancy clusters. More MD cascades are needed to get good statistics on cluster production. MD studies are needed to determine the characteristics of loop formation, especially sessile-glissile transformations, migration energies, glide direction changes and loop interaction characteristics. In the absence of this information, stochastic annealing simulations will be used to explore the consequences of variation in the relevant parameters.

FUTURE WORK

The effects of glissile SIA loop migration characteristics are being investigated in annealing simulations. Thermally-activated and interaction-stimulated changes in glide direction are being incorporated into the model along with sessile-glissile transformations. This work will be reported on at ICFRM-8.

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