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Statistical analysis of cluster production efficiency in MD simulations of cascades in copper

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Abstract

A large number of displacement cascades has been simulated using molecular dynamics (MD) for primary knock-on atoms (PKAs) of energy in the range 2–20 keV at 100 and 600 K. The defect production efficiency, intracascade clustering efficiency and fractions of self-interstitial atoms (SIAs) and vacancies contained in the clusters produced in the cascades were determined. The fractions of both SIAs and vacancies contained in clusters increased with increasing PKA energy and approached a saturation level at 20 keV. At both temperatures the clustered fraction of SIAs was higher than the clustered fraction of vacancies. This difference is larger at 600 K than that at 100 K and increases with increasing PKA energy. These results are compared with the corresponding values derived from experimental results. The comparison suggests that the single cascade simulations yield considerably higher values of the clustered fractions of both SIAs and vacancies primarily because the impact of intercascade defect reaction kinetics cannot be taken into account in the MD simulations.

1. Introduction

In recent years a large amount of effort has been made, particularly using the molecular dynamics (MD) simulation technique, to establish the details of intracascade interactions and the resulting nature of the primary damage state. These studies have provided a physics-based insight into processes involved in the transformation of atomic displacements into surviving lattice defects and their clusters. The simulations have also yielded the total number of defects that survive at the end of the cooling-down phase of individual cascades in the form of single self-interstitial atoms (SIAs) and vacancies and their clusters [1–4]. Undoubtedly, the quantitative results of these simulations are uniquely valuable since they can be used as the source term for damage production in the analytical treatment of the global damage accumulation under cascade damage conditions. However, to convert MD modelling into a tool, that produces not only qualitative information but also reliable quantitative data, it must be verified by a comparison with experimental results. This is not a trivial task because such a comparison can be made only with the predictions of a theoretical model that uses the MD data and the model itself may contain approximations and uncertainties. However, such comparison with the results of specially designed experiments (e.g. [5]) can help reduce the number of uncertainties and thus can enable a comparison to be made with reasonable accuracy. Furthermore, the MD results have to be made more realistic for the purpose of comparison with experimental results by carrying out a large number of cascade simulations and statistical analysis of the results. This is the task that has been attempted in the present work.

2. Computational model

Primary knock-on-atoms (PKAs) of energy in the range from 2 to 20 keV were simulated at temperatures of 100 and 600 K in fcc cube-shaped crystallites containing from about 130 000 to 2 000 000 mobile atoms. These crystallite sizes are large enough to allow

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all events to be taken into account without omitting widely spread cascades (see e.g. [6]). The usual periodic boundary conditions were applied because of their simplicity, which benefits calculation speed and simplifies cascade monitoring, for example, by keeping the centre of the kinetic energy distribution near the centre of the crystallite. Special attention was paid to cascade statistics and a large number of cascades (up to 45) of the same energy was simulated for the same conditions in order to allow a wide variety of defects to form. More than 400 cascades were simulated in total. A separate treatment of 'hot' and 'cold' regions, reported earlier [7], was used, which allows a very precise treatment of the ballistic stage without a significant increase of computational time. For example, for a 20 keV cascade simulated at 100 K, t_s varied from 0.002 to 4.5 fs.

Low-symmetry directions $\langle 1\,3\,5\rangle$ were chosen for PKAs to avoid channelling effects. In each particular simulation PKA was chosen randomly. Taking into account thermal displacements of atoms and a large number of simulations at each conditions such procedure covers a wide range of 'effective' PKA directions.

In order to test the sensibility of the results to details of the model, two interatomic potentials were used, namely the equilibrium, short-ranged, many-body potential (SRMBP) developed in [8] and the non-equilibrium long-ranged pair potential (LRPP) described in [9]. This choice is based on a previous study of vacancy and interstitial clusters in copper [10,11]. Both potentials were modified at short distance by fitting to a screened Coulomb potential following the Biersack formulation [12]. The LRPP describes a slightly bigger effective size of ion, which leads to a higher value of average threshold displacement energy, E_d, of 42 eV in comparison with 28 eV for the SRMBP. Because of the lack of space, the results obtained only with the LRPP will be reported. However comments will be made on the effect of the interatomic potential on the results obtained.

Finally, it should be noted that no treatment of electron–phonon coupling was applied, an approximation considered to be appropriate in the case of copper. Also, no heat was extracted from the crystallite during cascade evolution. Monitoring of temperature and its distribution in the crystallite showed that after cascade relaxation the mean temperature increased by a maximum of 80 K for the ranges of energy and crystallite size chosen. This increase does not affect the final result within the time of simulation.

3. Results and discussion

3.1. Defect production

Results on the number, N_F, of surviving SIAs (and an equal number of surviving vacancies) versus PKA energy, E_{PKA}, are presented in Fig. 1. All data obtained for N_F are presented, together with the mean value and error bars (estimated from the standard deviation analysis), and the total number of cascades simulated for each energy at 100 and 600 K are indicated in brackets. The variation in N_F is large, and the ratio of the maximum-to-minimum values for N_F for a given condition reaches a value of ~10. The standard error of the mean varies from 22% to 50%. The statistical properties are better at higher temperature possibly because of lower channelling. The averaged data were fitted to the commonly accepted formulation $N_F = A(E_{PKA})^m$ [1] and the following parameters were obtained for the LRPP: $A = 1.34 \pm 0.22$ and $m = 1.02 \pm 0.06$ with E_{PKA} in keV. The exponent is larger than that reported earlier [1] due to an increase in the number of surviving defects at $E_P = 20$ keV. Note that a similar phenomenon was reported for Fe [13].

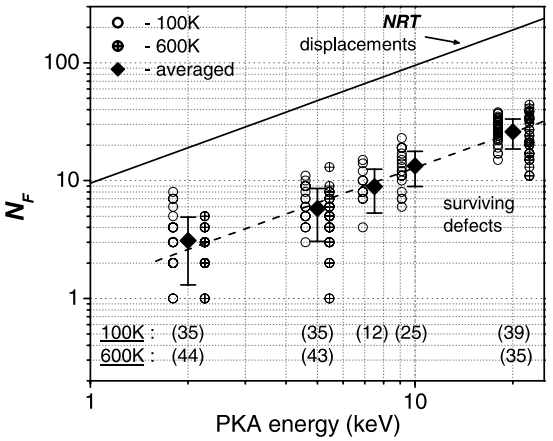

Fig. 1. Total number of surviving defects (vacancies or interstitials) versus PKA energy. All data obtained with the Cu LRPP at 100 and 600 K are presented together with the number of cascades simulated. The solid line presents NRT estimations of displacements for the average threshold energy $E_{th} = 40$ eV.

In general the defect production efficiency is found to be only ~16% of the number of NRT displacements both at 100 and 600 K for the PKA energies in the range of 2–20 keV. A similar statistical analysis has been made of the results obtained with the SRMBP showing the production efficiency to be ~27% of NRT (with the parameters $A = 1.31 \pm 0.30$ and $m = 0.99 \pm 0.07$). The difference in cascade efficiency can be explained by higher values of average threshold displacement energy, point defect formation energy and melting temperature in copper modelled by the LRPP. The low value of cascade efficiency in both models is consistent with experimental results reported for copper (see e.g. [14,15] and discussion at the end of this section) underlying the important role of the extensive intracascade recombination.

3.2. Intracascade clustering

Intensive cascade clustering was observed at all energies and temperatures. The averaged number of clusters containing $\geqslant 3$ defects (either SIAs vacancies) per cascade is presented as a function of the PKA energy in Fig. 2. It increases with increasing PKA energy for the both temperatures and is lower at the higher temperature. It should be noted that at a given PKA energy the number of SIA clusters per cascade is larger than that of vacancy clusters in general. For example at 20 keV each cascade yielded in general one vacancy and two SIA clusters. The average number of defects in clusters of size $\geqslant 3$ defects versus PKA energy is presented in Fig. 3, where it can be seen that the cluster size increases with increasing energy and this effect is stronger for SIA clusters. The effect of temperature is not very pronounced at low energy and cannot be estimated for intermediate energies due to the lack of data. However a significant increase, by a factor almost 2, in size of both types of clusters can be seen at the highest energy studied. It should be mentioned that the number of cascades simulated at the lowest and highest energy is large (see Fig. 1) and so the temperature effects for both the number of clusters and clusters size are statistically significant.

Both energy and temperature effects can be clearly seen in Fig. 4 for the total fraction of vacancies and interstitials formed in clusters of $\geqslant 3$ defects. The fraction of SIAs, ε_i, is higher than that of vacancies, ε_v, and the difference $(\varepsilon_i - \varepsilon_v)$ increases with PKA energy and temperature. The temperature effect on the difference is rather significant due to the opposing behaviour of ε_i and ε_v with temperature. This difference is the origin of the phenomenon known as 'production bias' and has been shown to explain the recoil-energy-dependent damage accumulation [5]. It is interesting to note that the temperature effect is even stronger for larger clusters. This can be seen in Fig. 5 where we present ε_i and ε_v for clusters having $\geqslant 6$ defects. Only three energies are presented due to the absence of such clusters at 2 keV and the lack of data for 600 K at 10 keV. Nevertheless, the increase in both fractions ε_i and ε_v with increasing energy and temperature can be assessed.

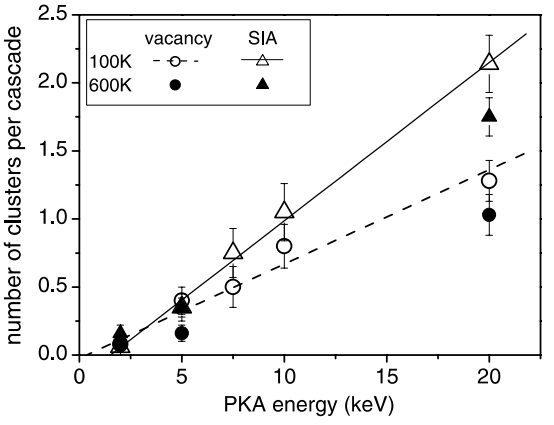

Fig. 2. The average per cascade number of vacancy and interstitial clusters having $\geqslant 3$ defects versus PKA energy.

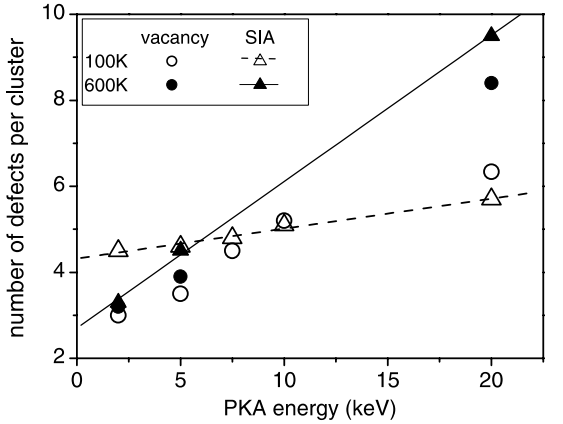

Fig. 3. The average number of defects in vacancy and interstitial clusters ($\geqslant 3$ defects) versus PKA energy.

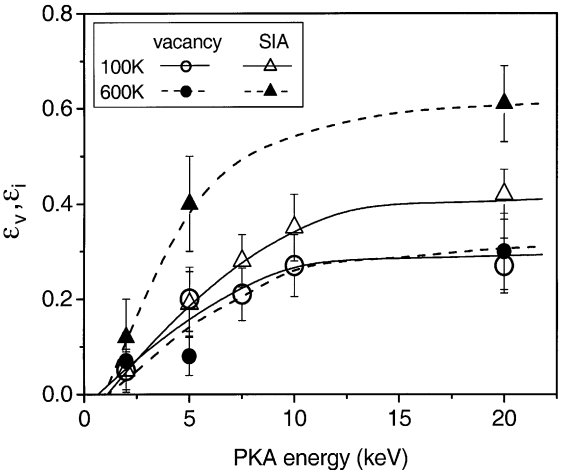

Fig. 4. The fraction of vacancies and interstitials surviving in clusters having $\geqslant 3$ defects versus PKA energy.

3.3. Cluster structure

The full statistics on cluster structure has not yet been estimated, but a wide variety of different configurations has been found. Thus, among SIA clusters we have observed clusters of up to 67 SIAs with the structure of perfect edge glissile dislocation loops with Burgers

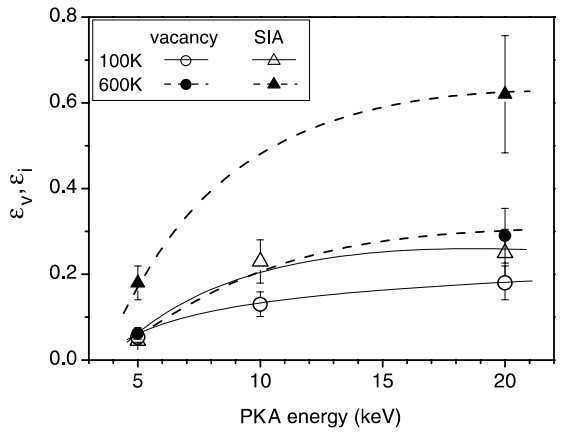

Fig. 5. The fraction of vacancies and interstitials surviving in clusters having $\geqslant 6$ defects versus PKA energy.

vector $\underline{b} = 1/2\langle 1\,1\,0\rangle$, faulted clusters of up to 38 SIAs with the structure of faulted sessile Frank loops with $\underline{b} = 1/3\langle 1\,1\,1\rangle$ and a variety of three-dimensional (3-D) clusters which are either combinations of differently oriented $\langle 1\,0\,0\rangle$ dumbbells or clusters forming intersecting stacking faults on inclined $\{1\,1\,1\}$ planes. In general more than about 60% of all primary interstitial clusters produced in cascades have the glissile configuration. Among vacancy clusters, the most frequent was a 3-D configuration consisting of two to four inclined and intersecting $\{1\,1\,1\}$ stacking faults. Some of these can be identified as regular or truncated stacking fault tetrahedra (SFTs) while others are partly dissociated vacancy dislocation loops. Small 3-D clusters of vacancies were also observed and some of them have the form of regular polyhedra (e.g. 5–6 vacancies). No large planar clusters which could be interpreted as vacancy dislocation loops were observed. More than about 30% of vacancy clusters contain $\{1\,1\,1\}$ stacking faults and the majority of these start to transform into SFT-like configurations during further (up to 250 ps) ageing beyond the thermal spike stage. In general, SFT-like configurations were more numerous at higher temperature. Some configurations of SIA and vacancy clusters are presented in [16].

3.4. Comparison of ε_i and ε_v with the values obtained from the analysis of experimental results in terms of PBM

The production bias model (PBM) explicitly assumes that there is imbalance in the intracascade production of clusters of SIAs and vacancies. This implies that the damage accumulation under cascade damage conditions must be sensitive to the clustering efficiency of SIAs. In order to test this hypothesis, the effect of recoil energy on damage accumulation in copper has been determined experimentally [5] and subsequently rationalized theoretically in terms of the PBM [15]. The damage data observed experimentally in the form of defect cluster density, density of SFTs and voids, void size distributions and void swelling have been explained quantitatively in terms of the PBM. In order to explain these results defect production efficiencies of 25 and 10% were assumed for 3 MeV protons (mean PKA energy of $\sim$1 keV) and fission neutrons (mean PKA energy of $>$20 keV), respectively. These values are reasonably consistent with the results of the present simulations for 600 K. Further, the ε_i and ε_v values in these calculations were taken to be 0.12 and 0.006, respectively, for 3 MeV protons and 0.30 and 0.06 for the case of fission neutron irradiations. It should be mentioned that the ε_i refers to clusters containing 6 or more SIAs and ε_v refers to the fraction of vacancies deposited in the visible SFTs, i.e. of size larger than about 1 nm ($\sim$21 vacancies).

Thus, it can be seen that the global values of ε_i and ε_v necessary to explain the experimental results are considerably lower than the average values obtained in the single cascade simulations. It should be noted, however, that the general trend that both ε_i and ε_v increase with increasing recoil energy is fully consistent with the results of the present simulations. The fact that the global values of ε_i and ε_v required to rationalize the microstructure data are smaller than the values obtained in the single cascade simulations is not surprising in view of the fact that during the long time period of irradiation experiments, the evolving microstructural components must experience many kinds of interactions between the mobile and immobile defect clusters. In other words, both SIA and vacancy clusters are likely to decay by absorption at sinks as well as by mutual recombination [17].

4. Concluding remarks

Simulations of displacement cascades in Cu with PKA energy within the range 2–20 keV have been performed, with sufficiently large statistics to conclude the following:

1. Defect production efficiency falls in the range 10–30% of NRT estimates of displacements.
2. The fraction of defects formed in clusters increases with increasing PKA energy and the ambient temperature.
3. The difference in fraction of clustered SIAs and vacancies increases with increasing PKA energy and the ambient temperature.
4. About one half of all defects formed in high energy cascades (e.g. $E_{PKA} = 10$–20 keV) are in clusters.
5. Most interstitial clusters have the form of glissile, perfect dislocation loops.
6. About 30% of vacancy clusters formed in high energy cascades have features of SFTs and dissociated dislocation loops.

These results are qualitatively consistent with experimental observations and results of treatment of experimental data using current models of microstructure evolution. It must be recognized, however, that the results of single cascade MD simulations carried out for $\sim 10^{-12}$–10^{-9} s cannot be used directly in the global treatment of damage accumulation, primarily because they cannot take into account the effect of intercascade reaction kinetics on the evolution of the surviving defect fractions during the long-term ageing of the cascade region. In other words, the results of single cascade simulations need to be calibrated against a combination of analytical calculations and specifically designed experiments.

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References

[1] D.J. Bacon, A.F. Calder, F. Gao, V.G. Kapinos, S.J. Wooding, Nucl. Instrum. and Meth. B 102 (1995) 37.
[2] D.J. Bacon, A.F. Calder, F. Gao, Rad. Eff. Def. Sol. 141 (1997) 283.
[3] D.J. Bacon, F. Gao, Yu.N. Osetsky, Nucl. Instrum. and Meth. B 153 (1999) 87.
[4] D.J. Bacon, F. Gao, Yu.N. Osetsky, J. Comput. Aided Mater. 6 (2000) 255.
[5] B.N. Singh, M. Eldrup, A. Horsewell, P. Ehrhart, F. Dworschak, Philos. Mag. A 80 (2000) 2628.
[6] E. Alonso, M.J. Caturla, T. Diaz de la Rubia, J.M. Perlado, J. Nucl. Mater. 276 (2000) 221.
[7] Yu.N. Osetsky, D.J. Bacon, MRS Symp. Proc. 650 (2001) R4.2.
[8] G.J. Ackland, G. Tichy, V. Vitek, M.W. Finnis, Philos. Mag. A 56 (1987) 735.
[9] Yu.N. Osetsky, A.G. Mikhin, A. Serra, Philos. Mag. A 72 (1995) 361.
[10] Yu.N. Osetsky, A. Serra, M. Victoria, S.I. Golubov, V. Priego, Philos. Mag. A 79 (1999) 2259, 2284;
Yu.N. Osetsky, A. Serra, M. Victoria, S.I. Golubov, V. Priego, Philos. Mag. A 80 (2000) 2131.
[11] Yu.N. Osetsky, D.J. Bacon, A. Serra, B.N. Singh, S.I. Golubov, J. Nucl. Mater. 276 (2000) 65.
[12] J.P. Biersack, Nucl. Instrum. and Meth. B 27 (1987) 21.
[13] R.E. Stoller, J. Nucl. Mater. 276 (2000) 22.
[14] S. Zinkle, B.N. Singh, J. Nucl. Mater. 199 (1993) 173.
[15] S.I. Golubov, B.N. Singh, H. Trinkaus, Philos. Mag. A 81 (2001) 2533.
[16] Yu.N. Osetsky, D.J. Bacon, B.N. Singh, B. Wirth, these Proceedings.
[17] H. Trinkaus, B.N. Singh, these Proceedings.