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Secondary factors influencing cascade damage formation

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Abstract

Primary cascade damage production in iron has been extensively investigated by molecular dynamics, and the average defect production as a function of cascade energy and temperature is well characterized. However, preliminary results indicate several factors alter 'normal' cascade evolution, leading to quite different defect production behavior. Further investigation of three such factors has been carried out: (1) primary knock-on atom (PKA) direction, (2) nearby free surfaces, and (3) pre-existing effects. Results of the investigation confirm these factors significantly impact damage production. Effects include: enhanced defect survival for PKA directions lying in close-packed {1 1 0} planes, increased point defect clustering and larger defect clusters in cascades initiated near a surface, and reduced defect survival in material containing defects. The origin and implications of these effects are discussed relative to the interpretation of certain experimental observations and parameters used in other modeling studies.

1. Introduction

A detailed understanding of primary damage formation in irradiated materials has been obtained from molecular dynamics (MD) simulations of atomic displacement cascades [1–10], with increased computational capability allowing the development of a statistically meaningful cascade database for iron [1,5]. Mean values have been derived for the number of stable point defects (interstitials and vacancies) created, the number of defects that cluster during the cascade event, and the size distribution of in-cascade defect clusters. Stable defects are those remaining after in-cascade recombination is complete. The NRT number of displacements [11,12] is often used as a normalizing factor for the MD results; this convention is followed here.

Ongoing research indicates several factors can cause cascade evolution (hence defect production) to deviate from the average behavior. Three of these factors are: PKA direction [3,13], presence of a nearby surface [8,9,14], and pre-existing defects in the material [15,16]. In some cases, simulations exhibited substantial differences from typical MD cascades that are carried out in an atomic simulation cell containing perfectly crystalline material. However, in other cases the results were ambiguous, limited in their parameter range, or an insufficient number of simulations were done to establish statistical significance. The quantitative impact of any cascade variable can only be determined by a systematic study with 'enough' events to capture inherent statistical variations [1,5]. The objective of the current investigation is to establish the degree to which these three factors influence cascade evolution and defect formation by carrying out additional simulations and extending the range of the previous work to higher PKA energies and temperatures.

2. Application of MD simulation method

The methods and models used are detailed in previous publications [1–5,13,14]. Briefly, the MD simulations employed the code MOLDY [17] and a modified version of the Finnis–Sinclair potential [3,18]. Periodic boundary conditions were imposed on a constant pressure ensemble of atoms, with the simulation cell size

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containing from 16 000 atoms for the low-energy investigations of PKA direction effects, up to 250 000 atoms for the 10 and 20 keV simulations employed to evaluate the impact of free surfaces and pre-existing damage. The iron cascade database mentioned above served as the basis for comparison with the new simulations [1,5].

3. Results of MD simulations

Only a brief summary of the new results can be included here; additional details will be published elsewhere.

3.1. Influence of PKA direction

A dramatic effect of PKA direction at low cascade energies was reported previously [1,13]. In order to avoid lattice effects such as channeling, MD cascade simulations are typically carried out using a high index PKA direction. For example, most of the simulations discussed in Ref. [1] were generated using a [1 3 5] PKA direction. A preliminary evaluation of PKA direction effects using 1 keV cascades indicated that mean values obtained with [1 3 5] PKA should be representative of the average behavior at this energy [4]. However, analysis of 300 eV simulations found that stable defect production was significantly increased for PKA directions lying in close-packed {1 1 0} planes. Some cascades initiated using a [1 1 4] direction developed completely within a single {1 1 0} plane. This planar channeling resulted in increased separation of vacancies and interstitials and therefore less in-cascade recombination.

This effect was not expected to persist for higher PKA energies because the higher energy recoils would be more likely to break out of the {1 1 0} plane. However, it was surprisingly observed that increased defect survival was detected at energies as high as 2 keV for [1 1 4] PKA at 100 K. The ratio of defect survival using [1 1 4] to that using [1 3 5] was 1.59, 1.12, 1.28, 1.15, and 0.995 for cascade energies of 0.3, 0.5, 1.0, 2.0, and 5.0 keV, respectively [13]. In order to obtain further information on the potential for this phenomenon to influence defect production, additional simulations have been carried out at 600 and 900 K to see if increased atomic thermal motion would act to minimize the planar channeling.

Some of the results of 600 and 900 K simulations are shown in Fig. 1(a) and (b), respectively. This figure compares stable defect formation as a function of energy for the [1 1 4] and [1 3 5] PKA directions. The error bars shown are the standard error of the mean. The error bars are somewhat larger than for the 100 K results [1,13] because the number of cascades at each condition is smaller. However, it appears that there is no systematic deviation between the two PKA directions at either temperature. Even at 300 eV, where the effect was strongest in 100 K cascades, the impact is small at 600 and 900 K.

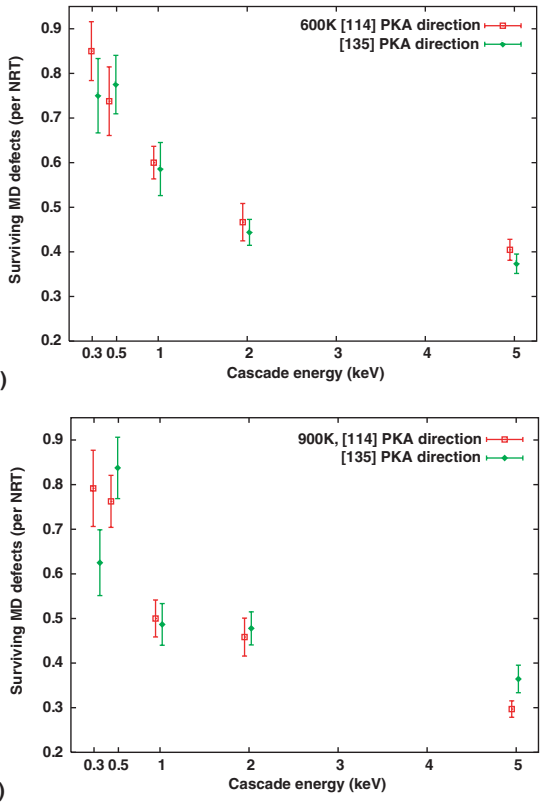

Fig. 1. Energy dependence of the PKA direction effect on stable point defect formation at (a) 600 and (b) 900 K.

3.2. Influence of free surfaces

The rationale for investigating the impact of free surfaces on cascade evolution was described previously, and results obtained from 10 keV cascades were presented [14]. The goal is understanding the reduced yield of visible defects from MD cascades simulations compared to thin foil ion irradiation experiments carried out in a transmission electron microscope [19–21]. The 10 keV simulations exhibited a significant effect of the free surface on both total stable defect production and in-cascade clustering [14]. However, the largest vacancy and interstitial clusters formed were too small to be observed by TEM.

Additional simulations have been carried out using 20 keV PKA, an energy where point defect clustering is increased and larger clusters have been observed in the database. The new simulations employed the method discussed in Ref. [14], with a free surface created by removing five layers of atoms from one face of a 250 000 atom cell. Periodic boundary conditions are otherwise

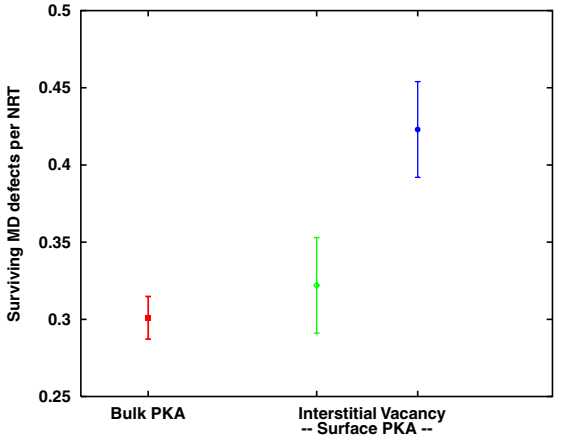

Fig. 2. Average stable defect production (per NRT) in 20 keV cascades at 100 K for cascades initiated by a PKA near the center (bulk) and at the free surface of the simulation cell.

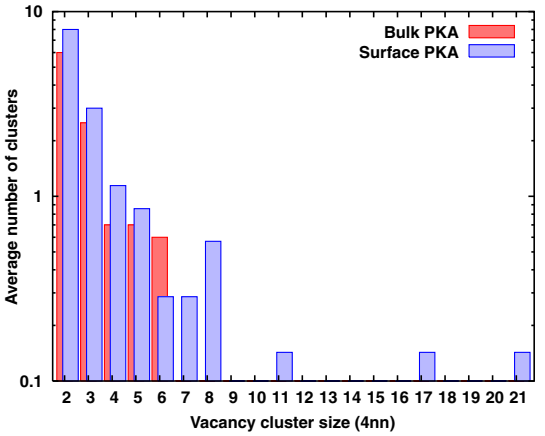

Fig. 3. Comparison of in-cascade vacancy cluster size distribution in 20 keV, 100 K cascades initiated by a PKA near the center (bulk) and at the free surface of the simulation cell.

imposed. Eight simulations were carried out at 100 K in which all the PKAs were surface atoms. Several PKA directions were used, with each of these directions slightly more than 10° off the [0 0 1] surface normal. The results of these simulations can be compared with 10 'bulk' cascades (PKA initiated near the simulation cell center) conducted previously.

The number of surviving point defects (normalized to NRT displacements) is shown in Fig. 2 for both bulk and surface cascades, with error bars indicating the standard error of the mean. The results are similar to those observed at 10 keV. Stable interstitial production in surface cascades is not significantly different than in bulk cascades; the mean value was slightly higher for the 20 keV surface cascades, whereas it was slightly lower for the 10 keV case. However, there is a substantial increase in the number of stable vacancies produced, and the change is clearly significant. The number of surviving interstitials and vacancies is no longer equal for cascades initiated at the surface because interstitials can be lost by sputtering or the diffusion of interstitials and small glissile interstitial clusters to the surface. Reducing the number of interstitials leads to a greater number of vacancies surviving since less recombination can occur.

In-cascade clustering at 20 keV was also similar to the 10 keV simulations, i.e. no significant change in the fraction of interstitials in clusters (~ 0.18/NRT) while the in-cascade vacancy fraction (based on a fourth-nearest-neighbor criterion [4]) increased from ~ 0.06/NRT to 0.25/NRT. Moreover, the vacancy cluster size distribution changed dramatically, with larger clusters produced at 20 keV. The size distributions obtained from 20 keV bulk and surface cascades are shown in Fig. 3. The largest vacancy cluster observed in the bulk cascades contained only six vacancies, while the surface cascades had clusters as large as 21 vacancies. This latter size is near the limit of visibility in TEM, with a diameter of almost 15 nm.

3.3. Influence of pre-existing damage

Cascade simulations were carried out at 10 keV and 100 K to expand the range of previous work carried out using 1 keV simulations in copper [15] and 0.40, 2.0, and 5.0 keV simulations in iron [16]. A 10 keV cascade energy is high enough to initiate in-cascade clustering, is near the plateau region of the defect survival curve, and involves a limited degree of subcascade formation. For these conditions, the database contains two independent sets of cascades, 7 in a 128k atom cell and 8 in a 250k atom cell that provide a basis of comparison. A cell size

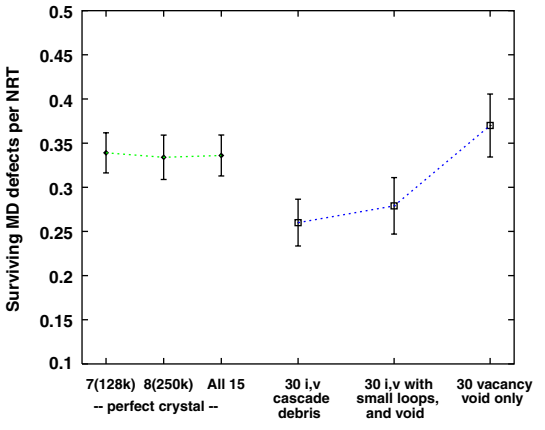

Fig. 4. Comparison of defect survival values for cascades in perfect crystal and material with pre-existing defects.

Table 1
Summary of defect production results from cascades with pre-existing damage

	Survival fraction (per NRT)	Standard error	Interstitial cluster fraction (per NRT)	Standard error
Perfect crystal (128k and 250k atoms)	0.336	0.0137	0.170	0.0155
Defective crystal				
30 i,v: cascade debris, with 1 di- and 1 7-interstitial cluster	0.260	0.0214	0.179	0.0119
30 i,v: cascade debris with 4 di-, 1 tri-, and 1 8-interstitial cluster; 6-vacancy void, 9-vacancy loop	0.279	0.0258	0.110	0.0191
30-vacancy void only	0.370	0.0288	0.190	0.0188

of 250k atoms was used for the cascade simulations with pre-existing damage.

Three types of pre-existing damage were investigated. The first was the as-quenched debris from a 10 keV cascade in perfect crystal. A total of 30 vacancies and interstitials were present, including one di-interstitial and one 7-interstitial cluster. The second case was similar, but the 30 vacancies included a 6-vacancy void and a 9-vacancy loop, and the interstitial clusters included four di-, one tri-, and one 8-interstitial cluster. The third configuration contained only a single 30-vacancy void. Eight simulations were carried out with different initial PKAs and a $\langle 1\,3\,5\rangle$ PKA direction. The same set of PKAs was used for all three defect configurations.

The influence of pre-existing damage on stable defect formation in 10 keV cascades is shown in Fig. 4, where results from the three defect configurations are compared with those obtained in perfect crystal. The variation between two sets of perfect crystal simulations is provided for comparison. Both defect survival and interstitial clustering results are summarized in Table 1. As expected, substantial variation was observed between the different simulations for any given pre-existing defect configuration, but a significant reduction in the average defect formation was observed for the two configurations most typical of cascade debris. A slight increase (that may not be statistically significant) in defect production was observed when the cell contained only a small void. Only the second defect configuration lead to a significant change in interstitial clustering.

4. Discussion and conclusions

A fairly complete picture of displacement cascade evolution in iron has been obtained through extensive simulation studies. The work reported here illustrates the impact of three cascade conditions that lead to deviations from the average defect production behavior. These are: PKA direction effects, the influence of free surfaces, and the impact of pre-existing defects or cascade debris. In each case, the results reported here extend the range of previous work by the current authors and others.

A planar channeling effect was observed to influence low energy cascades (up to 2 keV) at 100 K for PKA directions that lie in the close-packed $\{1\,1\,0\}$ planes in bcc iron. This phenomenon has now been investigated in MD simulations up to 5 keV at temperatures up to 900 K. Although dramatic differences are observed between cascades that exhibit planar channeling and 'normal,' more isotropic cascades, the overall effect of PKA direction on average defect formation in any practical sense appears to be limited within the range of cascade parameters examined. The maximum energy where an effect was seen at 100 K was 2 keV, and at 600 and 900 K essentially no effect was observed for any PKA energy.

In the case of surface-influenced defect formation, a sufficient number of 20 keV iron cascades were completed to statistically evaluate variations between these and bulk cascades. Relative to cascades initiated far from the free surface, stable vacancy defect production increased for surface-influenced cascades, while the number of surviving interstitials was nearly unchanged. The difference between the number of surviving vacancies and interstitials arises from sputtering and surface absorption of mobile interstitial defects. The fraction of vacancies contained in clusters increased substantially for cascades initiated at the surface, and larger vacancy clusters were formed. The impact on defect clustering increases with cascade energy, with the largest vacancy clusters from the 20 keV cascades approaching TEM visibility. These results imply that cascade defect production in bulk material is different from that observed in situ using transmission electron microscopy.

Although the approach in this investigation of pre-existing damage was slightly different, the results are consistent with the previous studies by English and coworkers [15] and Gao et al. [16]. They observed substantial reductions in defect production when a cascade was initiated in material containing defects. The reductions in defect production observed in this study (Fig. 4

and Table 1) are somewhat smaller. This difference may partially be due to the higher cascade energy employed here (10 keV vs. 0.4–5 keV); but the statistical nature of cascade evolution is also a factor. Gao et al. analyzed the results of several simulations as a function of distance between the center of mass (COM) of the new cascade and that of the pre-existing damage. A good correlation was found between this spacing and the number of defects produced. In this work, the distance between PKA location and the pre-existing damage was nearly constant. Since the morphology of each cascade was quite different, the COM spacings varied. The average behavior for a fixed initial separation can not be directly compared to Gao's results. Because of the reduced defect survival observed in defective material, these results and the earlier work suggest that it may be appropriate to develop a fluence-dependent cascade survival efficiency for use in kinetic radiation damage models.

Acknowledgements

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