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The effect of free surfaces on cascade damage production in iron

Roger E. Stoller *

Metals and Ceramics Division, Oak Ridge National Laboratory, Building 5500, P.O. Box 2008, Oak Ridge, TN 37831-6376, USA

Abstract

The first results have been obtained from a study intended to provide a quantitative measure of the effect of free surfaces on cascade damage evolution. A sufficient number of 10 keV iron cascades were completed to statistically evaluate variations between surface-influenced and bulk cascades. Two sets of near-surface cascades were completed; surface atoms served as the primary knockon atom (PKA) in the first set, while the PKA were located 10 lattice parameters below the surface in the second. Relative to bulk cascades, stable point defect production increased for cascades $10a_0$ below the surface. Stable vacancy production increased further for cascades initiated at the surface, while the number of surviving interstitials decreased. The difference between the surviving vacancies and interstitials arises from sputtering and surface absorption of mobile interstitial defects. The fraction of vacancies contained in clusters increased as the cascade initiation site approached the surface, and larger vacancy clusters were formed. Conversely, no significant change in the in-cascade interstitial clustering fraction or the interstitial cluster size distribution was observed.

1. Introduction

Our understanding of primary defect formation in irradiated materials has advanced as increased computational ability has permitted simulation methods such as molecular dynamics (MD) to explore larger atomic systems [1–9]. Although the results of these simulations cannot be directly confirmed by experimental measurements, they are broadly consistent with relevant data, including low-temperature electrical resistivity measurements and diffuse X-ray scattering studies. Perhaps the most influential body of experimental data on primary damage formation is that provided by experiments in which thin foils are irradiated by high-energy electrons and/or heavy ions [10–18]. In most cases, the experimental observations are carried out in situ by transmission electron microscopy. The results of MD simulations are also in general agreement with the data from these in situ irradiation experiments. For example, some material-to-material differences observed in the MD simulations, such as differences in in-cascade clustering between bcc iron and fcc copper, also appear in the experimental data [2,5,6]. However, the yield of large point defect clusters in the simulations is lower than would be expected from the thin foil irradiations, particularly for vacancy clusters. It is desirable to investigate the source of this difference because of the influence this data has on our understanding of cascade damage formation.

Previous theoretical [8,9] and experimental work [17,18] indicates that the presence of a nearby free surface can influence primary damage formation. For example, interesting effects of foil thickness have been observed in some experiments [17]. Unlike cascades in the bulk, which produce vacancies and interstitials in equal numbers, the number of surviving vacancies in surface-influenced cascades can exceed the number of interstitials. This could lead to the formation of larger vacancy clusters and account the differences in visible defect yield between the results of MD cascade simulations

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* Tel.: +1-865 576 788; fax: +1-865 574 0641.
E-mail address: rkn@ornl.gov (R.E. Stoller).

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conducted in bulk material and the thin-film, in situ experiments. The work reported in Refs. [8,9] has demonstrated the kinds of effects that can occur, but the magnitude of the effect has not been quantified in detail. Since displacement cascades are stochastic events, the quantitative impact of the free surface can only be determined by a systematic study with 'enough' events to capture inherent statistical variations in their behavior.

The results of an initial investigation intended to obtain a quantitative measure of the effect of free surfaces on cascade damage evolution are reported below. The work has focused on iron because the most extensive bulk cascade database exists for this material, and because the effect of free surfaces has not yet been evaluated in bcc materials. Currently, only 10 keV cascades at a temperature of 100 K have been completed in sufficient numbers to examine the statistical variations between surface-influenced and bulk cascades. The long-term objective of the investigation is to determine whether thin foil irradiations provide reliable estimates of fundamental radiation damage parameters such as total point defect production, the fraction of point defects in clusters, and the size of the in-cascade clusters.

2. MD method and computational approach

A substantial database of atomic displacement cascades in iron has been developed [1–4] using the MD code MOLDY [19] and a modified version of the Finnis–Sinclair potential [20,21]. The database covers cascade energies from 0.1 to 100 keV and temperatures from 100 to 900 K. This database provides an excellent basis for evaluating the effect of free surfaces. A cascade energy of 10 keV and a temperature of 100 K was chosen for this initial study. For these conditions, the database contains two independent sets of cascades, seven in a 128 000 atom cell and eight in a 250 000 atom cell. An energy of 10 keV is high enough for some in-cascade clustering to occur, is near the plateau region of the defect survival curve, and initiates a limited degree of subcascade formation. In addition, the required size of the simulation cell, 250 000 atoms, is relatively small. This permits multiple cascades to be carried out in a reasonable timeframe.

The new simulations were carried out using the same MD code and interatomic potential mentioned above. A free surface was created by removing five layers of atoms from one surface of a $(50a_o)^3$ atom cell, containing 250 000 atom sites. Atoms with sufficient kinetic energy to be ejected from the free surface (sputtered) are frozen in place just above the surface. Periodic boundary conditions are otherwise imposed. Two sets of nine simulations were carried out to evaluate the effect of the free surface on cascade evolution. In one case, all the primary knockon atoms (PKAs) selected were surface atoms, and in the other the PKA were chosen from the atom layer $10a_o$ below the free surface. 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 the two sets of 'bulk' cascades conducted previously in which cascades were initiated near the center of simulation cell.

3. Results of MD simulations

Fig. 1 provides a representative example of a cascade initiated at the free surface. The peak damage state at $\simeq$1.1 ps is shown in (a) with the final damage state at $\simeq$15 ps shown in (b). The large number of apparent vacancies and interstitials in Fig. 1(a) is due to the pressure wave from the cascade reaching the free surface. With the constraining force of the missing atoms removed, this pressure wave is able to displace the near-surface atoms by more than $0.3a_o$, which is the criterion used to choose atom locations to be displayed. A similar pressure wave occurs in bulk cascades, making the maximum number of displaced atoms much greater than the final number of displacements. Most of these displacements are short-lived, as shown in Fig. 2, in which the time dependence of the defect population is shown for three typical bulk cascades, one surface-initiated cascade, and one cascade initiated $10a_o$ below the surface. The effect of the pressure wave persists longer in surface-influenced cascades, and may contribute to stable defect formation.

The final displaced atom and vacancy positions obtained from each cascade were analyzed to determine the number of surviving point defects, the fraction of the point defects of both types contained in clusters, and the cluster size distributions. When compared to the bulk cascade database, several differences were observed. In Fig. 3, number of surviving point defects has been normalized to the number of displacements calculated using the NRT standard [22]. The error bars represent the standard error of the mean value for each population, indicating which differences are statistically significant. Results for two independent sets of 10 keV bulk cascades are shown separately and as a combined data set. For a similar number (9) of cascades, the larger standard errors indicate greater dispersion for the surface-influenced cascades.

As shown in Fig. 3, the number of stable defects increased for cascades initiated $10a_o$ below the surface. In this case, no atom sputtering was observed and the number of stable vacancies and interstitials was equal. This increase apparently arises from an effect of the pressure wave on in-cascade recombination in one of two ways. Either the final separation between vacancies and interstitials is somewhat greater in the surface-

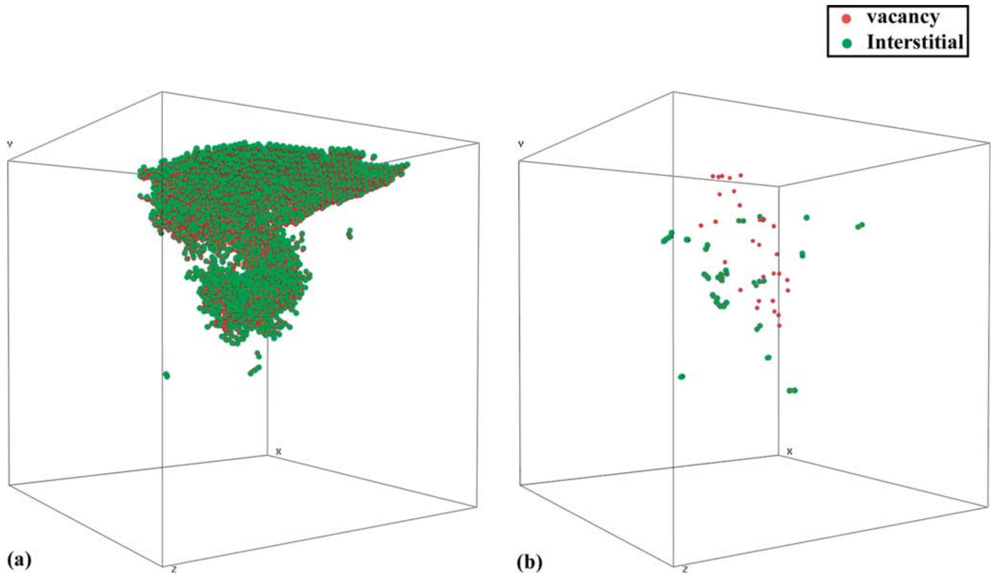

Fig. 1. Typical 10 keV cascade with surface atom PKA; peak damage state is shown at ≃1.1 ps in (a) and final damage state in (b) at ≃15 ps.

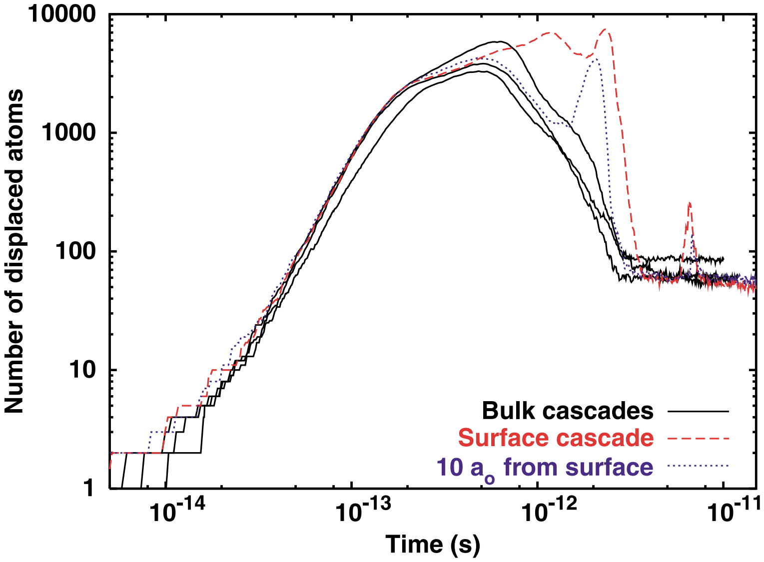

Fig. 2. Number of displaced atoms as a function of time for three typical 10 keV bulk cascades, cascades and cascades initiated at the free surface and $10a_o$ below the free surface.

influenced cascades, or the surface relaxation leads to a smaller effective recombination radius. In the case of cascades initiated at the surface, the number of surviving interstitials and vacancies is no longer equal. The number of vacancies continues to increase while the number of interstitials decreases. Interstitials are lost by two mechanisms; atoms are sputtered from the free surface and a few interstitials and small glissile interstitial clusters are absorbed by the surface. Sputtering can be detected by viewing cascade animations, but the relative contribution of these two mechanisms has not yet been determined. Reducing the number of interstitials leads to a greater number of vacancies surviving since less recombination occurs.

In-cascade clustering of vacancies and interstitials from surface-initiated and bulk cascades is compared in

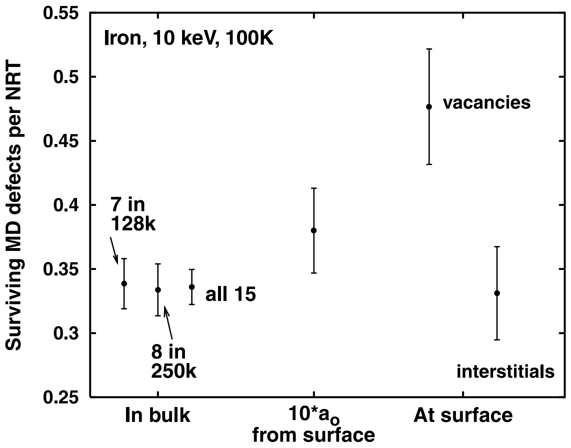

Fig. 3. Average stable defect production in 10 keV cascades: two sets of bulk cascades, cascades initiated $10a_0$ below the free surface, and cascades initiated at the free surface.

Figs. 4–6. The fraction of the total defects contained in clusters is shown in Fig. 4, and the cluster size distributions are shown in Figs. 5 and 6. The comparison of cluster size distributions is limited to the bulk and surface-initiated cascades to save space. As discussed previously [3,4], point defect clustering exhibits greater cascade-to-cascade variation than does total defect production (e.g. note the larger standard errors in Fig. 4 relative to Fig. 3). Thus, no clear statistically significant effect can be observed for interstitial clustering in Fig. 4(a). In contrast, the results shown in Fig. 4(b) indicate a continuous increase in vacancy clustering as the cascade initiation site approaches the free surface. The vacancy clustering results are shown for both a second (2nn) and fourth-nearest-neighbor (4nn) clustering criterion as discussed elsewhere [3,4]. The comparison of interstitial cluster size distributions shown in Fig. 5 indicates a

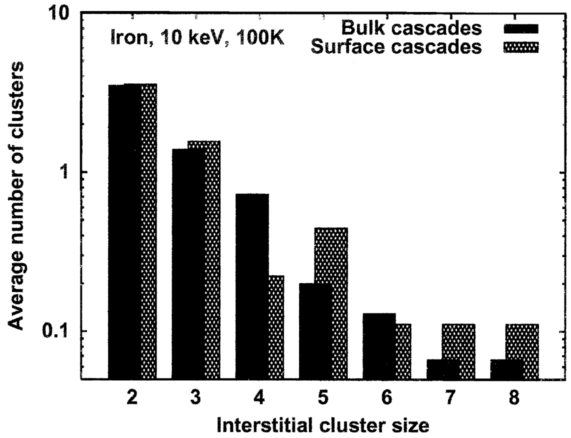

Fig. 5. Comparison of in-cascade interstitial cluster size distribution in 10 keV cascades initiated by a PKA near the center (bulk) and at the free surface of the simulation cell.

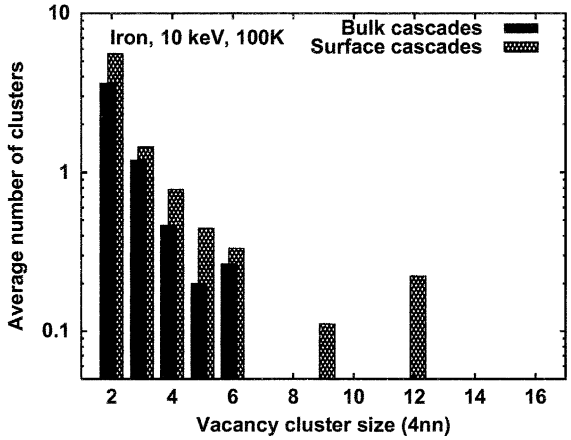

Fig. 6. Comparison of in-cascade vacancy cluster size distribution in 10 keV cascades initiated by a PKA near the center (bulk) and at the free surface of the simulation cell.

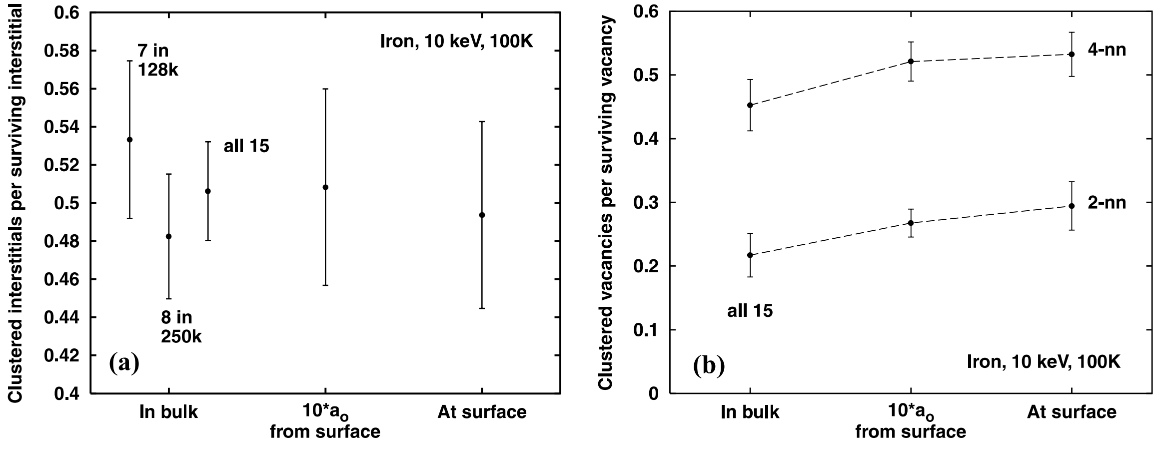

Fig. 4. In-cascade point defect clustering at 10 keV for bulk cascades, cascades initiated $10a_0$ below the free surface, and cascades initiated at the free surface. Interstitial clustering is shown in (a) and vacancy clustering in (b).

tendency for the interstitial clusters to be larger for surface-initiated cascades. However, the change is modest and does not appear to be statistically significant for the limited number of cascades in the database. The change in the vacancy cluster size distribution shown in Fig. 6 is significant, with larger vacancy clusters observed for the surface-initiated cascades.

4. Discussion and conclusions

An initial comparison of MD cascade simulations conducted at various distances from a free surface demonstrates a systematic influence of the surface on primary damage formation in iron. The results were obtained from two series of 10 keV cascade simulations conducted at 100 K, with cascades initiated at the free surface of the simulation cell in one series and 10 lattice parameters below the surface in the second. Average defect production values from the near-surface cascades were compared with those in a database of bulk cascades. The results can be summarized as follows:

1. Stable vacancy production increases as the cascade initiation site approaches the surface.
2. Stable interstitial production increases and then decreases as the cascade initiation site approaches the surface.
3. For cascades initiated very near the surface, the number of stable vacancies exceeds the number of interstitials due to atom sputtering and the glide of interstitial defects to the surface.
4. The fraction of vacancies contained in clusters increases and cluster sizes increase for near-surface cascades.
5. No significant change is observed for in-cascade interstitial clustering in near-surface cascades.

None of the in-cascade clusters obtained in these simulations would be large enough to be visible in the transmission electron microscope. Thus, the results are trivially consistent with the very low defect yield ($\simeq$0.001) observed experimentally, and the postulates that either cascade overlap [11,14] or very high damage energies [10,15] are required to obtain visible defects in iron. However, based on the increased tendency for vacancy clustering, the results obtained to date from 10 keV simulations are consistent with the expectation that the defect yield would be higher for in situ, thin foil irradiation experiments than for irradiation of bulk materials. For example, no vacancy clusters containing more than six vacancies (4nn) were observed in 15 bulk cascades, while the nine surface cascades yielded two 12-vacancy clusters (4nn). Therefore, further work is required to obtain the desired MD-based estimates for visible defect yield. This work will focus on both higher energy simulations and higher temperatures; the conditions for which larger in-cascade clusters are formed in bulk cascades [4]. Somewhat larger numbers of simulations are also required to improve the statistics since near-surface cascades seem to exhibit more variability than bulk cascades. This is particularly important for the defect clustering parameters.

Finally, two additional aspects of the simulations should be mentioned in the context of comparing experiments with MD simulations. First is the adequacy of the interatomic potential used. Although the potential has been fitted to and successfully predicts a broad range of physical parameters, it is only an approximation of iron. For example, as an isotropic potential, it does not account for bonding directionality arising from the iron d-orbitals. The second aspect is related to the first, viz. the fact that the simulations are conducted in a virtual material that is more pure than any found in nature. Solutes and impurities could alter total cascade damage production and the point defect cluster size distributions by trapping some of the mobile defect species.

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