The Effect of Free Surfaces on Cascade Damage Production in Iron - Roger E. Stoller (Oak Ridge National Laboratory)

Extended Abstract

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], leading to the expectation that more direct comparisons can be made between the simulations and experimental results. 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. Unlike cascades in the bulk, which produce vacancies and interstitials in equal numbers, the number of vacancies produced in the 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 conducted in bulk material and the thin-film, *in situ* experiments. The work reported in Refs. [8 and 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.

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 900K. This database provides an excellent basis for evaluating the effect of free surfaces. A cascade energy of 10 keV and a temperature of 100K was chosen for this initial study. For these conditions, the database contains two independent sets of cascades, 7 in a 128,000 atom cell and 8 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 discussed elsewhere [1-4,19-21]. A free surface was created by removing 5 layers of atoms from one surface of a $(50a_0)^3$ atom cell, containing 250,000 atom sites. In the course of the simulations, any atoms passing through free surface 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 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 [001] 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 either $(40a_o)^3$ or $(50a_o)^3$ atom cells.

Figure 1 provides a representative example of a cascade initiated at the free surface. The peak damage state at ~1.1 ps is shown in (a) with the final damage state at ~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 0.3 a_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. In contrast to the bulk cascades, the effect of the pressure wave persists longer in surface-influenced cascades, and may contribute to stable defect formation as discussed below.

The final displaced atom and vacancy positions obtained in 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 Figure 2, 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 values for each population, indicating that the differences observed are statistically significant. The two 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 Figure 2, 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 (or some combination of the two). Either the final separation between vacancies and interstitials is somewhat greater in the 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 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 interstitials are absorbed by the surface. The relative contribution of these two mechanisms in this set of 10 keV cascades has not yet been determined. This reduction in the number of interstitials and leads to a greater number of vacancies surviving since less recombination occurs.

Overall, the results of the current investigation 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

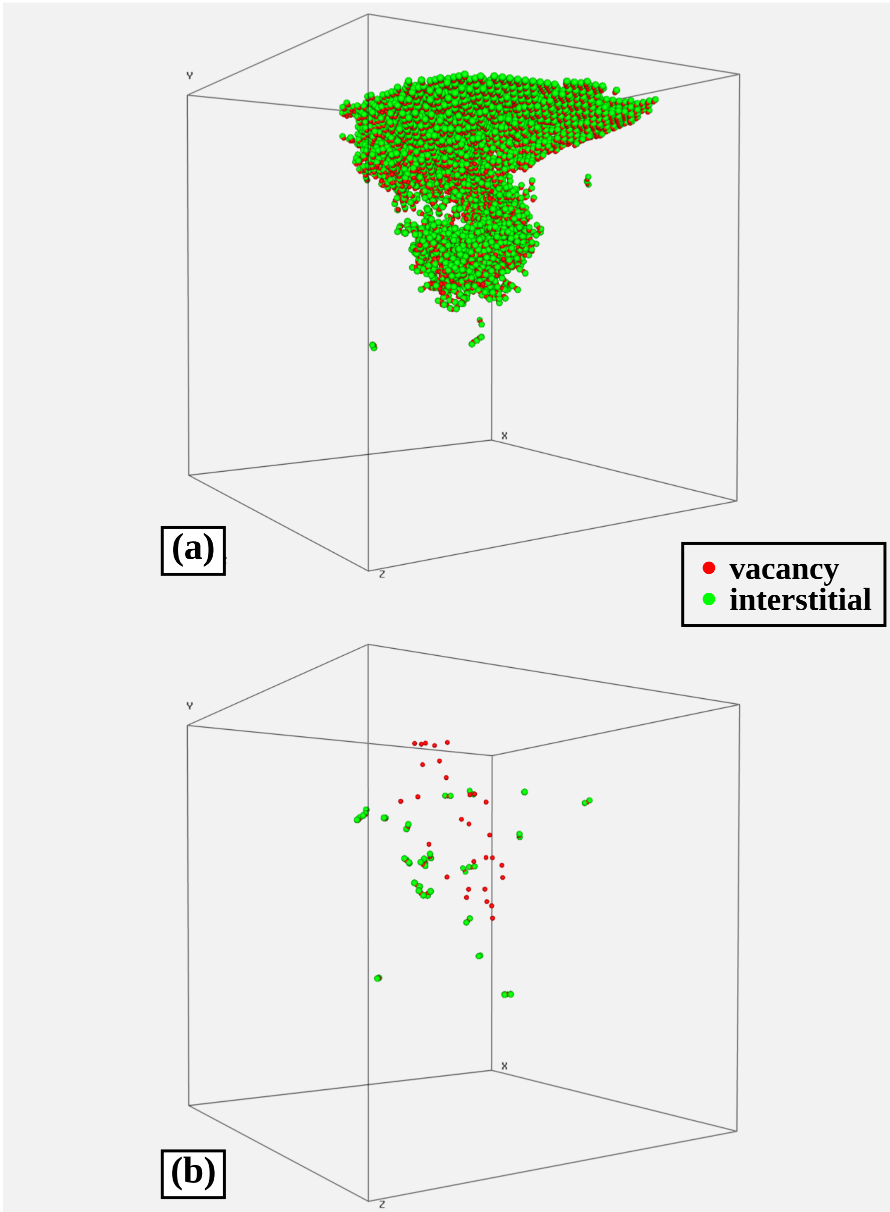

Figure 1. Typical 10 keV cascade with surface atom PKA; peak damage state is shown in (a) at ~ 1.1 ps and final damage state in (b) at ~15 ps.

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 some interstitials 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

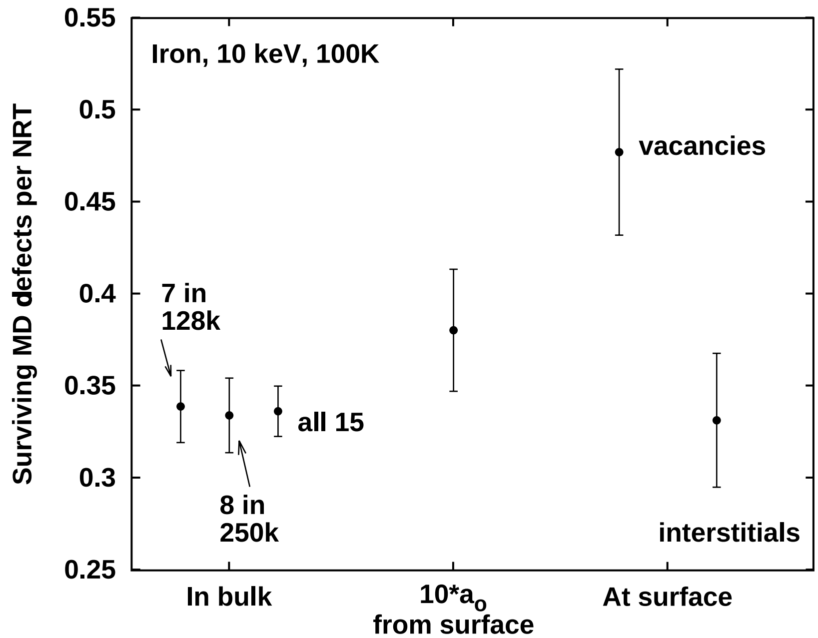

Figure 2. Average stable defect production in 10 keV cascades: two sets of bulk cascades, cascades initiated 10 a_o below the free surface, and cascades initiated at the free surface.

very low defect yield (~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. Therefore, further work is required to obtain the desired MD-based estimates for visible defect yield. The future 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 needed to obtain statistically significant variations in the defect clustering parameters.

Acknowledgments

This research was also sponsored by the Division of Materials Sciences and Engineering, U.S. Department of Energy under contract DE-AC05-00OR22725 with UT-Battelle, LLC.

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(Extended abstract of a paper to be published in the Journal of Nuclear Materials, presented at the 10th International Conference on Fusion Reactor Materials, Baden-Baden, Germany, October 2001)