AN ATOMISTIC STUDY OF HELIUM RESOLUTION IN bcc IRON - Roger E. Stoller[1] and David M. Stewart[1, 2] - [1]*Materials Science and Technology Division, Oak Ridge National Laboratory, Oak Ridge, TN 37831-6138, USA;* [2]*Center for Materials Processing, The University of Tennessee, Knoxville, TN 37996-0750, USA*

Objective

This research is part of a larger effort intended to reveal the fundamental mechanisms by which helium influences microstructural evolution and mechanical properties in in irradiated metals and alloys.

Summary

The evolution of gas-stabilized bubbles in irradiated materials can be a significant factor in the microstructural processes that lead to mechanical property and dimensional changes in structural materials exposed to high-energy neutrons. Helium generation and accumulation is particularly important under DT fusion irradiation conditions. Although the process of ballistic resolutioning of gas from bubbles has been long-discussed in the literature, there have been few computational studies of this mechanism. Resolutioning could limit bubble growth by ejecting gas atoms back into the metal matrix. A detailed atomistic study of ballistic He resolutioning from bubbles in bcc iron has been carried out using molecular dynamics. A newly-developed Fe-He interatomic potential was employed, with the iron matrix described by the potential of Ackland and co-workers from 1997. The primary variables examined were: irradiation temperature (100 and 600K), iron knock-on atom energy (5 and 20 keV), bubble radius (~0.5 and 1.0 nm), and He-to-vacancy ratio in the bubble (0.25, 0.5 and 1.0) in order to obtain an assessment of this dynamic resolutioning mechanism. The results presented here focus on the 5 keV cascades which indicate a modest, but potentially significant level of He removal by this process.

Progress and Status

Introduction

The evolution of gas-stabilized bubbles in irradiated materials is inherently a dynamic process, involving a balance of growth and shrinkage mechanisms. Simplistically, the bubble volume will grow in units of atomic volume by absorbing vacancies and shrink by either emitting vacancies or absorbing interstitials. Addition or emission of gas atoms will also change the volume, with the relative change dependent on the ratio of gas atoms to vacancies in the bubble. The kinetics of bubble evolution due to reactions with point defects and mobile gas atoms has been extensively investigated using various models of cluster dynamics [1-5]. However, the potentially important mechanism of dynamic ejection of gas due to elastic collisions with energetic knock-on atoms has received much less attention, even though it has been discussed in

the literature for many years [6-13]. Such resolutioning could limit bubble evolution by ejecting gas atoms back into the material matrix.

There are two related mechanisms by which gas atoms can be removed from bubbles and returned to the material matrix; these are schematically illustrated in Fig. 1. The so-called track resolutioning mechanism shown in Fig. 1a is primarily discussed in the context of nuclear fuel in which a highly energetic (up to ~100 MeV), heavy fission fragment deposits its kinetic energy over a short mean free path, μ_{FF}. All bubbles atoms that fall within a cylinder defined by this mean free path and an effective track diameter, R, are assumed to be dissolved due to the high energy density in the fission track and all (or some fraction of) the gas atoms contained in these bubbles is returned to the matrix. Ballistic resolutioning, as shown in Fig 1b, is a result of direct collisions between energetic particles and individual gas atoms. The energetic particle could be a fission fragment, a matrix atom recoiling from a collision with a high energy neutron, or (much less frequently) a neutron. Modern computational tools make the ballistic resolutioning mechanism well suited for investigation by molecular dynamics (MD). Recently the effects of ballistic resolutioning of helium from bubbles in UO_2 was examined in an MD study by Parfitt and Grimes [13]. In addition to the two mechanisms just mentioned, they identified a “damage assisted resolution” mechanism in which helium was incorporated into disordered regions of the lattice adjacent to the initial bubble.

Because of the importance of helium generation and accumulation under fusion irradiation conditions, a detailed atomistic study of dynamic He resolutioning from bubbles in bcc iron has been carried out using MD. The primary variables examined were: irradiation temperature (100 and 600K), iron knock-on atom energy (5 and 20 keV), bubble radius (~0.5 and 1.0 nm), and He-to-vacancy ratio in the bubble (0.25, 0.5 and 1.0) in order to obtain an assessment of this dynamic resolutioning mechanism. Varying the He to vacancy ratio is equivalent to varying the pressure in the bubble.

Computational Method and Approach

The basic computational approach employed in this investigation follows that published previously for simulation displacement cascades in iron using the MOLDY MD code [14-16]. A newly-developed Fe-He interatomic potential has been employed [17-19], with the iron matrix described by the potential of Ackland and co-workers from 1997 [20]. Constant pressure, periodic boundary conditions were employed in a cubic simulation cell of 70 or 80 lattice parameters, corresponding to 686k or 1024k atoms. Before initiating a ballistic event, a bubble of the desired size was created by removing the necessary number of adjacent iron atoms in an approximately spherical volume (see Table 1), and the number of helium atoms required to obtain the desired He-to-vacancy ratio (Table 1) were placed within the volume. Then, the atom block was thermally equilibrated at the specified temperature for approximately 25 ps. Consistent with the strong He-He and He-vacancy binding observed in previous work with these potentials, no thermal resolutioning was observed. The equilibrated atom configuration was saved and used as the starting point for the subsequent simulations. The ballistic cascade simulations were initiated by giving one of the Fe atoms located near the bubble a defined amount of kinetic energy and an initial direction oriented toward the bubble. As illustrated in Fig. 2, the distance from this primary knockon atom (PKA) to the bubble boundary was between 2 and 10 lattice parameters. Based on previous experience with the statistical nature of cascade

simulations, between ten and twenty simulations were carried out for each of the conditions listed in Table 1, with variability ensured by the different PKA locations and in some cases by further equilibration of the starting configuration. Since no temperature damping was done, a cell size large enough to minimize PKA-induced heating was used, 250k and 1024k atoms for 5 keV and 20 keV simulations, respectively.

Each MD cascade simulation was continued until variables such as the number of defects, and the average kinetic and potential energy had stabilized for a few ps. The time required to reach this new pseudo-equilibrium depended on the PKA energy and irradiation temperature, varying between about 15 to 25 ps. The final atom configuration was then characterized by locating the He atoms and measuring the distance between any ejected He atoms and the initial bubble, and by counting the number and distribution of the point defects (vacancies and interstitials) created by the cascade. The approach used to identify point defects is described in Refs. 14-16.

Results of Simulations

The results presented here will primarily focus on the 5 keV cascade simulations for which a greater level of characterization has been completed. Further simulations are still in progress to develop a more completely symmetric database than that shown in Table 1, which will permit comparisons at both 100 and 600K, and 5 and 20 keV with a range of bubble sizes and He/vacancy ratios. The bubble sizes chosen for this initial study are typical of those observed at low doses in irradiated materials, and values of the helium-to-vacancy ratio will produce conditions from under- to over-pressurized bubbles [17].

In addition to the He resolution behavior, the impact of the bubbles on defect creation is also of interest. The extensive database on cascades in perfect crystal iron developed using a similar interatomic potential [14-16] provide a basis for comparison. Results for the 5 keV cascades are summarized in Fig. 3, where the number of surviving Frenkel pair and the fraction of the interstitials found in clusters are shown. The data points indicate the mean values for a population of cascades at each condition and the error bars show the standard error about the mean. The open symbols in Fig. 3 represent data taken from the perfect crystal database and the filled symbols are the current results in simulation cells containing bubbles. For these particular conditions, there does not appear to be a systematic effect of the bubbles on primary damage formation. Defect survival is somewhat higher, and the interstitial clustering somewhat lower in the cascades with 0.5 nm bubbles at 100K. The larger error bars for the cascades with bubbles at this temperature indicates that the interaction with the bubbles introduces more variability than in perfect crystal material. In contrast, defect survival is unchanged by 0.5 nm bubbles at 600K while the interstitial clustering fraction is somewhat higher. Note that there appears to be less impact of the bubbles on the cascade statistics at 600K where effect of temperature already introduces greater variability than at the lower temperature. The larger, 1.0 nm bubbles also do not influence defect survival at 600K, but do influence interstitial clustering. Clustering is reduced for when the He/vacancy ratio is 0.5, but increased for a He/vacancy ratio of 1.0 as it was for the 0.5 nm bubbles.

The number of He atoms ejected by the 5 keV cascades, and the average distance (in Fe lattice parameters) is shown in Fig. 4. There is a modest effect of temperature, with fewer atoms ejected at 600K for the 0.5 nm bubbles. Although the data are not shown, a temperature effect was observed in 20 keV cascades with 1.0 nm bubbles. The effect of bubble size and He/vacancy

ratio are much stronger. For a He/vacancy ratio of 1.0, average number of He atoms ejected increases from 0.75 to 2.4 when the bubble size increases from 0.5 to 1.0 nm. Reducing the He/vacancy ratio by a factor of two in the larger bubbles reduces the number of He atoms ejected by more than a factor of two, 2.4 to 1.0. The average distance the He atoms are ejected is also slightly reduced at 600K compared to 100K for the 0.5 nm bubbles, and a strong effect of bubble size is observed at 600K. The average ejection distance increases by about a factor of two from the 1.0 nm bubbles, and does not appear to be significantly influenced by the He/vacancy ratio. The numbers shown in parentheses in Fig. 4 are the number of He atoms ejected by more than 5 lattice parameters. This value was chosen to give a simple measure of the tendency for rather long ejections to occur. It appears most sensitive to He/vacancy ratio. Although the average ejection distance was not affected, the number that were displaced over long distances increased from 7 to 18 when the He/vacancy ratio went from 0.5 to 1.0 in the 5 keV cascades at 600K with 1.0 nm bubbles.

The effect of He/vacancy ratio on He ejection from 1.0 nm bubbles is shown in Fig. 5 for 5 and 20 keV cascades at 600K. There is a systematic increase with He/vacancy ratio (bubble pressure) for both cascade energies, and the degree of He resolutioning is lower for the higher energy.

Discussion and Summary

The simulations completed demonstrate that a number of factors influence ballistic resolutioning of gas atoms from bubbles in a metal matrix. Some of these are consistent with one's intuition, e.g. more helium atoms are displaced from larger bubbles (Fig. 4) and from bubbles that have a higher He/vacancy ratio or bubble pressure (Figs. 4 and 5). The slightly reduced He resolutioning at 600K relative to 100K (Fig. 4) is probably an effect of the increased thermal motion which may reduce the probability of direct, high-energy-transfer collisions. This is similar to the reduction in stable defect formation at 600K shown in Fig. 3 for perfect material [17]. The reduction in He resolutioning for 20 keV cascades compared to 5 keV cascades shown in Fig. 5 is consistent with the lower scattering cross section for higher energy ions.

In the study reported in Ref. [13] of cascade-induced He resolutioning from bubbles in a UO_2 matrix, an additional He resolutioning mechanism (besides the purely ballistic process) was reported. The authors described a lower energy process that they call damage assisted resolutioning which involves He atoms that escape into and are trapped in the regions of heavily damaged crystal near the bubble. Analysis of animated visualization of the cascades in iron did not reveal any significant resolutioning of this type. This may be a result of the differences between a metal and a covalently-bonded oxide matrix, or the difficulty in determining the nature of the resolution mechanism for He atoms that were found within a few lattice parameters of the bubble. Further analysis of the simulations discussed here, as well as additional simulations that are in progress is planned to investigate this difference. As pointed out in Ref. [13], the number of He atoms removed on typical MD timescales (~tens of ps) may be an over-estimate of what would remain in the matrix over longer times. It is likely that He atoms located within a few lattice parameters of the original bubble will be recaptured. This was the motivation for indicating the number of He atoms that were ejected over longer distances in Fig. 4.

Ultimately, the motivation for this work is to provide input to coarser length scale models such as mean field reaction rate theory and Monte Carlo models that can be used to simulate

radiation-induced microstructural evolution over longer times [21]. Initial parametric studies (unpublished) of He bubble evolution using a mean field cluster dynamics model indicated that He resolutioning could have a significant impact on the bubble size distribution. While this preliminary MD study revealed a relatively modest degree of dynamic resolutioning for 5 and 20 keV cascades, a more thorough investigation is required for quantitative assessment and to develop a fundamental understanding of the underlying mechanisms. A change in interatomic potentials could alter the quantitative results presented here. However, the interatomic potential choice is unlikely to have a significant impact on the high-energy ballistic mechanism, although it may influence the rate at which ejected helium is recaptured by a bubble once the system as re-equilibrated, e.g. see the evaluation of potentials discussed in Ref. 19.

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Table 1. Simulation conditions investigated.

Temperature (K)	Cascade energy (keV)	Bubble size		He to vacancy ratio
		Number of vacancies	Radius (nm)	
100	5	40	0.48	1
	20	339	1.0	1
600	5	40	0.48	1
	5	339	1.0	0.5, 1
	20	339	1.0	0.25, 0.5, 1

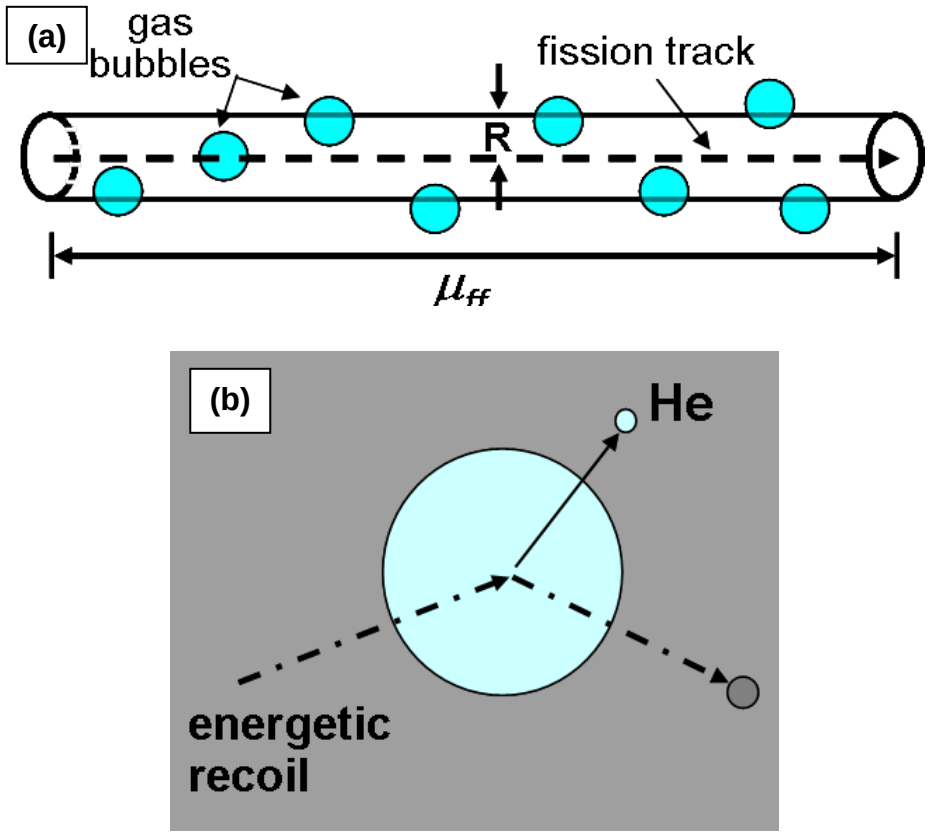

Fig. 1. Schematic diagram of gas resolution mechanisms: (a) “track” resolutioning with bubble dissolution, and (b) ballistic resolutioning from a bubble by an energetic recoil.

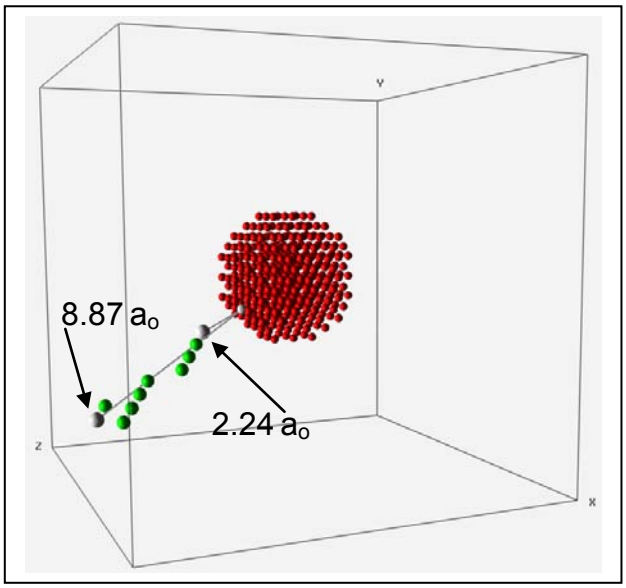

Fig. 2. Illustration of typical initial PKA locations relative to bubble formed by 339 vacancies, radius ~1 nm, in Fe matrix.

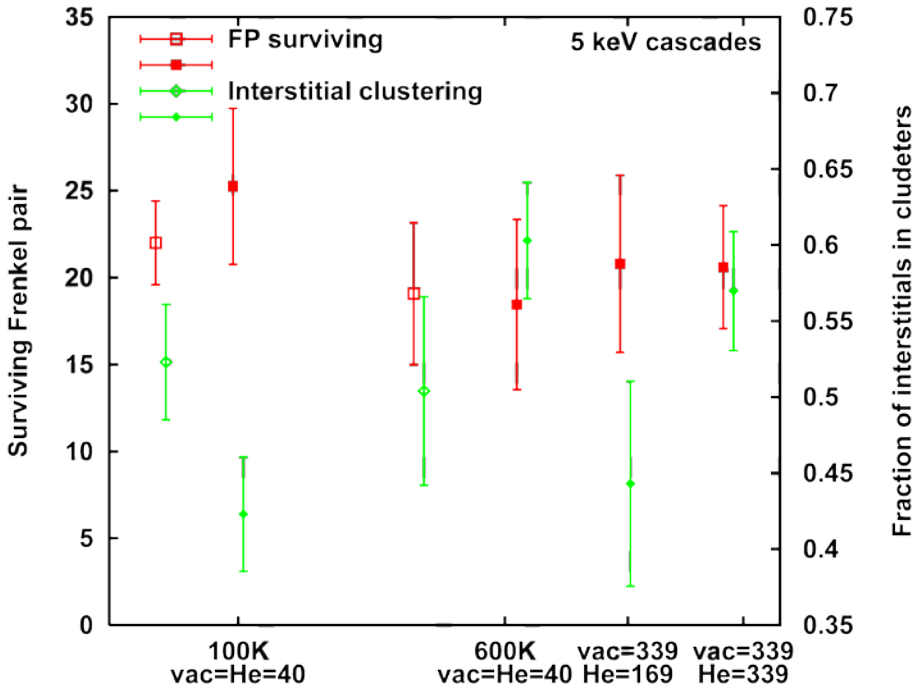

Figure 3. Impact of He bubbles on primary damage formation from 5 keV cascades. Open symbols are from perfect crystal database and filled symbols are current results in simulation cell containing bubbles.

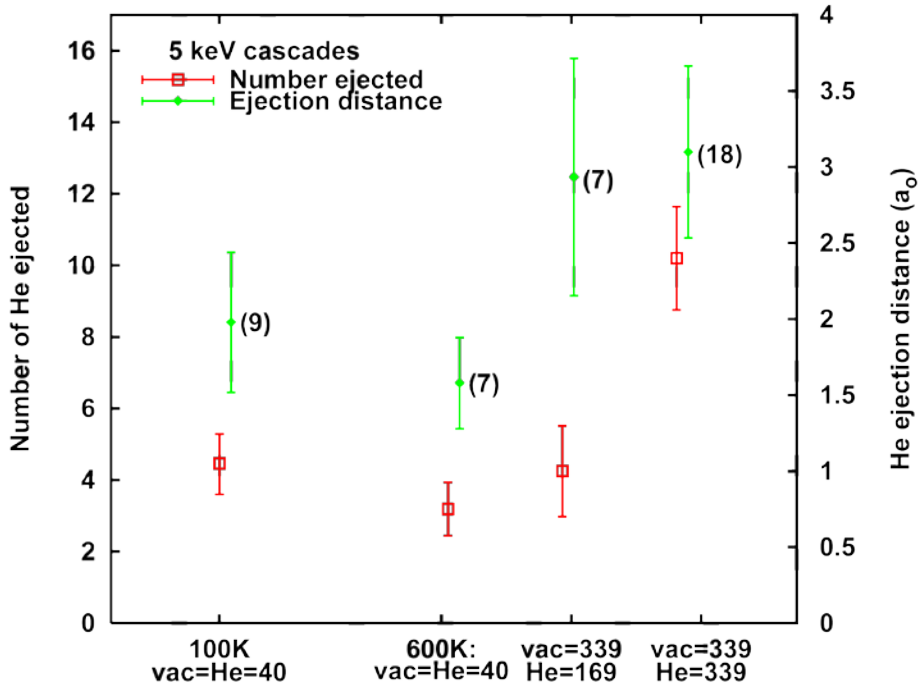

Figure 4. Number of He atoms ejected and average ejection distance from bubbles as a result of 5 keV cascades. Values shown in parentheses 4 are the number of He atoms ejected by more than 5 lattice parameters

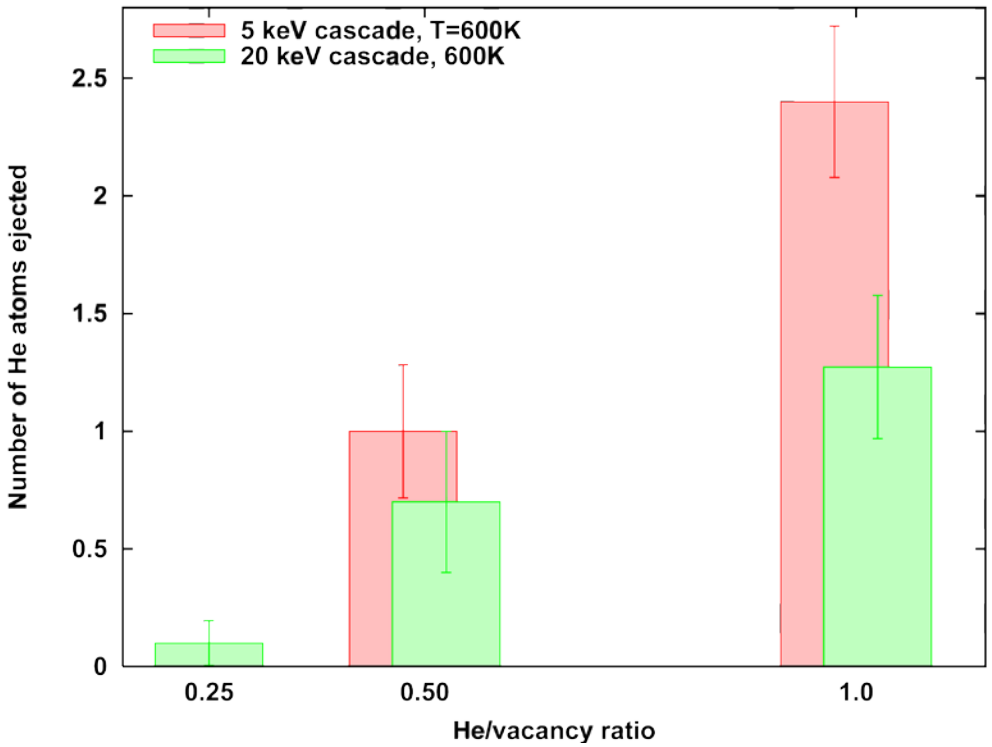

Figure 5. Influence of He/vacancy ratio on the number of He atoms ejected from 1.0 nm bubbles as a result of 5 and 20 keV cascades at 600K.