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A molecular dynamics simulation study of displacement cascades in vanadium

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Abstract

Molecular dynamics calculations were performed to simulate the primary state of radiation damage processes in vanadium. The interaction of atoms was described by the embedded atom method (EAM) potential that was modified at close atomic separation to merge smoothly with the universal ZBL-potential for description of the higher energy recoils in cascades. The initial development of displacement cascades was observed to consist of replacement collision sequences (RCSs) and subsequent molten structures at the centers of cascades. We evaluated the efficiencies of defect production in the displacement events initiated from recoils with kinetic energies of up to 20 keV. The Frenkel pair production is found to have a power-law dependence on the primary recoil energies. The energy dependence is obviously different between single and multiple Frenkel pair production. The efficiencies of defect production and cascade mixing are somewhat higher and lower in bcc vanadium cascades, respectively, than those of fcc metals.

1. Introduction

Changes in mechanical properties due to irradiation are generally supposed to result from irradiation-induced microstructural changes, such as the formation of point defects, dislocation loops, voids, surface craters and solute segregation. The microstructural evolution during irradiation is a non-linear function of initial defect production, which is also a non-linear function of irradiation conditions such as the energies and fluences of projectile particles and irradiation temperature. These complicated radiation-induced or -enhanced behaviors of materials make fusion material design and selection more difficult. A physical description of radiation damage processes involves the varying time- and length-scales from ballistic binary collisions to collective atomic motion in the cooling phase of cascades, followed by thermally activated processes. The time scale of the radiation damage processes relevant to the field of nuclear fusion materials ranges widely from 10^{-15} to 10^{7} s, and the length scale also ranges widely from 10^{-9} to 10^{-3} m [1].

Radiation effects are currently described in terms of an irradiation dose parameter 'dpa' (displacement per atom) of Norgett, [2], which is the calculated number of atoms displaced by irradiation particles. The irradiation dose parameter could play a significant role in the irradiation correlation problem, i.e., predicting material behavior under a non-existing irradiation environment from existing irradiation data of materials. This problem is of practical importance for development of nuclear fusion materials, because there are no facilities, at present, equivalent to nuclear fusion reactors. However, it is actually difficult to predict material behaviors in a fusion device by using the NRT dpa as the irradiation correlation parameter unless the property changes are proportional to dpa.

In the present study, we investigate the non-linearity of defect production from displacement cascades initiated from a recoil atom of kinetic energy of up to 20 keV using a large-scale molecular dynamics (MD) technique. In the simulations, the target material is vanadium, which has significant advantages for use as a structural material in nuclear fusion devices because of its low activation and high radiation resistance.

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2. Molecular dynamics model

In order to investigate defect production due to cascades in irradiated vanadium, we performed simulations with MD which is a well-defined microscopic description of many-body systems, computing the phase space trajectories of a collection of particles that individually obey classical laws of motion. Due to recent advances in computational facilities and development of realistic potentials, this technique has become a practical tool to the many-body problems in question. The simulations in the present study were done with the MD code, MDCASK [3], which runs efficiently on a Cray T3D massively parallel computer at Lawrence Berkeley Laboratory's National Energy Research Supercomputing Center. The code employs PVM message passing routines [4] for communication between processors.

An isotropic many-body potential of the EAM type developed by Johnson and Oh [5] was chosen in the present simulations to describe interactions between vanadium atoms. The potential was modified to connect the universal potential of Biersack, Littmark and Ziegler [6] to describe short range interactions, following the approach by Proennecke et al. [7]. The modified potential for vanadium provides a realistic description of defect properties, melting temperature and displacement characteristics very well [8].

Following the usual procedure of this kind of cascade damage simulation at the beginning of displacement events, kinetic energy was given to an atom in an well-equilibrated MD crystallite. In the present study, primary recoil energies were chosen to range up to 20 keV. The initial directions of motion of recoils were randomly chosen. The number of atoms in the simulation cell ranged up to 549 250, depending on the primary recoil energies. For all simulations, the computational crystallites employed periodic boundary conditions, and were equilibrated at 10 K prior to initiation of the recoil events.

3. Results

Fig. 1 shows the time development of displacement cascade processes in vanadium after an impact of a 20 keV recoil atom. The open and closed circles in the figure indicate a vacant lattice site and a displaced atom, respectively. Some key features of the cascade evolution [9] are clearly observed as replacement collision sequences (RCSs) and local melting at the center of the cascades. As shown in Fig. 1(a), where the defect configuration after the collisional phase of cascades is observed, the majority of stable self-interstitial atoms (SIAs) are associated with the RCS, which is the mechanism for the efficient separation of SIAs from the melting zone at the center of the cascade. During the subsequent cooling phase, significant rearrangement of

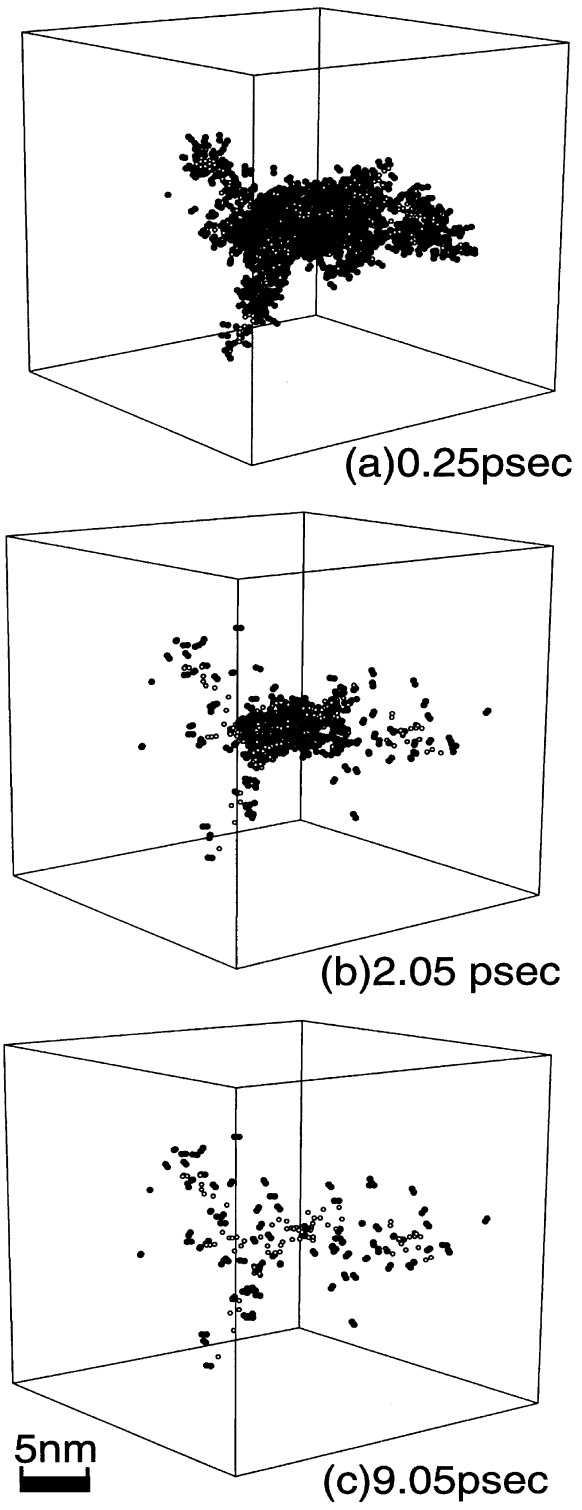

Fig. 1. The time development of displacement cascade processes in vanadium after an impact of a 20 keV recoil atom. The open and closed circles indicate a vacant lattice site and a displaced atom, respectively.

defects takes place: as the melt zone resolidifies, only SIAs outside the melt zone and an equal number of vacancies within the melt zone, can survive after the cascade cooling [10]. Fig. 1(c) is a snapshot of the defect configuration at the end of the cooling phase of cascades, showing that many defects are still left in the simulation cell. These defects might recombine with the other types of defects by normal, thermally activated processes during the subsequent thermal phase of cascades.

The numbers of Frenkel pairs produced by 1, 2, 5, 10 and 20 keV cascades in vanadium are only 78%, 66%, 62%, 43% and 35% of the NRT model [2], respectively. However, the fractions for bcc vanadium are still somewhat higher than those of the fcc metals reported in the literature [11]. The tendency for greater survivability in bcc vanadium is in good agreement with the work of Bacon et al., where the fraction for bcc iron is higher than those of other close-packed metals [12]. Fig. 2 shows a plot of number of Frenkel pairs produced by cascades as a function of primary recoil energy. The computed number of Frenkel pairs produced indicates a power-law dependence on the primary recoil energy, rather than a linear dependence like the NRT model prediction, i.e.

$$N_{\mathrm{FP}} = AE_{\mathrm{p}}^{m}, \tag{1}$$

where N_{FP} is the number of Frenkel pairs produced, A and m are constants, and E_{p} is the primary recoil energy. The power-law dependence is also reported for other metals [12]. It is interesting to note that the slope of the plot changes between higher and lower energy ranges as represented in the figure, showing 0.75 and 2.0, respectively.

The direct formation of defect clusters within cascades is of interest as an important part of the radiation damage process, because a difference in clustering efficiencies between vacancies and interstitials produces a bias effect that induces imbalance in defect behavior between interstitials and vacancies during the subsequent thermal and diffusional phases [13]. The present simulation results show that the production efficiencies of vacancy- and interstitial-clusters directly formed in cascades in vanadium are smaller than those of several other metals [11,12]. Even in the 20 keV events, only 6% and 2% of produced defects are taken up in vacancy- and interstitial-clusters, respectively. The smaller clustering efficiencies are consistent with experimental results that show no vacancy clusters are observed by transmission electron microscopy (TEM) in self-ion irradiated vanadium [14].

Fig. 3 shows the computed mixing efficiency of atoms due to cascades as a function of primary recoil energy. The cascade mixing efficiency, ξ, is a measure of rearrangement of atom configuration during the cascades, and is defined by the following equation:

$$\xi = \frac{\sum_{i=1}^{N}[r_i(t) - r_i(0)]^2}{6nE_{\mathrm{Dam}}}, \tag{2}$$

where E_{Dam} is the damage energy per recoil, n is the atomic density and r_i is the displacement of atom i at

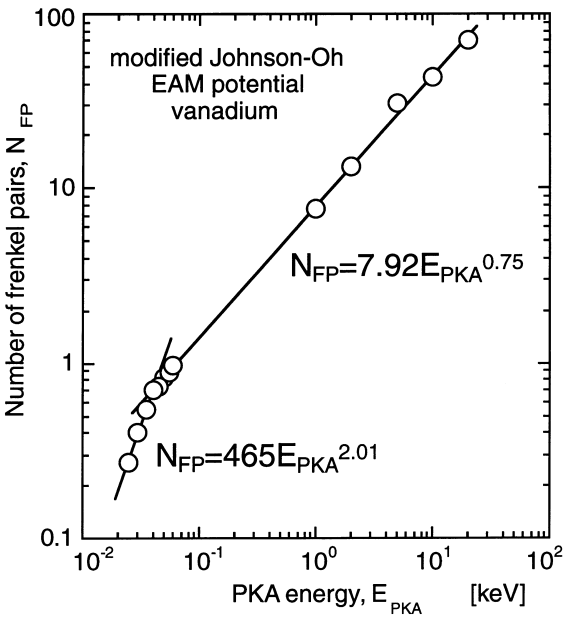

Fig. 2. A plot of a number of Frenkel pairs produced by cascades as a function of the primary recoil energy.

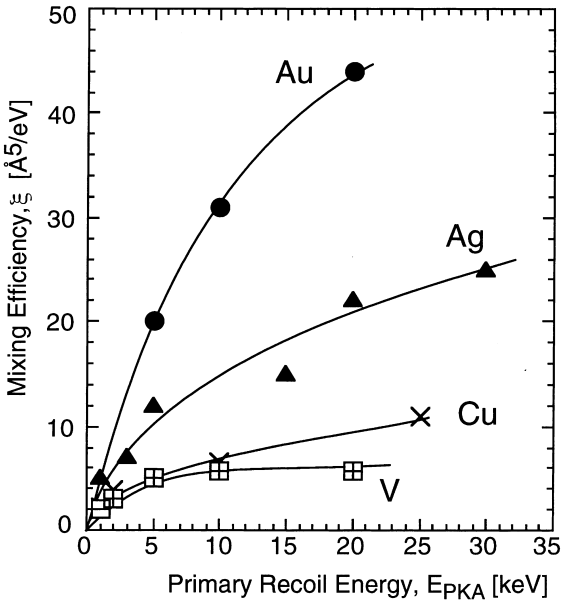

Fig. 3. The computed mixing efficiency for cascades in Au, Ag, Cu and V as a function of the primary recoil energy.

time t. Diaz de la Rubia et al. pointed out that the vast majority of atomic displacement, $\sum_{i=1}^{N}[r_i(t) - r_i(0)]^2$, take place during the cooling phase of cascades [11]. The cascade mixing efficiencies of other fcc metals [11] are also shown in the figure. The mixing efficiency is observed to increase with increasing recoil energy, and is almost saturated beyond the recoil energy of 10 keV for vanadium cascades. The efficiency of vanadium cascades is lower than those of other fcc metals, indicating that rearrangement of atom configuration during the cascades in vanadium is less than those in fcc metals.

4. Discussion

It is interesting to note in Fig. 2 that the energy dependence of defect production changes where the number of defects produced is unity. The knee in the plot, occurring at about 40 eV for vanadium, indicates that the physical description of Frenkel pair production is different below and above the energy. Above this energy, multiple frenkel pairs are produced. The NRT dpa was originally an attempt to calculate defect production in terms of the production of individual Frenkel pairs extrapolated into higher energy ranges. The NRT dpa is calculated from the displacement damage function, $\nu(E_D) = \kappa E_D/2E_d$, where E_D is the damage energy, E_d is the displacement threshold energy and κ is a constant of 0.8. The E_d is determined on a basis of a single Frenkel pair production. The NRT dpa is based on the criterion of whether a single atom is displaced or not. Because of cascades, the discrepancy between the NRT model prediction and actual defect production increases with increasing recoil energies. At present, the inconsistency is compensated for by introducing a parameter called 'production efficiency', which is defined as the fraction of the number of Frenkel pairs produced relative to the calculated NRT displacements. Above E_d, the defect production should be evaluated on the basis of a multiple Frenkel pair production mechanism that is, for example, the cascade molten zone model by Averback et al. [10]. Whether Eq. (1) and the molten zone model are consistent with each other could be an interesting subject for a more precise study of defect production due to cascades.

Fig. 4 shows a plot of cascade collapse efficiencies that are obtained from ion irradiation experiments in the literature [15–25], as a function of incident ion energies. The cascade collapse efficiency is defined as the number of TEM-observable vacancy clusters divided by the number of incident ions. It is interesting to note that the number of vacancy clusters also shows a power-law dependence on ion energies. Moreover, the knee in the plot is also located where the number of vacancy clusters produced is unity, indicating that the physical description of the energy dependence of the cluster formation is also different between single and multiple cluster formations.

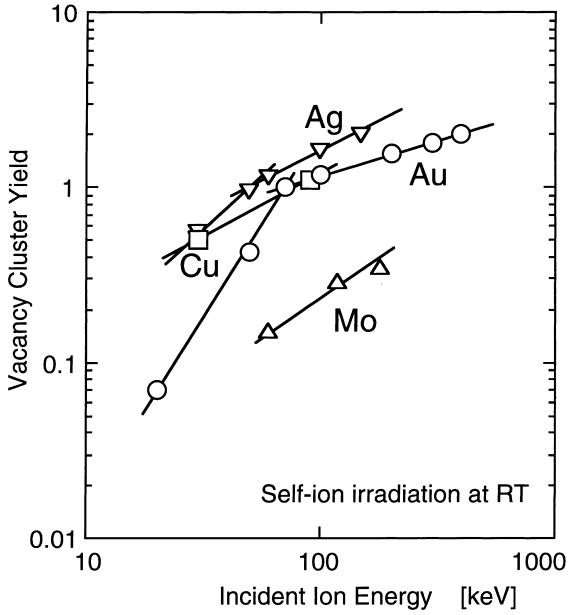

Fig. 4. A plot of experimental vacancy cluster yield as a function of incident ion energy. The yield is defined as the number of vacancy clusters observed by TEM divided by the number of incident ions.

Imagine a plot of the number of Frenkel pairs from Fig. 2 as a function of the inverse of the primary recoil energy that is proportional to a differential scattering cross-section, $d\sigma_s$. The plot shows that the Frenkel pair production is decreasing with increasing cross-section, exhibiting the fractal behavior [26] with a typical dependence. One may easily consider that the cascade collapse efficiency also shows the same kind of fractal behavior, as shown in Fig. 4. Heinisch [27] also found that the recoil energy dependence of vacancy densities of MARLOWE [28] cascades behaves fractally, showing a knee at the energy where subcascades are formed. The defect productions relevant to the displacement cascade events, i.e., Frenkel pair production due to cascades, cluster formation directly in cascades and a displaced atom production at the end of the collisional phase of cascades, all exhibit the fractal behavior against the inverse of the recoil energies or the scattering cross-section. It might indicate that, for a better expression of the irradiation correlation parameter, we not only consider the linear theory within the NRT expression but also include the fractal nature and power-law dependence.

A large fraction of Frenkel pairs produced from cascades are recombined within the cascade region during the cooling phase of cascades. The significant mixing of atoms also occurs during the cooling phase of cascades [11], as mentioned above. It indicates that higher mixing efficiency during the cascade cooling enhances more recombination, leading to lower

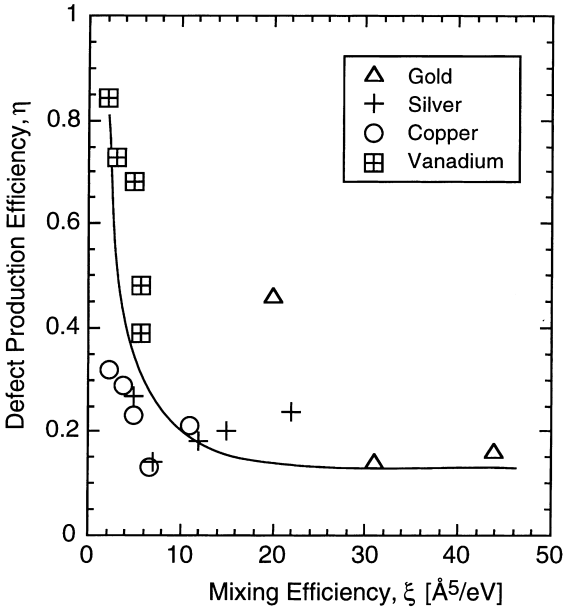

Fig. 5. The efficiency of Frenkel pair production as a function of computed mixing efficiency in several pure metals. Higher mixing efficiency provides lower production efficiencies.

production efficiencies of Frenkel pairs produced. Fig. 5 shows the production efficiencies of Frenkel pairs as a function of the cascade mixing efficiency in vanadium as well as several other pure metals [11]. Large fluctuation is seen in the figure, indicating that the production efficiencies cannot be explained by only the mixing efficiency. However, the Frenkel pair production efficiency decreases with increasing cascade mixing efficiency. One would say that the higher mixing efficiencies accompany the lower efficiencies of Frenkel pair production.

A more important point is why the mixing efficiency is smaller and the production efficiency is higher for the cascade in vanadium than those in other fcc metals. The answer is not simple, but it could be due to the differences in (1) lattice structure, i.e., bcc/fcc, (2) melting temperature of the metals, and (3) mobility of atoms during the cooling phase. The first might influence the energy branch during the collisional phase, which could be related to the RCSs and the spatial distribution of energy deposition in cascades. The second might determine the size of the molten zone produced by cascades. The third would be related to the nature of metal elements themselves.

5. Conclusion

We performed MD simulations of displacement cascades in vanadium initiated from a recoil atom of kinetic energy of up to 20 keV. The Frenkel pair production due to the cascades shows a power-law dependence on primary recoil energies, indicating a different physical description between below and above the displacement threshold energy, E_d. The cascades in bcc vanadium provide a higher production efficiency of defects and a lower mixing efficiency, compared to those in fcc metals.

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