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Synergistic effects of PKA and helium on primary damage formation in Fe–0.1%He ☆

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Abstract

Molecular dynamics (MD) simulation of displacement cascades in Fe–0.1%He are used to investigate the synergistic effects of primary knock-on atoms (PKA) and He on the irradiation damage production at temperatures between 10 K and 523 K. The results indicate that the vacancies produced by cascades are all in the central region of displacement cascade. The presence of He does not influence Frenkel pair production. During cascade evolution all displaced Fe and He atoms combine with each other to form Fe–He or Fe–Fe interstitials dumbbell as well as interstitial clusters. The number and size of interstitial clusters increase with the PKA energy and temperature. A few large clusters consisting of a large number of Fe interstitials with a few helium atoms are formed, the rest being Fe–He clusters at small and medium sizes. The interstitial dumbbell of Fe–Fe and Fe–He are in the ⟨100⟩ and ⟨110⟩ lattice direction, respectively.

1. Introduction

Reduced activation ferritic/martensitic (RAMS) steel is considered to be a candidate for use as a first wall structural material for future fusion reactors [1]. One concern with He, produced by transmutation from the 14 MeV fusion neutrons, is a loss of fracture toughness, leading to a DBTT shift, induced by irradiation at low temperature, e.g. room temperature to 300 °C [2]. The dose dependence of impact properties for different advanced ferritic/martensitic alloys at low irradiation temperature occurs in the low-dose range, and is mainly an effect of He generated by different levels of boron [3]. A complementary investigator [4] indicated that DBTT shift (ΔDBTT) increases with helium content at 250 °C at doses up to 0.2 dpa. The TEM observation of F82H irradiated in three ways, i.e. 590 MeV proton, 590 MeV proton + neutron, and neutron only were carried out. These conditions lead to different He levels. The dose ranged from 0.5 dpa to 10 dpa at 250–310 °C. The mean defect size and density increases with the damage dose, with the mean size ranging from 1.5 to 6.9 nm and density ranging from 2.5×10^{21} to 7.0×10^{22} m^{-3}. However, no cavities were observed, even at the highest studied dose [5]. JLM-1(9Cr–2WVTa) specimens implanted to 120 and 580 appm He and to doses from 0.048 to 0.226 dpa were annealed at 310 °C,

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450 °C, 503 °C and 555 °C. After annealing at 310 °C, no bubble or cavities were observed. But, after annealing at 450 °C, 503 °C and 555 °C some bubbles or cavities appeared both at grain boundary and in the matrix [6]. Reduced activation martensitic steel specimens were irradiated by dual-beams in a high voltage electron microscope H-1300 from 0.1 to 50 dpa at 250 °C and 300 °C with 10 appm/dpa He. In situ microstructure observation shows that bubbles were present after 1.7 dpa for 250 °C irradiation and after 0.1 dpa for 300 °C irradiation [7]. These results could imply that He atoms participate in the formation of interstitial clusters initially and, when enough vacancies are available, they could contribute to cavity formation. Because He from implantation and nuclear reactions could be first located in an interstitial site, a question can be raised regarding the role of cascades on interstitial He behavior. With this in mind, the synergistic effects of interstitial He and cascades are investigated by MD simulation to see whether the He contributes to interstitial clusters or vacancy clusters at temperature less than 300 °C. Results are compared to experimental ones.

2. Simulation method

Molecular dynamics (MD) calculations were performed to investigate displacement cascades, using pure Fe as a model for steels and Fe containing 0.1%He as uniformly distributed interstitials. The combination of interatomic potentials is described in [8]. The combined potentials give a formation energy for the He atom in the Fe matrix of 3.25, 5.34 and 5.29 eV when it is on a substitutional, tetrahedral and octahedral sites, respectively [9]. Ab initio calculations give 4.22, 4.39 and 4.58 eV for the same sites, respectively [10]. The empirical potentials thus overestimate the formation energy of the tetrahedral site, which should be most favorable interstitial. They give instead the octahedral site. However, there is at present no other available empirical potential. Moreover, these empirical potentials do predict the same self-trapping behavior of He predicted by ab initio, and do predict the substitutional site as the most favorable one.

Prior to initiating the cascade, a simulation box containing 0.1% interstitial He in a uniform distribution, as well as a box without helium, was equilibrated for 0.4–1.3 picoseconds (ps) at design temperature. These were then used as the starting points for the cascade simulation and defect identification. Each cascade was started by imparting a specified kinetic energy E_{MD} to the selected primary knock-on atom (PKA) along a high-index direction to avoid channeling. Cubic box size, simulated time and number of cascades versus E_{MD} are summarized in Table 1. Following common practice [11], all presented results were obtained working in the NVE microcanonical ensemble with periodic boundary condition. Temperature control is made in boundary regions which mimic a thermal bath. The final atomic configuration was analyzed to detect and count defects, using a Wigner–Seitz cell method. An empty cell corresponds to a vacancy and two atoms in the same cell correspond to one interstitial. Cluster size distributions were obtained considering first nearest, second nearest, third nearest and $2a_0$ nearest neighbor (nn) distances. The distribution of clusters for third nn is close to that for $2a_0$ nn. Considering most of interstitials in stable Fe–He dumbbell formation and the group containing two Fe–He dumbbell interstitials plus one matrix atom between two dumbbells in $\langle 1\,0\,0\rangle$ direction, as well as in $\langle 1\,1\,1\rangle$ direction, still belong to cluster, we defined the clusters using $2a_0$ criterion (nn) for interstitials. Due to the interstitials in dumbbell formation the cluster should containing more than six interstitials (equal to three dumbbell interstitials) to be counted as cluster.

3. Results and discussion

3.1. Defect number

The average number (N_{FP}) of surviving Frenkel pairs for various temperatures at the end of cascade in Fe–0.1%He resulting from the MD simulations is plotted as a function of E_{MD}, shown in Fig. 1(a). The points have been interpolated using the empirical power law proposed by Bacon et al., $N_{FP} = A \cdot E_{MD}^{m}$ [12]. Prefactors and exponents obtained in a least-square fit are agreement with previous work on Fe [13]. The exponent increases with the temperature from 0.7178 at 10 K to 0.8503 at 523 K. This means that the defect production efficiency is more sensitive to PKA energy at 523 K in Fe containing 0.1%He interstitial.

Traditionally, the number of Frenkel pairs produced per cascade is estimated using [14] the NRT formula, $N_{NRT} = 0.8E_D/2E_d$, where E_d is the average displacement energy for all crystallographic directions and E_D is the PKA energy. E_D is the

Table 1
Summary of number of cascades, simulation time and box size versus temperature and E_{MD}

Temperature (K)	E_{MD} (keV)	Number of cascades						Other parameters	
		Fe	Fe–0.1%HeI	Fe–1%HeI	Fe–0.1%HeS	Fe–1%HeS	Fe–0.2 * 10^{-5} V	Time (ps)	Box size (atoms)
10	3		10					25	50 094
	5		10	2	1			25	50 094
	7		10					25	50 094
	10		10					25	50 094
300	3		10					25	50 094
	5		10	2	1			25	50 094
	7		10					25	50 094
	10		10					25	50 094
523	3	1	10			1	1	25	50 094
	5	1	10	2	1		1	25	50 094
	7	1	10				1	25	50 094
	10	1	10				1	25	50 094

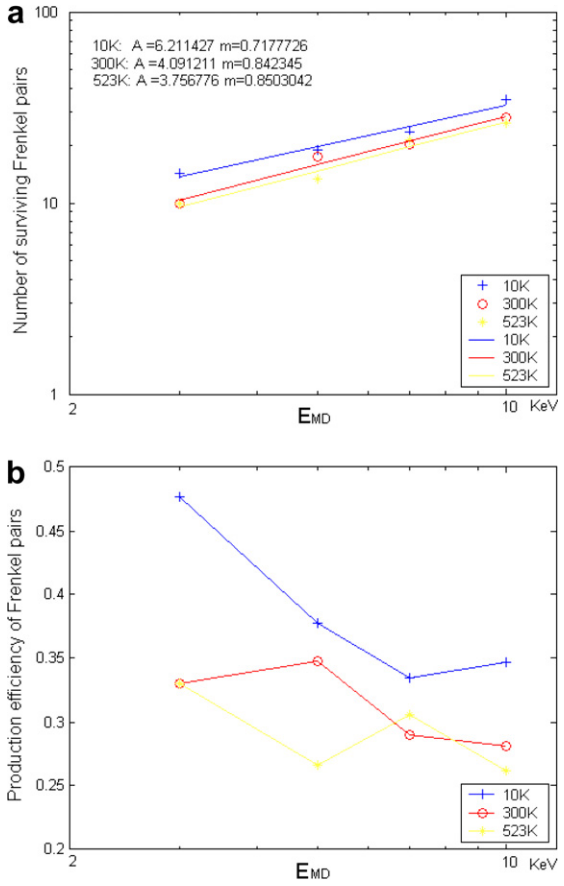

Fig. 1. Cascade energy dependence of number of surviving Frenkel pairs (a) and defect production efficiency compared to NRT (b) at the end of the cascade in Fe with 0.1% interstitial He at various temperatures. In (a) the power law proposed by Bacon et al., $N_{FP} = A \cdot E_{MD}^{m}$ has been used for interpolation.

fraction of recoil energy that goes into displacement damage, after subtracting the fraction dissipated for electron excitation. Since in the present MD simulations the interaction between ions and electrons is not included, it is assumed that $E_D = E_{MD}$. The defect production efficiency is defined as the ratio of the surviving MD Frenkel pairs to the number of NRT displacements (N_{FP}/N_{NRT}). For the sake of comparison, we took $E_d = 40$ eV to estimate the number of defects with the NRT formula. The defect production efficiency decreases with recoil energy and temperature down to a more or less asymptotic value of about 0.33 at 10 K and 0.26 at 523 K. These values are slightly higher in Fe containing 0.1% interstitial He than in pure Fe since more recoils recombine with He to form Fe–He interstitial dumbbells, reducing recombination with vacancies and increasing defect production.

Fe recoils in cascades can combined with He to form Fe–He interstitial dumbbells, thus leaving more vacancies in primary recoil sites and the defect production efficiency is slightly higher than that without interstitial He. For the interstitials, because of 0.1%He interstitial uniform distribution, most are of Fe–He dumbbell type, and only a few defects are pure Fe–Fe dumbbells, produced by the cascade. The direction of Fe–He dumbbell is along ⟨1 0 0⟩, and the direction of Fe–Fe dumbbell is along ⟨1 1 0⟩, which is in agreement with ab initio calculation [15]. In the case of 0.1%He in substitutional sites, the number of surviving Frenkel pairs after PKA event is slightly less than that for 0.1%He in interstitial sites and the total number of interstitials is the same as the number of vacancies. Most self-interstitials are pure Fe–Fe dumbbells, only a few are Fe–He dumbbell type. This indicates that He is very stable in the substitutional site and the probability of displacing He is rare in the case of 0.1%He in substitutional sites. Because the transfer of kinetic energy to the He atoms is 3.5 times less than that to Fe atoms and the displacement threshold energy of He in lattice is similar to Fe one, the probability of displacing a He atom is much lower than for an Fe atom. However, the He from transmutation reactions (or implantation) includes one energetic He and one residual host atom. This implies that He from transmutation reaction and implantation may be in interstitials sites.

3.2. Cluster formation

Previous papers on displacement cascades in Fe show that very little in-cascade vacancy clustering occurs in this material, while sizeable interstitial clusters are found [11–13,15,16]. Our results agree with these findings.

The number and size of interstitial clusters are shown in Fig. 2, which sums the number and size of interstitial clusters of 10 events for one case. The results presented are for a distance between neighbors of 0.573 nm (3rd nn). In this calculation, we consider a cluster those that contain more than 3 interstitials. This indicates that the number and size of interstitial clusters increases with the energy of PKA and temperature in the range 10–523 K. When the temperature increases, the mobility of defects increases therefore reducing the number of surviving Frenkel pairs. At same time the probability of interstitials meeting with each other increases, thus enhancing the number and size of interstitial clusters. Particularly for Fe containing 0.1% interstitial helium, the number of defects is much higher than that in Fe without helium, because the defects in the former includes the He interstitials already present before the cascade. When recoils meet with helium, they not only form interstitial dumbbells, but also bring some kinetic energy to these dumbbell interstitials to prompt their mobility and enhance the probability of He–SIA clustering. Therefore the number and size of interstitial clusters is much higher than that in Fe without helium. For the same reason, temperature increases the mobility of defects, again enhancing the He–SIA clustering in Fe containing 0.1%He in interstitial state. Therefore, the presence of He in interstitial sites does influence the fraction of defects in clusters and the number and size of interstitial clusters increase with temperature. No detailed analysis was conducted to detect a possible preferential association of vacancies and vacancy clusters with He atoms. On the contrary, a large portion of the interstitials in cluster in Fe–0.1%He are He atoms, and there are no vacancy near the clusters or substitutional He.

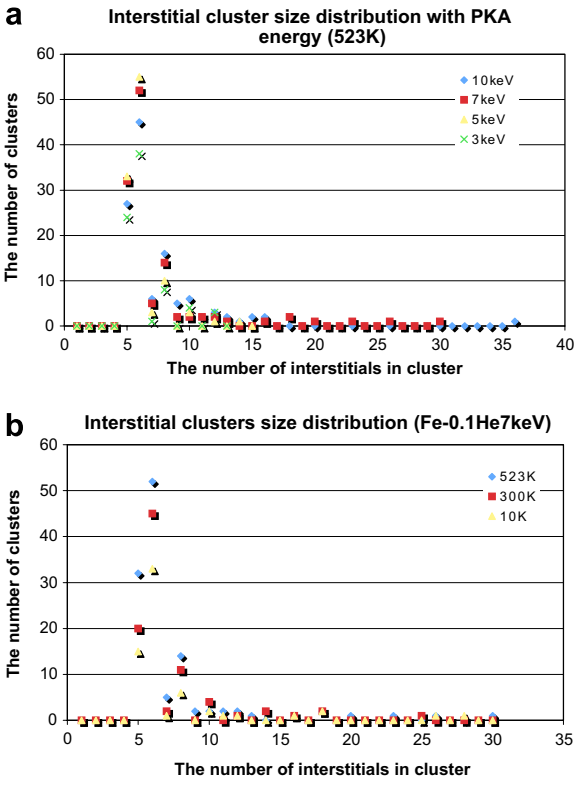

Fig. 2. The interstitial cluster size distribution (a) for various PKA energies at 523 K; (b) for various temperatures at 7 keV. The clustering criterion is third nearest neighbor, spacing $\leqslant$0.5733 nm.

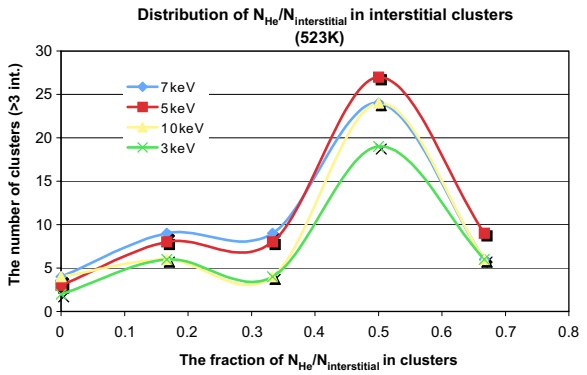

Fig. 3. The number of interstitial clusters versus the fraction of $N_{He}/N_{interstitial}$ in clusters (containing >3 interstitials) at temperature 523 K.

We expect that the formation of Fe–He mixed interstitial clusters, which are shown here to be stabilized by He, will have a large impact on the subsequent evolution of radiation damage. These He–Fe mixed interstitial clusters may be obstacles to dislocation motion, leading to hardening associated with a DBTT shift at low temperatures from room temperature to 300 °C [2]. The observed large amount of small sized Fe–He mixed interstitial clusters may help explain the experimental observations [5,6]. A high density of defects with mean size range in 1.5–6.9 nm was observed, but no cavities, even at the highest studied dose [5]. At higher temperature (>450 °C) the Fe–He mixed interstitial clusters will come unstable, evolving to bubble or voids by release of some of He atoms [17]. It is now important to prove the existence of these Fe–He interstitial clusters, by small angle neutron scattering (SANS) experiments, for example.

Fig. 3 presents the number of clusters as a function of ratio of He to Fe atoms in each cluster. This figure shows that most of the clusters have one He atom per Fe atom. For large sized cluster, the number of He is much smaller than the number of Fe interstitials. For example for 10 keV PKA at 523 K, there is a cluster including 36 interstitials without He and a cluster including nine interstitials with three He atoms; for 10 keV PKA at 300 K, there is a cluster including 10 interstitials with one He. In these calculation events, there is no vacancy nearby the clusters or substitutional He.

4. Summary

Displacement cascades up to 10 keV were simulated by molecular dynamics in Fe, Fe–0.1%He on interstitial sites and Fe–0.1%He in substitution sites, using a fitted and validated EAM many-body potential. A content of 0.1%He in interstitial or substitutional sites does not seem to affect the collisional stage of the cascade, but it does influence the distribution of dumbbell species during the post-collisional stage. Most interstitials are mixed Fe–He dumbbells species in Fe with 0.1%He interstitials and Fe–Fe dumbbells in Fe with 0.1%He in substitutional sites. The number and size of interstitial clusters in former is much higher than that in Fe with 0.1%He in substitutional sites as well as Fe without He.

(1) The defect production efficiency in Fe–0.1%He in interstitial sites is slightly higher than that in Fe–0.1%He in substitution sites and in pure Fe. The amount of Fe–He dumbbell seems to reduce recombination.
(2) The interstitial clusters of small and medium size contain a large percentage (50%) of He atoms, which stabilize them. This feature may drastically reduce the mobility of interstitial clusters in Fe–0.1%He interstitial state compared to pure Fe, with significant impact on the subsequent evolution of radiation induced defects in the alloy.
(3) The number and size of interstitial clusters increase with the PKA energy and temperature in the range 10–523 K. The small and medium clusters with a high amount of He, as well as large clusters may be hard obstacles to dislocation motion.
(4) The interstitial dumbbells for Fe–Fe and Fe–He are in the ⟨110⟩ and ⟨100⟩ lattice direction, respectively. There are no vacancies or substitutional He near the clusters.

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