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Atomistic studies of nucleation of He clusters and bubbles in bcc iron

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ABSTRACT

Atomistic simulations of the nucleation of He clusters and bubbles in bcc iron at 800 K have been carried out using the newly developed Fe–Fe interatomic potential, along with Ackland potential for the Fe–Fe interactions. Microstructure changes were analyzed in detail. We found that a He cluster with four He atoms is able to push out an iron interstitial from the cluster, creating a Frenkel pair. Small He clusters and self-interstitial atom (SIA) can migrate in the matrix, but He-vacancy (He-V) clusters are immobile. Most SIAs form <111> clusters, and only the dislocation loops with a Burgers vector of $\boldsymbol{b} = 1/2$ <111> appear in the simulations. SIA clusters (or loops) are attached to He-V clusters for He implantation up to 1372 appm, while the He-V cluster–loop complexes with more than one He-V cluster are formed at the He concentration of 2057 appm and larger.

1. Introduction

In a fusion reactor environment helium (He) is produced at high rates in steels by nuclear (n, α) transmutation reactions. Because of the extremely low solubility of He in metals, He atoms tend to be deeply trapped in small vacancy clusters and microstructural features, leading to the creation of He-stabilized bubbles, which can significantly degrade the mechanical properties of materials. Therefore, understanding of the nucleation of He bubbles in steels, both in the bulk and within microstructural features, is of fundamental importance to the development of fusion reactors. Multiscale modeling, especially including the atomic scale, provides a basis to obtain insight into and general understanding of the complex radiation damage process. Particularly, molecular dynamics (MD) methods have been widely employed to study the atomic-level processes of defects controlling microstructural evolution in advanced ferritic steels.

The behavior of He in Fe has been widely investigated [1–15]. MD simulations have been employed to yield important understanding of the behavior of He defects in bcc Fe using different interatomic potentials [2–4]. Recently, Stewart et al. investigated the formation and diffusion of He clusters and bubbles in bcc iron using a three-body Fe-He interatomic potential [2]. Gao et al. [14] developed a new interatomic potential for Fe–He interactions, which is based on the electronic hybridization between Fe *d*-electrons and He *s*-electrons. The diffusion properties of He interstitials and interstitial He clusters in the bulk of bcc Fe have been studied using MD with this newly developed Fe–He potential [15]. The low migration energy barrier for a single He interstitial in the bulk is consistent with that obtained using *ab initio* methods. It was also found that small He clusters can migrate at low temperatures, but at higher temperatures they can kick out a SIA and become trapped by the resulting vacancy, forming He-vacancy clusters. Also, the binding energies of small He-V and He–He clusters are in good agreement with those obtained by *ab initio* and other potential calculations. In the present study we investigate the nucleation of He clusters and bubbles, the emission of SIAs from the He clusters, and the formation of dislocation loops in the bulk of bcc Fe, as well as the effects of these phenomena on the microstructural changes.

2. Simulation procedure

In the present simulations a modified version of the MOLDY computer code [16] is used. The interatomic potentials of Ackland [17] and Beck [18] are used to describe the Fe–Fe and He–He interactions respectively, while the Fe–He interaction is described by the newly developed Fe–He potential [14].

The NVT ensemble [15] is used in the simulations, and periodic boundary conditions are applied along all three axes to avoid the influence of free surfaces. A MD box of $45a_0 \times 45a_0 \times 45a_0$ with 182,250 Fe atoms is used, where a_0 is the lattice constant of perfect bcc iron (2.8553 Å). Initially 125 He atoms are randomly distributed in the box, which corresponds to a mean He concentration of 685 appm. Then, the box is quenched to 0 K for 10,000 time

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steps, followed by a temperature rescaling to 800 K and annealing at 800 K for 1.2 ns, where there is no significant migrations or jumps of atoms that affect the size and distribution of He clusters. After the equilibrium state has been achieved, an additional125 He atoms are randomly inserted into the system, which gives a total of 250 He atoms. The same quenching and annealing approach is used, but the annealing time is about 0.6 ns, which is long enough for the system to reach its equilibrium state (i.e. the defect configurations remain without obvious changes). The same approach is repeated until a total of 500 He atoms have been added, i.e. the mean He concentration is 2743 appm, which is lower than the maximum He concentration of 3000 appm used in experiments [19]. It should be noted that helium atoms are gradually added from low to high concentrations, and thus, the nucleation mechanism and growth of helium clusters (bubbles) can be explored, at least at the early stage of nucleation. The highest concentration mimics the experimental condition, which will allow us to study up-limit effects on the nucleation of He clusters. After annealing at 800 K, the system is then quenched to 0 K for further analysis in terms of defect configurations, He bubble distributions and dislocation loop orientation.

3. Results and discussion

3.1. 125 He atoms (685 appm He)

The initial 125 He atoms are randomly distributed in the simulation box as single interstitials, either in tetrahedral or octahedral positions. These single He interstitials can easily migrate at 800 K because of their low migration energy barrier of 0.058 eV [14]. It is of interest to note that two He atoms can join together, forming a He dimer (He_2), but it sometimes dissociates into two single He interstitials because of its small binding energy (~0.3 eV with the present potential). Also, a He_2 cluster migrates easily in the Fe matrix because of its low migration energy of 0.09 eV and the emission of a self-interstitial from the He_2 cluster is not observed. Once a He atom migrates into a He_2 cluster, a He_3 cluster is formed. Similarly, the He_3 cluster can easily migrate with the migration energy of 0.097 eV [15], but He dissociation from the He_3 cluster does not occur due to its slightly higher binding energy [14]. Although a Fe atom can be displaced from its site, temporarily forming a He_3VFe complex, the emission of a self-interstitial does not occur, as shown in Fig. 1. The structure of the He_3VFe complex appears at 0.27 ns, but during the annealing process, this complex structure transfers to a He_3 cluster because of the recombining of the Fe atom and the vacancy. It is also noted that the He_3 cluster can fast migrate, which is clearly shown in Fig. 1, where interstitials and vacancies were identified and counted using a displacement analysis method [20]. When the He cluster reaches four He atoms, an Fe atom can be kicked out, forming a He-vacancy (He_4V) cluster and a SIA, as shown in Fig. 2. It is of interest to find that the He atoms in the cluster rotate quickly around the vacancy, but the centre of mass of the He_4V cluster does not change with time. This suggests that the helium atoms in the cluster still strongly bond with the vacancy, forming a stable configuration at 800 K. The emitted SIA forms a dumbbell with another Fe atom, and attaches to the He_4V cluster. With increasing simulation time, some of the SIAs are able to escape from their original He-vacancy clusters, and join with other SIAs, forming SIA clusters attached to He-vacancy clusters. A few He_4 clusters are observed to share one lattice vacancy with an attached Fe atom, forming He_4VFe at 800 K, which is similar to the configuration of He_3VFe. Thus, a cluster of four or more He atoms can emit a SIA, forming a Frenkel pair at 800 K. This is different from Stewart's results [2] in which it is demonstrated that Frenkel pair creation happens in clusters with six or more He at 800 K.

After annealing 1.2 ns, most He atoms become part of He-V defects. The He and defect distributions at 1.2 ns for the case of 125 He atoms are shown in Fig. 3, from which it is clearly shown that almost all the SIA clusters are attached to He clusters, and most SIA clusters are made up of <111> crowdions and split dumbbells. Some SIA clusters consist of <111> crowdions and <110> split dumbbells, while some form the mixture configurations of many Fe_3V, one of which is shown in the inset in Fig. 3(a). The formation of the mixture configurations of many Fe_3V may be due to the fact that the pressure of its nearest He cluster is not enough to push SIAs away to form <111> crowdions. The size distribution of He and SIA clusters is shown in Fig. 3(b). The largest He cluster includes 12 He atoms, while the largest SIA cluster contains only 7 SIAs. Both the size distributions of He and SIA clusters exhibit almost uniform distributions.

3.2. 250 He atoms (1372 appm He)

After randomly inserting the second 125 He atoms in the simulation box, corresponding to the mean He concentration of 1372 appm, the system is quenched to 0 K and annealed at 800 K. The distributions and configurations of the He-V and SIA clusters, as well as point defects, after 0.2 ns and 0.6 ns are shown in Figs. 4(a) and (b), respectively. It is found that the centers of the He-V clusters remain the same, while single He atoms and SIAs can migrate in the matrix. Since some He clusters already existed in the simulation box, the inserted He atoms can join the existing He-V

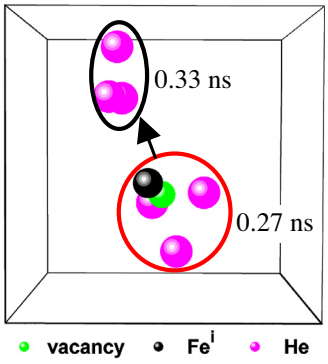

Fig. 1. Structure change of the He_3VFe complex, where the red circle indicates the He_3VFe complex at 0.27 ns. This configuration transfers to a He_3 cluster by the recombination of the SIA and the vacancy at 0.33 ns, indicated in the black circle. Green, black and magenta spheres represent vacancies, SIAs and He atoms, respectively. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

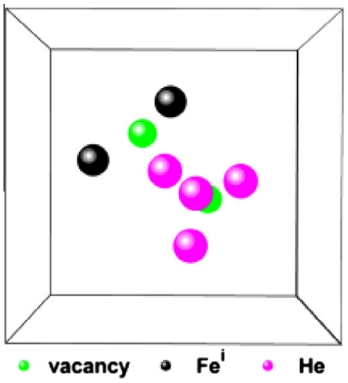

Fig. 2. Structure of a He_4V cluster and a SIA, where the representations of spheres are the same as those in Fig. 1.