Acta Materialia 97 (2015) 86–93

Contents lists available at ScienceDirect

Acta Materialia

journal homepage: www.elsevier.com/locate/actamat

Effects of local structure on helium bubble growth in bulk and at grain boundaries of bcc iron: A molecular dynamics study

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ARTICLE INFO

Article history:
Received 4 March 2015
Revised 21 June 2015
Accepted 24 June 2015
Available online 12 July 2015

Keywords:
Grain boundaries
Atomistic simulation
He bubble growth
Bcc iron
Dislocation loops

ABSTRACT

The nucleation and growth of helium (He) bubbles in the bulk and at $\Sigma3\langle110\rangle\{112\}$ and $\Sigma73b\langle110\rangle\{661\}$ grain boundaries (GBs) in bcc iron have been investigated using molecular dynamics simulations. The results show that a $1/2\langle111\rangle\{111\}$ dislocation loop is formed with the sequential collection of $\langle111\rangle$ interstitial crowdions at the periphery of the He cluster and is eventually emitted from the He cluster. Insertion of 45 He atoms into a He cluster leads to the formation of a $1/2\langle111\rangle$ dislocation loop in $\Sigma3$ GB. It is of interest to notice that the transition of a dislocation segment through the GB leads to the formation of a step at the GB plane following the loop formation, accounting for the formation of a residual GB defect. A $1/2\langle111\rangle$ loop, with a $\{110\}$ habit plane, is emitted with further increase of the He bubble size in the $\Sigma3$ GB. In contrast, the sequential insertion of He atoms in $\Sigma73b$ GB continuously emits self-interstitial atoms (SIAs), but these SIAs rearrange at the core of the inherent GB dislocation, instead of forming a dislocation loop, which leads the GB dislocation to propagate along the $[\bar{1}\bar{1}12]$ direction. In the bulk and $\Sigma3$ GB, the He bubble exhibits three-dimensionally spherical shape, but it forms longitudinal shape along the dislocation line in the $\Sigma73$ GB, a shape commonly observed at GBs in experiments.

1. Introduction

It is well known that grain boundaries (GBs) and interfaces are effective sinks for radiation-induced defects [1–3]. Bai and co-workers have investigated defect-GB interaction mechanisms in copper and shown that GBs capture interstitials and inject them back into the lattice to recombine with vacancies that are within a few nanometers of the GB, leaving a healed crystal [4]. Because of the extremely low solubility of helium (He) in metallic materials, high concentrations of He created by transmutation are known to induce the formation of He bubbles at pre-existing and radiation-induced defects, and significantly degrade the mechanical properties of first wall structural materials [2,5–7]. The formation of He bubbles in the bulk or at GBs remains one of the most important issues in nuclear fusion technology, and He accumulation, both in the bulk and at GBs, has major consequences for the structural integrity of nuclear materials. Sefta et al. have investigated the tungsten surface evolution due to helium bubble nucleation, growth and rupture using molecular dynamics (MD) simulations [8]. MD simulations of helium implantation into single-crystalline tungsten were performed and it was found that the clusters were nucleated via the creation of $\langle111\rangle$ crowdion interstitials and interstitial dislocation loop punching [9]. The growth of helium clusters in titanium crystals was simulated using MD, and defect escape from the helium cluster was observed at certain points in the growth process [10]. At the microscopic level, understanding the clustering mechanism of He atoms, the formation of He–vacancy (He–V) clusters and the growth of a He cluster into a bubble is a prerequisite for simulating more complex phenomena at longer time scales and larger He concentrations. Zhang et al. investigated the energetic landscape and diffusion of He at grain boundaries in α-Fe from first principles and found the diffusion details and precise paths are GB dependent [11].

The nucleation of He clusters and bubbles in the bulk and at the GBs in α-Fe has been previously studied to understand the formation and evolution of these defects, and the effects of these phenomena on microstructural changes [12–16]. These investigations showed that the evolution of GB structure, the accumulation of He atoms and the nucleation of He bubbles all depend on the He concentration, irradiation temperature and the original

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http://dx.doi.org/10.1016/j.actamat.2015.06.055
1359-6454/

GB structure. In our previous work we found that ⟨1 0 0⟩ dislocation loops can directly form in the region between large He clusters in the Σ3 GB at 600 and 800 K [8]. At high He concentrations, a large number of self-interstitial atoms (SIAs) are emitted from He clusters and these SIAs are able to rearrange to form an extra atomic plane within the Σ3 GB, resulting in the self-healing of the deformation induced by He accumulation and GB migration. In contrast, the large number of SIAs emitted by He clusters in a Σ73b GB causes climb of intrinsic GB dislocations parallel to the GB plane, and this climb becomes more significant with increasing temperature. In bulk Fe, at low He concentrations, SIA clusters (or dislocation loops formed from SIA clusters) are attached to He–V clusters, while at high He concentrations He–V cluster-loop complexes with more than one He–V cluster are formed. In these studies [13,14]. He atoms were randomly positioned at tetrahedral sites in the bulk or the stressed region near the GB plane of α-Fe, mimicking the He accumulation at different concentrations. In the present work, He atoms are inserted one by one to study the growth of a He cluster and the formation and evolution of SIAs in the bulk and at two GBs (Σ3 and Σ73b) in α-Fe, thus providing information on the detailed mechanisms controlling nucleation and growth of He bubbles. It should be noted that Frenkel pairs are also produced within fusion reactor conditions. There are two possible mechanisms that give rise the nucleation of He bubbles, i.e. vacancy-enhanced nucleation of He bubbles and one due to He accumulation in the bulk and at GBs. The present study focuses on the later case to understand the clustering mechanism of He atoms and the nucleation of He–vacancy (He–V) clusters.

2. Simulation procedure

The potentials used in this work are the same as those used in Ref. [8]. The Fe–Fe, Fe–He and He–He interactions are described by the interatomic potentials of Ackland et al. [17], Gao et al. [18] and Aziz [19], respectively. This set of potentials provides a good description of bulk properties and defect properties of the Fe–He system, which are in agreement with experimental data and *ab initio* calculations. The ⟨1 1 0⟩ Fe–Fe dumbbell configuration is the most stable interstitial. Dynamically, the ⟨1 1 0⟩ interstitial may change to the ⟨1 1 1⟩ direction, followed by faster one-dimensional migration. The Fe–He potential gives the binding energies of small He–V and He–He clusters in good agreement with those obtained by *ab initio* and other potential calculations.

An MD box of $30a_0 \times 30a_0 \times 30a_0$ with 54,000 Fe atoms is used to simulate the formation and growth of He clusters in bulk iron, where a_0 is the lattice constant of a perfect bcc iron (2.8553 Å). Periodic boundary conditions are applied along all three directions. Two symmetric tilt GBs with a common ⟨1 0 1⟩ tilt axis are constructed to investigate the formation and growth of He clusters in GBs that exhibit markedly different structure and energy. They are the Σ3 {1 1 2} Θ = 70.53° and the Σ73 {6 6 1} Θ = 13.44°, where the minimum energy atomic structures are described in Ref. [8]. The block sizes of the Σ3 and Σ73b GBs are 84.070 Å × 79.00 Å × 80.69 Å with 45,560 Fe atoms and 103.50 Å × 70.00 Å × 80.76 Å with 48,000 Fe atoms, respectively. For the two GBs, periodic boundary conditions are imposed along the directions parallel to the GB plane, but fixed boundary conditions are applied normal to the GB plane.

The NVT (constant number of atoms, volume and temperature) ensemble is applied in the present simulations with a time step of 1 fs. Helium atoms are continuously inserted one by one into the same region of the simulation box. After each He atom insertion, the configuration is quenched to 0 K, followed by temperature rescaling to 300 K and annealing for an additional 100 ps at that temperature. A similar procedure is repeated during the simulations. A similar approach was employed to study SIA emission by a He cluster [20], but with a much fast rate of introducing He atoms. We have carefully tested the rate of introducing He atoms, and found that the rate has small effects on the number of He atoms in a cluster that emits a self-interstitial atom (SIA). The nucleation and growth of He bubbles in the present simulations are closely associated with emission of SIAs, and thus the rate would not have significant effects on the results presented in this study. Interstitials and vacancies are identified and counted using a displacement analysis method [21]. If an Fe atom is displaced from its lattice site by at least $0.3a_0$, this atom is considered to be an SIA; if there is no atom within $0.3a_0$ of a lattice site, the site is taken to be a vacancy.

3. Results and discussion

3.1. He cluster evolution in bulk iron

The growth of He clusters (or bubbles) in bulk Fe is investigated first, and the results are compared with those at GBs. Recent studies show that single He interstitials and small He clusters (He_2, He_3) quickly diffuse through lattice at 300 K [7], and thus, the simulation is initialized with four He atoms (He_4) located at a tetrahedral interstitial site in the centre of the simulation box. The He_4 cluster is also able to migrate away from the initial site without creating a SIA. Upon adding the fifth He, the He_5 cluster pushes the nearest Fe atom off its lattice site along the ⟨1 1 1⟩ direction creating more space within the He cluster, but the emission of a SIA does not occur. After gradually inserting two more He atoms, the displacements of the Fe atoms around the He cluster increase, as shown in Fig. 1(a). However, Fig. 1(b) clearly shows that an Fe atom is kicked out from the cluster when the number of He atoms increases to eight, forming a He_8V cluster and an SIA. The SIA appears as a ⟨1 1 1⟩ crowdion with a distance of about 3.5 Å from the centre of the He cluster, but the ⟨1 1 1⟩ crowdion can also transform to the ⟨1 1 0⟩ dumbbell during annealing. We observed that the minimum number of He atoms in the cluster required to create an SIA is slightly different from that of Refs. [20–23] at 300 K. With different potentials, Morishita et al. [22] found that the pressure of a six He atom cluster was large enough to push an Fe atom off from its normal site and create a Frenkel pair. Gao et al. also predicted that a He_6 cluster would create an SIA with ⟨1 1 1⟩ crowdion configuration [20], but Guo et al. observed that three He atoms were sufficient to create an SIA in their simulations [23]. Our result is close to the prediction of Ref. [24], in which a Frenkel pair can be produced by a cluster containing ten He atoms at 200 K, and nine He atoms at 400 K. The insertion of the eleventh He atom followed by annealing at 300 K causes a second SIA to be emitted with a ⟨1 1 0⟩ dumbbell configuration, as shown in Fig. 1(c). These SIAs collect at the periphery of the He–V cluster. Fig. 1(d) illustrates that after insertion of 13 He atoms, all the SIAs move to the same side of the cluster, rather than remaining uniformly distributed over the cluster surface, which is consistent with Morishita's observations in Fe [22] and Wilson's results in Ni [25]. A fourth SIA is emitted from a 15 He atom cluster and then the four SIAs spontaneously transform into $[1\bar{1}1]$ crowdions during annealing.

Insertion of additional He atoms results in the accumulation of more SIA atoms at the periphery of the cluster. Several snapshots of the configurations produced by continuously adding He atoms are shown in Fig. 2, where all the insets are viewed normal to the ⟨1 1 1⟩ direction. When the 23rd He atom is introduced, eight SIAs align into the same ⟨1 1 1⟩ direction as crowdions, forming a well-defined SIA cluster attached to the He–V cluster, as shown in Fig. 2(a), where the inset is the configuration of the SIAs, vacancies and He atoms projected on the {1 1 1} plane. A 1/2⟨1 1 1⟩ dislocation loop with $N_I = 16$ (the number of SIAs) is formed after the insertion of 35 He atoms. The inset in Fig. 2(b) shows the 16 SIA