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Atomistic simulations of helium clustering and grain boundary reconstruction in alpha-iron

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Abstract—The accumulation and clustering of He atoms at Σ3 ⟨110⟩ {112} and Σ73b ⟨110⟩ {661} grain boundaries (GBs) in body-centred cubic Fe, as well as their effects on GB reconstruction, have been investigated using atomic-level computer simulations. The accumulation of He atoms and the evolution of the GB structure all depend on local He concentration, temperature and the original GB structure. At a local He concentration of 1%, small He clusters are formed in the Σ3 GB, accompanied by the emission of single self-interstitial Fe atoms (SIAs). At a He concentration of 5%, a large number of SIAs are emitted from He clusters in the Σ3 GB and collect at the periphery of these clusters. The SIAs eventually form ⟨100⟩ dislocation loops between two He clusters. It is likely that impurities may promote the formation of ⟨100⟩ loops and enhance their stabilities in α-Fe. At a He concentration of 10%, the large number of emitted SIAs are able to rearrange themselves, forming a new GB plane within the Σ3 GB, which results in self-healing of the GB and leads to GB migration. In contrast to the Σ3 GB, He clusters are mainly formed along the GB dislocation lines in the Σ73b, and the emitted SIAs accumulate at the cores of the GB dislocations, leading to the climb of the dislocations within the GB plane. As compared to bulk Fe, a higher number density of clusters form at GBs, but the average cluster size is smaller. The product of cluster density and average cluster size is roughly constant at a given He level, and is about the same in bulk and GB regions, and varies linearly with the He concentration.

Keywords: Grain boundary; Atomistic simulation; Nucleation of He clusters; bcc Fe

1. Introduction

Because of the extremely low solubility of helium (He) in metallic materials, high concentrations of He created by nuclear transmutation reactions are known to induce the formation of He bubbles, which are associated with the pre-existing and radiation-induced defects and microstructural features such as dislocations and grain boundaries (GBs). In sufficient concentration He can significantly degrade the mechanical properties of materials. In particular, the nucleation and growth of He bubbles at GBs may lead to high-temperature embrittlement, which is manifested by severe loss of creep–rupture strength [1,2]. Production of He in structural materials employed in the fusion nuclear environment will be significant and represents one of the most challenging materials problems to resolve for fusion power generation. The nucleation of He bubbles at microstructural features in metals has been studied using experimental approaches and theoretical methods. Evans et al. observed the formation of He clusters at ⟨100⟩ GB dislocations of low-angle tilt GBs in irradiated molybdenum [3]. Baskes et al. [4] investigated the trapping of He at GBs in nickel using atomistic computer simulations and found that GBs are important trapping sites for He atoms. Furthermore, the size of bubbles at GBs in copper formed by He implantation was found to increase with increasing GB energy [5], which underscores the importance of GB structure. The nucleation behavior of He bubbles in single-crystal and nanograin body-centred cubic (bcc) molybdenum has been investigated using molecular dynamics (MD) simulations [6]. These simulations showed that strong precipitation of He occurs at the GBs in nanograin molybdenum and the density, size and spatial distribution of He bubbles vary with GB structure. Helium bubble nucleation at low-angle twist boundaries in gold has been investigated [7,8] and it was found that GBs with well-defined structures (pure twist boundaries) could trap He atoms, forming a superlattice of small bubbles at GB dislocation nodes.

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Reduced-activation ferritic/martensitic (RAF/M) steels are leading candidate structural materials for future fusion reactors and He concentrations on the order of 2000 at ppm will be produced by end-of-life. Consequently, the formation of He bubbles at GBs in steels or Fe has been widely investigated. Lane et al. [9] studied He bubble nucleation at GBs in a ternary austenitic steel after He implantation using transmission electron microscopy (TEM), while Randle et al. [10] investigated the interaction mechanisms of He bubbles with GBs in an austenitic steel using TEM. Dynamic interaction of He bubbles with GBs in Fe and Fe–9Cr was also studied by in situ electron microscopy and thermal desorption spectroscopy (TDS) [11]. Novel experimental techniques have been employed to characterize the transport and fate of He in RAF/M steels under fusion relevant He injection coupled with neutron-induced displacement damage [12]. In addition, MD methods have been extensively employed to study the atomic-level processes of He behavior at GBs in bcc Fe. We have previously studied the diffusion mechanisms of He interstitials and di-He clusters at GBs in α-Fe using MD simulations [13–15]. We discovered that He atoms diffuse one-dimensionally along specific directions in the Σ11 GB, whereas in the Σ3 GB they migrate one-dimensionally at low temperature, two-dimensionally at intermediate temperature, and three-dimensionally at high temperature. The different migration behavior of He in the two GBs indicates that He diffusivity is sensitive to the GB structure. Furthermore, the binding energies of He atoms to GBs in bcc Fe were found to increase with increasing excess volume of GB [16]. Terentyev et al. also studied the migration of a He interstitial in ⟨1 1 0⟩ tilt GBs in α-Fe [17], finding that GB atomic structure plays an important role in the accommodation, migration mechanism and diffusivity of He atoms.

In our previous work, the clustering of interstitial He in bulk Fe as a function of He concentration was investigated using MD simulations [18]. At low He concentrations, self-interstitial atoms (SIAs) emitted from He clusters remain attached to the clusters, while at higher He concentrations cluster–interstitial dislocation loop complexes with more than one He–vacancy (He–V) cluster are formed, in good agreement with experimental observations [19]. In the current study, we explore He accumulation in a Σ3 ⟨1 1 0⟩ {1 1 2} and a Σ73b ⟨1 1 0⟩ {6 6 1} GB compared to bulk bcc Fe using MD simulations at different local He concentrations and temperatures. Here we focus on the effects of GB structure, temperature and He concentration on He clustering and the formation of dislocation loops, as well as GB structural evolution.

2. Simulation procedure

In the present simulations, a modified version of the MOLDY computer code [20] is employed. The interatomic potentials by Ackland et al. [21] and Aziz [22] are used to describe the Fe–Fe and He–He interactions, respectively, while the Fe–He interaction is described by the potential developed by Gao et al. [18], which is based on the electronic hybridization between Fe *d*-electrons and He *s*-electrons. The migration energy of a single He interstitial, as calculated with this potential, is consistent with that obtained by ab initio methods, and the binding energies of small He–V and He–He clusters are also in good agreement with those obtained by ab initio and other interatomic potential calculations. The diffusion properties of He interstitials and their clusters in bulk Fe have been studied using MD [23]. The results showed that small He clusters can migrate at low temperatures, but they can emit SIAs and are trapped by the resultant vacancy, forming He–V clusters, at higher temperatures.

Two symmetric tilt GBs are considered in this work, both with a common ⟨1 0 1⟩ tilt axis. The two GBs are Σ3 {1 1 2}, $\Theta = 70.53°$ and Σ73b {6 6 1}, $\Theta = 13.44°$. These two grain GBs have the same tilt axis, but different angles. Σ3 ⟨1 1 0⟩ {1 1 2} with $\Theta = 70.53°$ and Σ73b {6 6 1} with $\Theta = 13.44°$ represent high- and low-angle tilt GBs, respectively. Their atomic structures are shown in Fig. 1a and b, respectively. Periodic boundary conditions are imposed along the x and z directions, where crystal coordinates corresponding to the x, y and z directions in the two GB models are indicated in Fig. 1. Atoms in region F are rigidly fixed in their original positions, whereas the atoms in region T are used to control temperature by performing a velocity rescaling every 200 time steps according to the difference between the current temperature and the desired temperature. Regions V indicate vacuum slabs. The thickness of each slab, except for region T, is larger than the cut-off distance of the interatomic potential (0.53 nm). A bulk Fe model is also simulated for comparison. The dimensions and the number of Fe atoms in each model are given in Table 1, along with the number of He atoms included to simulate 1%, 5% and 10% local concentrations (see below).

The NVT (constant number of atoms, volume and temperature) ensemble is used in the present simulations with a time step of 1 fs. Initially, GB models are relaxed at 0 K and zero pressure with a molecular statics (MS) approach, and then the excess atomic volume (Ω_{xs}) of the two GBs is determined. The excess atomic volume (EAV), which can provide information about the width of the grain boundary, is found to affect the He binding energy and accumulation in MD simulations. In addition, EAV may be also used in other simulation methods such as kinetic Monte Carlo. As shown in Fig. 1c and d, the excess atomic volume of the Σ73b GB is about 10 times larger than in the Σ3 GB, particularly at the cores of the GB dislocations. Due to its extremely low solubility He is strongly attracted to regions of high EAV. From Fig. 1c and d it is apparent that regions with elevated EAV occur within ±0.4 nm of the Σ3 GB plane, and ±0.5 nm of the Σ73b GB. He atoms are inserted at random positions within these EAV regions to create local He concentrations of 1%, 5% or 10%. Here the local He concentration is defined as the ratio of the number of He atoms to the number of Fe atoms within the EAV region. In order to compare results from the Σ3 GB with those in bulk Fe, He atoms at approximately the same concentration are randomly added to a region with a thickness of ~0.8 nm along the [0 0 1] direction in bulk Fe. The numbers of He atoms for all the models are given in Table 1. After He insertion, the cell is relaxed at 0 K to achieve a minimum energy configuration, followed by temperature rescaling to the required annealing temperature and held there for 1 ns. Note that the simulation cell volume is held constant at the 0 K value, which is expected to introduce a negligibly small error in the results. Models are annealed at temperatures of either 300, 600 or 800 K. Helium clustering is monitored by counting the number and size of He clusters defined as those that are within a distance of 0.2 nm of each other.