8.5 ATOMISTIC STUDIES OF GROWTH OF HELIUM BUBBLES IN α-FE—F. Gao, L. Yang, and R. J. Kurtz (Pacific Northwest National Laboratory)

OBJECTIVE

The clustering and growth of He bubbles in bcc Fe are simulated using molecular dynamics. We found that the creation and evolution of defects due to the growth of He bubbles depends on the separation distance between helium bubbles. A 1/2<111> interstitial loop is always created by a single helium cluster, but a <100> loop is formed under special conditions.

SUMMARY

Experimental results and atomistic simulations demonstrate that nucleation and growth of He bubbles in α-Fe will contribute to embrittlement and is a significant material problem for development of fusion power. Understanding the nucleation growth of He bubbles in steels is one of the most important issues in nuclear fusion technology. In order to understand the formation and evolution of self-interstitials (SIAs) caused by He bubble growth under He-rich/vacancy-poor conditions, the clustering of He and growth of He bubbles in bulk Fe is currently being investigated by inserting He atoms one by one into one or two He clusters. We find that a 1/2 <111> dislocation loop is formed and eventually punched out by a single He cluster, but when two He clusters are present the configuration of the SIA loop formed depends on the distance between the two He clusters. A <100> cluster is created when the distance between the He clusters is less than about $6a_0$ and eventually grows into a <100>{100} dislocation loop.

PROGRESS AND STATUS

Introduction

Ferritic/martensitic steels and alloys have been used for structural materials in current nuclear fission reactors and proposed as candidate first wall materials in future fusion energy facilities. Thus, a large number of experimental and computer simulation studies have focused on the irradiation damage of Fe-based bcc materials [1,2], including the production of self-interstitial atoms (SIAs), vacancies, He and H atoms. The behavior of He in irradiated materials is of interest because high concentrations of He created by transmutation are known to induce the formation of He bubbles with existing and radiation-induced defects, and significantly degrade the mechanical properties of first-wall structural materials.

Modeling of the nucleation and growth of He bubbles in irradiated metals began a few decades ago. Atomic simulation is a useful tool to explore the kinetic process of initial clustering of He to a nanometer sized bubble. Processes of He bubble growth in bcc Fe and Fe-alloys have been simulated at the atomistic level. By loading He atoms into a pre-existing bubble seed in pure Fe and an Fe-Cr alloy, 1/2<111> interstitial dislocation loops are emitted during the relaxation procedure [3]. After randomly adding He atoms in bcc Fe, which simulates a He-rich and vacancy-poor condition, SIAs or 1/2<111> interstitial dislocation loops appear due to the clustering of He [4]. In addition, the formation of 1/2<111> dislocation loops during He cluster growth has also been observed by inserting He atoms one by one. The transformation of the SIA clusters into a 1/2<111> dislocation loop is similar to the previous study in [3]. However, the numbers of SIAs created differ using different Fe-Fe, Fe-He and He-He interatomic potentials [5]. Up to now, only 1/2 <111> dislocation loops, due to the clustering of He, were reported in bulk Fe and Fe alloys in the simulations. However, after He implantation, two sets of interstitial dislocation loops, with Burgers vectors of 1/2<111> and <100>, were identified in oxide dispersion strengthened ferritic steel using transmission electron microscopy (TEM) [1]. Also, the predominant types of loops found in irradiated reduced activation ferritic/martensitic (RAFM) steels were <100> {100} and ½<111>{111} [5]. We have studied the accumulation and clustering of He atoms at grain boundaries (GBs) in bcc iron. Some <100> dislocation loops are directly formed at the GB that are confined between He clusters, and they are very stable, even at high temperatures. However, the formation mechanism of those <100> dislocation loops is unclear.

Simulation Methods

The potentials used in this work are the same as those used in Ref. 6. The Fe-Fe, Fe-He and He-He interactions are described by the interatomic potentials of Ackland et al., Gao et al., and Aziz, respectively. An MD box of $50a_0 \times 50a_0 \times 50a_0$ with 250,000 Fe atoms is used to simulate the formation and growth of He clusters in bulk iron, where a_0 is the lattice constant of perfect bcc Fe (2.8553Å). Periodic boundary conditions are applied along all three directions. The NVT (constant number of atoms, volume and temperature) ensemble is chosen in the present simulations with a time step of 1 fs. At first, the growth of one He bubble in the box is simulated by the following procedure: He atoms are inserted one by one into the He cluster. After each He atom insertion, the configuration is quenched to 0 K, followed by a temperature rescaling to 300 K and annealing at that temperature for 100 ps. For the insertion of the next He atom, the configuration is quenched back to 0 K and the procedure is repeated. The growth of two He bubbles in the simulation box are investigated using a method similar to the one above, but He atoms are inserted one by one into the two He clusters, respectively.

Results and Discussion

<u>One He ClusterEvolution</u>

We firstly investigated the growth of one He cluster (or bubble), which corresponds to the condition of a low density of He bubbles growing in bulk Fe. Recent studies show that single He interstitials and small He clusters (He_2, He_3) quickly diffuse through the lattice at 300 K, and thus, the simulation is initialized with four He atoms (He_4) located at a tetrahedral interstitial site in the center of the simulation box. It is found that the He_4 cluster is also able to migrate away from the initial site without creating an SIA. Upon adding the fifth He, the He_5 cluster pushes the nearest Fe atom off its lattice site along the <111> direction, creating more space for the He cluster, but the emission of an SIA does not occur. After inserting two more He atoms, the displacements of the Fe atoms around the He cluster increases. However, 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 <111> crowdion at a distance of about 3.5 Å from the center of the He cluster, but the <111> crowdion can also transform to the <110> dumbbell during annealing. We observed that the minimum number of He atoms in the cluster required to create an SIA is slightly different from other simulation results. With different potentials, Morishita et al. [7] 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. [8] also predicted that a He_6 cluster would create an SIA with <111> crowdion configuration, but Guo et al. [9] observed that three He atoms were sufficient to create an SIA in their simulations. Our result is close to the prediction of Stewart et al. [10], 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 <110> dumbbell configuration. These SIAs collect at the periphery of the He-V cluster. 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 [7] and Wilson's results in Ni [11]. 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 SIAs at the periphery of the cluster. Several snapshots of the configurations produced by continuously adding He atoms are shown in Figure 1, where all the insets are viewed normal to the <111> direction. When the 23rd He atom is introduced, eight SIAs align along the same <111> direction as crowdions, forming a well-defined SIA cluster attached to the He-V cluster, as shown in Figure 1(a), where the inset is the configuration of the SIAs, vacancies and He atoms projected on the {111} plane. A 1/2<111> dislocation loop with N_I = 16 (the number of SIAs) is formed after the insertion of 35 He atoms. The inset in Figure 1(b) shows the 16 SIA cluster configuration on the {111} plane, which arranges to form part of a hexagonal dislocation loop. The open circles represent empty positions in this loop. It should be noted that the dislocation loop is still attached to the He cluster. After the 41st He atom is inserted into the cluster, a dislocation loop with N_I =

18 is able to move away from the He cluster along a <111> direction. The configurations of the He cluster and nearby defects following 10 ps annealing are shown in Figure 1(c).

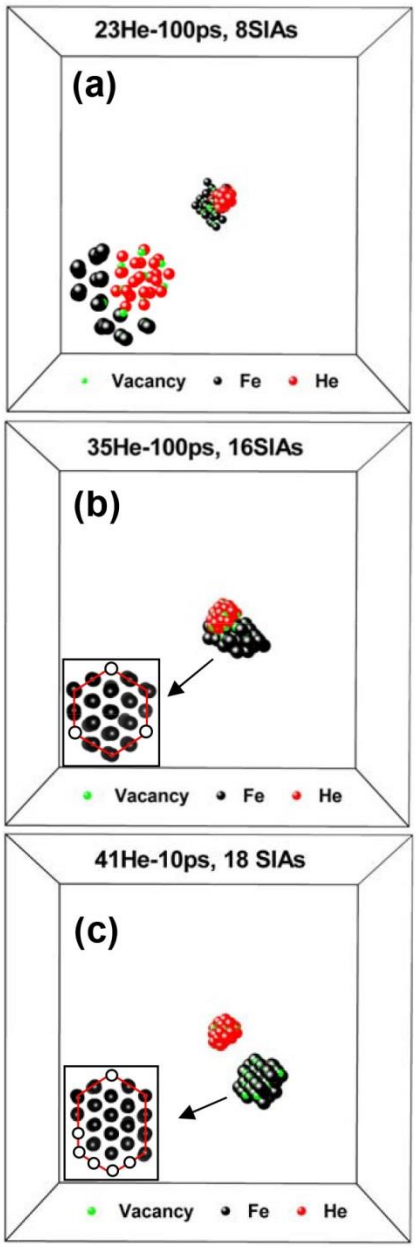

Figure 1. Atomic configurations of the He cluster and it's nearby displaced Fe atoms after adding He atoms followed by annealing 0.1 ns at 300 K: (a) 7He, (b) 8He, (c) 11He and (d) 13He, where large black, medium red and small green spheres present SIA, He and vacancies, respectively.

Two He Bubbles

Three different models involving two He clusters separated at different distances are considered. Two He clusters are inserted along the [001] direction with separation distances of $10a_0$ and $6a_0$, respectively, and another is inserted along the [111] direction with a separation distance of $6a_0$. Figure 2 shows three snapshots of the atomic configuration with two He clusters including 3 He atoms (a), 30 He atoms (b) and

62 He atoms (c) in each cluster, where the initial distance between the two He clusters is $10a_0$. Without special notation, the number of He atoms in the following text means the number in each He cluster. We find that by adding 30 He atoms, some SIAs are created with either <111> crowdion or <110> dumbbell orientation, resulting in SIA clusters on one side of the He clusters. After insertion of 62 He atoms, a large <111> SIA cluster with 28 SIAs forms near the lower He cluster, but an SIA cluster composed of <111> and <110> SIAs is attached to the top He cluster. The formation and evolution of SIAs are similar to those observed for growing He clusters in bulk Fe. It is of interest to note that adding one more He in each He cluster induces the SIAs near the He cluster (A) in Figure 3 to migrate along a <111> direction, joining the SIAs near the He cluster (B) to form an interstitial loop with 55 SIAs. The interstitial loop is much larger than that created by one He cluster consisting of 63 He atoms in bulk Fe. The Fe atoms at the periphery of the top He cluster almost return to their lattice sites. The configuration of defects around the lower He cluster, which is marked by the yellow circle in (a), is shown on the (100) and (111) planes in Figs. 3 (b) and (c), respectively. It is clear that the SIA cluster is a <111> dislocation loop. During growth of the He clusters, the centers of the two He clusters remain near their initial positions.

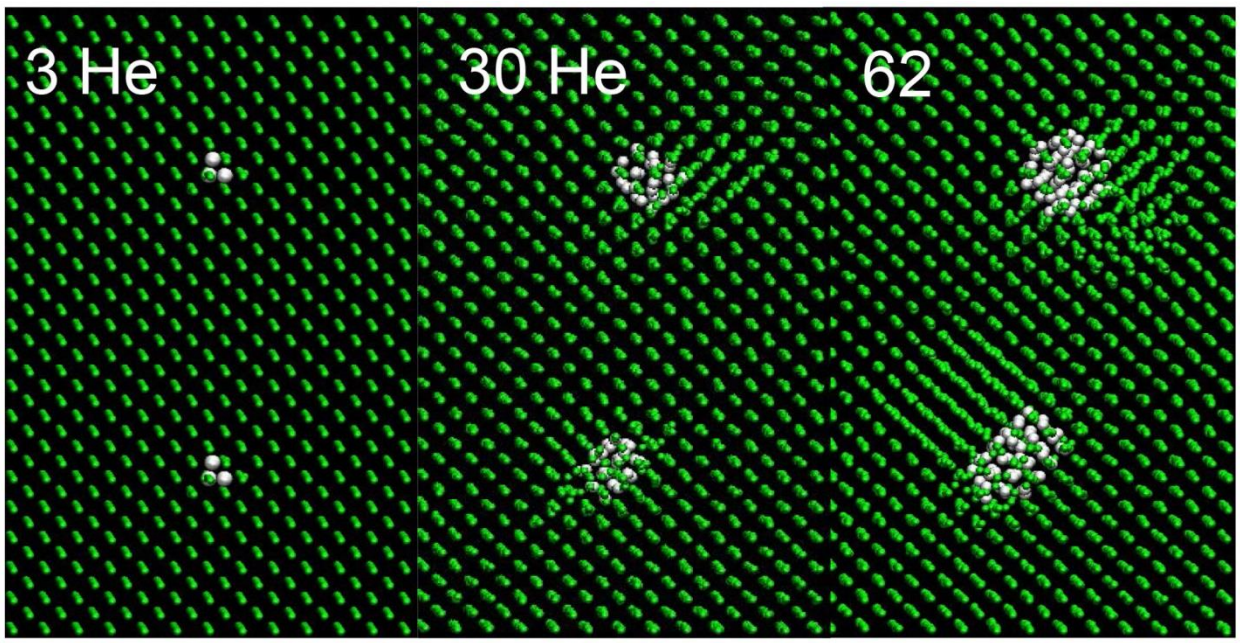

Figure 2. Configurations of the He clusters in bulk Fe, as viewed in the (001) plane, where green spheres are Fe atoms and grey He atoms: (a) 3 He atoms, (b) 30 He atoms, and (c) 62 He atoms in each cluster.

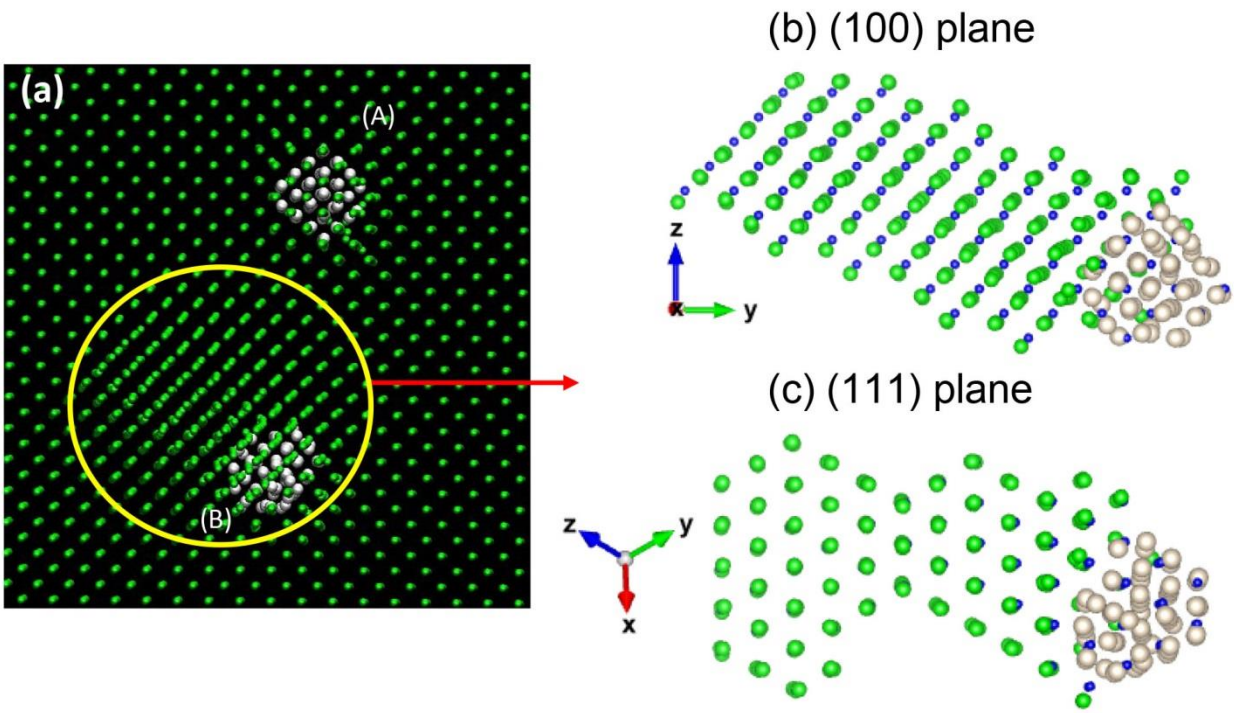

Figure 3. (a) Configurations of He clusters with 63 He atoms in bulk Fe, where green spheres are Fe atoms and grey He atoms. Configurations of defects in the region marked by yellow circle in (a): (b) view on the (100) plane and (c) on the (111) plane, where green spheres are Fe interstitial, blue and grey spheres are vacancies and He atoms, respectively.

By decreasing the initial distance between the two He clusters, a different defect evolution process is observed. Figure 4 shows the configurations of the He clusters and SIAs created for an initial distance of $6a_0$ between the two He clusters, as viewed on the (001) plane, where green spheres are Fe interstitials, and blue and grey spheres are vacancies and He atoms, respectively. It is clearly seen that after adding even He atoms to each He cluster, one of the He_7 clusters emits a Fe atom, forming a He_7V cluster and a <110> crowdion SIA, but the other cluster pushes the nearest Fe atoms off their lattice sites along the <111> direction and creates more space within the He cluster without emitting a SIA, as shown in Figure 4. The SIA initially appears as a <111> crowdion, but transforms to a <110> dumbbell during annealing. The SIA moves around the He-V cluster as the next He atom is added to both clusters. After the insertion of the 11th He atom, the two He clusters seem to attract each other and move closer. A third SIA is emitted to form a <110> dumbbell configuration. From Figure 4, it is of interest to note that most SIAs congregate in the region between the two He clusters after adding 13 He atoms. Three SIAs at the center between the two He clusters transform to <100> dumbbells, but the other surrounding SIAs maintain <111> and <110> configurations. Simultaneously, the lower He cluster further moves towards the top He cluster, decreasing the separation distance between the two clusters to 1.5 nm. With increasing numbers of He atoms, more <111> crowdions transform to <100> dumbbells in the region between the two He clusters, but most SIAs remaining at the periphery of the two clusters are still <110> and <111> crowdions, which can be seen in the snapshots of 21 and 43 He atoms in Figure 4. As a result, a <100> SIA loop with 30 SIAs is directly formed after the insertion of 43 He atoms. The size of the <100> SIA loop increases with increasing numbers of He atoms in the clusters, because more <111> crowdions transforms to the <100> dumbbells. However, the emission of a dislocation loop does not occur even if the number of He atoms in the clusters reaches 63 atoms.

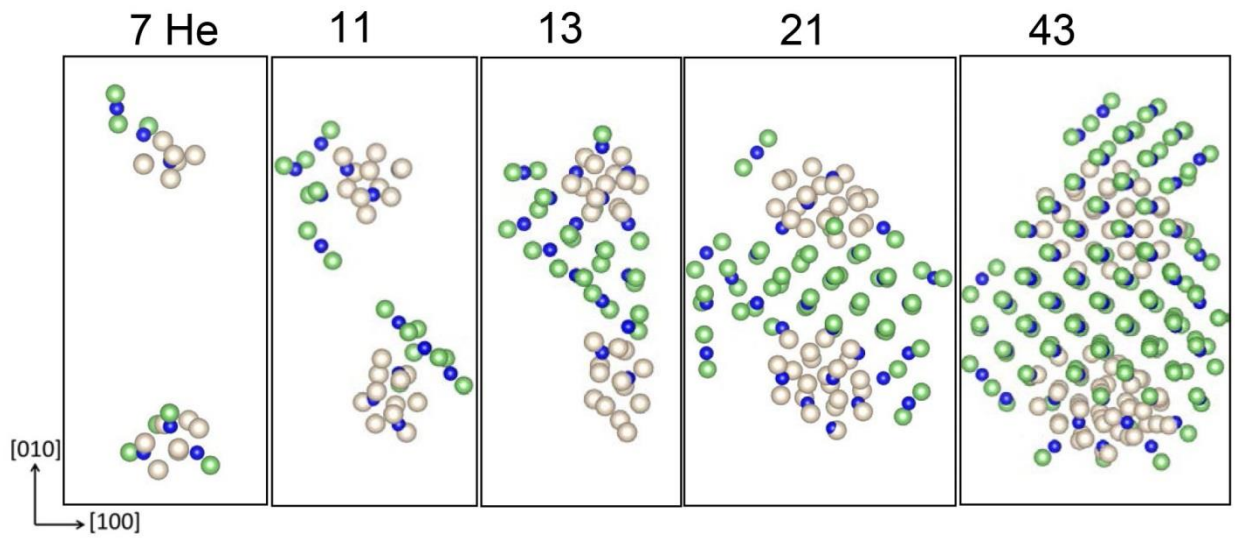

Figure 4. Configurations of He clusters and SIAs viewed on the (001) plane, where green spheres are Fe interstitials, blue and grey spheres are vacancies and He atoms, respectively.

CONCLUSION

The growth of He clusters or nano-scale bubbles in bulk Fe is studied by molecular dynamics approach. The evolution of SIA defects associated with the growth of He bubbles is analyzed in detail. It is found that the formation and evolution of SIA dislocation loops strongly depends on He cluster spacing. A 1/2 <111> dislocation loop forms at the periphery of a single He cluster and the loop is eventually emitted with increasing numbers of He atoms in the cluster. The growth and defect evolution of two He clusters is explored using a similar method, but He atoms are inserted one by one at the same time into both of the clusters. It is interesting to observe that the configuration of the SIA loops formed depends on the initial separation distance between the two clusters. A <100>{100} dislocation loop is created when the distance between the two He clusters is about $6a_0$, whereas a <111> loop forms when the initial cluster separation distance is $10a_0$.

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