ATOMISTIC STUDIES OF PROPERTIES OF HELIUM IN BCC IRON: COMPARISON OF FE–HE AND FE MATRIX POTENTIALS—David M. Stewart, Stanislav Golubov (Oak Ridge National Laboratory and the University of Tennessee), Yuri Ostesky, Roger E. Stoller, Tatiana Seletskaia, and Paul Kamenski (Oak Ridge National Laboratory)

SUMMARY

In fusion applications, helium caused by transmutation plays an important role in the response of RAFM steels to neutron radiation damage. We have performed atomistic simulations using a new 3-body Fe–He inter-atomic potential combined with the Ackland iron potential. The results are compared with older (Wilson) and more recent (Juslin-Nordund) Fe-He pair potentials, and with alternate iron matrices. With the ORNL potential, interstitial helium is very mobile and coalesces together to form interstitial clusters. We have investigated the mobility of these clusters. If the He cluster is sufficiently large the cluster can push out an Fe interstitial, creating a Frenkel pair. The resulting helium-vacancy cluster is not mobile. The ejected SIA is mobile, but is weakly trapped by the He–V cluster. If more helium atoms join the He–V cluster, more Fe interstitials can be pushed out, and they combine to form an interstitial dislocation loop. The reverse process is also studied. Multiple helium atoms can be trapped in a single vacancy, and if there are few enough, the vacancy can recombine with an Fe interstitial to create a helium interstitial cluster. These mechanisms are investigated together in larger simulations that examine the nucleation of He defects. Results are compared based on temperature, interatomic potentials used and helium concentration.

PROGRESS AND STATUS

Introduction

Helium produced in neutron irradiated iron plays an important part in its mechanical properties. The growth, migration and coalescence behavior of helium bubbles is very sensitive to the properties of individual He interstitials and helium-vacancy clusters [1]. A new He–Fe inter-atomic potential has been developed at ORNL, based on extensive fitting to first-principles calculations of point defects and clusters [2–5]. This potential has been used to investigate the properties of helium and helium-vacancy clusters in MD and MS simulations.

Helium diffuses very fast in the matrix, but is easily trapped in vacancies [6]. It is possible for a self-interstitial to recombine with the vacancy, kicking the helium back into an interstitial position. Previous calculations [5] showed that recombination is possible not only for a single substitutional He, but even when the vacancy contains multiple Helium atoms. If the vacancy contains 5 or less atoms, it is found to be energetically favorable for it to recombine with the SIA to form a helium interstitial cluster. For 6–8 atoms there is no clear winner, and for more than 8 it is more favorable for the SIA to be trapped close to a He–V cluster without recombining with it.

A dynamic simulation of a He_8 interstitial cluster at 600K showed that the reverse process (i.e. Frenkel pair formation) can happen—an iron atom is pushed out of its lattice site, creating a He_8V cluster and an SIA. The SIA was trapped beside the cluster.

Simulation Method

The general procedure followed is: Generate perfect BCC lattice. Introduce the defect(s) to be studied. Relax at constant volume using a mixture of conjugate gradient and simulated annealing, and save the atom positions in units of the lattice constant. Start the MD simulation.

The MD simulations used NVE dynamics. The lattice constant and initial velocities were chosen to give close to zero pressure and the desired initial temperature. The boundary conditions are periodic in X, Y and Z, which are $\langle 100 \rangle$ directions. The velocity Verlet algorithm with a timestep of 0.3fs is used. As volume and temperature correction are not used, when processes that release energy are simulated the temperature and pressure both rise during the simulation.

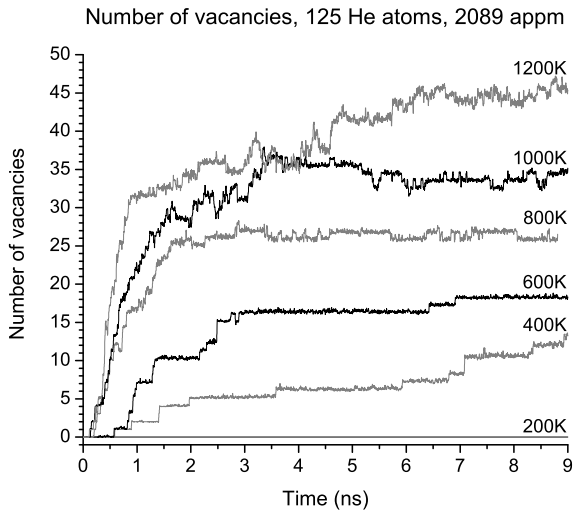

Figure 1. Vacancy production

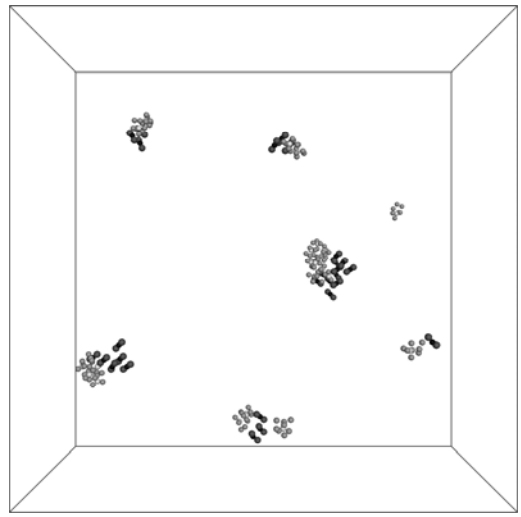

Figure 2. Clusters at 800K at t = 3.2ns

Coalescence

In order to study the process of Frenkel pair formation by helium interstitial clusters, dynamic simulations were run with 125 helium atoms in a 31×31×31 BCC iron matrix (60,000 iron atoms). At 200K, there was insufficient kinetic energy to break up even a pair of He atoms, so the helium slowly and inexorably coalesced until it formed interstitial clusters too big to be mobile, He_4 or bigger. The largest observed cluster was He_8. No vacancies or SIAs were observed. At 400K, He_2 and He_3 were still mostly stable but more clusters were mobile so coalescence happened faster. When clusters reached 8 or 9 helium atoms, a single SIA was ejected. None of the ejected SIAs escaped their HeV cluster. At higher temperatures, smaller clusters like He_2 and He_3 were short lived, reducing the number of surviving clusters. The clusters that did form were bigger since the number of available He atoms was fixed at 125. The higher the temperature, the less He atoms were needed to eject an SIA. Higher temperatures also led to more SIAs escaping the HeV cluster where they were created. These SIAs were usually captured by larger clusters that had SIAs. The SIAs were observed to form small dislocation loops.

The number of vacancies (equal to the number of SIAs ejected) is plotted as a function of time in Fig. 1. A snapshot of the 800K simulation after 3.2 nanoseconds is shown in Fig. 2. All the helium has coalesced into 9 clusters, all of which have pushed out from 1 to 6 SIAs. The SIAs have formed interstitial loops beside some of the clusters.

Similar simulations were run with different potential combinations. Results for Ackland [7] + ORNL [2–5], Mendelev [8] + ORNL [2–5] and Ackland [7] + Wilson [9] are shown in Fig. 3. With Ackland + Wilson, Frenkel pairs formed much faster than they did with the ORNL potential, and nearly twice as many were formed. This is because an SIA can be ejected from an interstitial cluster of only 2 or 3 Heliums. The Mendelev + ORNL combination formed bubbles at a rate and quantity that was between the other two combinations. For Ackland + Juslin–Nordlund [10] (not shown on the graph), the helium remained separated as interstitial atoms and Frenkel pairs were not created.

In order to see the effect of the helium concentration, additional simulations were run with the same number of helium atoms (125) in larger box of 40×40×40. This gives 128,000 iron atoms and thus a concentration of 976 appm He. The number of vacancies created for the 600K run is shown in Fig. 4. The bubbles took longer to form when the concentration was lower, but the number and size distribution was similar.

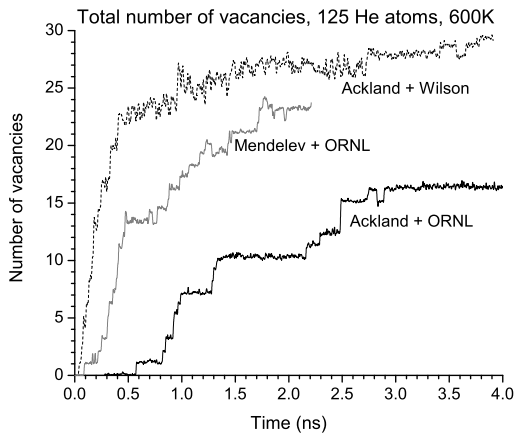

Figure 3. Coalescence for different potentials.

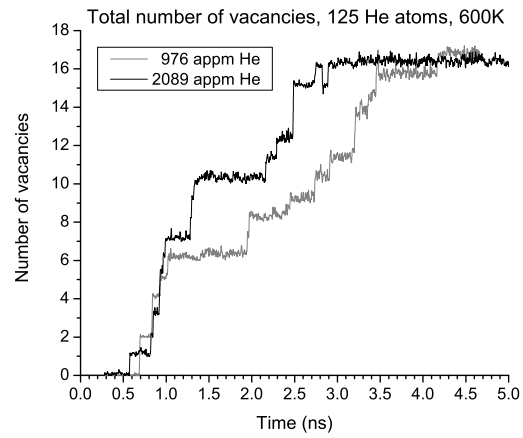

Figure 4. Coalescence for different box sizes.

Cluster Diffusion

The rate of diffusion of helium interstitial clusters was also measured. Simulations of clusters consisting of up to 6 He atoms were run for 15ns in a 10×10×10 box at a range of different temperatures. In all cases, the clusters do one of three things:

- remain as a cluster;
- dissociate into smaller clusters and/or atoms; or
- eject one or more SIAs.

The cluster diffusion rate is considered to be the diffusion rate of the centre of mass of the He atoms. An Arrhenius plot of the rates is shown in Fig. 5. When a cluster remains intact, the diffusion rate is calculated using the whole simulation, and shown as a solid symbol in Fig. 5. In many cases, a cluster dissociates or ejects an SIA only after a considerable amount of time. In these cases, the diffusion rate is calculated from the simulation up to that point, and shown as empty symbols in Fig. 5.

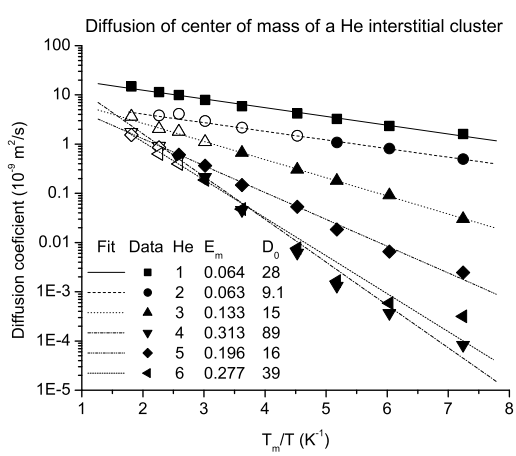

Figure 5. Arrhenius plot for Ackland + ORNL.

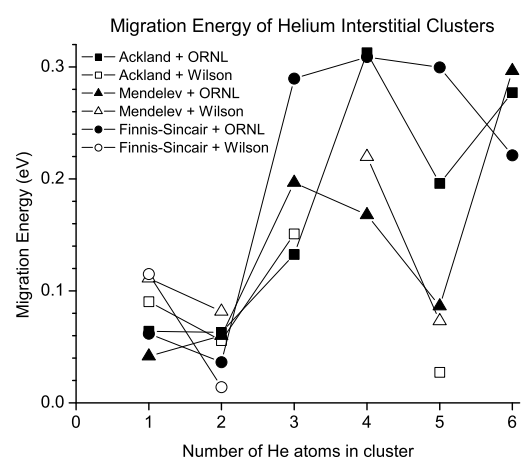

Figure 6. Migration energy results.

Arrhenius fits are done for several different combinations of matrix and He–Fe potentials, and the resulting energy barriers are plotted in Fig. 6. The test matrix for this measurement was 2 He–Fe potentials times 3 Fe matrix potentials times 6 He interstitial cluster sizes times 9 different temperatures.

The Juslin–Nordlund He–Fe potential predicts that all clusters will dissociate into individual interstitial Helium atoms, so there are no cluster diffusion rates for this potential with any matrix.

The Ackland matrix potential with the ORNL He–Fe potential showed almost identical barriers of about 0.06eV for a single He and the He_2 pair. He_3 had the next lowest barrier and then He_5. He_4 and He_6 both had the highest barriers of about 0.3eV.

The Mendelev matrix with the ORNL He–Fe potential showed different behavior. The barrier for a single He was only 0.04eV while for He_2 remained at 0.06eV. But the next lowest was He_5 with a barrier of only 0.09eV. He_3 and He_4 showed very close diffusion rates and barriers of about 0.18eV. The highest barrier was He_6 with 0.3eV.

With the Finnis–Sinclair matrix [11] and the ORNL He–Fe potential, the He_2 cluster had a lower barrier than a single He. Clusters of size 3, 4 and 5 showed very close diffusion rates and barriers of about 0.3eV, while He_6 had a barrier closer to 0.2eV.

The Ackland matrix potential with the Wilson He–Fe potential also showed the He_2 cluster with a lower barrier than a single He. Surprisingly, the He_5 cluster had an even lower barrier of only 0.03eV, based on simulations below 400K. At 400K and higher, it ejects an SIA too quickly to calculate a diffusion rate. The He_3 cluster shows very strange behavior, repeatedly ejecting an SIA and recombining with it. This suggests that the two states have similar energy. That the SIA was always available to recombine is an artifact of the simulation, as it has periodic boundary conditions and a small size. The He_4 cluster remained intact for a long time, especially at low temperatures, but would eject an SIA rather than move. At all temperatures, the He_6 cluster ejected an SIA too quickly to calculate a diffusion rate.

The Mendelev matrix potential with the Wilson He–Fe potential again shows decreasing barriers from the single He to the He_2 cluster to the He_5 cluster. The He_4 cluster showed very slow diffusion—the barrier was 0.2eV, but it had an extremely low pre-exponential factor. Above about 400K He_4 and He_5 ejected an SIA too quickly the gather diffusion data. At all temperatures, the He_3 and He_6 clusters immediately ejected an SIA.

References

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