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Stability of helium bubbles in alpha-iron: A molecular dynamics study

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ABSTRACT

Molecular dynamics simulations were performed to estimate the dissociation energies of helium interstitials, vacancies and self-interstitial atoms from small helium–vacancy clusters. Several sets of empirical potentials have been tested and compared with available *ab initio* calculations in order to provide the best combination of potentials to study the stability of small helium bubbles. The behavior of the cluster seems to be better described using Ackland potential for the Fe–Fe interactions and Juslin potential for the Fe–He interactions. From the calculations, it appears that the dissociation energies mainly depend on the helium-to-vacancy ratio rather than the cluster size. The helium/vacancy crossover slightly varies with increasing number of vacancies, but the crossover defining the loop-punching regime decreases strongly with increasing cluster sizes.

1. Introduction

Ferritic steels are possible structural materials for future fusion reactors. During the operation of these reactors, materials are subjected to 14 MeV neutron irradiation, generating helium by transmutation reactions and simultaneously energetic displacement damage. High helium concentrations are known to induce the formation of He bubbles and consequently enhance void swelling. Helium may also lead to the modification of microstructural and mechanical properties such as high temperature embrittlement, surface roughening and blistering [1]. These deteriorations may result from the insolubility of He, which therefore tends to precipitate into vacancies, voids or grain boundaries.

Small He_nV_m clusters may play an important role in the nucleation of He bubbles. However the atomistic properties of He in metal are difficult to identify experimentally. Thus atomistic simulations such as molecular dynamics provide useful tools to study the formation and the stability of these clusters and their impact on moving dislocations, vector of plasticity [2].

Here we present the results of an empirical molecular dynamics study on the formation of small helium–vacancy clusters in bcc iron, which will provide insight into the growth of the bubble in materials.

2. Computational method

The modified molecular dynamics code MOLDY [3] has been used to study the formation of small helium–vacancy clusters. The calculations were carried out on a cluster of Apple Mac OS X computers with dual G5 2.0 or 2.5 Ghz processors.

The formation energies of the helium–vacancy clusters He_nV_m are evaluated using different empirical potentials. To describe Fe–Fe interactions, the potentials developed by Ackland et al. [4] or Dudarev et al. [5] are employed. The He–He interactions are described using the Beck potential [6]. And finally two different potentials have been used the Fe–He interaction, namely the Wilson–Johnson potential [7] and the newly developed Juslin potential [8]. The latter potential is a purely repulsive pair potential, which has been specifically designed to reproduced formation and migration energies of small helium–vacancy clusters in iron obtained by *ab initio* calculations. Here, the formation energies are defined as the difference in total energy between a crystal containing a defect and a perfect crystal of the same number of iron atoms with the corresponding number of helium atoms in a fcc structure. In the present calculations, the box size was set to $10a_0 \times 10a_0 \times 10a_0$, where a_0 is the lattice parameter. For all calculations periodic boundary conditions and constant volume were used. The clusters have been generated with the following procedure. Starting from a single vacancy, the iron atom with the highest potential energy is removed from the system. The formation energy of the divacancy is then calculated. By iterating this scheme, the size of the void V_m is successively increased up to m equal 15. For each void size, n helium atoms are introduced randomly up to a helium-to-vacancy ratio equal to 5. The system is subsequently relaxed using a gradient conjugate algorithm. For each ratio several initial random configurations are tested and the one with the lowest formation energy is kept. From the formation energies of the helium–vacancy clusters, the vacancy, helium and self-interstitial iron atom (SIA) binding energies have been calculated as previously

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defined by Morishita et al. [9,10]. The dissociation energies have been assumed to be the sum of the binding energy and the migrations energy of the corresponding defect.

3. Results

Fig. 1 shows the dissociation energies of a vacancy, an helium and a SIA from a helium–vacancy cluster He_nV_m for the different combinations of empirical potentials. All combinations of empirical potentials reveal the same trends, as revealed in previous studies [9,11], that is to say that the stability of He_nV_m clusters against thermal emission of vacancies or interstitial helium mainly depends on the relative proportion of helium and vacancies in a cluster. The dissociation energy of a vacancy from a helium–vacancy cluster increases with the helium density. It is the more likely at the lowest densities. On the contrary the dissociation of an helium atom decreases with the helium content. For a helium-to-vacancy ratio of 5, this energy remains in all cases largely positive, which indicates the tendency of helium to aggregate [12]. The dissociation of SIA's from He_nV_m clusters is clearly unfavored at low helium density, but when the density is high enough the dissociation of SIA becomes more favored than the dissociation of helium. Three different dissociation regimes can be defined. In the first one, corresponding to low n/m ratios, the dissociation of vacancies is the most favored process. The second regime occurs when the dissociation energy of helium becomes lower than the one of vacancy. The intersection of the two curves defines an optimal ratio with respect to the emission of vacancy or helium. From this point helium dissociation is highly favored against thermal emission of vacancies. At high helium density as the dissociation energy of SIA goes below the dissociation energy of helium, a third regime can be defined. It corresponds to the emission of SIA's, also called loop-punching regime [2]. The thermal stability of clusters is then a competition between the emission of vacancies, helium interstitials and SIA's, depending on the helium-to-vacancy ratio. When comparing potentials together, it can be clearly seen that Ackland potential gives dissociation energies for the SIA's about 1 eV higher than Dudarev potential. We can also notice that Juslin potential gives lower dissociation energies for the helium compared to Wilson–Johnson potential by about 1 eV. Consequently the intersections of the different curves change considerably from one potential to another one. Therefore as the domains of existence of each dissociation regime differ, the behavior of small helium–vacancy clusters vary a lot depending on which potential set is used. *Ab initio* calculations on small He_nV_m clusters (n and m up to 4) have pointed out that the crossover corresponding to the intersection of the dissociation energy curves for helium and vacancy should occurred for a ratio n/m around 1.3 and a dissociation energy around 2.6 eV [11]. According to this calculation, the Juslin potential seems more adequate than the Wilson–Johnson potential to describe this optimal ratio, although it underestimates it a little bit. Other recent DFT calculations have shown that the dissociation energy of SIA's remains high even for high helium content [13]. Hence only Juslin potential associated with Ackland potential gives satisfactory results concerning this issue, the loop-punching regime appearing for n/m ratio above 4–5 depending on the number of vacancies in

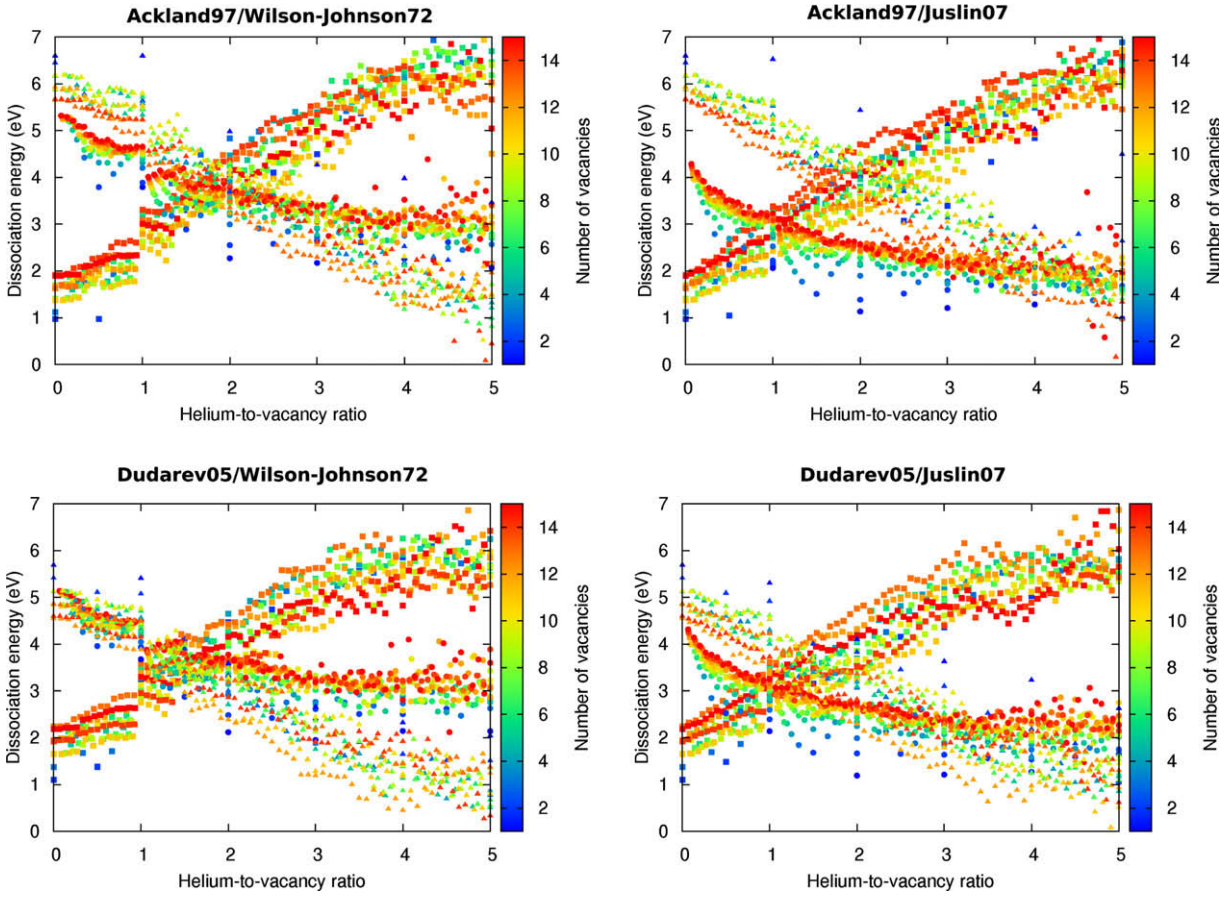

Fig. 1. Dissociation energies in eV of a vacancy (square), an helium interstitial (circle), and a self-interstitial atom (triangle) from a He_nV_m cluster as a function of the helium-to-vacancy ratio n/m. Several combinations of empirical potentials have been tested: Ackland97 [4] and Dudarev05 [5] for the Fe–Fe interactions, and Wilson–Johnson72 [7] and Juslin07 [8] for the Fe–He interactions.

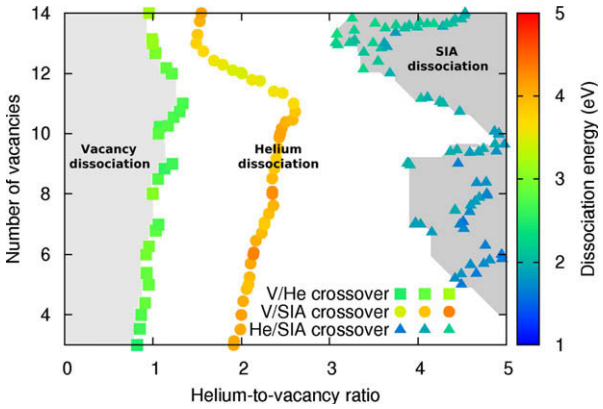

Fig. 2. Positions of the different crossovers as a function of the number of vacancies contained in a He_nV_m cluster using Ackland97 and Juslin07 potentials. The three different dissociation regimes have been highlighted.

the cluster. Actually, this last combination of potential compares in overall fairly well with the DFT calculations and seems particularly meaningful to describe helium–vacancy clusters.

Fig. 2 shows the position of the different crossovers of the dissociation energy curves as a function of the number of vacancies in a cluster for the Ackland97/Juslin07 potentials. The position of the vacancy/helium crossover varies slightly with increasing number of vacancies, with a maximum n/m ratio of 1.3 reached for 11 vacancies. From this point the ratio decreases. The helium density should normally decrease with increasing cluster size as suggested by a theoretical work [14] and experimental observation in electron energy loss spectroscopy [15]. The position of the vacancy/SIA crossover varies in the same way as the vacancy/helium crossover, with a maximum n/m ratio of 2.7 for 11 vacancies. The frontier between the helium dissociation regime and the SIA dissociation regime, i.e. the position of the helium/SIA crossover is fuzzy. It simply comes from the fact that when the helium density is too high, loop-punching occurred during the relaxation procedure of the cluster, leading to complexified formation energies. However the trend is clear, in that the helium density needed to favor the emission of SIA decreases strongly with increasing number of vacancies, from about 5 for 3 vacancies to 3 for 14 vacancies. According to the equation of state proposed by Trinkaus, the equilibrium helium pressure within a helium bubble must decrease when its size increases [15]. Therefore in order to keep an acceptable pressure within a cluster, it is expected that a lower n/m ratio is needed to emit a SIA when the cluster size increases. This points out that when a helium–vacancy cluster grows it becomes significantly easier to emit SIA's or SIA loops from the cluster.

4. Conclusion

The stability of helium–vacancy clusters in bcc iron has been investigated using a molecular dynamics method. The results show the importance of the choice of the potential to describe the different kinds of interactions in the system. When compared to recent DFT calculations, only the Ackland potential for Fe–Fe interactions combined with newly developed Juslin potential for Fe–He interactions gives a correct agreement. The results show that the dissociation energies of the different species to the helium–vacancy clusters greatly depend on the helium-to-vacancy ratio. Thus the thermal stability of the clusters is a competition between the emission of vacancies, helium interstitials and SIA's. At low helium density, the emission of vacancies is clearly favored. Above an optimal helium/vacancy ratio, helium dissociation is highly favored. This optimal ratio appears to slightly vary with increasing cluster sizes. When the helium density increases again, the emission of SIA's becomes more favored and loop-punching can occur. But contrary to the optimal helium/vacancy ratio, the helium/SIA crossover decreases strongly with the size of the helium–vacancy clusters.

Acknowledgments

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