Available online at www.sciencedirect.com
ScienceDirect
Journal of Nuclear Materials 367–370 (2007) 311–315
journal of nuclear materials
www.elsevier.com/locate/jnucmat

Atomistic modeling of helium interacting with screw dislocations in α-Fe

H.L. Heinisch *, F. Gao, R.J. Kurtz

Materials Science Division, P8-15, Pacific Northwest National Laboratory, P.O. Box 999, Richland, WA 99352, USA

Abstract

Formation energies, binding energies, and migration energies of interstitial He atoms in and near the core of an $a/2\langle 1\,1\,1\rangle$ screw dislocation in α-Fe are determined in atomistic simulations using molecular statics employing conjugate gradient relaxation and the dimer method for determining saddle point energies. The set of interatomic potentials employed is the same as used in many recent static and dynamic He–Fe simulations. Interstitial He atoms have a maximum positive binding energy of about 1 eV to the screw dislocation core relative to interstitial He atoms in the perfect crystal, which is about half that of binding energies of He to the edge dislocation. However, interstitial He atoms diffuse along the screw dislocation core with a migration energy of 0.4–0.5 eV, which is the same range as migration energies of interstitial He atoms along the edge dislocation.

1. Introduction

One of the most unique and important aspects of fusion materials development is the need to cope with the tremendous amounts of helium that will pervade the structure of the first wall and blanket regions of a fusion reactor. He impurities are well known for their adverse effects on mechanical properties of metals and alloys in a nuclear environment, such as the role of He in void swelling and high temperature embrittlement [1–4]. Thus, it is extremely important that we understand quantitatively the fate of He atoms with respect to the microstructural features with which they can interact. Toward that end, we have been employing molecular statics, molecular dynamics and the dimer method of potential surface mapping [5] to study the fate of He atoms in the vicinity of dislocations [6] and grain boundaries [7,8] in α-Fe, which is taken to be a first-order model for the advanced ferritic steels that are promising materials for fusion reactors.

Recently, Morishita et al. [9], using molecular dynamics, determined the migration energy of a single interstitial He atom in Fe to be 0.08 eV, a value that was also corroborated in our earlier calculations [6] using the dimer method and the same interatomic potentials. These results indicate that an interstitial He atom easily migrates through a perfect α-Fe lattice at temperatures below 100 K. Also, atomistic calculations by Morishita et al. [10] and Kurtz and Heinisch [7] showed that interstitial He is strongly trapped in a single vacancy in α-Fe

* Corresponding author. Tel.: +1 509 376 3278; fax: +1 509 376 0418.
E-mail address: hl.heinisch@pnl.gov (H.L. Heinisch).

0022-3115/$ - see front matter © 2007 Elsevier B.V. All rights reserved.
doi:10.1016/j.jnucmat.2007.03.035

as a substitutional atom with a binding energy of about 2.0 eV. Also, multiple He atoms were found to be strongly bound into vacancy clusters, increasing the lifetime of the vacancy clusters depending on the He-vacancy ratio [9]. We continue to use atomic-scale computer simulations to investigate the interactions of He atoms with extended defects, including dislocations [6] and grain boundaries [7,8]. In all these investigations it appears that the activation energies and diffusion mechanisms of interstitial He are strongly correlated to the binding properties of He atoms, which for interstitial He atoms is also correlated to the excess atomic volume available to the He. In the present paper we report the results obtained from atomistic modeling of substitutional and interstitial He in and around the core of the $a/2\langle 1\,1\,1\rangle$ screw dislocation in α-Fe.

In the present investigation the locations of the most stable configurations of the He atom–screw dislocation interaction are determined by calculating the formation energies of He atoms in interstitial and substitutional positions near the core of the screw dislocation. Also, the migration energies and migration mechanisms of interstitial He atoms near the dislocation and trapped within the dislocation core are determined using the dimer method for finding transition state energies.

2. Procedure

The computational model is similar in concept to that employed in the earlier calculations for He interactions with an edge dislocation [6]. A cylindrical cell of Fe atoms, periodic along the cylinder axis, with body-centered cubic structure is oriented such that the cylinder axis lies along the [1 1 1] direction. The cell is about 5 nm in diameter and 3 nm long, containing about 5400 atoms. An $a/2\langle 1\,1\,1\rangle$ screw dislocation was created along the central axis of the cell by displacing all the atoms, including the boundary atoms of the cell, according to the anisotropic elastic displacement field of the dislocation [11], then fixing the boundary atoms and relaxing the remaining moveable atoms to allow the dislocation core field to develop. Then He atoms or other defect complexes were placed within the cell and additional relaxations were performed to obtain the energies of the various configurations.

The interatomic potentials used for all the simulations were the set due to Ackland et al. [12], Wilson and Johnson [13], and Beck [14] for the Fe–Fe, Fe–He and He–He interactions, respectively. This set of potentials has been used in a number of recent studies [6–10], and their implementation is described in detail elsewhere [9,10]. More recent Fe–Fe interatomic potentials have been developed by Mendelev et al. [15]. They fitted parameters of their embedded atom type potentials to a number of experimental and calculated quantities including the results of *ab initio* calculations of point defect properties [16] which indicate that the $\langle 1\,1\,0\rangle$ dumbbell interstitial in Fe is more stable than the $\langle 1\,1\,1\rangle$ crowdion. Moreover, in contrast to the Ackland potential, the Mendelev potential predicts the core structure of the [1 1 1] screw dislocation obtained by *ab initio* methods [17]. Our preliminary testing of the Mendelev potential shows that numerical values of the results for the He–dislocation interactions may be somewhat different from those with the Ackland potential. However, we have used the Ackland potential for the present work reported here in order to compare results systematically with our earlier results for point defects, edge dislocations and grain boundaries [6–10].

Very recently, density functional theory calculations of some properties of He atoms in Fe [18] have produced He defect energies somewhat different from the potentials used here, and a new empirical He–Fe potential is being developed [19]. Unfortunately, the density functional calculations do not lend themselves easily to investigating such extensive, nonsymmetrical defect configurations as dislocations and grain boundaries. See Ref. [6] for further discussion of this point.

Binding energy calculations were performed for He atoms placed at substitutional and interstitial positions in the dislocation–distorted lattice at a number of locations along a line through the center of the dislocation. Conjugate gradient relaxations were performed to determine the relaxed configurations of the He and surrounding Fe atoms, as well as the energy of the relaxed configuration. The binding energies of the He atom to the dislocation are defined as the differences between the formation energy of a He atom in a perfect Fe lattice and the formation energies of the He atom at various locations in the 'dislocated' lattice.

Atomic volumes at atom sites at various distances from the dislocation were determined by the Voronoi volume method [20]. The Voronoi volume about an atom is defined as the volume that contains all points closer to the atom than to any other atom. It can be obtained by determining the volume of the polyhedron formed by the planes that

are perpendicular bisectors of the lines connecting the central atom to all other atoms (typically 2–3 neighbor shells). The excess volume is the difference between the atomic volume in the dislocated crystal and the atomic volume in the perfect crystal.

Migration energies of interstitial He atoms were determined using the dimer method [5] to determine saddle point energies for possible transitions of interstitial He atoms, especially for migration to other locations, starting from various initial positions near the dislocation. Both the energy barrier heights and saddle point atom configurations are obtained from the dimer calculations.

3. Results

Substitutional He. The binding energy of a substitutional He atom to the dislocation was determined as a function of the distance of the He atom from the center of the dislocation along the $[1\,0\,-1]$ direction, on a line running through the center of the dislocation. In Fig. 1 the substitutional He atom binding energies and the dislocation-induced excess atomic volumes in the same positions are plotted as a function of their distance from the center of the dislocation. There is a strong correlation between the excess atomic volume at the substitutional site and the binding energy of the substitutional He atom. The maximum binding energy occurs at positions near the center of the core with a value of about 0.25 eV, which is about half the value of the maximum binding energy of a substitutional He atom to the $a/2[1\,1\,1]$ $\{1\,1\,0\}$ edge dislocation in α-Fe [6].

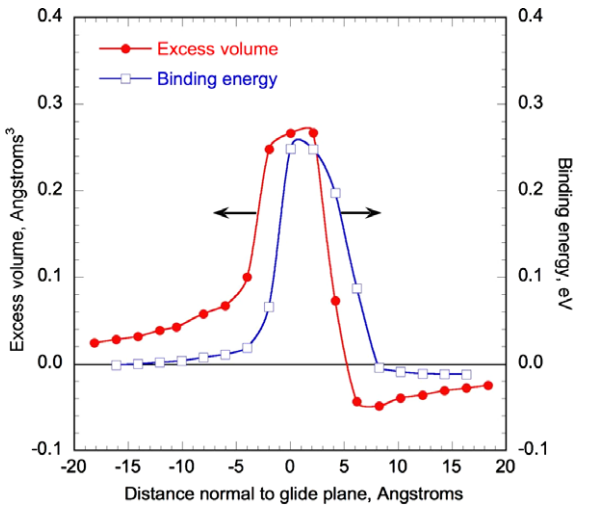

Fig. 1. Binding energies (right ordinate) of substitutional He atoms to the $a/2\langle 1\,1\,1\rangle$ screw dislocation in Fe as a function of their distance from the glide plane along a line through the center of the dislocation normal to the glide plane. Excess atomic volumes (left ordinate) along the same line through the dislocation are plotted for comparison.

Interstitial He. The binding energies of interstitial He atoms to the screw dislocation are shown in Fig. 2, plotted as a function of the distance of their initial unrelaxed positions relative to the dislocation line, similar to Fig. 1. The maximum binding energy of an interstitial He atom to the screw dislocation, which occurs in the core of the dislocation, is about 1 eV. This value is about four times larger than the maximum binding energy of substitutional He atoms to the screw dislocation (0.25 eV). However, the binding energy of interstitial He to the $a/2\langle 1\,1\,1\rangle$ screw dislocation is less than half the maximum binding energy of interstitial He to the $a/2[1\,1\,1]$ $\{1\,1\,0\}$ edge dislocation [6]. Also, unlike octahedral interstitial He in and near the edge dislocation, there are no locations near or in the screw dislocation core where octahedral interstitial He atoms spontaneously assume a crowdion configuration. Undoubtedly, this is related to the much smaller excess volume associated with the screw dislocation.

It is clear that He interstitials are significantly trapped in the vicinity of the core of $a/2\langle 1\,1\,1\rangle$ screw dislocations, and it is unlikely they can easily migrate away from the screw dislocation (based on a minimum migration energy equal to the 1 eV binding energy plus saddle point energies on the order of tenths of an eV). However, it is possible

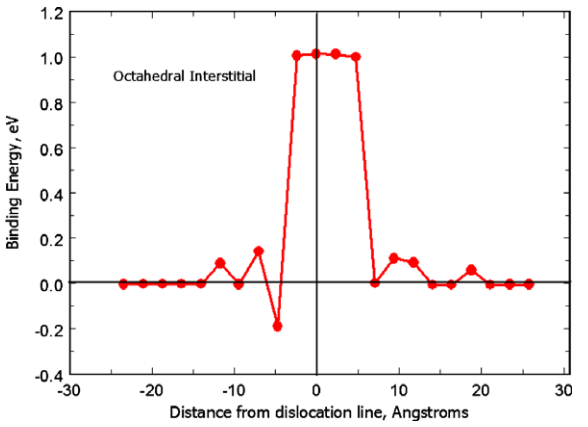

Fig. 2. Binding energies of interstitial He atoms to the $a/2\langle 1\,1\,1\rangle$ screw dislocation in Fe as a function of their distance from the glide plane along a line through the center of the dislocation normal to the glide plane.

for interstitial He atoms to migrate along the screw dislocation core by so-called 'pipe diffusion' at a significantly lower temperature. The dimer method was used to determine the migration pathways and saddle point energies for diffusion of a He interstitial initially placed at various octahedral interstitial positions about the screw dislocation, chosen with regard to the symmetry of the dislocation stress field and the core field, as shown in Fig. 3. This set of locations is not exhaustive, but it is representative of the various symmetries of octahedral sites, including their various orientations relative to the dislocation strain field, as well as extending radially from the core.

In general, a single 'hop' transition of a He atom from its initial position to the next stable position has components both longitudinal and transverse to the dislocation line. Thus, the migration path along the dislocation consists of a zig-zag trajectory, with the barrier for each successive hop varying somewhat with position. An exhaustive study has not been performed, but the initial dimer results indicate that interstitial He atoms within a radius of about 0.3 nm from the center of the dislocation migrate as octahedral interstitials generally along the screw dislocation with a migration energy of

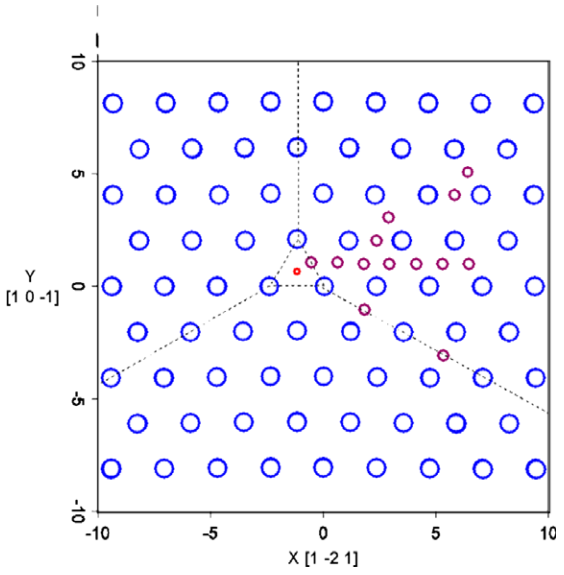

Fig. 3. Locations of the initial position of a single interstitial He atom prior to relaxation. The larger circles are Fe atom rows, and the smaller circles are the various initial He atom positions. The dislocation line is along the z axis (into the page) at the center of the dashed triangle. The z-components of all the He locations are approximately the same. Dimensions of the axes are in Angstrom units.

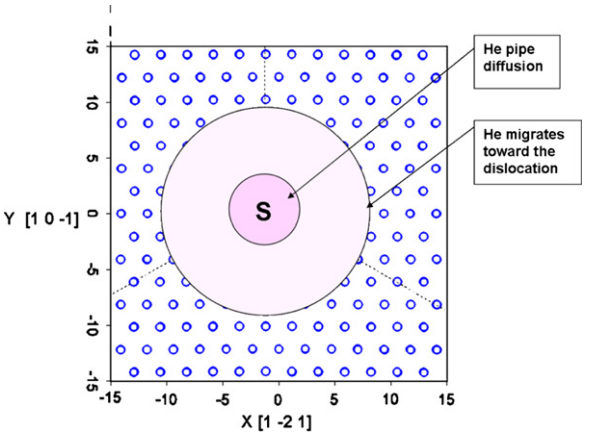

Fig. 4. The cylindrical volumes centered on the screw dislocation line (into the page) within which an interstitial He atom (a) migrates preferentially toward the dislocation and (b) migrates along the dislocation, i.e. pipe diffusion.

$E_m \sim 0.4$–0.5 eV, while He atoms at a greater radius, up to within about 1.0 nm from the center of the dislocation, tend to migrate preferentially toward the center of the screw dislocation with a migration energy of $E_m \sim 0.2$–0.4 eV. Thus, there is a roughly cylindrical region about the screw dislocation within which interstitial He atoms are attracted to the dislocation (see Fig. 4). In general, interstitial He atoms can become trapped near the dislocation, and while trapped, they can migrate along the dislocation relatively easily compared to migration away from the dislocation.

4. Discussion

Both substitutional and interstitial He atoms have significant binding energies to the $a/2\langle 1\,1\,1\rangle$ screw dislocation within about 0.5 nm (or about 2 Burgers vectors) from the center of the dislocation, as shown in Figs. 1 and 2. The binding energies drop off sharply beyond that, essentially defining the extent of the dislocation core within which the excess atomic volume is sufficient to trap the He atoms.

With a migration energy in Fe of 0.08 eV in the perfect crystal, the highly mobile He interstitials produced under irradiation have a high probability of being trapped at both screw and edge dislocations, where they can migrate by pipe diffusion along the dislocations with a migration energy of about 0.4 eV. Interstitial He atoms are bound to the screw dislocation with a maximum binding

energy of 1.05 eV in these simulations, about half the binding energy of interstitial He to the edge dislocation. Still, substantial temperatures are required to dissociate a He from the screw dislocation. A rough estimate is that an interstitial He atom is at least 10^5 times more likely to migrate along the screw dislocation than to dissociate from it at 600 K. It is interesting to note that the He migration energy of 0.4–0.5 eV along the dislocation is in the same range as the migration energy for interstitial He atoms within $\Sigma 3$ and $\Sigma 11$ grain boundaries determined from results of long-time molecular dynamics simulations [8] using the same interatomic potentials used here.

The binding energies of interstitial He atoms to edge [6] and screw dislocations differ by a factor of 2, and they correlate strongly with the excess atomic volume in and near the dislocation core regions. It is interesting that the values of the interstitial He migration energies along edge and screw dislocations are essentially the same, 0.4–0.5 eV. However, the stable He configurations near the dislocations are quite different. Near the edge dislocation, on the tension side, He atoms are most stable in a crowdion configuration, and they migrate along the dislocation line from one close-packed row to the next, normal to the direction of the crowdion [6]. Near the screw dislocation, which lies along the [1 1 1] close-packed direction, He atoms never relax into crowdion configurations, rather, they remain in the octahedral or tetrahedral interstices. Overall, the excess interstitial volumes near the screw dislocation are less than those near the edge dislocation.

5. Conclusions

The results of these simulations indicate that interstitial He atoms are attracted to and trapped at $\langle 1\,1\,1 \rangle$ screw dislocations in α-Fe. Within about 1 nm from the screw dislocation core He atoms migrate as octahedral interstitials preferentially toward the dislocation core, and in this form He atoms can migrate along the dislocation with a migration energy of 0.4–0.5 eV.

Acknowledgement

This work was supported by the US Department of Energy, Office of Fusion Energy Science under contract DE-AC06-76RLO 1830.

References

[1] E.E. Bloom, J. Nucl. Mater. 85&86 (1979) 795.
[2] H. Ullmaier, Nucl. Fusion 24 (1984) 1039.
[3] R.L. Klueh, D.J. Alexander, J. Nucl. Mater. 218 (1995) 151.
[4] D.S. Gelles, J. Nucl. Mater. 283–287 (2000) 838.
[5] G. Henkelman, H. Jónsson, J. Chem. Phys. 111 (1999) 7010.
[6] H.L. Heinisch, F. Gao, R.J. Kurtz, E.A. Le, J. Nucl. Mater. 351 (2006) 141.
[7] R.J. Kurtz, H.L. Heinisch, J. Nucl. Mater. 329–333 (2004) 1199.
[8] F. Gao, H.L. Heinisch, R.J. Kurtz, J. Nucl. Mater. 351 (2006) 133.
[9] K. Morishita, R. Sugano, B.D. Wirth, J. Nucl. Mater. 323 (2003) 243.
[10] K. Morishita, R. Sugano, B.D. Wirth, T. Diaz de la Rubia, Nucl. Instrum. and Meth. B 202 (2003) 76.
[11] J.P. Hirth, J. Lothe, Theory of Dislocations, McGraw-Hill, New York, 1968.
[12] G.J. Ackland, D.J. Bacon, A.F. Calder, T. Harry, Philos. Mag. A 75 (1997) 713.
[13] W.D. Wilson, R.D. Johnson, Interatomic Potential and Simulation of Lattice Defects, Plenum, 1972, p. 375.
[14] D.E. Beck, Mol. Phys. 14 (1968) 311.
[15] M.I. Mendelev, S. Han, D.J. Srolovitz, G.J. Ackland, D.Y. Sun, M. Asta, Philos. Mag. 83 (2003) 3977.
[16] C. Domaine, C.S. Bequart, Phys. Rev. B 65 (2001) 024103.
[17] S.L. Frederiksen, K.W. Jacobsen, Philos. Mag. A 83 (2003) 365.
[18] T. Seletskaia, Y. Osetsky, R.E. Stoller, G.M. Stocks, Phys. Rev. Lett. 94 (2005) 046403.
[19] T. Seletskaia, Yu.N. Osetsky, R.E. Stoller, G.M. Stocks, J. Nucl. Mater., these Proceedings, doi:10.1016/j.jnucmat.2007.03.018.
[20] G. Voronoi, J. Reine Angew. Math. 134 (1908) 198.