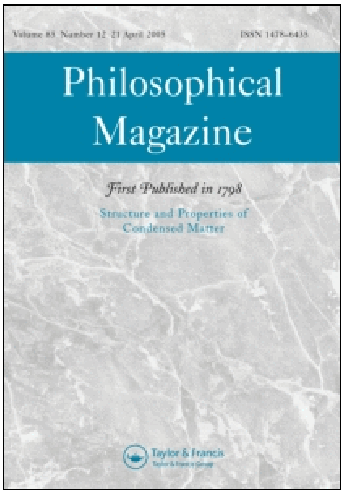

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Simulation of cascades in tungsten–helium

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Helium (He) is present in fusion reactor wall materials, and its effect on radiation damage must be taken into account. The effect of He on displacement cascades in tungsten (W) has been studied using molecular dynamics simulations. Three different W–W potentials were compared and found to differ, especially for the clustering of the vacancies formed in the cascades. While there are differences in the amounts of damage depending on the potential, the overall effect of He in interstitial positions was to increase the amount of damage, while He in substitutional positions reduces it, due to the effect He has on the recombination of tungsten interstititals.

Keywords: tungsten; helium; radiation damage; displacement cascade

1. Introduction

Tungsten (W) is considered as a prime candidate for the divertor material in future fusion reactors, such as the ITER test reactor [1,2], due to its suitable mechanical and thermal properties and a low sputtering yield. Due to deposition of helium (He) from the fusion plasma, as well as energetic neutron radiation induced (n,α)-transmutation reactions [3], tungsten in fusion reactors will contain small amounts of He [4]. The effect of He on material properties, He retention and diffusion [5–7], trapping [8], bubble formation [9,10], blistering [11], etc., has been studied previously; however, the effect on the primary damage formation due to displacement cascades is not known.

Molecular dynamics (MD) simulation is a valuable tool for studying the onset of primary damage due to displacement cascades. The length and time scales are in the range of tens of nm and tens of ps, respectively, which is well within reach of MD calculations, while not easily observed experimentally. Displacement cascade damage in pure W has been studied previously [12–14]. Several different interatomic potentials have been used to investigate cascade damage in W [15–18], but the amount and clustering of the formed defects is similar.

The effect of helium on displacement cascades in iron [19] and iron–chromium [20] has been studied recently. At the relatively high concentrations of 1 atomic percent (at.%) interstitial He, a significant increase in damage formation is seen,

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Table 1. The number of Frenkel pairs with He initially in interstitial positions. The results for pure W are, for the D potential from Ref. [12] and for the AT and J potentials from Ref. [13].

2 keV	Pure	0.1% He	1.0% He
AT	2.6 ± 0.1	3.4 ± 0.1	4.1 ± 0.1
D	3.6 ± 0.3	4.2 ± 0.1	6.1 ± 0.2
J	2.8 ± 0.1	3.8 ± 0.1	10.3 ± 0.2
5 keV			
D	7.5 ± 0.7	11.8 ± 0.2	11.8 ± 0.2
20 keV			
D	17.9 ± 1.1	–	44.7 ± 1.7

while a decrease is observed for substitutional He. At 0.1 at.% He, the effect is not statistically significant. As these are body-centered cubic metals like tungsten, similar results can be expected. There are, however, large differences in He, vacancy and self-interstitial defect properties and thermal properties between iron and tungsten, which can affect primary damage formation.

Most of the cascade studies in pure W have been done in the low keV range. As the effect of He can be expected to be more pronounced at higher energies, due to more total damage, we have compared how He affects cascades in tungsten for three W potentials for 2 keV recoils, which is the highest energy studied in the literature for pure W for these potentials. For the potential by Derlet et al. [18], for which higher energy recoils have been studied, we also studied 5 keV and 20 keV recoils.

Many different interesting initial He positions could be considered, and in a fusion reactor, many factors will determine the He content and microstructure. In addition, one would always find deuterium and other defects in a real divertor. While He will be found in both interstitial or substitutional positions and, in particular, He bubble formation is an issue in a real material, in this paper, we focus on all He either in interstitial or all He in substitutional positions initially, with a relatively short equilibration time, in order to study the basic effect of He. The He concentrations used were 0.1 at.% and 1.0 at.%.

2. Methodology

Three W–W interatomic potentials were compared in this work. The Ackland–Thetford (AT) [16], the Juslin et al. (J) [17] and the Derlet et al. (D) [18]. The AT potential is an improved version of the Finnis–Sinclair potential [15]. For D the repulsive part is by Björkas et al. [12]. The AT and D potentials are Finnis–Sinclair type potentials, which are similar to the embedded atom method, and the J potential is a bond order potential of Tersoff type. The potentials have all been developed with different applications in mind, yet perform relatively similarly for low keV cascades in pure W (see Tables 1 and 2).

For W–He, the potential developed by Henriksson [11] was used. It was developed to describe He interstitials and bubbles in W for the AT potential, but can

Table 2. The fraction of vacancies (V) and self-interstitials (SI) that are in clusters, as described in the text, for 2 keV cascades. The results for pure W are, for the D potential from Ref. [12] and for the AT and J potentials from Ref. [13].

	AT		D		J	
	Pure	1.0% He	Pure	1.0% He	Pure	1.0% He
V	0.28 ± 0.04	0.04 ± 0.01	0.24 ± 0.08	0.23 ± 0.02	0.22 ± 0.04	0.07 ± 0.01
SI	0.24 ± 0.04	0.21 ± 0.03	0.31 ± 0.07	0.29 ± 0.03	0.24 ± 0.04	0.20 ± 0.01

Table 3. The interstitial and substitutional formation energies for He in W for the different potentials and DFT. Formation energies are given in eV. The tetrahedral interstitial is unstable with the AT potential.

	AT	D	J	DFT [8,21]
Tetrahedral	–	8.8	8.3	6.2
Octahedral	7.8	9.0	8.2	6.4–6.5
Substitutional	6.3	6.2	4.2	4.7–5.0

be expected to perform similarly for other potentials that describe W well. Since its development, recent *ab initio* results [8,21] indicate that it overestimates the formation energy of interstitials and the ground state position depends on the W potential used (see Table 3). There is also an old W–He potential from 1972 by Wilson [22] which underestimates the formation energy and gives a ground state position that does not agree with density functional theory (DFT) calculations. For He–He, the Beck potential [23] was used.

The simulations were performed with the MD code PARCAS.[1] The simulation cells were constructed as a BCC lattice of W atoms with He atoms randomly distributed in either octahedral interstitial or substitutional positions. Cells with 0.1 at.% or 1.0 at.% He were used. The simulation cells were at first relaxed for 25 ps at zero pressure and 300 K, after which an atom near the center was given a recoil energy of 2 keV, 5 keV or 20 keV in a random direction. The temperature dependence was not investigated in this work, but can be expected to be similar to that found in iron–helium by Lucas and Schäublin [19], where the effect of He is more pronounced with increasing temperature. The temperature was controlled using the Berendsen method [27]. A variable time step, periodic boundaries in all three dimensions and electronic stopping were used. Temperature scaling with a constant of 100 fs was applied in the cell's border regions, which have a thickness of one lattice parameter. The simulation was stopped if the kinetic energy of a border atom was larger than 10 eV and restarted with the recoil atom placed farther away from the border. In the simulation of a 2 keV recoil in a cell with 0.1% He, a border energy threshold of 15 eV was used, as energies around 10 eV were observed at the beginning of the initial equilibration. While only an issue for the first few time steps, the threshold was unchanged when the recoil was introduced. During the recoil and

Table 4. The simulations performed with interstitial He. For substitutional He, the same simulations were performed for 2 and 5 keV, with 59 582 and 148 176 atoms, respectively.

Recoil energy [keV]	% He	Number of events D	J	AT	Time [ps]	Box size $\times a_0$	No. of atoms
2	0.1	100	100	100	50	31	59 641
2	1.0	100	100	100	50	31	60 183
5	0.1	100	–	–	50	42	148 324
5	1.0	100	–	–	50	42	149 672
20	1.0	10	–	–	50	67	607 602

cooling down of the cascade, the energies of border atoms were below the 10 eV threshold as in the other cases. Information about the amount of He in the simulation cells, the simulation time and the size for the simulation cells for each recoil energy and potential, is given in Table 4.

The simulations were analyzed using Wigner–Seitz cells in order to find defects in the atom lattices. Thus, a cell without a W atom corresponded to a vacancy, while cells with more than one W atom were defined as self-interstitials. A vacancy and self-interstitial clustered fraction were calculated. The vacancy cluster analysis used the second nearest neighbor distance as a cutoff radius, while the third nearest neighbor distance was used for interstitials, in order to be comparable with earlier work on pure W [12,13].

3. Results and discussion

During the 25 ps equilibration before the recoil is introduced, the He atoms migrate and some cluster together. The amount of clustering is, however, limited, as the He atoms cannot easily displace a W atom from its lattice point due to the high formation energy of a W Frenkel pair, creating space for He bubbles (as opposed to Fe and FeCr where this occurs [20]). For 1% He initially in interstitial positions, 1–3 He generally occupy the same Wigner–Seitz cell as a W atom. Clusters spread over several Wigner–Seitz cells also form, but the sizes are still limited to less than 10 He atoms. For substitutional He, some of the He migrated from the vacancy they initially occupied, and ended up as interstitial He or often getting trapped by other substitutional He, forming He-vacancy clusters containing a small number of He atoms. The amount of He clustering is, however, much smaller starting from substitutional He than from interstitial He, as the substitutional He atoms are relatively stable on this time scale.

We compared the formation energies of an interstitial and a substitutional He with the different potentials (Table 3). The ground state formation energy of the He interstitial is relatively high compared to density functional theory (DFT) results, and within 1 eV for all potentials. The difference in formation energy between the octahedral and the tetrahedral interstitial is small, except for the AT potential where the tetrahedral position is unstable. Only the D potential gives the ground state position as tetrahedral as predicted by DFT, though significantly more unstable. The formation energy of the substitutional position is about 6.2 eV for AT and D and 4.2 eV for the J potential, which is due to the low formation energy of the vacancy for that potential [17]. Compared to DFT results, the formation energy of the substitutional interstitial is closest for the J potential results, the difference in energy between interstitial and substitutional is best with the AT potential and the ground state position of the interstitial is correct only for the D potential. Thus the basic defect properties do not show that one potential would be superior to the others for studying He in W.

The three different W–W potentials were compared for 2 keV cascades with 1% He. This energy was chosen, as it is the highest energy for which results for pure W exist for all the potentials [12,13]. A higher energy means more damage and thus a more discernable difference between the potentials due to the effect of He on the damage.

As can be seen in Table 1, in the case of 1.0% He initially in interstitial positions, all three potentials agree that there is more total damage in the W matrix, given by the number of Frenkel pairs formed due to the cascade. This can be understood by the binding of He to vacancies and W interstitials, reducing the mobility of the interstitials and making it less energetically favorable for W interstitials to recombine with the vacancies, as He atoms go into vacancies and a kick-out mechanism is needed to recombine. From the time the heat spike recrystallizes, almost all remaining vacancies are filled with one or more He. Lucas and Schäublin explain the similar effect seen in Fe by He strongly binding to and stabilizing intersterstitial clusters [19], but as discussed in the following paragraph, in tungsten–helium the clustering of self-interstitial atoms (SIA) is close to that in pure W. In FeCr, where the clustering of self-interstitial atoms is increased as in Fe, a fast binding of He to vacancies formed in the cascade was also observed [20]. In all these materials, the effect of He on the recombination mechanism seems to be governed by binding of He to all defects in the metal matrix, though the binding to and stabilizing of SIA clusters is different in Fe and W. It should, however, be noted that the cooling down and recombination of a heat spike is a complicated combination of several processes over a large temperature scale. With the J potential, the effect is considerably higher than for the other potentials. This is due to the stability of the substitutional, which means that the He atoms are more prone to bind to the vacancies.

An interesting difference between the potentials appears in the clustering of vacancies and self-interstitials in the W matrix for 2 keV cascades. For pure W, the clustering is similar for the potentials, 20–30% for all cases as shown in Table 2. With 1.0% He, the amount of vacancy clustering for the AT and the J potentials is reduced to about 4% and 7%, respectively, while it remains the same for D. The interstitial clustering is almost the same as for pure W, though slightly reduced. This difference in vacancy clustering between the potentials is not easily explained by the W–W potentials or the behavior of single He defects, and analyzing how the He atoms affect the clustering during the cooling down of the heat spike is not trivial. Thus it is difficult to estimate which potential describes the clustering best.

For substitutional He, the number of Frenkel pairs decreases for 1% He (Table 5). As the heat spike cools down, all the He atoms will not recombine with a vacancy to form substitutional He and thus there are more vacancies for the W

Table 5. The number of Frenkel pairs with He initially in substitutional positions. The results for pure W are, for the D potential from Ref. [12] and for the AT and J potentials from Ref. [13].

2 keV	Pure	0.1% He	1.0% He
AT	2.6 ± 0.1	1.8 ± 0.1	0.3 ± 0.05
D	3.6 ± 0.3	3.5 ± 0.2	1.5 ± 0.1
J	2.8 ± 0.1	3.6 ± 0.1	1.7 ± 0.1
5 keV			
D	7.5 ± 0.7	7.8 ± 0.2	2.0 ± 0.1

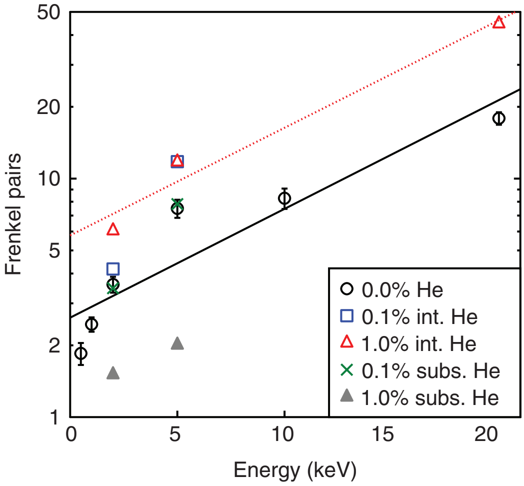

Figure 1. The number of Frenkel pairs formed for different recoil energies and He contents using the D potential. The results for pure W are from Ref. [12]. The error bars for the results in this work are of the size, or smaller, than the markers, and have been left out for clarity. The errors are given in Tables 1 and 5.

atoms to recombine with. This reduces the overall number of Frenkel pairs formed in the cascade. For the AT potential almost all damage recombines. For 0.1% He, the effect is much smaller. For the D potential the results are within the statistical error of pure W, and for the J potential the number of Frenkel pairs is even increased to a similar value to that for 0.1% interstitial He, though in this case the result for pure W appears to be quite low.

For the D potential, the behavior at higher recoil energies was studied. As expected, the number of Frenkel pairs increases with the recoil energy for 1% interstitial He initially. The slope in a log plot remains almost the same as for pure W (Figure 1) and the number of Frenkel pairs is significantly higher than in pure W, especially at higher energies. The clustering of vacancies (Figure 2) and self-interstitials (Figure 3) is close to that in pure W for the recoil energies studied. For 1% substitutional He, we note that the number of Frenkel pairs for 5 keV cascades is reduced to almost the same number as for 2 keV cascades, 2.0 and 1.5 respectively.

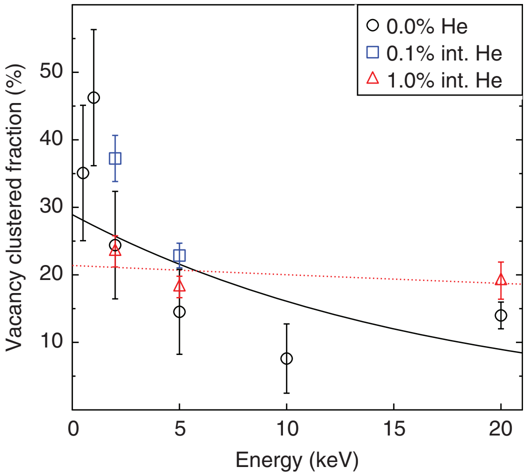

Figure 2. The clustered fraction, in %, of vacancies in the W matrix for the D potential. A vacancy belongs to a cluster if it is within second nearest neighbor distance of another vacancy. The results for pure W are from Ref. [12].

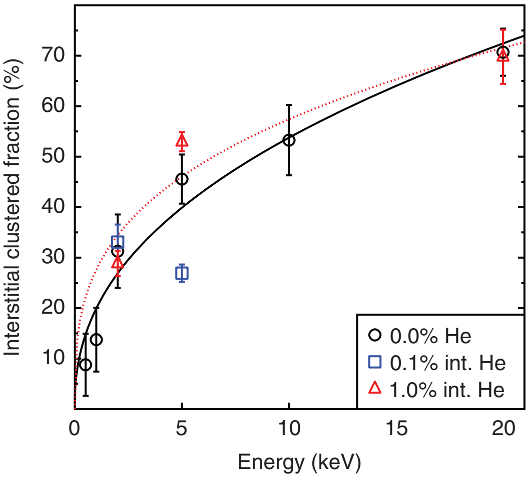

Figure 3. Self-interstitial clustered fraction, in %, for the D potential. An interstitial belongs to a cluster if it is within third nearest neighbor distance of another interstitial. The results for pure W are from Ref. [12].

Generally the effect of He concentration is that the more He in the system, the larger the difference compared to pure W. Both for interstitial and substitutional He, the results are close to those in pure W for 0.1% He and significantly different for 1.0% He. There is a rather surprising exception to this. The number of Frenkel pairs for the 5 keV cascade with 0.1% interstitial He is the same as with 1% He down to two decimals of accuracy. The interstitial clustering, on the other hand, is much lower in this case than for both 1% and 0% He. The recombination results in a more spread out interstitial configuration for the 0.1% case, though the mechanism behind this is unclear. It can be noted that for pure W [12] the number of Frenkel pairs is also quite high for 5 keV cascades, almost as high as for 10 keV, with an apparent change in the recoil energy dependence between a low and a high energy region. This indicates that there could be a change in the recombination mechanisms at about 5 keV for the D potential. This is an interesting venue for future studies, both for the pure material and for the effect of He concentration.

While the composition and structure of a real divertor material is more complicated, with He bubbles, other impurity atoms, surfaces and grain boundaries, the mechanisms of how He affects the recombination in W for this simplistic view of initially interstitial or substitutional He can be expected to exist in a real material. As the He concentrations approach 1 at.%, the effect on cascades in a real material is likely to be appreciable, as the effect is dramatic for both cases studied here, He initially in interstitial and in substitutional positions.

4. Conclusions

We have simulated the effect of helium on the primary damage formed in displacement cascades in tungsten. Three W–W potentials have been compared and while there are differences, it is not clear which potential describes He in W best. The present He–W potential overestimates the formation energy of He interstitials for all the W–W potentials, in comparison with DFT data from the literature. The W–W potentials especially differ when looking at the clustering of vacancies from a cascade with He present, with large differences compared to the results for pure W, where it is relatively similar.

All potentials predict that the overall effect of He is that He in initially interstitial positions increases the number of Frenkel pairs in the W matrix and substitutional He reduces it. Especially at higher He concentrations and recoil energies the effect is significant. The increase due to interstitial He is explained by He entering the vacancies produced as the heat spike cools down, reducing the recombination of tungsten interstitials. For substitutional He, the opposite takes place, as some He become interstitial or clusters with other He, thus leaving more vacancies for the W interstitials to recombine with.

Note

1. K. Nordlund: PARCAS computer code. The main principles of the molecular dynamics algorithms are presented in [24,25]. The adaptive time step and electronic stopping algorithms are the same as in [26].

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