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A comparison of interatomic potentials for modeling tungsten–hydrogen–helium plasma–surface interactions

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ABSTRACT

We compare the hydrogen and helium clustering characteristics of three interatomic potential energy models intended for simulation of plasma-facing materials for fusion applications. Our simulations compare a Finnis–Sinclair potential and two different Tersoff-style bond order potentials created by Juslin et al. (2005) and Li et al. (2011), respectively, with respect to both helium and hydrogen clustering behavior in tungsten. We find significant differences between the Juslin and Li potentials in terms of both hydrogen and helium clustering behavior as well as the spatial distribution of hydrogen below the surface. These simulations are an important test on the road to more accurate models of gas clustering and surface evolution of tungsten divertors in ITER and other plasma devices.

1. Introduction

Tungsten is currently the material of choice for the divertor in prototype fusion reactors such as ITER due to its high melting temperature, high thermal conductivity, and low sputtering yield [1]. However, experiments in linear plasma devices [2–4] and tokamaks [5] have shown that a fuzz-like structure forms on the tungsten surface between approximately 900 K and 2000 K after exposure to helium-containing plasma with helium energies as low as 12 eV [3,6]. Such "fuzz" is absent at surface temperatures outside this range and/or much higher incident helium energies above hundreds of electron-volts [6,7]. The precise mechanisms involved in tungsten fuzz formation remain the target of significant research. Understanding this phenomenon will require a combination of high- and low-flux and fluence experiments combined with insights from both atomistic and coarse-grained modeling.

Molecular dynamics (MD) is a useful simulation tool that has been used by several research groups, including ours, to investigate this tungsten surface deformation phenomenon [8–12]. Previous MD simulations have involved pure helium as well as mixed helium/hydrogen exposure conditions, though most work to date has focused on the helium–tungsten system. Sefta [13] observed that these two types of simulations differed: in mixed hydrogen/helium simulations, the helium clusters were notably smaller and more numerous than in pure helium plasma simulations. The most obvious explanation for this was the interatomic potential, because the helium/hydrogen simulations used a different tungsten–tungsten potential: a Tersoff bond order potential (BOP) [14] created by Juslin et al. [15] was used to model the tungsten–tungsten interactions in the mixed helium/hydrogen simulations, while a Finnis–Sinclair potential [16] was used in the helium simulations. The differences in helium clustering seen with the Juslin BOP relative to that seen with the Finnis–Sinclair potential may indicate a deficiency in one or both of these potentials for the purposes of simulating gas clustering.

Recently, another hydrogen–tungsten Tersoff-style potential was developed by Li et al. [19] which was designed to yield hydrogen–tungsten interactions more consistent with density functional theory calculations, particularly near defects, by re-fitting with a longer cutoff distance for both the W–W and W–H parts of the model. There are many differences that emerge as a result, the most notable being the hydrogen interstitial formation energies. This work attempts to compare these three different tungsten–tungsten potentials with respect to gas clustering in the bulk as well as sub-surface clustering dynamics for pure helium, pure hydrogen, and a 90% hydrogen/10% helium mixture. We find that the two BOP models give drastically different hydrogen clustering behavior, especially near surfaces, suggesting the need for further work to determine which behavior, if any, are physically meaningful and/or relevant to experimental results.

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Table 1
Select hydrogen properties for the Juslin BOP, Li BOP, and DFT (data from Li et al. [19]).

Potential	H tetra. (eV)	H oct. (eV)	H sub. (eV)	W vac. (eV)
Juslin	1.03	1.39	4.05	1.68
Li	0.85	1.17	4.02	3.52
DFT	0.88	1.26	4.08	3.56

2. Simulation method

All molecular dynamics (MD) simulations were performed using LAMMPS [20]. Three tungsten potentials were compared: a Finnis–Sinclair potential [16] modified at short range by Ackland and Thetford [17] and by Juslin and Wirth [18]; and two different Tersoff-type [14] bond-order potentials (BOP), one developed by Juslin et al. [15] and one developed by Li et al. [19]. The Finnis–Sinclair potential was not used in simulations containing hydrogen. Table 1 shows the formation energies of hydrogen interstitials and tungsten vacancies with these potentials. A helium–tungsten potential fit by Juslin and Wirth [18] and Beck's potential for helium–helium [21] as modified at short distances by Morishita et al. [22] were used in all helium-containing simulations. Helium–hydrogen interactions were described by a Lennard-Jones 12-6 potential [23] with $\sigma = 1.333$ Å and $\varepsilon = 5.9225 \times 10^{-4}$ eV [24].

We performed two different types of simulations to study clustering with each interatomic potential. The first simulates clustering in the bulk, and was started by creating a BCC tungsten ($a = 0.31689$ nm) block with periodic boundary conditions in all three dimensions. Thermal equilibrium was established by selecting velocities from a Maxwell–Boltzmann distribution at 1200 K and running for 5 ps with a 1 fs time step using velocity re-scaling every 100 time steps, followed by 15 ps without temperature control. Gas atoms (helium and/or hydrogen) were inserted at random positions every 6–8 ps. After a gas atom was inserted, the system was evolved without a thermostat for 30000 time steps followed by 5000 time steps with velocity using the same parameters used during equilibration. A variable time step was used during the simulations in case an atom was inserted very close to another one, producing a large force. The step size was recomputed to a value between 10^{-5} fs and 1 fs every ten time steps so that no atom would move farther than 0.001 nm during one step (a typical step size is 0.1 fs). The size of the simulation cell was $20a$ (about 6.3 nm) in each dimension.

The second type of simulation was used to evaluate differences between the two Tersoff potentials related to free surfaces. Simulation conditions were identical to the bulk simulations, except that the z (001)-direction was a free surface; the box was $40a$ (12.6 nm) long in the z-direction, and the gas implantation profile in the z-coordinate was sampled from the SRIM [25] depth distribution for 60 eV. This method saves considerable time over direct energetic implantation because direct bombardment requires much smaller time steps and results in most of the atoms reflecting from the surface, as well as requiring significant temperature control due to the kinetic energy dissipation of the energetic gas atom. The results should be comparable to direct bombardment studies provided the flux and fluence are corrected for reflected atoms. This approximation is possible because 60 eV is well below the sputtering threshold (105–110 eV for helium and roughly twice as much for deuterium). The first nine insertions in a sequence of ten were hydrogen, with the tenth being helium, so as to mimic a 90% hydrogen, 10% helium plasma. The nominal flux is $3.6 \times 10^{27}\,\mathrm{m^{-2}\,s^{-1}}$ (excluding reflected ions). At this flux, 1000 insertions corresponds to a fluence of $3.6 \times 10^{19}\,\mathrm{m^{-2}}$ (again excluding reflections).

3. Results and discussion

Figs. 1 and 2 show the observed differences in both helium and hydrogen clustering for the three different potentials. The helium clustering appears qualitatively similar across all three potentials, but the Juslin BOP does seem to produce more small helium clusters than either the Finnis–Sinclair or the Li BOP. The cluster size distributions are shown in Fig. 3, and provide a more quantitative assessment of the differences in helium clustering behavior. Atoms are considered to be in the same cluster if they are within 0.32 nm of each other. As seen in Fig. 3a, the Juslin BOP has over three times as many helium monomers as either the Finnis–Sinclair potential or the Li BOP. While the Li BOP does not produce helium clusters as large as those with the Finnis–Sinclair potential, the results with the Li potential are more comparable to the Finnis–Sinclair results than to the Juslin BOP results. On the other hand, the hydrogen clustering shown in Fig. 2 is drastically different between the Juslin and Li BOPs. There are obvious hydrogen clusters in the simulation using the Juslin BOP that are oriented in the ⟨1 1 1⟩ direction, while there is virtually no clustering with the Li BOP. The hydrogen cluster size distributions are shown in Fig. 3b, which confirm the visual observation: the Juslin BOP gives a varied distribution of different sized clusters while the Li BOP hydrogen clustering consists almost entirely of individual hydrogen atoms mixed with a few two and three atom clusters.

The only differences between the three helium simulations were the choices of tungsten–tungsten potential, and therefore the variability in the cluster size distribution is attributable to the tungsten–tungsten interactions. One explanation for the helium clustering behavior is the tungsten vacancy formation

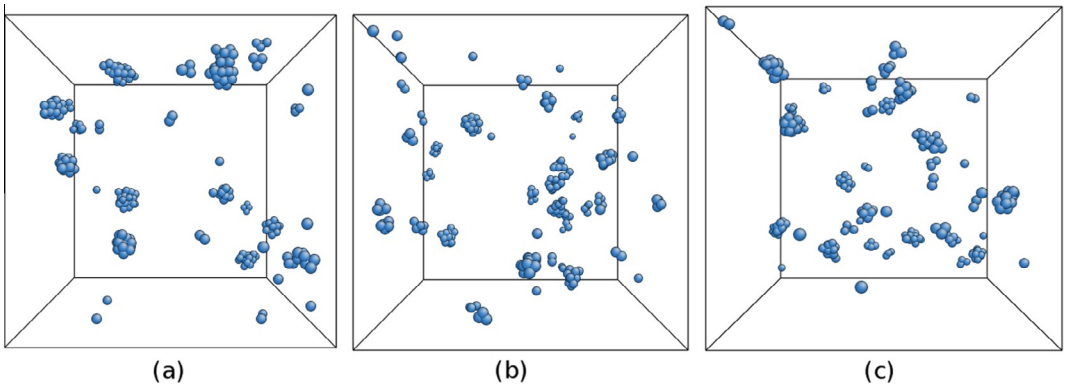

Fig. 1. Snapshots of simulations of for helium implantation in a periodic box at 1.5 atom% for (a) Finnis–Sinclair, (b) Juslin BOP, and (c) Li BOP. Helium atoms were randomly inserted every 10 ps into a $20 \times 20 \times 20$ lattice unit box with periodic boundary conditions.

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