Sputtering Yields of Pure and Helium-Implanted Tungsten Under Fusion-Relevant Conditions Calculated Using Molecular Dynamics

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Abstract

This study uses atomistic simulations to calculate the sputtering yield of tungsten in the presence and absence of near-surface helium clusters. We find that near-surface helium in the concentrations tested (consistent with 100 eV implantation fluences of 1.0 and $2.2 \times 10^{19} \text{ He} \cdot \text{m}^{-2}$ at a flux on the order of $10^{25} \text{ He} \cdot \text{m}^{-2}\text{s}^{-1}$) has a negligible effect on the sputtering yield, suggesting that any changes in sputtering that might be observed experimentally are due either to much higher near-surface concentrations of helium or due to other structural changes in the tungsten that would occur only at much higher helium fluences than we are able to probe using molecular dynamics simulation. We do find a dependence on surface orientation and incident helium energy: comparing $\{001\}$, $\{110\}$, and $\{111\}$ surfaces, we find that $\{110\}$ surfaces at high energy show the most sputtering, while $\{111\}$ surfaces at low energy show the least. We attribute these differences in surface orientation to the more closely-packed nature of $\{110\}$ surfaces compared to the other two orientations.

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1. Introduction

Tungsten is the primary candidate material for the divertor in future nuclear fusion reactors such as ITER [1]. While one reason for choosing tungsten is its lack of chemical sputtering, physical sputtering is possible at sufficiently high incident energies. For helium, the main product of fusion reactions, the threshold energy to sputter tungsten is approximately 105–110 eV, which is in the range of expected energies of particles in the baffle region of the ITER divertor [2]. Over time, the cumulative atomic-scale processes of physical sputtering can produce erosion and shorten the divertor lifetime. Current expectations are that physical erosion should be negligible in the detached-divertor regime, with expected ion energies of ≈ 15 eV [2]. However, components such as those near the baffle region of the divertor, which are near the interface of the hot scrape-off layer (SOL) of the plasma edge, may be exposed to particles with energies significantly higher than 100 eV, creating physical sputtering. Expected ion energies at the midplane are expected to be approximately 500 eV [2]. Consequently, it is of interest to quantify the sputtering yields caused by helium at energies relevant to ITER, and to determine whether the sputtering yield will change significantly as the device ages and/or is modified by implanted gas atoms.

Experiments in linear plasma devices demonstrate that a very high density of nanometre-sized helium bubbles form in near-surface layers of the tungsten matrix [3–6]. This has been confirmed to occur on much smaller time and length scales in molecular dynamics simulations [7–10]. Therefore, it is also necessary to understand whether sputtering rates change with increasing levels of helium retention in the material.

This study compares the sputtering yield of tungsten due to helium ions in the presence and absence of near-surface helium atoms using molecular dynamics. Specifically, we determine whether the near-surface “layer” of substitutional helium atoms and small helium atom clusters observed in our previously reported atomistic simulations [8] have an effect on the sputtering yield. We find that helium in the concentrations so far observed in such simulations has a

negligible effect on the sputtering yield, suggesting that any changes in sputtering are due either to much higher near-surface concentrations of helium, or to other structural changes in the tungsten that would occur only at much higher fluence.

2. Procedure

Molecular dynamics (MD) simulations were performed using LAMMPS [11]. The interatomic potentials used were the Ackland–Thetford modification of the Finnis–Sinclair N -body potential for tungsten [12, 13], as modified at small interatomic separations for sputtering and cascade simulations by Juslin and Wirth [14]; the tungsten–helium potential of Juslin et al. [14]; and Beck’s potential for He–He interactions [15, 16] as modified at small interatomic separations by Morishita et al. [17].

Molecular dynamics simulations of sputtering were performed on pure tungsten surfaces and helium-implanted tungsten surfaces. Helium-implanted simulations were bombarded with helium ions with energies between 200 eV and 500 eV. The set-up of each kind of simulation is detailed in the following subsections.

It should be noted that molecular dynamics simulations neglect electronic stopping, which can be a significant fraction of the energy loss. Electronic stopping is small at low ion energies, and neglecting it is a good approximation for heavy ions such as iron or tungsten; however, helium is light enough that nuclear stopping is also small, so the net effect is that electronic stopping cannot be neglected without introducing some error into the simulation. For example, TRIM calculations using 10,000 helium atoms impinging on a tungsten surface suggest that 56 percent of the stopping power comes from electronic stopping (ionization) for 200 eV incident helium, while 59 percent comes from electronic stopping for 500 eV incident helium. As such, things like sputtering yields and depth distributions as calculated from molecular dynamics simulations should be interpreted with this in mind. In particular, it is fine to make comparisons of the

relative differences in depth across different surface orientations, but the actual depth distributions will be too “deep” due to neglect of electronic stopping.

2.1. Set-Up for Pure Tungsten Case

In the case of pure tungsten, three different surface orientations were used, namely (001), (011), and (111). The x - and y -directions of the simulation box had periodic boundary conditions, while the z -direction was a free surface (with one surface exposed to the plasma). The depth of the simulation cell was approximately 44.5 nm in the z -direction. The height and width varied from 3.8 to 5.4 nm in the x - and y -directions, depending on the surface orientation. The system was subsequently thermalised to 933 K using a Nosé–Hoover thermostat [18–20] with a time constant of 0.05 ps and a time step of 0.5 fs, and the system state was saved in a restart file.

A sputtering simulation consists of reading the state from the aforementioned restart file, then introducing a helium atom above the free surface with the appropriate kinetic energy, aimed normal to the surface. Each simulation was then time-integrated using a variable time step for 5 ps, which was observed to be sufficient for helium even at high energies to thermalise and for any sputtered tungsten atoms to be ejected from the free surface. The time step during the sputtering simulation was chosen to be in the range 1.0×10^{-5} fs to 0.5 fs, with the step size re-sampled every ten steps such that no atom moved more than 0.01 Å during the first of the ten steps. At the end of each sputtering simulation, any atoms more than two lattice units above the free surface were considered to have sputtered. Owing to the rarity of a sputtering event, the simulation was repeated between 4000 and 8000 times to ensure satisfactory statistics. Each of these 4000–8000 simulations introduced the helium atom at a different random position in the x – y plane above the free surface. The sample sizes for each surface orientation in the pure tungsten cases are shown in Table 1.

2.2. Set-Up for Helium Pre-Implanted Tungsten Case

In the case of tungsten pre-implanted with 100 eV helium, a large box ($50 \times 50 \times 23$ nm) of pure tungsten was created with a (111) surface orientation.

Table 1: Number of individual, independent sputtering simulations on pure tungsten with varying surface orientations and helium energies. All simulations performed at 933 K.

Surface Orientation	Energy (eV)			
	200	300	400	500
(0 0 1)	8000	8000	8000	8000
(0 1 1)	4000	4000	4000	4000
(1 1 1)	4000	6000	6000	6000

This was *cumulatively* implanted with thermalised helium introduced at depths sampled from a cumulative distribution function associated with an incident ion energy of 100 eV at a temperature of 933 K and a flux of $7.9 \times 10^{25} \text{ He} \cdot \text{m}^{-2}\text{s}^{-1}$. The state of this simulation was sampled at fluences of $1.0 \times 10^{19} \text{ He} \cdot \text{m}^{-2}$ or $2.2 \times 10^{19} \text{ He} \cdot \text{m}^{-2}$. In this simulation, helium very close to the surface causes trap mutation, creating an adatom–vacancy pair and resulting in a “layer” with locally high helium concentration consisting of substitutional helium as well as small helium–vacancy clusters, as shown in Figure 1. For further details on how this simulation was carried out, please refer to Ref. 8.

For the purpose of this study, a smaller, more tractable simulation was created by sampling a $5.2 \times 5.2 \times 23.1 \text{ nm}$ block from the upper corner of the large simulation. This block of tungsten and helium was then thermalised to 933 K (660 °C) using the same thermostat as in the pure tungsten case, and the state was subsequently saved in a restart file. An identical procedure to the case of pure tungsten was used to estimate the sputtering rate, with the only difference being the starting state. The numbers of both tungsten and helium atoms sputtered from the free surface were tracked for each simulation, and each simulation was repeated between 4000 and 6000 times using a different x – y position, as in the pure tungsten case.

3. Sputtering of Pure Tungsten

3.1. Sputtering Yield

The predicted sputtering yields of tungsten from helium irradiation as a function of surface orientation and helium energies are shown in Figure 2. The results show that $\text{W}\{110\}$ surfaces have a significantly higher sputtering yield

Figure 1: (a),(c): Size distributions of helium clusters for helium implantation fluences of $1.0 \times 10^{19} \text{ He} \cdot \text{m}^{-2}$ and $2.2 \times 10^{19} \text{ He} \cdot \text{m}^{-2}$, respectively. (b),(d): Frontal views of helium atoms in the starting configurations for our pre-implanted tungsten sputtering simulations (for 1.0 and $2.2 \times 10^{19} \text{ He} \cdot \text{m}^{-2}$, respectively). All visible atoms in parts (b) and (d) are fewer than four lattice units below the (111)-oriented surface.

Figure 2: Sputtering yields of pure, single-crystal tungsten as a result of helium irradiation as a function of incident energy for three different surface orientations. Error bars correspond to 95% confidence intervals. Data for $\{100\}$ and $\{110\}$ surfaces in the absence of helium from Sefta et al. [21] are shown for comparison.

than $W\{100\}$ or $W\{111\}$ surfaces. This is qualitatively reasonable, because $\{110\}$ surfaces are the most closely-packed of the three, and hence most likely to produce a direct helium-tungsten collision. This observation is consistent with experimental results for single-crystal BCC metals [22], which showed the same difference between $Mo\{100\}$ and $Mo\{110\}$ surfaces. $\{111\}$ surfaces are the least closely-packed of the three orientations studied, and also show the lowest sputtering yields. Voitsenya and coworkers [23] came to the same conclusions for largely the same reasons based on experimental sputtering results. This is also supported by the respective depth distributions of helium at the end of each sputtering simulation, as shown in Figure 3. The sputtering yields also increase with increasing incident energy, but the sputtering yield begins to level off for the $\{111\}$ surface at 500 eV. This behaviour is consistent with the behaviour seen experimentally [24], but it should be noted that our previous simulations [21] showed no such decrease even at energies as high as 1000 eV, suggesting this observation may be due at least in part to sampling error. We emphasize again that these simulations neglect electronic stopping, which is a significant source

Table 2: Percentage of incident helium atoms that remain below a tungsten surface without reflecting or rapidly escaping as a function of helium energy and surface orientation.

Surface Orientation	Energy (eV)			
	200	300	400	500
(1 0 0)	53.4%	59.7%	62.0%	62.2%
(1 1 0)	40.0%	43.2%	47.5%	48.9%
(1 1 1)	63.3%	67.2%	68.9%	71.0%

of error, particularly at high energies. In particular, this is believed to be the primary cause of the discrepancy between the sputtering yields calculated here and by Sefta et al. [21] compared to those measured experimentally, as shown in Figure 4.

3.2. Depth Distributions

The incident energy of the helium atoms and the orientation of the free surface affect the fraction of helium atoms which embed in the metal. The fraction of atoms that do not reflect or rapidly escape into the gas phase are shown below in Table 2.

The depth distributions of helium after thermalisation are shown in Figure 3. Unsurprisingly, the mean depth increases as a function of incident energy. The depth is also highly dependent on the surface orientation, consistent with our previous findings for 100 eV helium [8] and with the recently published results of Borovikov [25]. In the (1 1 1) case, helium atoms are embedded substantially deeper than in the (1 0 0) case, presumably due to atomic channelling.

We would like to re-emphasize at this point that electronic stopping is neglected in these simulations. As such, it is perhaps inappropriate to consider the *absolute* depth distributions at face value. It is, however, acceptable to compare *relative* depths between different surface orientations, as electronic stopping is neglected similarly in such situations.

4. Comparison between Pure and Pre-Implanted Tungsten

4.1. Tungsten Sputtering

There appears to be no striking difference between the sputtering yield of pure tungsten and helium pre-implanted tungsten ($\Phi = 1.0 \times 10^{19}$ He.m⁻² and

Figure 3: Cumulative depth distributions of helium embedded in tungsten as a function of incident energy for three surface orientations. The ordinate of the middle three plots is $y = \log_{10}(1-u)$, where u is the ordinate of the corresponding upper plot. The bottom three plots show a sample of ~ 500 trajectories at 500eV from a normalised starting position for each surface orientation.

Figure 4: Sputtering yields as calculated in this paper and the work of Sefta [21] and select experimental data as collected in Ref. 24. Figure adapted from Refs. 21 and 24.

$\Phi = 2.2 \times 10^{19} \text{ He} \cdot \text{m}^{-2}$), within the uncertainty limits, as shown in Figure 5. It should be noted that $W\{100\}$ and $W\{110\}$ surfaces, unlike $W\{111\}$, do not show a significant concentration of near-surface helium atoms. As such, we have no reason to expect sputtering on such surfaces to be any different than for the non-implanted surfaces.

4.2. Helium Ejection

For the case of pre-implanted tungsten, the number of helium atoms ejected was also tracked. The overwhelming majority of cases resulted in the incident helium atom being either embedded or reflected from the free surface without dislodging any existing tungsten or helium atoms, with similar reflection probabilities as in the pure tungsten scenarios. This results in a net change of 0–1 helium atoms. However, in $\approx 1\%$ of cases, a net loss of helium was observed. The majority of such cases resulted in a net loss of one helium atom, with a maximum loss of 16 helium atoms observed in once instance. Large amounts of helium release occur when the impinging helium atom impacts the surface

Figure 5: Comparison between sputtering yield of pure and pre-implanted tungsten as a function of helium incident energy for a (1 1 1) surface orientation at 933 K. The pre-implanted “layer” of helium near the surface has very little, if any, effect on the sputtering yield.

directly over a near-surface bubble, locally increasing the kinetic energy and causing it to burst, as shown in Figure 6.

Ejection of more than one helium atom after a single bombardment was relatively rare, with the overall expected change in the number of helium atoms per incident helium atom ($\Delta\text{He}/\text{irradiation event}$) being around 0.6–0.7/event (see Table 3).

The differences as a function of fluence appear small until one considers the trend with respect to the pure tungsten cases as shown in Table 2. It is clear that the fraction of ion bombardments that result in embedding at 500 eV goes from 71% at no fluence to 70% at $\Phi = 1.0 \times 10^{19} \text{ He m}^{-2}$ to 69% at $\Phi = 2.2 \times 10^{19} \text{ He m}^{-2}$. A similar trend holds at all other energies: 400 eV, $69 \rightarrow 68 \rightarrow 66$; 300 eV, $67 \rightarrow 65 \rightarrow 64$; 200 eV, $63 \rightarrow 62 \rightarrow 61$. While not definitive (the differences are not substantially higher than the standard error), these trends suggest that the effective reflection coefficient rises slightly as a function of fluence.

Figure 6: Demonstration of a near-surface helium bubble bursting as a result of an ion impinging on the surface directly above it. Red (and smaller) atoms are tungsten, blue (and larger) are helium. (a),(b): Frontal and side snapshot prior to helium bombardment; (c),(d): Frontal and side snapshot after collision, showing sub-surface helium atoms ejected, (e),(f): Frontal and side snapshot showing removal of helium from a burst bubble.

Table 3: Probabilities of net loss (negative ΔHe), net gain (positive ΔHe), and no net change (zero ΔHe) of helium retention following a single helium-irradiation, and the expected value of ΔHe per irradiation event, for two different fluences for a pre-implanted, $(1\ 1\ 1)$ -oriented surface.

Fluence: $1.0 \times 10^{19} \text{ He} \cdot \text{m}^{-2}$				
Energy (eV)	Loss	No Change	Gain	$\langle\Delta\text{He}\rangle$
200	0.9%	37.3%	61.8%	0.60
300	0.9%	33.9%	65.2%	0.64
400	1.0%	31.1%	67.9%	0.66
500	1.1%	29.2%	69.7%	0.68

Fluence: $2.2 \times 10^{19} \text{ He} \cdot \text{m}^{-2}$				
Energy (eV)	Loss	No Change	Gain	$\langle\Delta\text{He}\rangle$
200	1.2%	37.8%	61.0%	0.59
300	1.4%	34.8%	63.8%	0.61
400	1.3%	32.4%	66.3%	0.64
500	1.6%	29.7%	68.7%	0.65

5. Conclusion

The sputtering yields calculated from molecular dynamics simulations of pure and pre-implanted tungsten from helium irradiation under ITER-relevant conditions are not strongly dependent on the presence of relatively low concentrations of helium near the surface. We find that sputtering yields depend on surface orientation and incident helium energy, from a maximum of 0.036 W/He for $\{1\ 1\ 0\}$ surfaces at 500 eV incident energy to a minimum of 5×10^{-4} W/He for $\{1\ 1\ 1\}$ surfaces at 200 eV incident energy. The $\{1\ 1\ 0\}$ orientations are the most prone to sputtering, followed by $\{1\ 0\ 0\}$ and lastly, $\{1\ 1\ 1\}$. We attribute these differences in surface orientation to the more closely-packed nature of $\{1\ 1\ 0\}$ surfaces compared to the other two orientations.

For a W(111) surface pre-implanted with 100 eV helium at a flux of $\Gamma = 7.9 \times 10^{25} \text{ He} \cdot \text{m}^{-2} \text{s}^{-1}$ to either a fluence of $\Phi = 1.0 \times 10^{19} \text{ He} \cdot \text{m}^{-2}$ or $2.2 \times 10^{19} \text{ He} \cdot \text{m}^{-2}$, the change in sputtering yields due to 200–500 eV helium bombardment is negligible (within the 95% confidence intervals).

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