

Modelling of W-Be mixed material sputtering under D irradiation

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Abstract. We present an atomistic study on the D irradiation on W-Be mixtures, including a comparison between Molecular Dynamics and Binary Collision Approximation methods. We compared the D reflection and Be erosion yields after the non-cumulative D impacts, concluding that both methods agree qualitatively, but low energy irradiation related chemical effects can be recognized in MD. We also followed the evolution of W-Be mixtures under cumulative D irradiation. At low energies, the surface deuterates, quickly saturating the D reflection and suppressing the Be erosion.

Keywords: *Tungsten, Beryllium, Deuterium, sputtering, atomistic simulations, Fusion reactor materials, ITER*

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1. Introduction

ITER is planned to be the first Tokamak fusion device producing more energy than it consumes, based on a deuterium (D)-tritium (T) plasma. Due to its design, the conditions (particle flux, thermal loads and temperature), and hence the material requirements, vary with location in the machine. Tungsten (W) will be used for the divertor and the main wall will be made of beryllium (Be). On the other hand, deuterium will be the most abundant plasma element. Erosion, transport and subsequent re-deposition of the plasma facing materials will lead to mixing of them, calling for a complete understanding of the interaction between W, Be, their mixtures and the D reaching from the plasma.

Aiming to understand such complex systems, different experiments have been carried out exposing W to impurity (Be, He)-seeded D plasmas. These experiments explore the W–Be material mixing and its properties under fusion reactor like conditions, such as the formation of different surface structures (Be deposition, W-Be alloying or *W fuzz* nano-morphologies) [1, 2, 3] and the effect of Be on the long-term D retention [4, 5] and erosion yields [6]. Besides the experimental work, particle balance models for W–Be material mixing [3], dynamic Monte Carlo simulations of Be deposition-erosion processes [6] and empirical equations accounting for the influence of the substrate temperature, recoil energy and the D to Be ratio [5] can be found in the literature. However, a detailed computational study of the above mentioned processes and a reliable database for the erosion, deposition and reflection yields is lacking; a single nano-scale study exists, focused on the Be erosion from Be_2W surfaces [7].

We present an atomistic study of W–Be mixtures under D irradiation, using both Molecular Dynamics (MD) and Binary Collision Approximation (BCA) methods. MD is a computational method able to simulate chemical effects, alloying and other many-body interaction related processes. Therefore, it is suitable for the study of low-energy irradiation effects in materials, such as material mixing and quantifying erosion and reflection yields, key to understand the plasma-wall interactions. However, MD requires complex set-ups and it is computationally expensive, being limited to short time and length scales, and consequent high fluxes. Also BCA method works on the atomic scale, but without considering the many-body interactions. It results in a method orders of magnitude faster computationally, allowing to form a complete database even at lower fluxes and higher fluences, but at the expense of missing out an accurate description of the low-energy interactions. It should always be possible to parametrize a BCA code to match the MD data, as otherwise it will be missing part of the physics. In the present work, we present a first comparison between these codes for W-Be-D compounds, aiming to improve the description of the composite substrates in the BCA code and to determine the energy threshold for the use of MD-BCA, together with finding the processes that govern the evolution of the mixed material surface under continuous D irradiation.

Further, in fields such as fusion, a direct comparison between the atomistic simulations and experiments is not always possible, as sputtering processes in fusion reactors often involve different effects such as redeposition, surface segregation, thermal diffusion and desorption [8]. Instead, our calculations help to identify and understand the key processes happening in the plasma facing materials, offering a more reliable database to the large scale codes. For example, the structure and depth profiles of the mixed materials will be used by rate equation codes [9], and the reflection and sputtering yields will be provided to impurity transport codes (ERO [10], WallDyn [11]).

2. Method

The Molecular Dynamics work was carried out using the PARCAS [12] code. A reactive bond order potential [13] was used to calculate the W–Be–D interactions.

The MD simulations were performed in two main steps. First, the W-Be mixture was prepared and then the D impacts were simulated.

The cells used for the non cumulative irradiation were created by randomly placing 3000 atoms in a $3.2 \times 3.2 \times 4.0$ nm box, at least 2 Å apart, with the different W:Be ratios (2:1 for W_2Be , 1:1 for WBe and 1:2 for WBe_2). Then, the cells were relaxed at 500 K, including the opening of the positive z-axis oriented surface. The cell used for the cumulative D irradiation was created by Be deposition on crystalline (100) W surface, by 3000 cumulative Be impacts at 50 eV, obtaining a $3.2 \times 3.2 \times 5.5$ nm cell: a 3.0 nm layer of pure crystalline W at the bottom and a 2.5 nm mixed W-Be layer on top, with an average 1:5 W:Be ratio and a density of 6 g/cm^{-3} . This structure was then repetitively quenched (heated up to 2000 K, kept at that temperature for 20 ps, cooled down to 500 K at a rate of 0.05 K/s and kept at 500 K for 30 ps) until its potential energy remained constant.

The amorphous W–Be mixtures were then exposed to 10000 non cumulative D impacts at 10, 50, 100 and 200 eV. The Be deposited W cell was irradiated by 2500 cumulative, low energy (10, 30, 50 and 100 eV) D impacts, resulting in a total simulated time of 17.5 ns, a flux of $2 \cdot 10^{28} \text{ m}^{-2}\text{s}^{-1}$ and a fluence of $2.5 \cdot 10^{20} \text{ m}^{-2}$. In all the cases, each impact lasted 7 ps: for the first 2 ps, while most of the physical interaction happened, the temperature was scaled to 500 K only at the periodic boundaries (3 Å in x and y borders). During the next 5 ps the whole cell was cooled down to 500 K in order to release the energy introduced by the recoil.

Further details regarding these relaxations, as well as a more thorough description of the simulation parameters, such as temperature and pressure control, treatment of sputtered and backscattered species, use of the periodic boundary conditions and electronic stopping, can be found in Ref. [14].

The analogous simulations were run using the BCA method too. The SDTrimSP [15] code was chosen for these calculations, in static mode (non-cumulative impacts), at normal incidence and the energy range as in MD, 10–200 eV. The target densities were taken from the MD simulation cells, and surface binding energies were set to the cohesive energies of the MD simulations: 14.50 g/cm³ and 7.55 eV for W₂Be; 12.32 g/cm³ and 7.10 eV for WBe; and 9.70 g/cm³ and 6.55 eV for WBe₂, respectively. The Ziegler-Biersack-Littmark (ZBL) potential and the surface binding model for composite targets were chosen. The inelastic losses were calculated using an equipartition between the non-local *Lindhard-Scharff* and the local *Oen-Robinson* models.

3. Results and discussion

3.1. A comparison between Molecular Dynamics and Binary Collision Approximation

In the first part of our study, we compare the two atomistic simulation methods commonly used to calculate reflection and sputtering yields, BCA and MD. Our goal is to determine whether and at which energy the BCA and MD results agree, together with studying how different parameters affect the comparison.

Regarding the W erosion, the MD simulations showed no W sputtering within the simulated 10–200 eV range. The BCA calculations also gave a zero yield below 200 eV. At 200 eV the BCA yield was about 10⁻⁵, which is out of reach for the MD statistics as only 10000 impacts were simulated.

As for the Be erosion yields, the MD and BCA results are in good agreement (Fig. 1 right). The small differences in the Be erosion trend arise from the enhanced Be erosion at low irradiation energies due to the Swift Chemical Sputtering (SCS) [16, 17] present in MD. This explains the higher Be sputtering in MD at low energies (10 eV), whereas at high energies (100 – 200 eV) the BCA calculations result in larger Be erosion yields than the MD simulations. Also, the W₂Be (50–100 eV) and WBe₂ (50 eV) cases show higher sputtering yields than those expected from purely physical sputtering (the BCA calculations shape).

In contrast, the D reflection yields (Fig. 1 left) are in qualitative, but not quantitative agreement. At low energies (10 eV) the BCA and MD results match. At higher energies (50–200 eV), both methods show the same trend; the quantitative differences are still reasonable considering the high sensitivity of the D reflection to the physical description of the material, as seen in Fig. 2.

To understand this quantitative mismatch in the D reflection yields – specially concerning the high energies – and the effect of the different parameters in the characterization of the mixed materials in the BCA code, we tested different set-ups. Besides the choice described in the Methods section, we run the BCA simulation using the de-

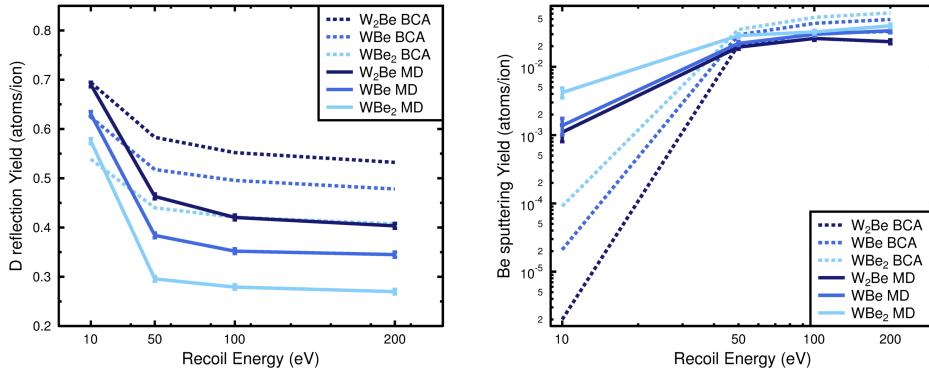


Figure 1. A comparison between the D reflection (left) and Be erosion (right) yields given by MD (solid line) and BCA (dashed line), as a function of the recoil energy, after 10000 non cumulative D impacts on amorphous W₂Be, WBe and WBe₂ surfaces, at normal incidence. The Be erosion is plotted in a log-log scale magnify the differences at low energies.

fault target density value in SDTrimSP (a linear interpolation between the densities of the elements). Different surface binding energies (SBE) were also tested, including half and double of the cohesive energy given by the MD simulations, and the default setting in the code (a linear interpolation between the SBEs of the elements). All these parameters lead to small quantitative changes. However, as seen in Fig. 2 (left), the largest differences in the D reflection arise from changing the atomic interaction potential and the surface binding model (isbv). The Be erosion yields (Fig 2, right), instead, are much less sensitive to all these changes. We conclude that the ZBL potential, also used for the repulsive interaction in MD, together with the surface binding model for composite materials described best the W-Be-D system.

In summary, within our irradiation energy range (10 – 200 eV) the MD-BCA comparison is qualitatively understood. The Be erosion yields converge at high energies and the effect of the SCS is highlighted at low energies. In contrast, the D reflection yields do not match at high energies. In the BCA, the D reflection strongly depends on the characterization of the material, whereas the Be erosion is not as sensitive to this parametrization. The ZBL potential with the surface binding model for composite materials, and the density and SBE taken from MD characterizes best the W-Be-D system. However, further BCA calculations are ongoing for a more accurate physical description of the composite materials, such as a new surface binding model.

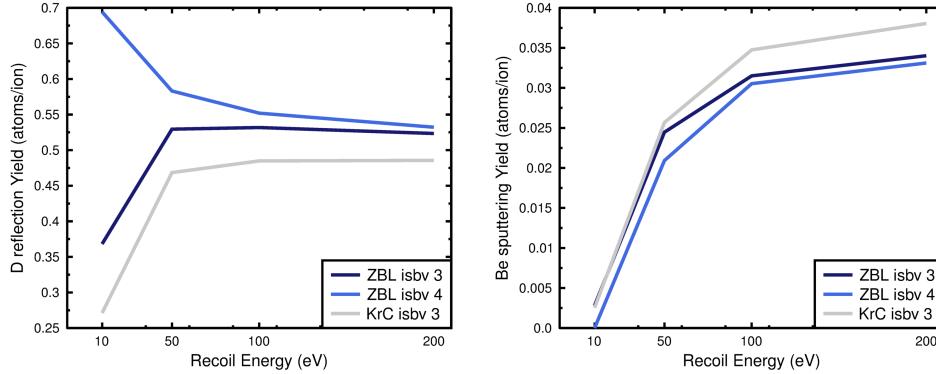


Figure 2. A comparison between the D reflection (left) and Be erosion (right) yields given by the BCA simulations for the surface binding model developed for pure elements (isbv 3) combined with the KrC and the ZBL potentials, the surface binding model for composite targets (isbv 4) together with the ZBL potential. This last case is described in detail in the Methods and used to compare to MD.

3.2. Effect of the surface composition on the sputtering yields

The effect of the composition and evolution of the surface are studied in what follows, focusing on the MD results. The BCA yields are included in the Be erosion analysis as a reference of the purely physical sputtering trend. First, the effect of the Be concentration is analysed in the non-cumulative simulations, as for each D impacts the substrate conditions (surface composition, morphology, etc.) are identical. Then, the surface evolution under the cumulative impacts is analysed and compared to the non cumulative results, to understand the effect of the surface deuteration and continuous Be erosion.

As for the non cumulative results, within the error bars of the MD yields, the D reflection drops linearly with the Be concentration at the surface (Fig. 3 left), at approximately the same rate for all the energies. In contrast, the Be erosion (Fig. 3 right) shows different trends depending on the energy. At low Be concentrations (W_2Be and WBe) and energies (10–50 eV), the Be sputtering is independent of the Be content of the cell. However, this erosion grows when further increasing the Be concentration (WBe_2). At higher energies (100–200 eV), the Be sputtering yields increase linearly with the Be concentration in the cell, as in the BCA calculations, indicating a purely physical sputtering.

We also analysed the effect of the surface evolution, focusing on the continuous Be erosion and deuteration. At low energies (10–30 eV), the D concentration quickly saturates (10^{20}m^{-2} for 10 eV, $2 \cdot 10^{20}\text{m}^{-2}$ for 30 eV) as the surface deuterates (Fig. 4 left). This quick pile up might partially be caused by the very high D flux in MD, missing out the effect of the D diffusion. Within the short statistics of the cumulative

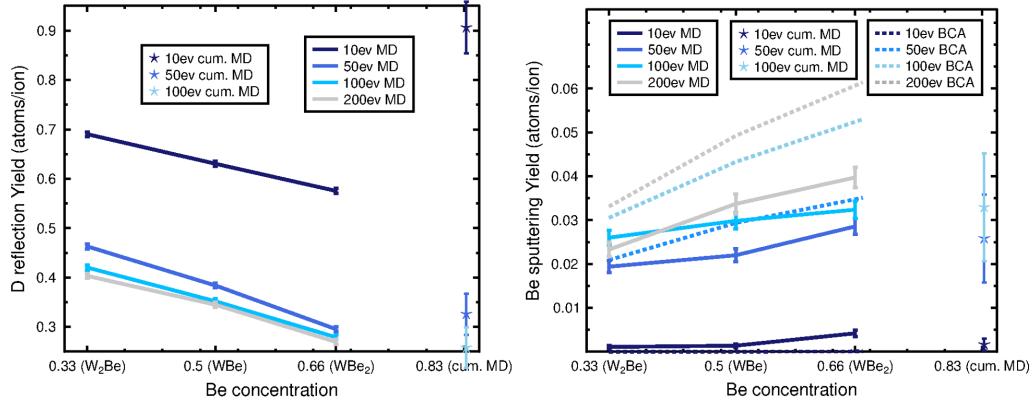


Figure 3. The D reflection (left) and Be erosion (right) yields given by MD for different recoil energies, as a function of the Be concentration in the substrate, after 10000 non cumulative D impacts on amorphous W₂Be (0.33), WBe (0.5) and WBe₂ (0.66) surfaces, at normal incidence. For a comparison, we include the cumulative irradiation MD results, simulated over a surface with a Be:W ratio of 5:1 (0.83). The Be erosion yields by BCA (dashed lines) are included as a physical sputtering trend reference.

simulations, the surface coverage with D does not show any effect on the evolution of the Be erosion (Fig. 4 right). However, the non cumulative results(Fig. 3 right) suggest that in the absence of D, at a 5:1 Be:W ratio the sputtering yields would be higher. Thus, we conclude that the Be erosion is suppressed by the large deuterium pile-up at the surface, an effect already reported for C [18]. At higher energies (50–100ev), the D is implanted in or beneath the deposited Be layer and the surface remains poorly deuterated. Within the MD fluences, the surfaces contain enough Be available not to alter the erosion. Hence, the D reflection and Be sputtering yields do not change over the cumulative, high energy impacts.

4. Conclusions

We presented an atomistic study of D irradiation on W-Be mixtures, at low-energies, using both MD and BCA methods, cumulative and non-cumulatively. We conclude that the methods agree; the Be erosion yields converge when increasing the irradiation energy, showing the chemical effects at low energies. In contrast, the D reflection yields –very sensitive to the surface characterization— agree only qualitatively. Further work is ongoing to optimize the BCA parametrization. The evolution of the surfaces due to the low energy D irradiation shows a D pile-up at the surface, suppressing the Be erosion.

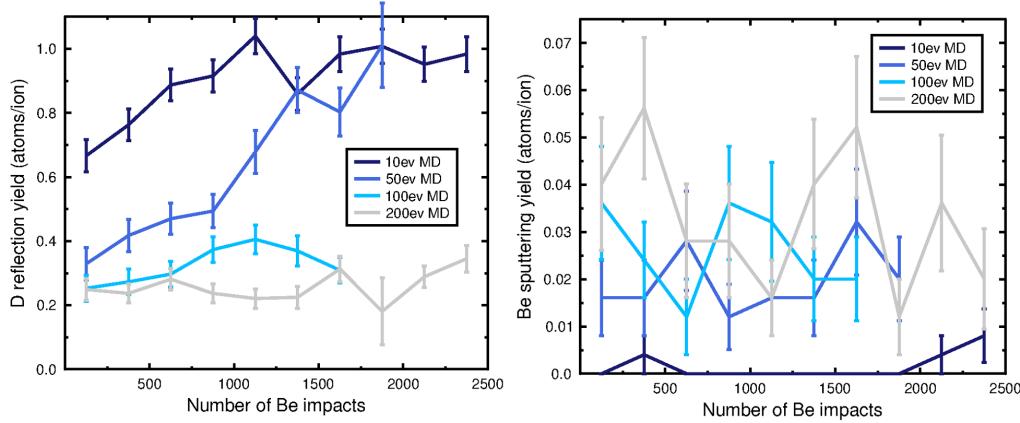


Figure 4. Evolution of the D reflection (left) and Be erosion (right) yields given by MD during the 2500 cumulative D impacts on the Be deposited W substrate, at normal incidence, for different recoil energies.

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