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Modeling of beryllium sputtering and re-deposition in fusion reactor plasma facing components

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Abstract

Quantitative characteristics of Be-sputtering by hydrogen isotope ions in a magnetron sputtering system, the microstructure and composition of the sputtered and re-deposited layers were studied. The energies of H^+ and D^+ ions varied from 200 to 300 eV. The ion flux density was $\sim 3\times10^{21}$ m^{-2} s^{-1}. The irradiation doses were up to 4×10^{25} m^{-2}. For modeling of the sputtered Be-atom re-deposition at increased deuterium pressures (up to 0.07 torr), a mode of operation with their effective return to the Be-target surface was implemented. An atomic ratio O/Be $\cong$ 0.8 was measured in the re-deposited layers. A ratio D/Be decreases from 0.15 at 375 K to 0.05 at 575 K and slightly grows in the presence of carbon and tungsten. The oxygen concentration in the sputtered layers does not exceed 3 at.%. The atomic ratio D/Be decreases there from 0.07 to 0.03 at target temperatures increase from 350 to 420 K.

1. Introduction

A protective armour will coat the plasma facing components (PFC) of the International Thermonuclear Experimental Reactor (ITER) – the first wall, the divertor and the limiter [1,2]. Currently, beryllium is being considered as the candidate material for fabricating the ITER first wall protective armour. The materials of the PFC armour will be subjected to combined plasma effects. Inside the ITER vacuum vessel, sputtered erosion products of different elements will re-deposit onto the armour which covers the first wall and other PFC. This may significantly affect its surface condition and properties.

To model possible patterns of plasma interaction with the first wall armour material under expected ITER conditions, we constructed a MAGRAS test facility [3] equipped with a magnetron sputtering system (MSS) in which different targets are exposed to a flux of high-energy deuterons. The key benefits offered by this facility are the ability to adjust the ion energy and ion flux density ranges and the possibility of obtaining required irradiation doses much quicker than they actually will occur within ITER, thus shortening the time of testing.

We modeled: (a) erosion of Be and W armour of the first wall and the divertor baffle as well as hydrogen isotope accumulation therein under a lengthy stationary effect of deuterium plasma and (b) formation of re-deposited layers in conditions close to those expected at the ITER first wall.

It is for the purpose of examining these issues, that experiments with the MSS providing a flux of low-energy (hundreds of eV) protons and deuterons in a wide range of gas pressures were used for modeling various regimes.

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2. Experimental facility and diagnostic techniques

MSS is a low-temperature plasma generator in which a discharge develops within crossed electric and magnetic fields. The ring-shaped part of the MSS cathode is bombarded by ions coming perpendicular to its surface. We used two MSS modifications: MSS-1 with electromagnetic coils and MSS-2 with a permanent magnet [3]. The key discharge parameters (gas pressure, current and voltage) are fixed using the data acquisition and processing system based on IBM PC/AT.

The experiments were performed using cathode-targets of two different designs. Compound targets used with MSS-1 (CT-1) consisted of six pellet-probes, located uniformly along the circumference, in the zone of maximum ion flux effect. Four pellets, later examined with surface and structural analysis methods, were fabricated from Be of different grades (TGP-56, TGShT, S-200F, hot pressed sheet beryllium) and polished. Two remaining targets were intended for measuring temperature. In our experiments, the hydrogen ion energy was about 200 eV, the average irradiation dose over the sputtering ring was from 1×10^{25} to 3×10^{25} m^{-2}, and the pellet temperature was around 700 K. The hydrogen pressure was $\sim5\times10^{-3}$ torr.

MSS-2 operates under higher H and D pressure and produces a higher intensity H^+ and D^+ ion flux onto the investigated target. Therefore on MSS-2 we used another type of compound target (CT-2) with a diameter of 40 mm, consisting of four 2 mm thick polished central sectors, pressed to the magnetron cathode by a beryllium ring. The sectors were either all-beryllium or made of different materials. We studied targets composed of beryllium and tungsten (another PFC armour material). In this case, the targets were combined differently in terms of Be/W area ratios [4] and used for studies of mutual re-deposition of the erosion products. Due to higher pressure the cathode erosion products scatter on the ambient gas and predominantly return back to the target surface. In cases of mixed-material targets, we examined the re-deposition patterns. The average irradiation dose in the experiments with MSS-2 was 4×10^{25} m^{-2}, and the temperature of exposed specimens was 350–420 K. Sectors made of different materials were exposed to ion fluxes of the same energy (200–300 eV).

A set of diagnostic techniques was used to analyze the eroded areas and re-deposited layers, including: gravimetric analysis – to measure the weight change of tested targets with an accuracy of 10^{-4} g; scanning electron microphotography – to examine the topography of eroded materials and re-deposited layers; optical and transmission electron microphotography – to determine the density and sizes of erosion products; profilometry of targets – to measure the erosion depth and thickness of layers re-deposited on the target; X-ray diffraction and electron-diffraction analyses – to examine the phase composition of eroded surface and re-deposited layers; elastic recoil detection method – to determine the distribution of hydrogen isotope atoms in the depth of the surface layers; Rutherford backscattering – to study chemical composition of different target zones and erosion products.

3. Results and discussion

3.1. Beryllium erosion products and deposited films

In a series of experiments with MSS-1 and CT-1, the density of the 200-eV proton flux was 3×10^{21} $m^{-2}\,s^{-1}$, and irradiation doses varied from 2×10^{25} to 3×10^{25} m^{-2}. Microphotographs of some typical parts of polished TGP-56 Be targets' irradiated surfaces show that ion etching has made visible differently oriented Be-grains of 8–15 μm. Pores with diameters of $\leqslant1$ μm have formed at zone boundaries and broken blisters are seen here and there.

Each of the four specimens made of different grades of Be showed roughly the same weight loss of about 7×10^{-4} g, corresponding to the Be-sputtering yield $Y\approx0.02$, which is consistent with theory and experimental results reported elsewhere. Transmission electron microscopy shows that the erosion products are Be-films and mostly tiny particles of 0.03–1.0 μm. The observed film scraps and particles of various forms could be fragments of blisters broken on the surface eroded by protons.

The function of Be in ITER is to provide a low-Z wall surface that getters oxygen. Be is reported to diffuse through BeO at temperatures above 700 K, yielding fresh surface layers for gettering. Thick BeO layers will eventually accumulate. Large area of oxide coverage on the first wall will lead to oxygen release by sputtering [1]. Therefore, it is of interest to study accumulation of the co-deposited hydrogen under growth of oxidized Be-layers.

In our experiments, the residual gas pressure after the chamber pumping-out was $\sim10^{-5}$ torr. The Be/O atomic ratio in the Be-films deposited on the Si-collectors, whose temperature during the experiments was $\leqslant100$ °C, corresponds to that of $BeO_{0,8}$.

The distribution of H-atoms in a ~100-nm deposited layer is fairly uniform. Cumulative H-concentration in the deposited film was 2.4×10^{17} cm^{-2} whilst the H/Be atomic ratio was around 0.2.

3.2. Re-deposition of Be-erosion products under deuteron bombardment

In modeling the re-deposition process, the idea was to cause sputtered Be-atoms to effectively return to the surface of the TGP-56 beryllium target exposed to high fluxes of deuterons [5]. In these experiments, we used MSS-2 and an all-beryllium target (CT-2-1). The deu-

terium pressure in the chamber was 0.07 torr, the irradiation dose of a 200-eV deuteron flux was 2.2×10^{25} m^{-2}, and the target temperature was kept at 420 K.

Fig. 1 shows schematically the post-irradiation structure of the target sector. There are fairly well-defined annular centrally symmetric zones. Deposition of sputtered material took place in zone 1. In zone 2 both Be-sputtering and re-deposition of the sputtered atoms occurred. It is separated from the re-deposition zone 4 by a narrow dark zone 3, adjoining the discharge boundary. During the ion bombardment, a screen shielded the exterior ring of sector 5.

Weighing results have demonstrated no change in the target weight after irradiation – the eroded atoms had effectively re-deposited back to the target. Part of the sputtered material is re-deposited outside the area of the beryllium target exposed to the deuteron flux (zones 1 and 4).

The co-deposited layers contain 50 at.% Be and 40 at.% O. The O/Be atomic ratio of ~0.8 prevails throughout most of re-deposited layers. This suggests that under the temperatures that existed during formation of the re-deposited layers in our experiment, sputtered Be-atoms trap oxygen from the residual gas while depositing onto the target surface.

The integral D-concentration in beryllium re-deposition zones is $\sim2\times10^{21}$ m^{-2}. The ratio of D-atom number to number of BeO molecules (D/BeO) there is ~0.15. It is determined by an elevated oxygen content in the amorphous re-deposited layers. Unlike the re-deposition area, in the sputtering zone, where surface is cleaned under the bombardment by intensive deuteron beam, oxygen concentration is only ~3 at.%, and O-presence is limited to the narrow (several tens of nm) near-surface layer. In the sputtering zone, D-atom distribution profiles have a table-like shape, usually attributed to high doses of ion implantation. Here, the D-concentration is much lower (2.5–4.0 at.%) than in the re-deposited layers.

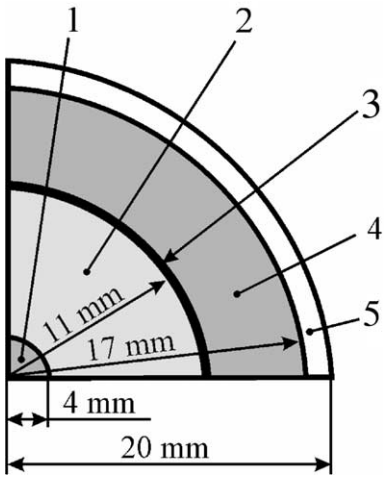

Fig. 1. The post-irradiation structure of a target sector: (1) deposition zone; (2) the zone of Be-sputtering and sputtered atoms re-deposition; (3) dark zone; (4) re-deposition zone; (5) the exterior sector ring shielded during the ion bombardment by a screen.

The Be-sputtering zone surface topography bears evidence of blistering (blisters are 0.5–1 μm across). The formation of separate cones, visible in the photographs, is probably due to different sputtering rates of beryllium and its oxide. X-raying proved that the sputtering zone surface layer subjected to D-ion bombardment consists mainly of α- and β-beryllium, and a non-essential amount of BeO (predominantly of α-type).

3.3. Effect of carbon on Be-behaviour

Carbon is one of the candidate materials for high-heat-flux divertor elements in ITER. Its effect on the Be-behaviour was studied by placing C-based materials at about 30 mm from Be-targets. CT-2-2 and CT-2-3 targets were exposed to D^+-ion doses of 8×10^{24} and 2×10^{25} m^{-2} with temperatures (controlled by thermocouples) of 350 and 410 K, respectively.

We measured geometric characteristics of sputtering and re-deposition zones. Zone 2, which is predominantly a sputtering area (Fig. 1), thinned in the CT-2-3 target by 3.75 μm, according to profilometry measurements. The deposit thickness in zone 4, which is a beryllium re-deposition area, increases with growth of irradiation dose and reaches ~80 nm and ~170 nm on average for CT-2-2 and CT-2-3 targets respectively. It varies radially and reaches around 230 nm within the space of 1 mm in zone 4 of the CT-2-3 target.

The microstructure analysis of the irradiated target surfaces has shown that blistering had also occurred in the sputtering zones. The blisters were 0.5–5.0 μm across. The analysis of recoil atom spectra revealed thin C- and D-enriched surface layers present in each zone of the target [6]. Fig. 2 shows the distribution of chemical elements in the re-deposition zones of CT-2-3 target.

D-atom distribution profiles for sputtering zones of CT-2-2 and CT-2-3 targets have a table-like shape like for the case of pure beryllium. The integral D-concentration in the sputtering zone decreased with temperature growth, and the D/Be atomic ratio reduced from 0.07 at 350 K to 0.04 at 410 K. This may be due to re-emission of deuterium from the target driven by opening of large blisters. The D-distribution in the sputtered parts of the target is confined to the narrow near-surface area (< 50 nm). In the zone, where Be-atoms are re-deposited, integral D-concentration increases with increasing thickness of the re-deposited Be-layer; D-distribution in depth of the layers is fairly uniform; along most of the deposit depth atomic ratio is O/Be ≈1.

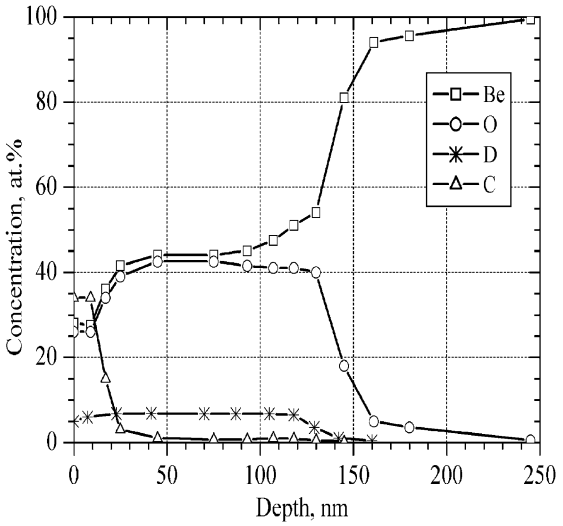

Fig. 2. Depth distributions of chemical elements in the re-deposition zones of CT-2-3 target.

In the bombardment-cleaned sputtering zones, the O/Be ratio is much lower than in the re-deposition area, but it is essentially higher than in the absence of carbon. The O-distribution profile has maximum in narrow, 10–15 nm-deep, near-surface layers. Carbon concentration (10–12 at.%) there is also lower in comparison with 24–35 at.% in re-deposition zones.

3.4. Co-sputtering and co-deposition of beryllium and tungsten

The experiments have shown that W-content in the re-deposited layers is practically independent of the Be/W area ratio in targets. The following are some of experimental results for a compound target with the Be/W area ratio of 1:3 [4]. A ~340 nm-deep near-surface layer of the Be-sector has a low (<1 at.%) concentration of tungsten, which is connected with relatively low sputtering and return rates of W. Its sputtering yield is 100 times as low as that of Be exposed to the same deuteron flux, and the return to the target surface is hampered by low efficiency of its heavy particles' thermalization on light D-gas.

But for the presence of ~1 at.% W in layers re-deposited on Be-sectors of compound targets, the chemical composition of such layers is very similar to that in layers re-deposited on homogeneous all-Be targets. The O/Be atomic ratio of ~0.8 persists across most of the re-deposited layer thickness. The integral D-concentration in the re-deposited layers on Be-sectors is $\sim 2\times 10^{21}$ m^{-2}. The D/BeO ratio there is slightly greater than that for an all-Be target. The thickness of a layer co-deposited with deuterium on either Be or W sectors of the target grows with increasing both irradiation dose and relative portion of Be-surface area on the target.

The integral W-concentration in the near-surface layers of Be-sector sputtering zones did not exceed 10^{19} m^{-2}. The D-concentration there (2.5–4.0 at.%) was lower than in the re-deposited layers. At the same time, D-concentration in the sputtering zones of the W-sectors was 0.2–0.3 at.%, that is, by 1–2 orders of magnitude lower than in the re-deposited layers.

Because of low rate of W-sputtering by D^+ ions with a 200–300 eV energy, close to the W-sputtering energy threshold, the surface microstructure of the sputtering and re-deposition zones on the W-sectors is practically identical with the initial one. Even at higher ion energies, changes in the all-W target surface microstructure are insignificant. The only difference is that the grain boundaries in the sputtering zone of such a target are more clearly distinguishable.

Fig. 3 shows that the atomic ratio D(H)/Be strongly depends on temperature. It decreases from 0.15 at 375 K to 0.05 at 575 K, being essentially lower than atomic ratios D(H)/C at the same temperatures in the layers formed by carbon co-deposition with D or H-atoms [2].

In the re-deposited Be-based layers containing carbon, the D-concentration is generally, somewhat higher than in those carbon-free. Likewise, the presence of W in such layers causes an increase – albeit at a lesser (~20%) extent – in the accumulation of deuterium. Where Be occupies the same vacuum volume as W and D, it evidently plays the dominant role in retaining D. Apparently, this is due to Be ability to getter O and thus give

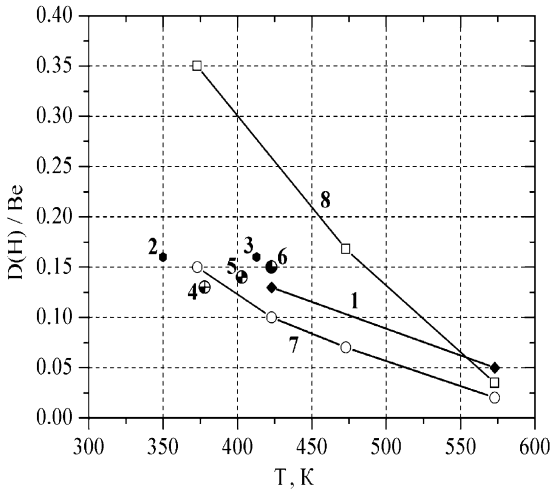

Fig. 3. Atomic ratio D(H)/Be in re-deposited layers: 1 – all-Be target; 2, 3 – the same, but with higher C-content (2 – target CT-2-2; 3 – target CT-2-3); 4, 5, 6 – compound Be–W targets with different Be/W area ratio (4 – 3:1, 5 – 1:1, 6 – 1:3), 7, 8 – data by Causey [7].

rise to the formation of hydroxyl compound $Be(OD)_2$ involving D-retention.

Fig. 3 also presents D(H)/Be atomic ratios from the MAGRAS experiments plotted together with two sets of data obtained by Causey in TPE-facility [7]. The similarity is particularly strong with Causey's results obtained under a controlled (relatively low) oxygen content. It should be noted that in our experiments re-deposition was accompanied by implantation of 200-eV deuterons, while Causey used ions with energies of $\leqslant 30$ eV.

4. Conclusions

1. The D/BeO ratio in the re-deposited layers decreases from 0.15 at 375 K to 0.05 at 575 K. The high D-concentration values are apparently due to the Be-ability to getter oxygen and thus give rise to the formation of hydroxyl compound $Be(OD)_2$. The layers re-deposited on the Be-sectors of the target contain $\sim$50 at.% Be and $\sim$40 at.% O.
2. Admixed carbon and tungsten enhance hydrogen isotope retention in the re-deposited layers. Deuterium accumulation in the layers re-deposited on the W-sectors of the target is due to its co-deposition with re-depositing Be-atoms.
3. The oxygen and deuterium content in the near-surface layers of the sputtering zones on the target is several times lower than in the re-deposited layers.
4. Physical sputtering of tungsten can be greater in the case where tungsten re-deposits with beryllium, than where it re-deposits on its own. In ITER such conditions may take place near the divertor baffle.

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