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Study of hydrogen isotope interaction with beryllium and carbon surfaces under their simultaneous exposure to stationary and powerful pulsed plasma

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Abstract

Formation of films on ITER first wall and divertor surfaces as a result of simultaneous interaction of H-isotopes with Be and C atoms was investigated in experiments simulating normal ITER operation and plasma disruptions. Under normal conditions, one can observe a continuous formation, growth and flaking of mixed Be + C films. Their Be/C atomic ratio varies from 1/2 to 2 (Be_2C). At high doses of deuteron irradiation (10^{24} m^{-2}) ratio D/(Be + C) ≈ 0.02–0.06. Under plasma disruption simulation, the D-concentration in mixed Be + C films is an order on magnitude smaller than under exposure to a stationary D-plasma.

1. Introduction

The presence of three different materials – beryllium (Be), tungsten (W) and carbon fiber composite (CFC) – in the ITER vacuum chamber inevitably results in their sputtering, redeposition, codeposition with hydrogen isotopes and formation of mixed layers.

According to Federici et al. [1], ~20 g of Be will be transported onto the CFC-covered divertor target area during each 400-s pulse under normal ITER operation. The interaction of Be^+-ions with C will result in the formation of mixed C + Be layers, and may cause the synthesis of Be-carbides. The fact of Be-carbide synthesizing on a substrate during magnetron sputtering of Be in CH_4 + Ar medium has been reported [2]. The formation of Be-carbide was also observed during PISCES plasma experiments at temperatures >300 °C [3,4]. As reported earlier [5,6], differently structured C-films with a varying content of H-isotopes can be observed in tokamaks.

The purpose of our experiments was to see how mixed layers (Be + C and Be + C + W) on a Be-substrate would affect the structure of films developing on Be-surface, and to investigate their adhesion and the ability to accumulate H-isotopes during normal ITER operation and plasma disruptions.

2. Experimental

Three sets of experiments were conducted to investigate mixed Be + C layers on Be and their ability to accumulate H-isotopes. To obtain the Be + C + H mixed layers, we exposed Be-targets to three irradiation doses (1×10^{23}, 2×10^{23} and 10^{24} m^{-2}) of acetylene (C_2H_2) plasma using the stationary plasma accelerator VITA with closed electron drift and lengthy acceleration zone [7]. To obtain the Be + C + W mixed layers, we directed a

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$C_2H_2^+$ plasma beam onto the conically shaped W-target with an apex angle of 45° surrounding a Be-target. The 300-eV plasma beam had a 10^{21}-$m^{-2}\,s^{-1}$ intensity. The Be-target temperature was 670 K.

In the second set of experiments, carried out using the PLAST beam-plasma discharge facility [8], a Be-target coated with a mixed Be + C layer was exposed to stationary 100-eV deuterium (D) plasma flux of a 10^{21}-$m^{-2}\,s^{-1}$ intensity to a dose of 10^{24} m^{-2}. To model the expected heat load on the ITER first wall (5 $MW\,m^{-2}$), we enhanced the applied power with an electron beam. Temperature was controlled by a thermocouple and was kept at preset $T = 670$ K by adjusting the water coolant flow rate V (prior to the experiments, all the appropriate calibration tests were made, and the $T = f(V)$ function was defined). A mixed Be + C layer on Be was formed under a simultaneous bombardment of Be and graphite surfaces with 20-keV Ar^+-ions, which caused a simultaneous sputtering of Be and C atoms and their deposition on the Be-substrate. The products of the Be + C layer sputtering by D-plasma were collected on a monocrystalline Si-plate.

The purpose of the third set of experiments was to investigate the impact of plasma disruptions on the erosion behaviour of the mixed Be + C layers. To this end, Be and CFC (specifically, the UAM-92-5D) targets were installed in the MKT electrodynamic accelerator [9] and exposed to ten 60-μs pulses of D-plasma with energy density of 900 $kJ\,m^{-2}$ per pulse. The plasma had a 10^{21}-cm^{-3} density and ion energy of up to 1–2 keV.

Following each plasma shot, a microstructure analysis of the target and the collector surfaces was performed using a JEOL scanning electron microscope (SEM). The chemical composition of the surface layers was determined by Rutherford backscattering (RBS) analysis using the Van de Graaf accelerator in which 1.6-MeV He^+-ions backscattered at 170° were detected with a surface barrier detector. D-distribution in irradiated targets was measured using the elastic recoil detection (ERD) method, in which a 2.2-MeV He^+-ion beam hit a sample under study at 15° to its surface. Beam trace was about 1.5×7 mm^2. The recoil atoms were detected at 30° relative to the direction of the incident He^+-beam. The D-atomic concentrations in absolute terms were determined by a calibration measurement of the energy spectra of standard samples.

3. Results

I. Surface microstructures of the Be-target subjected to different doses of C_2H_2 plasma exposure are shown in Fig. 1. A 10^{23}-m^{-2} exposure dose gives rise to a film on the Be-surface, consisting of small (~1 μm) aggregates, sometimes clustered in larger structures of up to 5 μm (Fig. 1(a)). A similar result was observed in experiments involving the synthesis of globular films on a W-substrate during exposure to a C_2H_2 plasma [7].

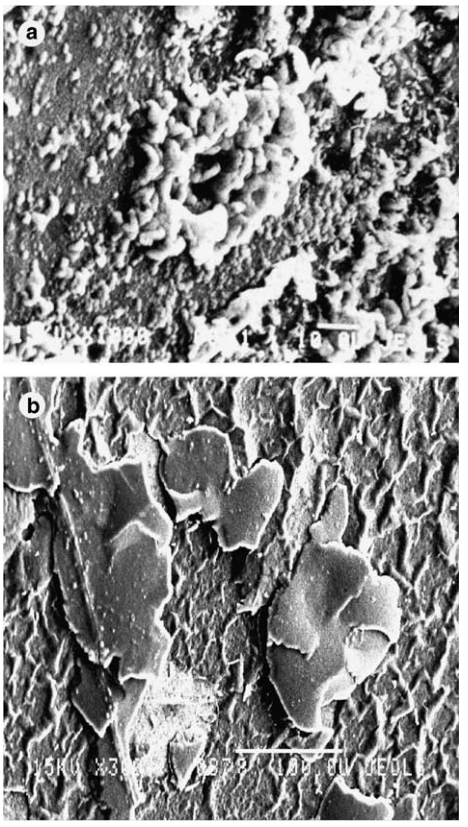

Fig. 1. Microstructure of Be-surface after its irradiation by C_2H_2 plasma flux with different irradiation doses (D): (a) $D = 10^{23}$ m^{-2}, magnification ×1000; (b) $D = 2 \times 10^{23}$ m^{-2}, magnification ×300.

The element distribution in the surface layer of the Be-target exposed to a 10^{23}-m^{-2} irradiation dose, determined by RBS technique in several zones of every ~15-mm specimen and averaged, is shown in Fig. 2. The RBS ion beam diameter of 1.5 mm was much larger than the above-mentioned sizes of aggregate clusters forming globular films (up to 5 μm). As the profiles suggest, a rather typical, ~1.5 μm thick, Be + C mixed layer has formed on the Be-surface, dominated by carbon near surface. It has moderate oxygen content of 2.7 at.%. Besides Be, C and O, the distribution of other elements (N, Fe, W), except hydrogen, was also measured using RBS technique. H-distribution was estimated as a difference between 100% and the sum of all the element concentrations (except H) in at.%. Hydrogen (~20 at.%) is found up to the depth of ~900 nm, but not deeper. The H/(Be + C) atomic concentration ratio of 0.27–0.28 is practically constant over the entire layer depth. So high H-concentration makes such estimation acceptable.

With a two-fold increase in the exposure dose, the C-atom concentration in the mixed layer grows, while the Be- and O-content goes down, the H-concentration

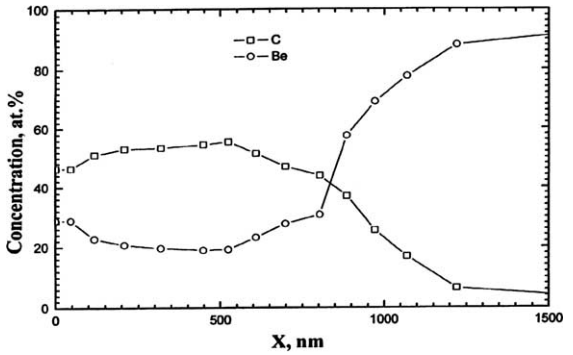

Fig. 2. Be and C distribution in the near-surface layer of the Be-target irradiated by $C_2H_2^+$-ions at a dose of 10^{23} m^{-2}.

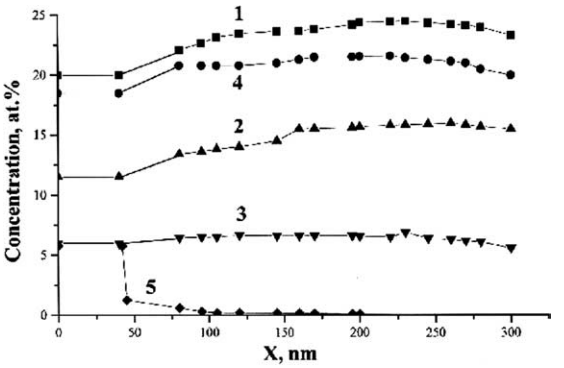

Fig. 3. Distribution of hydrogen atoms in depth of mixed layers at different doses of irradiation by $C_2H_2^+$-ions (D): for Be + C layer (curve 1: $D = 10^{23}$ m^{-2}; curve 2: $D = 2\times10^{23}$ m^{-2}; curve 3: $D = 10^{24}$ m^{-2}) and for Be + C + W layer (curve 4: $D = 2\times10^{23}$ m^{-2}). Curve 5 shows hydrogen distribution before irradiation.

falls to 12–14 at.%, and the H/(Be + C)-ratio reduces to 0.16–0.17. The films on the Be-surface flake off (Fig. 1(b)). This effect is different from that observed in experiments with W, where a dose increase led to the development of a cauliflower-shaped globular film structure [7].

In cases where the irradiation with C_2H_2-molecules happens concurrently with the deposition of sputtered W-atoms on the Be-surface, W can only be found in the near-surface ~300-nm layer. However such a limited occurrence of W is large enough for the film growing on the Be-substrate to considerably modify its structure, properties and composition as well as dramatically change the H-accumulation behaviour. The presence of 7.5 at.% W in the mixed Be + C layer, as determined by the RBS-technique, promotes the synthesis of a less brittle film on the Be-surface.

Exposure of a Be-surface to a 2×10^{23}-m^{-2} dose, in contrast to Fig. 1(b) case, caused only a local flaking of the film. It is possible to distinguish up to three layers of flakes in SEM microphotographs. The latter fact suggests that the flaking-off and growth of the film proceeded continuously during the irradiation. The H-content in the Be + C + W film is ~18 to 20 at.% (Fig. 3, curve 4). This value is 1.33 times greater than in the Be + C coating formed on Be under the same C_2H_2 plasma irradiation conditions in the lack of the W-impurities (Fig. 3, curve 2).

The hydrogen depth distribution profiles for films formed on Be, as determined by the ERD-method and shown in Fig. 3, suggest that under any C_2H_2 plasma irradiation dose, H-distribution in the 300-nm near-surface zone of mixed layers is uniform, and that H-concentration decreases with increasing irradiation dose. At a dose of C_2H_2 plasma exposure of 10^{24} m^{-2}, the H-concentration drops to 6 at.% across the surface layer due to development of a sponge-like film structure. Consequently the H/(Be + C)-ratio in the surface layer decreases to ~0.08. Fig. 3 also shows that before the exposure to plasma (curve 5), H occurred in the Be-target only in the narrow near-surface layer of ~45-nm at a concentration of ~6 at.% – as that observed after exposure to a 10^{24} m^{-2} dose (curve 3). Apparently, under the impact of the irradiation, hydrogen leaves the Be + C film almost entirely.

Approximately uniform H-distribution profiles may be explained by constant codeposition rate during the film formation at low irradiation doses and by development of a sponge-like structure in the 300-nm near-surface zone of mixed layers where H(D)-distribution has a table-like shape [10] at higher irradiation doses.

II. Similar results were obtained in the PLAST facility experiments involving the irradiation of a Be + C coated Be-target. Whereas before the exposure the Be + C coating has a fine grain structure, after the exposure it appears to be covered with a large number of small holes. The layer deposited on the Si-collector is not continuous either.

Fig. 4 shows the chemical composition of the layer redeposited on the Si-collector. The distribution of Be (~60 at.%), C (~26 at.%) and O (~10–12 at.%) in this ~1-μm layer is uniform.

An electron diffraction pattern of the deposit showed, along with the intensive reflections from the polycrystalline Be, some extra lines corresponding to interplanar spacings typical of beryllium oxide (BeO) and beryllium carbide (Be_2C). The Be_2C-reflections are blurred, have low intensity and a diffusion halo – the features characteristic of a dispersed, nearly amorphous structure. The Be + C films, including Be_2C, are brittle, non-continuous and readily flaking-off, similar to the films observed in the acetylene plasma experiments.

Fig. 5 presents the D depth distribution profiles for a Be + C layer on Be (670 K) exposed to a 10^{24} m^{-2} dose (curve 1) and for a layer redeposited on the Si-collector (curve 2).

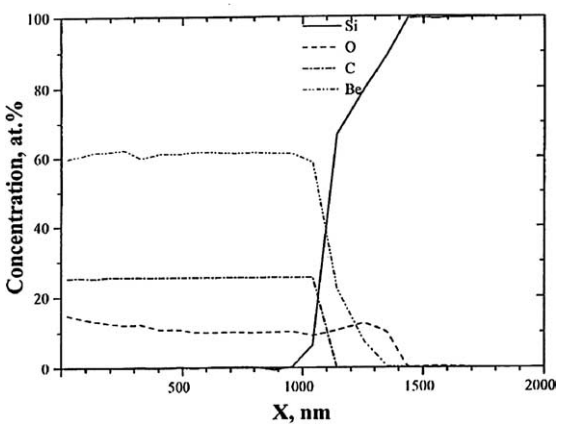

Fig. 4. Chemical composition of the mixed Be + C layer redeposited on the Si-collector.

The D/(Be + C) atomic concentration in the Be + C film on the Be-target was 0.04. A comparison with curve 3 in Fig. 3 demonstrating H-distribution in the Be + C layer with a Be/C ≈ 1/2 suggests that, under conditions similar to those that existed in the C_2H_2 plasma exposure experiments (at the same dose and temperature), the atomic ratio D/(Be + C) in a Be + C layer with majority Be-content (Be/C ≈ 2.3) is half as high. This value is a bit higher than the atomic ratio D(H)/Be in redeposited Be-layers reported by Zimin et al. [11] and Causey and Walsh [12]. Zimin provided a dependence of D(H)/Be, measured by his team (up to a dose of 2×10^{25} m^{-2}) and Causey and Walsh, against temperature. According to his measurements, D/Be ratio in the redeposited layers decreases from 0.15 at 375 K to 0.05 at 575 K. Above-mentioned value of D(H)/Be (0.04), according to his and Causey's studies corresponds to a temperatures 520–580 K, but Zimin notes that admixture of carbon promotes the higher hydrogen isotope content in redeposited layers what is equivalent to a shift

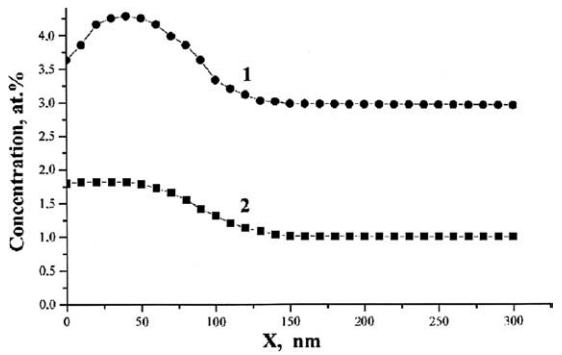

Fig. 5. Profiles of deuterium atom distribution at irradiation temperature of 670 K: in Be + C layer on Be at dose of 10^{24} m^{-2} (curve 1) and in a redeposited layer on a Si-collector (curve 2).

of the D(H)/Be dependence towards the higher temperatures. Guseva et al. [13] has found that H/Be atomic ratio at 370 K decreases with irradiation dose growth from 0.16 at doses of 5×10^{22}–1×10^{23} m^{-2} to 0.04 at 1.5×10^{25} m^{-2}.

Due to its codeposition with Be and C-atoms, D accumulates throughout the thickness of the layer redeposited on the Si-collector during sputtering of the Be + C coating on the Be-target by D-plasma. As in the experiments on Be-irradiation by $C_2H_2^+$-ions, the mixed layers contain D in small quantities: it escapes through the pores in the layers. The D/(Be + C)-ratio in the Be + C layer redeposited on Si-collector was 0.017.

III. The Be-surface, exposed to 10 pulses of a high-power (900 kJ m^{-2} per pulse) D-plasma flux in experiments involving a simultaneous irradiation of CFC and Be-targets under simulation of plasma disruptions, is cracked along the grains boundaries. The grains are richly covered with 0.25–2.0-μm Be-droplets. Some of the droplets are on the cracks: apparently, they returned to the target from the shielding plasma. The droplets can also be observed on the cracked CFC-surface. The surface of the Be-target is covered by ~60 nm thick mixed layers containing Be (39.0 at.%), C (32.7 at.%) and O (28.3 at.%) on the layer surface. Integral concentrations of D-atoms in the CFC and Be-targets are the same: they are about an order of magnitude less than those in the Be + C layers after exposure to a stationary plasma [14].

In these experiments temperature of Be-target reached Be-boiling temperature. According to measurements using infra-red pyrometer, temperature of the CFC-target was up to 3650 K.

4. Conclusions

Experiments simulating normal ITER operation and plasma disruptions have been performed to study the formation of films on ITER first wall and divertor surfaces as a result of a simultaneous interaction of H-isotopes with Be and C-atoms.

1. The Be/C-ratio in Be + C mixed layers varies from ~1:2 to ~2:1 (Be_2C). Under high irradiation doses (10^{24} m^{-2}), the concentration of H-isotopes in the films is small: D/(Be + C) ≈ 0.02–0.06. The films flake off readily. During exposure to a D-plasma, films grow and flake off continuously. H-isotopes do not permeate beyond the mixed layers.
2. The formation of mixed Be + C layers on plasma-facing ITER-components would protect those components from erosion and H-accumulation.
3. Simultaneous irradiation of Be and CFC under simulation of plasma disruptions causes the formation of mixed layers containing Be, C and O-atoms. The Be-surface cracks along the grain boundaries. D-con-

centration in mixed Be + C layers on Be and CFC-substrates is an order of magnitude smaller than following exposure to stationary plasma.

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