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Modification of optical properties of Be mirrors under bombardment by deuterium ions

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Abstract

Effects of D ion bombardment on Be mirror reflectance, $R(\lambda)$, in $\lambda = 220$–650 nm were studied as modelling the impact of charge exchange atoms in a fusion reactor. Without any mass change of mirror, a sharp drop of $R(\lambda)$ was observed for different kinds of beryllium at high ion energies (600–1350 eV) after ion fluence $\sim 10^{18}$ ions/cm^2. It is hypothesized that under deuterium ions the BeO surface film was transformed into the Be(OD)$_2$ film accompanying by changing the film optical properties. Effects of ion energy and ion fluence variation on $R(\lambda)$ of Be mirror are discussed.

1. Introduction

Beryllium was selected (e.g., [1]) to be the main plasma facing material in ITER. Eroded under plasma impact beryllium will be transported along the vacuum chamber and deposited on surfaces remote from the plasma confinement volume where deposition prevails over erosion. The first mirrors (FM) used for plasma diagnostics will be among remote components and thus highly probable that with time they become coated by Be film. As can be estimated, the Be film of ~20 nm thick will transform the reflectance of any metal mirror to values typical for reflectance of a bulk Be mirror. Thus it is not excluded that just beryllium should be used as the FM material. For safe ITER operation the behavior of the in-vessel Be mirrors in the ITER environment has to be analyzed. The data for such analysis can be obtained in modelling experiments with Be mirrors treated in conditions close to ITER conditions where FM surfaces will be bombarded by charge exchange atoms (CXA) of hydrogen isotopes with a wide energy distribution. This will occur in a residual background containing working gases (deuterium and tritium) and also hydrocarbon and oxygen-containing molecules.

Modelling experiments were begun in Kharkov three years ago and some results were previously presented [2,3]. The most important result was observation of the sharp drop of reflectance in the wavelength 220–650 nm after beryllium mirrors were subjected to bombardment by keV ions of deuterium plasma. This was observed at a low ion fluence when no mass loss of Be was measured and the surface was not modified owing to ion sputter erosion. The drop of reflectance at low ion fluence was shown to depend on the ion energy, i.e., the higher the energy the deeper the drop of reflectance. Such change of reflectance was hypothesized to be a surface effect: due to partial transformation of the upper oxide layer that naturally coats the Be mirror into a hydroxide film. It was found that the following exposure to low energy ions ($\leqslant 60$ eV) leads to gradual recovering of reflectance close to the value measured before the drop. In this paper we present some additional data obtained in similar experiments.

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2. Experimental

The majority of data on the behavior of Be mirrors exposed to ions of deuterium plasma were obtained using the experimental equipment described in [4]. For deuterium plasma generation ($n_e \leqslant 10^{10}$ cm^{-3} and $T_e \leqslant 5$ eV) a cw ECH (Electron Cyclotron Resonance) discharge at frequency 2.37 GHz in a double-mirror magnetic configuration was used. Plasma ions were accelerated by negative biasing of a copper holder with the mirror installed (discs with diameter 22, ~3 mm thick, weight ~2 g). The water cooled holder was placed in the path of the plasma streaming out of the magnetic mirror along the magnetic axis. With fixed biasing voltage in the range 20–1350 V the ion current density was ~1 mA/cm^2. The time of exposure of mirror to ion impact varied from 2 min to 3 h. After each exposure the measurements were provided of the reflectance at normal incidence (five measurements for every of 10 wavelengths in the range $\lambda = 220$–650 nm with the spread of R values in the limit ±1.5%) and the mass loss of the Be samples (by weighing to an accuracy of ~20 μg).

In addition, several Be mirrors were exposed to deuterium ions of a plasma reflex discharge in the longitudinal magnetic field 0.125 T. The Be sample was installed as one of the water-cooled cathodes with the second cathode made of stainless steel. For the discharge voltage 800 V the ion energy distribution has a maximum near 650 eV with a half width ~120 eV [5]. The ion current density to cathodes can be varied in the range 0.4–4 mA/cm^2.

In both devices the ion flux was not mass-separated and consisted not only of deuterium atomic and molecular ions but a small percentage of light impurity ions also. This resulted in an increase of the sputtering rate of mirrors compare to published data [6]. In the ECH case the increment was found to reach ~60% for Cu and Mo samples.

Some results of experiments on Be mirrors with ECH discharge plasmas were discussed in papers [2,3] for one kind of beryllium. In contrast, the results of experiments in a reflex discharge were not analyzed yet. In the present paper data from both devices are compared for mirrors of different kinds of beryllium, and emphasis is placed on the reflectance recovery owing to low energy ions bombardment.

3. Results

In Fig. 1 the results obtained in both experiments are presented as the dependence of reflectance on ion fluence. In the case of ECH plasma the biasing voltage was −1350 V and minimum exposure time was 120 s, and the minimum ion fluence ($F \approx 7 \times 10^{17}$ ios/cm^2) much exceeded the minimum for the reflex discharge case ($F \approx 1 \times 10^{17}$ ions/cm^2). The method of Be sample preparation and their structure were also quite different: the sample exposed in reflex discharge was the TShP-56 type, and the sample bombarded by ECH plasma ions was prepared by TGP-technology from powder. Nevertheless, the reflectance behavior under bombardment by deuterium plasma ions is qualitatively identical: namely, there are two scales of reflectance degradation; initially rapid for fluence $F < 10^{18}$ ion/cm^2 and then much slower with increasing fluence. Similar two-stage dependence of $R(F)$ was observed for mirrors of other kinds of bulk beryllium and for Be films deposited on glass and copper (results were not shown here). At the first stage the $R(F)$ dependence shows a fast drop without any mass loss measured, and at the second stage, with the orders of magnitude longer exposures to similar high energy ions, the mass loss becomes significant and was found to increase proportionally to the total ion fluence but the $R(F)$ dependence decays slowly (Fig. 1). The rate of $R(F)$ decay at the second stage of experiments depends strongly on the structure of mirror material: the lowest rate R degradation was observed for Be films deposited on the copper substrate. Such two-stage $R(F)$ dependence indicates different reasons of R degradation at low and high ion fluences: corresponding to chemical transformation of the upper oxidized layer and physical sputtering.

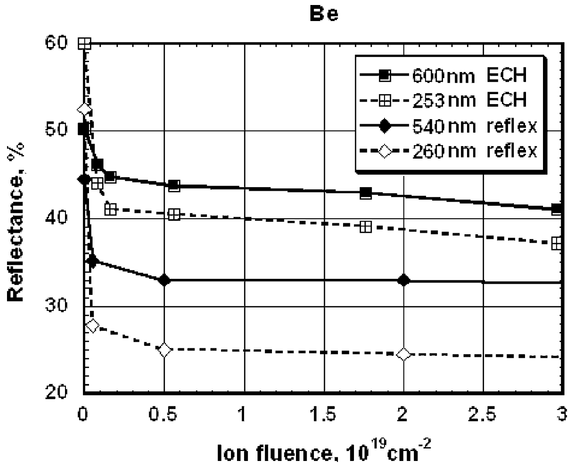

Fig. 1. Comparison of behavior of Be mirrors exposed to ions of deuterium plasma of ECH (squares) and reflex (rhombuses) discharges.

As for the low ion fluence effects, it has been shown earlier [3] that the result of ion bombardment on modification of optical properties of Be mirrors increases with increasing ion energy. In [3] to obtain the ion energy dependence of reflectance drop five Be mirror samples with almost similar initial optical properties were bombarded for similar exposure time (2 min) by ions of different energies (50, 100, 200, 650, and 1350 eV). As distinct from that experiment, in the present study the effect on R recovering under low energy ion bombardment was used in practice. Namely, the same Be sample (TGP-56) was in series exposed for short times (10 min) to ions accelerated by fixed voltage applied to the sample holder in the range 200–1350 V resulted in reflectance drop. After that the mirror was exposed to low energy ions (the holder biasing −20 or −60 V) for much longer times (several hours) until the reflectance has fully recovered. In Fig. 2 the dependences of the R decrement, $\Delta R(E_i) = R_{E_i} - R_0$, for three wavelengths are shown (R_0 is reflectance before and R_{E_i} – after exposure to ions with energy E_i).

The increase of the $\Delta R(E_i)$ decrement with increasing ion energy can be explained by differences in thickness of the transformed layer at each ion energy. The smaller energy ions have shorter mean projected range in the BeO film and accordingly a thin outer BeO layer is (presumably) transformed into the hydroxide film. With increasing ion energy the thickness of the transformed layer rises due to increase of the mean range of D^+ ions in BeO film and thus the portion of transformed oxide layer also grows. Fig. 3 shows the energy dependence of the calculated mean range of deuterium ions in BeO. It follows from data of Fig. 2 that the energy of bombarding ions which induce the reflectance drop (after 10-min exposure) trends to saturate and the saturation level is in the range 1.2–1.5 keV.

The mechanism of restoration of the Be mirror reflectance owing to exposure to low energy ions after the fast initial drop caused by high energy ion bombardment is not clear yet. However, the restoration effectiveness is demonstrated by data in Figs. 4 and 5. In Fig. 4 the recovery of reflectance at $\lambda = 253$ nm is shown

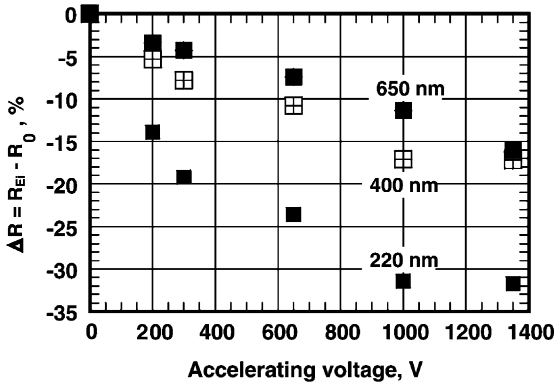

Fig. 2. Degradation of Be mirror reflectance at indicated wavelengths on bombardment with ions of indicated energy (in V).

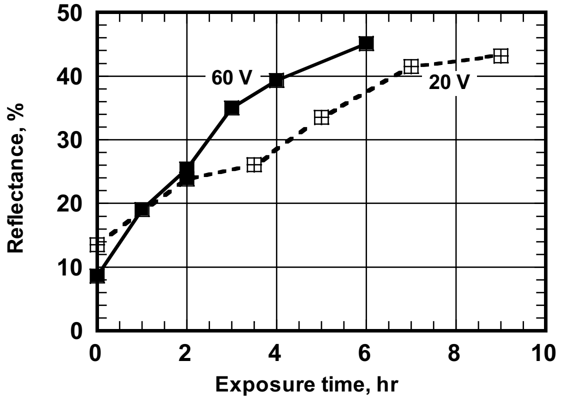

Fig. 4. Restoration of Be mirror reflectance due to bombardment by 20 and 60 eV ions of deuterium plasma following 10 min exposure to ions with energy 1350 eV.

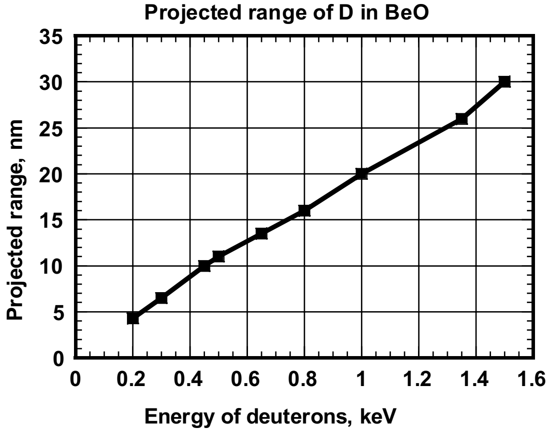

Fig. 3. Energy dependence of calculated mean projected range of D in BeO.

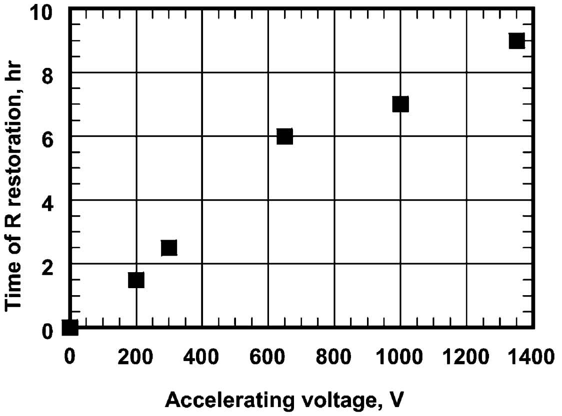

Fig. 5. Time of Be mirror reflectance restoration during exposure to 20 eV energy ions of deuterium plasma after drop due to bombardment by ions of indicated energies: 200, 300, 650, 1000 and 1350 eV.

for two mirror holder biasing voltage: −20 and −60 V. In Fig. 5 the time of R recovery (at $\lambda = 253$ nm) for the 20 V case is plotted as a function of higher ion accelerating voltage, which resulted in an initial drop of R at low ion fluence, Fig. 2. The exposure time values for the graph in Fig. 5 are those that needed for the dependences similar to presented in Fig. 4 to achieve the saturation level.

4. Discussion

Results shown in Figs. 1 and 2 as well as those published earlier [2,3] agreed with the concept that the drop of Be mirror reflectance under short exposure times to high energy deuterium ions is a result of partial chemical transformation occurring inside the BeO film. The results supporting this concept are:

1. The fast drop of reflectance was observed for mirrors fabricated of different kinds of beryllium and was never accompanied by any mass loss (present).
2. Annealing in vacuum partly restores the reflectance [2,3].
3. Ellipsometry shows an increase of the extinction coefficient after film transformation [3].
4. The procedure with drop and restoration of reflectance was repeated several times for the same Be samples without changing qualitatively the character of R behavior ([3] and present).
5. With fixed ion fluence (even less than 10^{18} cm^{-2}) the absolute R drop increases with increasing ion energy ([3], present).
6. This effect was observed at two different plasma devices and for Be samples with different structure (present).

The candidate chemical process, hypothesized in [2,3] and here, is transformation of BeO film (intrinsic to any beryllium surface) into a Be(OD)$_2$ film according to the reaction:

$$2\mathrm{BeO} + 2\mathrm{D} \Rightarrow \mathrm{Be(OD)_2} + \mathrm{Be} + 0.7\ \mathrm{eV/D}. \qquad (1)$$

The role of this reaction in the process of interaction of hydrogen isotopes with beryllium was discussed in [7,8]. In particular, it was suggested that this reaction was partly responsible for growth of the oxide film on Be surfaces exposed to D ions [9] or D atoms [7] when there are oxygen-containing molecules present in the vacuum atmosphere. Several mechanisms were discussed in [7,8,10] for oxide film to grow when Be is bombarded by D ions (or atoms) in a poor vacuum, and one mechanism is the release of a free Be atom for each hydroxide molecule created. These vacuum conditions were realized in our experiments and at least two experimental observations indirectly supporting the gradual growth of the oxide film. (i) The mass loss measurements for the given ion fluence shows that the ion sputtering erosion of Be mirrors is close to that estimated for molybdenum mirror samples exposed to ion bombardment under similar conditions, in spite of factor ∼5 difference between sputtering coefficients for these metals [6]. (ii) The gradual increase of R drop, caused by exposure to higher energy ions, was observed in the course of long experiments with the same Be sample, i.e., when the total time of exposure to deuterium plasma ions was increased.

Under these conditions the direct sputtering of metal beryllium is impossible because of the oxide film covering, and the sputtering process would be realized through sputtering of BeO and Be(OD)$_2$. That is, the development of surface micromorphology will be defined by the surface inhomogeneity of BeO formation but not by inhomogeneity of sputtering of metal beryllium due, for example, to different crystallographic orientation of individual grains, as was observed in [4] for Cu mirrors. In this sense the behavior of Be mirrors under long-term sputtering in vacuum with traces of oxygen has to be different, compared to the behavior of other metal mirrors where the metallic surface is directly sputtered by projectiles.

Coming back to results in Fig. 2, two cases are possible for the $\Delta R(E_\mathrm{i})$ trend to saturate. The first is that the thickness of the BeO film is less than the mean D ion range in the film, and the film partly transforms according to reaction (1), then covers the whole oxidized layer. With this, the well-defined saturation has to be observed starting from the definite ion energy. The second case, when the thickness of BeO film exceeds the mean ion range, would be characterized by a gradual approach to a saturation level, and the comparison of experimental data with data in Fig. 3 leads to an estimate of the minimum limit for thickness of the oxygen-containing layer. The data shown in Fig. 2 correspond exactly to this second case. By comparing Figs. 2 and 3 the thickness of the BeO film is in the range ∼20–25 nm, which is to be a reasonable estimate.

The effect of reflectance restoration of Be mirrors exposed to low energy ions allows optimism that in a fusion reactor the low energy portion of CXA flux will considerably exceeding the high energy portion [11], and a similar effect would counteract any reflectance degradation of Be mirrors caused by the higher energy tail in CXA energy distribution.

5. Conclusions

1. The effect of reflectance degradation in UV and Visible parts of the spectrum found earlier for Be mirrors subjected to low fluence D ion bombardment was confirmed by experiments on another plasma device and for Be samples with different structures. This ef-

fect is hypothesized to be due to partial transformation of the uppermost BeO layer into a $Be(OD)_2$ film.

2. Practically full reflectance restoration was observed after Be mirrors with transformed top layer were given a long-time exposed to low energy D^+ ions (20–60 eV).
3. The chemical processes on the Be surface, subjected to bombardment by deuterium plasma ions in an atmosphere with oxygen-containing molecules, result in decrease of the Be sputtering yield to the value measured for Mo sample bombarded by ions in similar experimental conditions.

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