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Letter to the Editors

Codeposition of deuterium with BeO at elevated temperatures

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Abstract

The trapping of energetic deuterium codeposited with BeO has been measured on a silicon collector probe in the temperature range from room temperature to 800 K. Deuterium codeposits with BeO at room temperature at a ratio of 0.36. At elevated temperatures the amount of codeposited deuterium decreases. At 573 K the ratio D/BeO is 0.17 and decreases to 0.07 at 773 K. The thermal release of deuterium codeposited at room temperature begins at 400 K. At temperatures above 800 K all deuterium is released.

1. Introduction

Beryllium, carbon and tungsten are currently anticipated as plasma facing materials for the International Thermonuclear Experimental Reactor (ITER). The plasma facing materials are subject to erosion, deposition and implantation by bombardment with ions or energetic neutrals from the plasma [1]. There are erosion dominated areas, such as the plasma facing parts of the vessel walls [2–5] and the divertor plates in the region of the plasma strike point [6]. The eroded material is redeposited at some other areas of the machine. Such deposition dominated areas are found in all thermonuclear fusion experiments [7–10].

A great concern is the ability of these redeposited layers to trap large amounts of hydrogen isotopes including tritium by codeposition [11,12]. Codeposited layers grow about linearly with the discharge time. Several kg of tritium are anticipated to be codeposited with carbon in ITER [12].

The phenomenon of codeposition has been largely investigated and is reasonably understood for the buildup of hydrogen rich carbon layers [13–15]. The hydrogen concentration of these plasma deposited layers depends on the ion energy and the target temperature. At room temperature and ion energies above about 100 eV the ratio of hydrogen to carbon is about 0.4 [16].

In a previous paper we have investigated the codeposition of energetic deuterium with carbon, beryllium and tungsten at room temperature [17]. We observed codeposition both with carbon and BeO at a ratio of 0.41 D-atoms/C-atom and 0.38 D-atoms/Be-atoms, respectively. No codeposition of deuterium with tungsten was observed at room temperature. In this paper the measurements with Be are extended to higher temperatures up to about 800 K. The results obtained at elevated temperatures are compared with results from thermal desorption spectroscopy (TDS) of layers codeposited at room temperature.

2. Experimental

The experimental setup for the codeposition experiments was described earlier in full detail [17]. The experiments were performed at the high current ion source at the Max-Planck-Institut für Plasmaphysik, Garching [18]. A Be-target was bombarded with a mass analyzed beam of 4.5 keV D_3^+ (1.5 keV per D atom) at normal incidence. The target current was about 0.1 mA on a beam spot with 6 mm diameter. The amount of atoms removed by sputtering was determined by the weight loss of the target measured with a micro balance.

In order to measure the codeposition of deuterium with sputtered beryllium a silicon collector with dimension 10 × 10 mm^2 was placed at an angle of about 160° and a distance of about 15 mm from the target. Be-atoms sputtered from the target are deposited on the silicon collector.

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In addition, the silicon collector is bombarded with energetic deuterium atoms backscattered from the target. The maximum energy of D backscattered from Be at 160° is 621 eV and the mean energy is 286 eV. The particle reflection coefficient of D backscattered from Be is 0.03 while the sputtering yield is about 0.02–0.04, resulting in a simultaneous flux of D and Be at the collector with a flux ratio of about 1.

The collector could be heated to a maximum temperature of about 800 K. The temperature of the collector was measured with an infrared pyrometer. The error in the temperature measurement was about ±20 K.

TDS (thermal desorption spectroscopy) measurements after the codeposition were performed in the same target chamber with a linear heating ramp of 5 K/s. Mass 3 (HD) and mass 4 (D_2) were detected with a quadrupole mass spectrometer [18].

The vacuum in the chamber during the codeposition experiments was 1×10^{-7} mbar. The residual gas was mainly H_2O and CO.

The layers deposited on the Si-collectors were transported through air to the analysis chamber and were analyzed with MeV ion beam techniques. The analyzing beam was collimated to 1×0.5 mm^2. Several spots on each collector were measured. The amount of deposited deuterium was determined with the D(^{3}He, p)α nuclear reaction at 790 keV. The absolute error in the measurement of the amount of D is about 5%.

Deposited Be and O were detected with 1.65 MeV proton backscattering at a scattering angle of 165°. The cross-sections for backscattering of protons from Be and O are enhanced compared to the Rutherford cross-section. Deposited carbon was measured with proton backscattering at 1.74 MeV at a scattering angle of 165°. The resonant cross-section at 1.74 MeV was calibrated against the well-known cross-section at 1.5 MeV. The absolute error in the measurement of the amount of Be and O is about 12% and about 7% for C.

3. Results and discussion

Fig. 1 shows the total amount of retained deuterium on the collectors versus the total amount of beryllium for different irradiation fluences and temperatures of 293 K, 573 K and 773 K. The amount of D trapped with Be increases linearly with the amount of deposited Be. The slope of the linear increase gives the ratio of trapped D/Be. At room temperature (293 K) the ratio D/Be is about 0.36, which is in good agreement with our previous measurements [17], where a ratio of 0.38 was obtained.

At elevated temperatures the amount of trapped deuterium decreases. At 573 K the ratio D/Be is 0.17, which is about one half of what is measured at room temperature. At 773 K the ratio D/Be has decreased to 0.07.

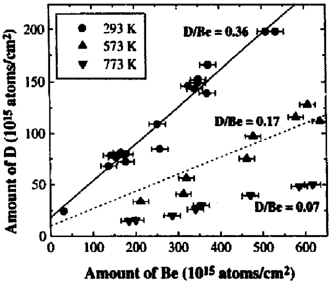

Fig. 1. Total amount of codeposited D versus the total amount of Be for different irradiation fluences and three different temperatures.

The ratio of 0.36 D/Be at room temperature is in good agreement with ion implantation experiments, where deuterium ions with keV energies are implanted into Be or BeO. Behrisch et al. [19] observed a saturation concentration of 0.34 D/Be when implanting 5 keV deuterium into BeO at 143 K, and 0.25 D/Be if the implantation was performed at room temperature.

The offset of the linear increase for the amount of Be close to zero is due to the implantation of deuterium into the silicon substrate at the beginning of the layer growth. This is clearly visible in the deuterium depth profiles [17]. The amount of D trapped in the Si substrate is in the range $1\text{–}2 \times 10^{16}$ cm^{-2}.

Due to the small growth rate of the codeposited layers (about 1 monolayer in 120 s) the deposited layers contain oxygen with a ratio O/Be ≈ 1. In the limits of the uncertainties of the measurement, the Be forms stoichiometric BeO. This is most likely due to reactions with water vapor and CO from the residual gas. It is well known that Be reacts rapidly with oxygen or oxygen bearing compounds such as H_2O and CO through the reactions $Be + H_2O = BeO + H_2$ and $Be + CO = BeO + C$. A saturated BeO layer has been observed to form on clean beryllium surfaces for oxygen exposures of only 10 Langmuirs [20]. Under particle bombardment with hydrogen or helium ions, the oxidation rate may even increase due to radiation-enhanced diffusion [21,22]. To prevent the codeposited layer from oxidation under the given growth conditions, a background pressure below 10^{-9} mbar would be necessary.

In addition also carbon is detected in the codeposited layer. The ratio C/Be was about 0.15. Also in fusion experiments such as JET, where Be evaporation is regularly applied for wall conditioning, the elements Be, C and O are always found in the surface layers of the vessel walls with varying compositions [23,24,4].

The amount of H in the codeposited layers was about $1.5\text{–}2 \times 10^{16}$ atoms/cm^2 in all analyses and did not increase with increasing layer thickness. The presence of H

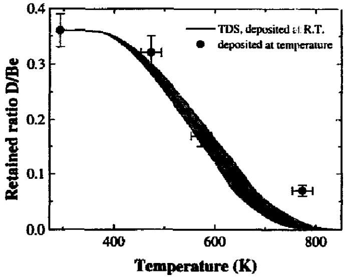

Fig. 2. Ratio of retained D/Be in the codeposited layers at different temperatures. Solid line: TDS, layer deposited at room temperature, heating ramp 5 K/s. The shaded area is the uncertainty range of the TDS measurement. Dots: Layer deposited at temperature.

is most likely due to adsorption of water from air during the transport to the analysis chamber.

A comparison of the retained amount of D in a TDS measurement with a linear heating ramp of 5 K/s of a layer codeposited at room temperature with the retained amount of D codeposited at the temperature is shown in Fig. 2. The shaded area is the uncertainty of the TDS measurement due to the uncertainty in the temperature calibration. At 573 K the ratio D/Be has decreased to 0.17, which is about half of the amount retained at room temperature. At 773 K nearly all deuterium is released from the sample codeposited at room temperature, but there is still deuterium trapped if the codeposition is performed at this temperature.

The thermal release of trapped deuterium from the layer codeposited at room temperature is in good agreement with TDS measurements of 1 keV deuterium ions implanted into BeO [25]. Deuterium implanted into clean Be with high fluences until saturation shows a different behavior: the deuterium is released in two stages at about 450 K and a broad stage around 700 K [26,25,27]. The low temperature stage at 450 K is not observed in ion implantation experiments with low fluences ($H/Be < 0.14$ [28]) and is associated with the formation of deuterium gas bubbles and blistering [26,25,27]. The high temperature stage is due to thermal detrapping of deuterium from damage sites. The activation energy for thermal detrapping was measured to be between 1.7 eV and 2.3 eV [26,29,28].

The formation of gas bubbles and blisters is not observed in ion implantation experiments of deuterium into BeO. Behrisch et al. [19] implanted 5 keV and 7 keV D into BeO until fluences of 3×10^{18} cm^{-2} and observed no blisters at −130°C and room temperature. The saturation concentration was 0.24–0.34 D/BeO. The TDS measurements of Möller et al. of 1 keV D implanted into BeO [25] and our TDS measurements of the thermal release of D from the codeposited layers (Fig. 2) do not show the low temperature stage. This implies that deuterium does not form gas bubbles in BeO but is bound at damage sites with a spectrum of different activation energies. Chemical binding, such as the formation of the hydroxide $Be(OD)_2$, seems unlikely because of the low amount of trapped deuterium and oxygen.

The results of the measurements are in agreement with the mechanism of coimplantation proposed in Ref. [17]. The growing layer is bombarded with reflected deuterium atoms with energies up to several hundred eV. Energetic deuterium atoms are implanted into the growing BeO layer until saturation. This model explains why the properties of the codeposited layer (ratio of D to BeO, thermal release) are similar to ion beam experiments where energetic deuterium ions are implanted into BeO.

4. Conclusions

The codeposition of D with BeO was investigated in the temperature range between room temperature and 773 K. At room temperature, D codeposits with BeO at a ratio of 0.36 D/BeO. Layers codeposited at elevated temperatures show a lower ratio of D/BeO. At 573 K a ratio D/BeO of 0.17 is measured, at 773 K the ratio is 0.07. Layers codeposited at room temperature can be outgassed by heating to 850 K. It is assumed that the deuterium is trapped by coimplantation at damage sites with a spectrum of different activation energies.

From the viewpoint of tritium inventory trapped in codeposited layers in next step fusion experiments such as ITER beryllium has only minor advantages compared to carbon. If oxygen is present then BeO will be formed. The D/C-ratio in codeposited deuterium/carbon layers is about 0.4–0.42 at room temperature. The desorption of deuterium from codeposited carbon layers starts around 700 K. Temperatures around 1200 K are necessary to remove hydrogen from carbon [30]. The amount of tritium trapped in codeposited BeO layers at room temperature is only slightly lower than in codeposited carbon layers. At elevated temperatures the amount of hydrogen trapped in codeposited BeO layers is lower than in carbon layers; however it is still high. Temperatures above 800 K are necessary to release the codeposited D from the BeO layer. These temperatures are too high to be applicable in ITER.

Trapping of large amounts of tritium by codeposition/coimplantation at deposition dominated areas of the vessel walls of ITER may be unavoidable if carbon based materials or beryllium are used as plasma facing materials. The amount of tritium trapped by codeposition may become critical [12]. Therefore applicable (in respect to maximum temperature and time) and reliable methods for tritium removal from beryllium and carbon containing codeposited layers have to be developed.

Acknowledgements

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