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Insight into the co-deposition of deuterium with beryllium: Influence of the deposition conditions on the deuterium retention and release

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ABSTRACT

A systematic study of the influence of the deposition conditions on the deuterium retention by and release from co-deposited beryllium layers has been carried out in PISCES-B. Experimental parameters such as the beryllium deposition rate, the incident particle energy and the substrate temperature are shown to affect the level of hydrogen isotope retention in the layers. In addition, the pressure during deposition and the presence of Ar in the plasma are monitored since they influence the layer morphology and the ease of fuel removal by thermal desorption. Consequences for both the tritium inventory and effectiveness of tritium removal during bake-out procedures in ITER will be discussed.

1. Introduction

Fuel retention will be one of the more challenging problems to be solved for the success of ITER, where the in-vessel retention is to be limited to 1 kg of mobilisable tritium [1]. Different mechanisms will contribute to the overall fuel retention [2]. For the foreseen long pulse operations of ITER, co-deposition of tritium with material eroded from the plasma-facing components (PFC) is expected to be the main source of tritium accumulation in the vessel. Since the ITER scrape-off layer plasma will contain a significant amount of Be eroded from the first wall, co-deposition of beryllium with isotopic hydrogen may dominate the global co-deposition process. A clear understanding of the retention properties of co-deposited layers is needed for prediction of the in-vessel retention and for the development of tritium removal techniques [3].

Co-deposition of hydrogen with carbon has been extensively studied, and the retention in co-deposited carbon layers depends on the energy of the incident particles and on the substrate temperature during deposition [4]. Until recently, the retention in co-deposited beryllium layers was poorly understood and large discrepancies were found between the D/Be ratios reported from different experiments. An empirical scaling equation has been proposed to describe the retention in Be co-deposits formed in PISCES-B [5]. It was found that the D/Be ratio is a function of the Be deposition rate, the energy of the incident D particles and the substrate temperature during deposition.

This paper describes the evolution of the layer morphology as a function of the pressure and temperature during deposition, which is discussed in terms of a structure zone diagram. It is observed that the desorption kinetics of deuterium is strongly affected by the same parameters. In addition it is shown that the influence of the deposition conditions is relatively well described by the model presented in [5].

2. Experimental

Beryllium co-deposits were produced in PISCES-B [6], the experimental setup is described in [7]. A beryllium target is exposed to a high-flux deuterium plasma (1–3 $\times$ 10^{22} ions m^{-2} s^{-1}). During plasma exposure, the target temperature is fixed by the plasma parameters and is in the range 400–800 K, but is not found to influence the retention properties of co-deposits as they are formed far from the target. A negative bias (-50 V), applied to the target, controls the energy of the impinging ions. A beryllium impurity fraction in the plasma is generated by an evaporative atomic beam source (Veeco/Applied EPI). The beryllium fraction in the plasma is controlled by adjusting the effusion cell temperature and is determined spectroscopically. The plasma parameters ($n_e \sim 0.5$–3×10^{18} m^{-3}, $T_e \sim 6$–10 eV) are measured by a reciprocating double Langmuir probe.

Co-deposits are collected on polished tungsten substrates installed with a line of sight to the target. Substrates are installed on a movable deposition probe assembly and are shielded from cross-field plasma transport. The substrate can be independently heated to about 600 K, the temperature being monitored by a thermocouple installed at the back of the sample.

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The total D and Be amounts in the layer were determined through nuclear reaction analysis (NRA) utilizing the $D(^3He,p)^4He$ and the $Be(^3He,p)^{11}B$ reactions. Samples were probed at two primary energies, 0.8 and 1.6 MeV.

The thermal desorption of deuterium was studied using thermal desorption mass spectrometry (TDS). Each of the samples was separately desorbed in a TDS chamber of typical pressure better than 10^{-6} Pa. An infrared heat source is programmed to linearly increase the sample temperature at a rate of 18 K/min up to 1100 K and then held at that temperature for a further period of 10 min before cooling, also at 18 K/min. Prior to deposition, the samples are ultrasonically cleaned using acetone then alcohol, and then outgassed using the same procedure used for TDS measurements. This ensures there is no naturally occurring residual mass 4 inventory in samples prior to forming the co-deposits.

Surface morphology of the layers was studied by means of a JEOL-JSM 6830 scanning electron microscope (SEM).

3. Results

3.1. Morphology of the layers

To investigate the influence of the deposition conditions on the layer morphology, experiments were made with two different neutral pressures of deuterium in the vacuum chamber (0.2 and 0.8 Pa), two substrate temperatures (373 and 573 K) and with different fractions of argon in the gas mixture. The argon fraction (c_{Ar}) was determined by measuring the relative partial pressures of D_2 and Ar. The choice of Ar was driven by its potential use in ITER to radiate power and reduce the divertor heat loads [8]. Fig. 1 shows the layer morphology obtained under different deposition conditions. For a neutral pressure of 0.8 Pa, at low temperatures, the layer exhibits a high level of porosity (Fig. 1(a)) which makes it appear dark despite a low roughness and impurity content, as was described in [9]. When using a mixture of D_2 and Ar (Fig. 1(c)), the layer appears much denser and no specific surface features can be observed. The layer appears grey and shiny, similar to bulk beryllium. It should be noted that an argon fraction of $c_{Ar} = 9\%$ is already sufficient to notably change the layer morphology. A similar effect is observed when reducing the neutral pressure in the case of a pure deuterium plasma (Fig. 1(e)). On the contrary, the morphology of the layers formed at high temperature appears to be independent of the neutral pressure and gas composition, at least in the range of values investigated here. The morphology of those layers is relatively similar to that described in [10] for magnetron sputtered Be layers. The evolution of the morphology with the deposition conditions is consistent with the structure zone model [11] describing qualitatively the effects of pressure and temperature on the structure of a vacuum deposited film. The formation of porous layers is favoured at high working gas pressure because of the reduction in surface mobility (due to a higher density of working gas atoms on the surface) of the deposited atoms during the film growth, hence the layer in Fig. 1(a) is more porous than the layer in Fig. 1(e). It has also been reported in [12] that the bombardment of the growing layer by energetic heavy particles tends to densify the resulting layer, mainly by sputter-induced redistribution of the deposited material. This explains the production of denser layers when Ar is present in the plasma.

3.2. Influence of the deposition conditions on the thermal desorption of deuterium

Not only do the deposition conditions affect the co-deposits morphology but also the thermal desorption of deuterium. Fig. 2(a) shows the TDS spectra of Be layers prepared with different fractions of Ar, at 373 K. For $c_{Ar} = 0\%$ (curve i), the TDS spectrum is characterized by a sharp peak with a maximum at about 450 K. For the highest argon fraction (curve iii), the peak appears broader and the maximum of desorption is shifted towards higher temperatures, at 550 K. For $c_{Ar} = 9\%$ (curve ii), the desorption curve is evidently a mix between these two cases, with both a sharp release peak at low temperature and a broader peak at high temperature. As indicated in the figure, the amount of retained deuterium, as determined by NRA, tends to decrease when the Ar fraction increases.

As seen in Fig.1(c) and (e), the morphology of the layer deposited with $c_{Ar} = 15\%$ and that of the layer deposited at low pressure, are similar. The desorption curves for these layers are shown in Fig 2(b). The deposit formed at lower pressure (curve iv) shows the high temperature D release peak whereas the deposit made at higher pressure (curve i) does not. In addition, the layer formed at lower pressure and the layer formed with $c_{Ar} = 15\%$ have similar morphology (Fig. 1(c) and (e)) and both exhibit the high temperature D release peak. All the deposits formed at high temperature show only the high temperature release peak, an example of $c_{Ar} = 0\%$ at 573 K is shown (curve v, Fig. 2(b)).

The shape of the low temperature peak is similar to that reported in [13] for implanted beryllium. This peak is attributed to a binding energy of a Be–D bond corresponding to adsorption of D on Be. The maximum of the second peak (550 K) coincides with the decomposition temperature of BeD_2, evidence for which comes from XPS analyses and comparison of the present TDS traces with that from a reference BeD_2 powder [14].

3.3. Influence of the deposition conditions on the retention

The co-deposited layer formation conditions have been found to affect the thermal desorption of deuterium. In [5], a scaling equation has been proposed to account for the influence of the Be deposition rate, r_d, (in 10^{15} at.% $cm^{-2}\,s^{-1}$), the average energy (E_n in eV) of the deuterium particles bombarding the growing layer, and the substrate temperature (T in K), on the deuterium content (referred to as D/Be ratio) in the co-deposited layer:

$$\frac{D}{Be} = 2.94 \times 10^{-5} \times r_d^{-0.59} \times E_n^{1.34} \times \exp\left(\frac{1306}{T}\right). \tag{1}$$

In [5] however, the layers were formed in a pure deuterium plasma with a typical neutral pressure of 0.6–0.8 Pa. To confirm the validity of this approach, Eq. (1) is used to determine the predicted D/Be ratios of the layers investigated here. Fig. 3 shows a comparison of the values predicted by the scaling equation with the experimentally measured values. Data obtained in [5] are indicated for reference. The good agreement between the experimental and predicted values indicates that although the presence of Ar, and the neutral pressure, have an effect on the Be deposition rate, Eq. (1) is still able to reproduce the experimental observations. The deposition conditions are thus determining the level of retention observed in the layers.

4. Implications

The results described here have two important implications. First, co-deposition conditions (pressure, temperature, presence of Ar) will affect the deuterium release behaviour which in turn has consequences on the amount of tritium which can be recovered during a bake-out of the ITER vessel. The deuterium release behaviour of two beryllium co-deposits formed at the same surface temperature, but exhibiting different levels of porosity (due to changes in the background neutral density) are compared in Fig. 4. In this case argon has been added to the deuterium plasma

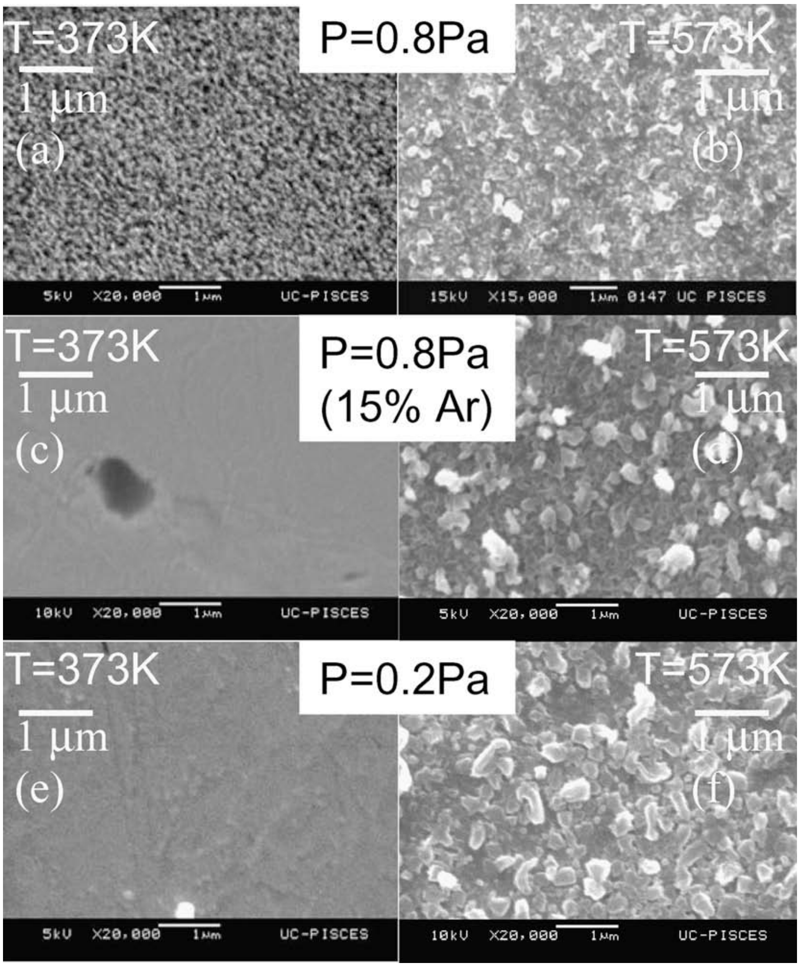

Fig. 1. Surface morphology of co-deposited beryllium layers formed with different neutral pressures, gas composition and substrate temperatures.

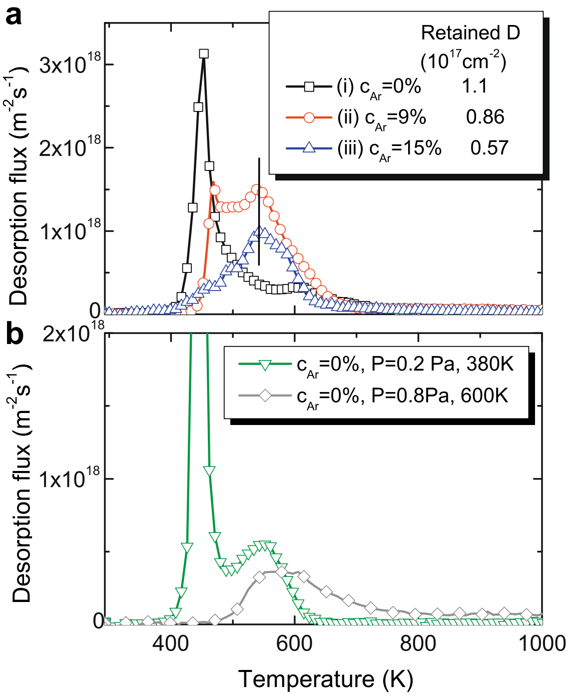

Fig. 2. Thermal desorption behaviour of deuterium from (a) co-deposited beryllium layers formed at 373 K with three different fraction of Ar in the plasma, and (b) with a low neutral pressure (0.2 Pa) at 373 and 573 K in pure deuterium.

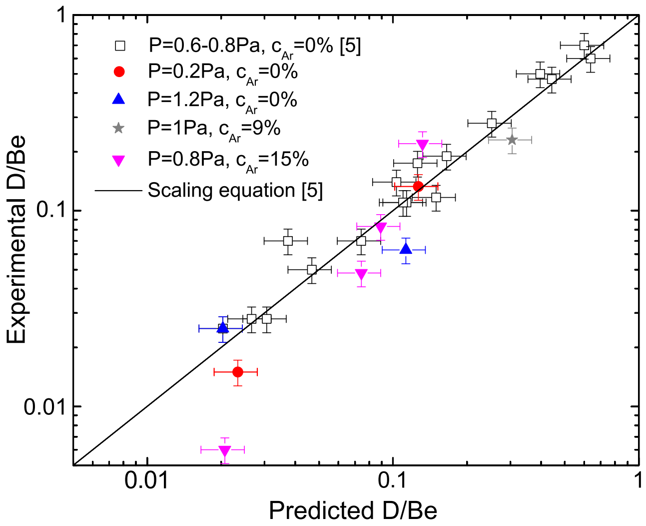

Fig. 3. Comparison between experimentally measured D/Be ratios of co-deposited beryllium layers produced under different conditions and values predicted by Eq. (1) using the investigated deposition conditions.

in PISCES-B, resulting in the formation of a dense layer. Also indicated in Fig. 4 is the presently envisioned bake-out temperature for all areas of the ITER vessel [15]. In case of a dense layer, only about 30% of the initial deuterium is removed at 510 K whereas

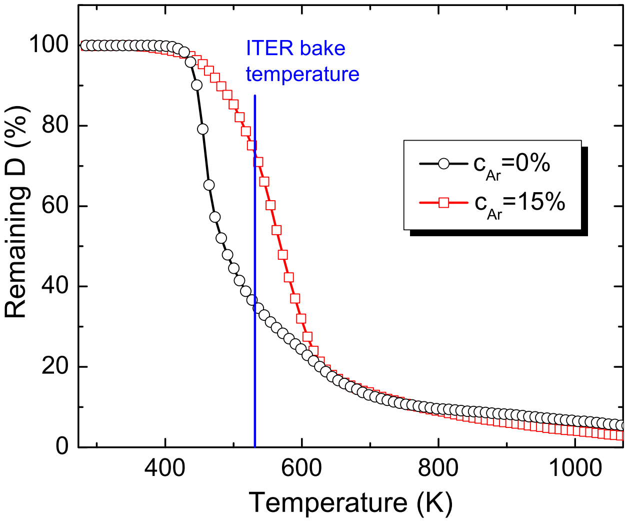

Fig. 4. Deuterium release characteristics of two beryllium co-deposits created at 373 K with two different levels of porosity. Also indicated is the present ITER bake temperature.

for a porous layer up to 70% of the deuterium inventory can be recovered without any additional treatment (like oxygen baking, surface flash heating, etc.). Those results suggest that in ITER, dense co-deposits may form in the divertor if Ar is to be used, but also in the deposition-dominated areas of the main chamber where the neutral pressure will be much lower than in the divertor.

Finally, the retention level in Be co-deposits will strongly depend on the deposition conditions such as deposition rate, and temperature, as demonstrated here and also energy of D particles as shown in [5]. The retention level will therefore depend on the location where such layers form. The traditional approach of using a fixed T/Be ratio for predictions of the retention in ITER [16] may therefore be refined by using the proposed scaling equation.

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