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Journal of Nuclear Materials 254 (1998) 84–86

journal of nuclear materials

Letter to the Editors

Codeposition of deuterium with beryllium

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Received 1 August 1997; accepted 27 October 1997

Abstract

We have measured the codeposition and/or coimplantation of deuterium with beryllium. Beryllium, sputtered from a target disk, was collected on a heated silicon catcher plate where it was simultaneously bombarded by reflected deuterons. Oxygen to beryllium ratios in the layer varied between 0.03 and 0.13. Deuterium to beryllium ratios dropped from 0.15 at 373 K to 0.02 at 573 K. © 1998 Elsevier Science B.V.

1. Introduction

The codeposition of energetic hydrogen isotope ions with beryllium has received much attention lately [1–3]. The first wall of the International Thermonuclear Experimental Reactor (ITER) will be lined with beryllium. Beryllium eroded from the first wall will be redeposited in different regions of the tokamak including cooler areas of the diverter. During the deposition process, the redeposited beryllium is continuously bombarded with energetic hydrogen isotope ions and neutrals. As operation of the tokamak continues, the initial layers of the redeposited material will become buried. Depending on the quantity of beryllium eroded from the first wall and the amount of tritium retained in the redeposited material, this process could significantly increase the tritium inventory in ITER.

The term codeposition has been used to describe the process in a tokamak fusion reactor where carbon is sputtered from one surface onto another along with the tritium and deuterium escaping from the plasma [4]. When carbon deposits in relatively cool areas of the vessel, it is able to trap substantial amounts of the hydrogen isotopes in the layer. For temperatures below about 700 K, hydrogen to carbon ratios of 0.4 are typical. Codeposition is known to be the principal source of hydrogen isotope buildup in most carbon containing tokamaks, and is anticipated to be the primary source of tritium retention in ITER [5–8]. Because ITER will contain both beryllium and carbon, the question arises whether beryllium will also codeposit with tritium, adding to the tritium inventory.

While several studies have been performed on the codeposition of beryllium with hydrogen isotopes [1–3], experimental conditions in those studies resulted in the experiments actually determining the codeposition of hydrogen isotopes with beryllium oxide. It was the purpose of the experiments reported here to examine the amount of deuterium retained in redeposited beryllium films where the oxide content is very small. The Tritium Plasma Experiment (TPE) was used to deposit sputtered beryllium onto polished silicon substrates heated to temperatures between 373 and 573 K. The oxygen to beryllium ratios of the films varied between 0.03 and 0.13. It was the high sputter flux in TPE that resulted in the achievement of the relatively oxygen free films. A high sputtering rate reduces the length of time required to produce the films at the same time that the sputtered beryllium provides oxygen gettering in the rest of the system.

2. Experimental

The experiments were performed in the Tritium Plasma Experiment (TPE), presently located in the Tritium System Test Assembly at Los Alamos National Laboratory. This device has been described earlier [9]. Beryllium was sputtered from a 5 cm diameter S-65 beryllium target disk by

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PII S0022-3115(97)00344-9

100 eV deuterons. The sputtered beryllium was collected on a heated 0.5 cm diameter polished silicon catcher plate located 5 cm in front of the beryllium disk and 5 cm from the centerline of the disk. For experiments performed with the silicon catcher plates at 373, 473, and 573 K, the 100 eV deuteron flux was maintained at 3.3×10^{21} D/m^2 s uniformly over the 5 cm diameter target disk. For the additional experiment performed with the catcher plate at 423 K, the particle flux on the target disk was tripled to 9.9×10^{21} D/m^2 s. According to the TRIM computer code [10], approximately 17% of the deuterons were reflected from the target disk with an average energy of about 30 eV. All of the experiments were run for 1 h. The target disk was actively cooled, but still reached a temperature of approximately 473 K. During the plasma exposure of the sputter disk, the deuterium pressure in the system was 3.9×10^{-2} Pa. The background pressure in the system was less than 1×10^{-5} Pa and consisted primarily of water vapor.

After the sputter deposition was completed, the samples were removed from the vacuum vessel and transferred in air to the Ion Beam Materials Research Laboratory (IBMRL) at Sandia National Laboratories in Albuquerque. Here quantitative profiles of beryllium, deuterium, oxygen, and carbon in the redeposited layer on the silicon disks were measured. The samples were also examined for other impurities, but none were found.

The samples were placed in the standard scattering analysis chamber of the EN Series Tandem Van de Graaff accelerator facility in the IBMRL. The working pressure of the analysis chamber is less than 1×10^{-5} Pa. The samples were analyzed at room temperature ($\sim$300 K). Rutherford backscattering analysis (RBS) [11] with a 2.2 MeV incident He^+ ion energy was performed first to profile oxygen in the samples. The RBS spectra were also used to check for other possible impurities. The RBS was performed using a 45° sample tilt and a 164° scattering angle.

Elastic recoil detection (ERD) [12] was then used to depth profile the amounts of beryllium, deuterium, hydrogen, and carbon on the samples. The incident ion used was Si^{5+} at two energies: 24 MeV to measure the hydrogen, and 28 MeV to measure the other elements. The standard ERD analysis geometry of 75° sample tilt, a 30° recoil angle, and a 12 μm Mylar range foil, was used. All three sets of analyses were performed consecutively in the same chamber under the same vacuum conditions.

3. Results and discussion

The experimental results for the beryllium, deuterium, and oxygen content of the redeposited films are shown in Table 1. The carbon concentration in the different films averaged approximately 1%, but never exceeded 2%. It can be seen in the table that the oxygen to beryllium ratio in the redeposited films varied from 0.03 up to 0.125. Whereas the experiment at 423 K represented an attempt to reduce the amount of oxygen in the film by increasing the beryllium sputtering (which also reduces the amount of oxygen in the system by active gettering on the chamber walls), the oxygen concentrations in the other three films likely reflect the order in which the experiments were performed. Both the anode and cathode of the plasma source were cleaned immediately prior to these experiments. Past experience has shown the vacuum conditions in TPE to improve during the first few experiments after cleaning. This improvement is then reversed as the conditions deteriorate while the anode and cathode again become contaminated.

Table 1
Sample thickness and oxygen and deuterium content

Sample temp.	Thickness	O/Be	D/Be
373 K	120 nm	0.125	0.15
473 K	120 nm	0.125	0.07
573 K	150 nm	0.06	0.02
423 K	320 nm	0.03	0.10

The deuterium concentration in the films as a function of temperature are plotted in Fig. 1 where they are compared to the earlier TPE results [1] using tritium and those produced by Mayer [2]. The hydrogen isotope concentrations of the most recent data are significantly lower than those in the films produced earlier (the protium content was below 0.4% in all cases). It is believed the primary reason for the difference between the older results and the newer ones is the amount of oxygen present in the different films. In the report by Mayer [2], it was stated that accelerator analysis showed his films to consist primarily of beryllium oxide instead of beryllium. In the earlier TPE

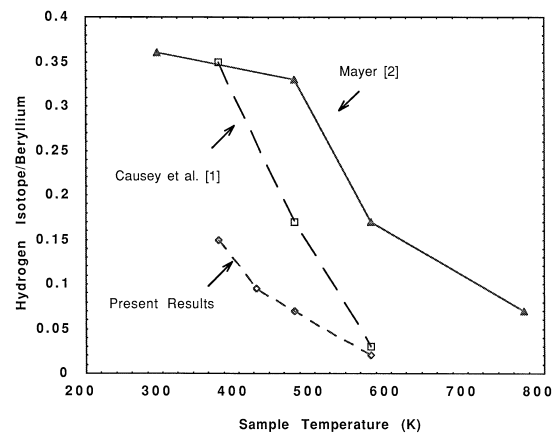

Fig. 1. Comparison of hydrogen isotope content of different redeposited beryllium films. The films produced by Mayer [2] consisted primarily of beryllium oxide.

experiments [1], the oxygen content of the redeposited films was not measured, but was believed to be higher than that in the present films due to the different vacuum conditions that existed for the two experiments. In the earlier experiments, the partial pressure of water in the vacuum vessel was several times higher than that of the recent experiments. While it is not known definitively how beryllium oxide retains hydrogen, it can be suggested that the mechanisms might include binding at defects in the beryllium oxide and/or the formation of chemical bonds between the beryllium oxide and hydrogen.

While the amount of deuterium in the redeposited beryllium layers of the present experiments is lower than that of the previous experiments, it is still substantial (see Table 1). The DIFFUSE computer code [13] was used to model the deuterium retention in the redeposited layer for the lower flux samples. Based on the thickness of the redeposited layer, the arrival rate of sputtered beryllium on the catcher plate was approximately 3×10^{18} Be/m^2 s. The arrival rate of reflected deuterium ions onto the redeposited layer for these lower flux experiments was calculated by the TRIM [10] code to be approximately 1.3×10^{19} D/m^2 s. Using trapping energies of 1.0 eV and 1.8 eV and trap densities of 0.31/Be and 0.05/Be as recommended by Wampler [14], it was not possible to predict inventories as high as the measured values using reasonable values for the recombination rate coefficient at the surface. The agreement between experiment and model became better when it was assumed that the traps were not saturable. The density of traps were assumed to remain unchanged, but the traps were assumed to be able to hold more than one deuterium atom. This improved agreement could suggest that atoms were coming out of solid solution, forming bubbles. Scanning electron microscopy showed this to be the case. For the thinner films, the 373 K sample showed a very high density of bubbles and blisters with a diameter of approximately 1 μm. At 473 K, the density of these bubbles and blisters was much lower, and none were apparent for the 573 K sample. The thicker sample (320 nm) was different in that the blisters were much larger (10 to 30 μm in diameter). The evolution of bubbles and blisters in beryllium and beryllium oxide exposed to hydrogen isotope implantation has been reported by several other authors [15–17].

The implications of these results for ITER depend upon the conditions that will exist during operation. Comparison of the present results with those measured earlier [1,2] shows the codeposition (or coimplantation) of tritium along with sputtered beryllium to strongly depend upon the amount of oxygen in the plasma. The cleaner the plasma, the lower the tritium retention. This lower retention may be partially offset by the sputtering rate being higher for pure beryllium than for beryllium oxide. With or without oxygen present, the retention of tritium in the redeposited beryllium layer will be less than that of redeposited carbon. This was clearly shown in the experiments performed by Hsu [18]. In his experiments, Hsu initiated a Penning plasma discharge in a closed vessel and monitored deuterium pressure changes. When graphite electrodes were used, the system deuterium pressure steadily decreased with time due to the codeposition process. The beryllium electrodes produced no sustained deuterium removal.

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