Tritium Retention in the Codeposited Beryllium–Tritium Layer

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The Tritium Plasma Experiment (TPE) has been used to measure the amount of tritium codeposited with beryllium in a film produced by the energetic sputtering of beryllium by a tritium plasma. In these experiments, a plasma consisting of 90% deuterium and 10% tritium was directed onto a biased beryllium disk. A catcher plate located approximately 7 cm away from the beryllium disk was used to collect the sputtered beryllium along with the deposited hydrogen isotopes. A typical experiment used an impinging flux of 3.3×10^{17} (D+T)/cm^2-s and lasted for 1 hour. After the collection of the layer, the small catcher plate was removed and outgassed in a special system designed for thermal spectroscopy of tritium laden samples. Data was collected for the catcher plate heated to 373, 473, and 573 K. At 373 K, the ratio of hydrogen isotopes to beryllium was approximately 0.35. At 573 K, this ratio had dropped to about 0.03. Implications for tritium inventory in the ITER reactor are discussed.

1. Introduction

Beryllium is a candidate material for plasma facing applications in tokamak fusion reactors. In such an application, the beryllium will be sputtered by energetic ions and neutrals escaping from the plasma. Many of these sputtered beryllium atoms will land in cooler areas of the tokamak where they will be protected from further sputtering. If these atoms codeposit with tritium from the plasma, the tritium inventory generated by this process could have an impact on the overall tritium inventory and serve as a safety problem for operation. Even if the tokamak using the beryllium does not use tritium, this codeposition process could still be important due to effects on overall plasma recycling and density control.

The codeposition of carbon with hydrogen isotopes is a known process [1-3]. It is believed to be the primary source of tritium inventory in the presently operating TFTR tokamak [4]. The limited use of beryllium in tokamaks and the problems associated with performing measurements with the toxic material have reduced the number of measurements on this codeposition process. The first attempt at measuring the codeposition of beryllium and hydrogen isotopes was performed by W. Hsu in 1990 [5]. Hsu used a Penning discharge to compare the pumping effect of sputtered carbon to that of beryllium. The sputtered carbon was seen to strongly pump hydrogen from the closed vessel in which the discharge was operated. No pumping was seen when the carbon electrodes were replaced by beryllium electrodes. More

recently, Mayer et al. [6] used accelerator analysis of films produced by sputtered beryllium in a deuterium environment to show that the two elements do codeposit. The experiments presented in this paper were performed to independently determine whether the codeposition process exists for beryllium and hydrogen.

2. Experimental Procedures

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 [7]. Beryllium was sputtered from a 5 cm diameter beryllium disk by 100 eV tritons and deuterons. The sputtered beryllium was collected on a 0.5 cm diameter heated aluminum catcher plate located 5 cm in front of the sputter disk and 5 cm from the centerline of the disk. Experiments were performed with the catcher plate held at 373, 473, and 573 K. During the beryllium sputtering process, the 100 eV hydrogen isotope ion flux was maintained at 3.3×10^{17} ions/cm^2-s uniformly over the entire beryllium disk. In each of the experiments, this flux was held steady for 1 hour. The ratio of deuterium to tritium in the plasma was 10 to 1. The combined deuterium and tritium partial pressure was 3.9×10^{-2} Pa. The base pressure in the system was approximately 1×10^{-5} Pa and consisted primarily of water vapor.

After the sputtering process was completed, the catcher plate was removed from TPE and transferred to an outgassing system. This system consisted of a furnace, ionization chamber, copper oxide catalysis bed, and glycol bubblers. A combination of 99% He and 1% hydrogen was swept through the furnace at 100 cc(stp)/min as the furnace was ramped up to 1073 K. After passing through the furnace and collecting the released deuterium and tritium, the sweep gas passed through an ionization chamber where the tritium was detected. After the ionization chamber, oxygen was added to the gas and sent through a copper oxide bed. The oxygen was added to replenish the oxygen depleted from the copper oxide bed as it converted the hydrogen into water. The water from the copper oxide bed was then collected in the series of glycol bubblers. Liquid scintillation counting of the glycol was used as a backup to the ionization chamber results. The total hydrogen isotope retention in the codeposited layer on the collector plate was calculated by correcting the tritium results for the deuterium to tritium ratio.

To be able to calculate the hydrogen to beryllium ratio in the codeposited layer, it was necessary to know the amount of beryllium actually sputtered and collected on the catcher plate. Because of the errors associated with calculating this amount of beryllium, it was measured experimentally. One catcher plate was used in an experiment identical to that described above except for the use of pure deuterium instead of the deuterium-tritium mixture. Copper was used for the catcher plate to increase the ability of the detection system to differentiate between the substrate and the deposited materials.

3. Results and Discussion

In the discussion below, the term "hydrogen" will often be used to describe deuterium as well as tritium. The terms "deuterium" and "tritium" will typically be used only where it is necessary to describe a process specific to those isotopes. Results of the accelerator analysis of the sample produced using pure deuterium showed the amount of beryllium on the catcher plate after the one hour exposure to be 6.1×10^{16} Be/cm^2. Because the samples produced for the tritium analysis were produced under very similar conditions (the ratio of

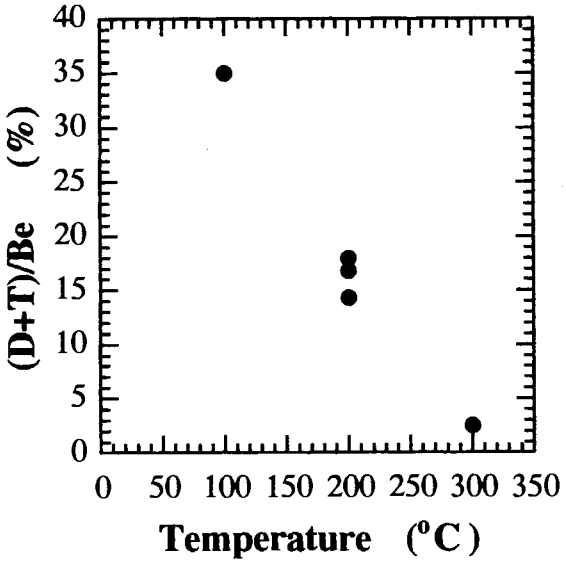

Figure 1. Hydrogen to beryllium ratio measured for the deposited film.

deuterium to tritium was 10 to 1), this amount of beryllium was assumed to exist for all of the samples.

The experimental results for the ratio of deuterium plus tritium to beryllium on the catcher plates are shown as Figure 1. The ratio drops from 0.35 at 373 K down to about 0.03 at 573 K. From this data it must be concluded that codeposition (or coimplantation) of hydrogen with beryllium (or beryllium oxide) does occur.

For discussion, the data above is repeated in Figure 2 along with the results from Mayer et al. [6] for codeposition and results from Wampler [8] for saturation of beryllium with implanted deuterium. It can be seen that there is fairly good agreement between all three sets of data. The agreement with the results of Mayer et al. [6] is not unexpected due to the similarities of the two different studies. Both sets of experiment used the sputtering of beryllium to generate the beryllium layer and both experiments had reflected hydrogen isotope ions that may have played a role in the final results. Mayer et al. used 0.1 mA of 4.5

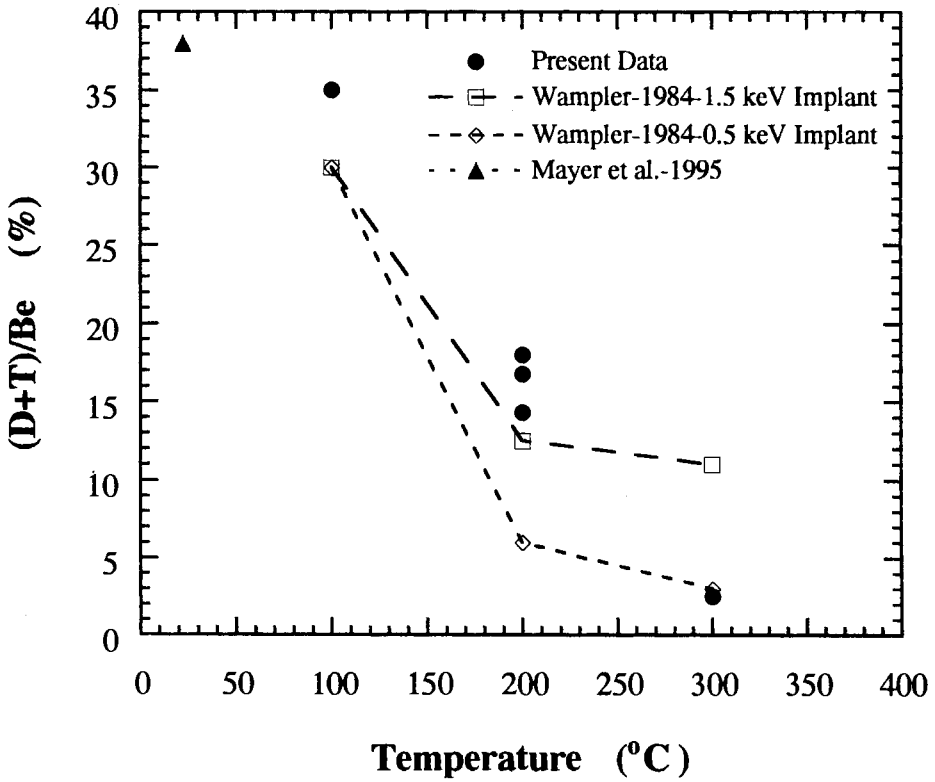

Figure 2. Comparison of the hydrogen retention in codeposited films with that for saturated films.

keV D_3^+ at normal incidence to produce their film. The number of reflected deuterons was approximately equal to the number of sputtered particles. The average energy of the reflected deuteron was listed as 286 eV. For the experiments reported in this report, the TRIM computer code [9] was used to calculate the fraction of the 100 eV hydrogen ions striking the beryllium sputter plate that were reflected and the energy of those reflected particles. Of the particles striking the beryllium target, 14% were reflected with an average energy of approximately 30 eV. The number of reflected particles was significantly higher than the number of sputtered particles. For both sets of experiments, there is the possibility that the hydrogen retention was driven by the coimplantation of beryllium and hydrogen as apposed to the codeposition. In a fusion reactor, the question of whether the layer is codeposited or coimplanted is only of academic interest. There are sufficient quantities of energetic hydrogen ions and neutrals in almost all locations of an operating tokamak to produce the layer regardless of the process. In reality, the codeposition of hydrogen with carbon which has been well studied [1-3] could also be caused by coimplantation.

The role of oxygen on the codeposition process is unknown, but thought to be very important. Mayer et al. [6] determined by accelerator analysis that the beryllium deposited on their catcher plate was completely oxidized. While not measured in the present study, the arrival rate of water molecules due to the approximate 1×10^{-5} Pa background pressure was more than sufficient to also oxidize the thin beryllium film produced in the TPE device. Due to the difficulty producing beryllium hydride [10], it is likely the retention of hydrogen in the film is due to some sort of bonding of hydrogen to beryllium oxide.

The agreement of the present results for hydrogen retention in a sputter deposited film with that of Wampler [8] where the layer was produced by the saturation of the beryllium surface with energetic particles is very interesting. A similar agreement between hydrogen retention in the saturated and codeposited films was reported by Causey et al. [11] for carbon. It was stated in that report that both types of carbon films have very low densities with many of the carbon atoms connected through porosity to the surface. The hydrogen retention was thought to be controlled by the bonding of the hydrogen to the carbon atoms. It is probable that a similar effect is controlling the retention of hydrogen in beryllium. Recent experiments by Chernikov et al. [12] have shown deuterium bombardment of beryllium to cause open porosity in the implant zone. In this zone or layer, a large fraction of the beryllium atoms (or beryllium oxide molecules) are connected to the external surface. There may be direct bonding of the hydrogen to these atoms (or molecules). The fact that the fractional retention of hydrogen in the two types of films drops rapidly as the temperature is increased above 373 K shows this bonding to be fairly weak (less than 2 eV). This can be compared to the carbon/hydrogen layer where the layer does not release the hydrogen until the temperature is elevated above about 750 K.

Above it was stated that Hsu et al. [5] compared the hydrogen pumping of sputtered beryllium with that of sputtered carbon to conclude that codeposition of beryllium and hydrogen does not occur. In the experimental study presented here, it was determined in the accelerator analysis of the deposited layer that the amount of beryllium found on the catcher plate was approximately a factor of ten less than expected. Whether this reduced apparent sputtering rate was due to protection of the beryllium surface by oxygen or other unknown effects, a similar reduced sputtering rate in the experiments by Hsu would have resulted in a pumping rate too small for detection during the time he monitored the rate.

The implications of the results presented here to the tritium inventory in tokamak fusion reactors are variable depending on the design of the reactor. For large tritium inventories to occur due to the beryllium codeposition process, it is necessary that beryllium be used in areas of relatively intense particle fluxes and for there to be relatively cool areas nearby for the layer deposition. An example would be the use of beryllium as a divertor material with surrounding areas kept at temperatures below 300 K. The use of beryllium as a first wall material would not be expected to result in substantial tritium inventories unless the particle fluxes were sufficiently non-uniform to generate areas of net erosion and net deposition.

4. Conclusions

When beryllium is sputtered onto surrounding surfaces, it codeposits with hydrogen if there is energetic hydrogen also striking the surface. It cannot be determined from the experiments presented here whether molecular hydrogen also codeposits with sputtered beryllium. At 373 K, the H/Be ratio in the codeposited/coimplanted layer is 0.35. It drops to 0.03 at 573 K. Oxygen was also present in the films produced in this experimental study, and may be necessary for the codeposition process to occur. For the codeposition of beryllium and tritium to be a problem for tokamak reactors, it is necessary that areas of relatively intense flux to be surrounded by cool areas onto which the deposition can occur.

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