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Tritium retention in S-65 beryllium after 100 eV plasma exposure

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Abstract

The tritium plasma experiment (TPE) has been used to measure the retention of tritium in S-65 beryllium under conditions similar to that expected for the international thermonuclear experimental reactor (ITER). Beryllium samples 2 mm thick and 50 mm in diameter were exposed to a plasma of tritium and deuterium. The particle flux striking the samples was varied from approximately 1×10^{17} (D + T)/cm^2 s up to about 3×10^{18} (D + T)/cm^2 s. The beryllium samples were negatively biased to elevate the energy of the impinging ions to 100 eV. The temperature of the samples was varied from 373 K to 973 K. Exposure times of 1 h were used. Subsequent to the plasma exposure, the samples were outgassed in a separate system where 99% He and 1% H_2 gas was swept over the samples during heating. The sweep gas along with the released tritium was sent through an ionization chamber, through a copper oxide catalyst bed, and into a series of glycol bubblers. The amount of released tritium was determined both by the ionization chamber and by liquid scintillation counting of the glycol. Tritium retention in the beryllium disks varied from a high of 2.4×10^{17} (D + T)/cm^2 at 373 K to a low of 1×10^{16} (D + T)/cm^2 at 573 K. For almost every case, the tritium retention in the beryllium was less than that calculated using the $C = 0$ boundary condition at the plasma facing surface. It is believed that this lower than expected retention is due to rapid release of tritium from the large specific surface area created in the implant zone due to the production of voids, bubbles, and blisters.

Keywords: Plasma–wall interaction simulator; Low Z material; Wall particle retention; Tritium inventory and economy

1. Introduction

It is presently anticipated that beryllium will be used as a plasma facing material in the ITER fusion reactor. As such, it will be exposed to varying amounts of energetic tritium and deuterium. As a first wall material, the D + T particle flux will be in the range of 10^{16} to 10^{17} (D + T)/cm^2 s. As a divertor material, the particle flux will increase to 10^{18} to 10^{19} (D + T)/cm^2 s. For both applications, the tritium permeation through the beryllium into the water coolant as well as the tritium inventory are of importance for the safety of the device. The tritium migration parameters for beryllium (diffusivity, solubility, surface recombination rate coefficient, and trapping) are so poorly known that results of calculations of the permeability and trapping can only be considered as rough estimates. The work presented here was performed with the intention of making these calculations more accurate by lowering the error bounds on some of the migration parameters.

S-65 beryllium disks were placed in the tritium plasma experiment (TPE) where they were exposed to plasmas consisting of tritium and deuterium. The 100 eV particle fluxes were varied between 1×10^{17} and 3×10^{18} (D + T)/cm^2 s with exposure times held at 1 h. The temperature of the samples during the exposures was 373 to 973 K. Subsequent to the plasma exposure, the samples were thermally outgassed through an ionization chamber and a catalyst bed into ethylene glycol bubblers.

2. Experimental procedures

The tritium plasma experiment (TPE) was used as the plasma source for all of the experiments. TPE is now

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located in the Tritium System Test Assembly (TSTA) at Los Alamos national laboratories. This experimental facility has been summarized elsewhere [1], but it can be briefly described as an arc reflex plasma source. During implantation, the sample was negatively biased to increase the energy of the deuterium and tritium ions striking the sample surface to 100 eV (the particles are primarily D_1^+ or T_1^+). The plasma intensity was varied by varying the voltage and temperature of the cathode.

The heating of the 2.54 cm radius samples was provided entirely by the plasma itself. The sample holder uses an outer copper ring to press the sample disk and two accompanying disks against the water-cooled back plate. The disk located immediately behind the sample disk is copper and is used to help flatten the temperature distribution in the sample. The third disk (the one in contact with the cooled plate) is varied in composition (copper, aluminum, or steel) and geometry (grooves or holes were used to reduce the effective thermal diffusion area) to allow different temperatures to be obtained for the same heat flux. Some temperature variation was also obtained by changing the tightness of the screws holding the outer copper ring. All of the disks including the samples were 2 mm thick.

The feed gas for the plasma was introduced at a rate of 30 cc(stp)/min. This gas consisted of 3% tritium and 97% deuterium. Within first order effects, the total hydrogen isotope retention is not dependent on the ratio of the individual isotopes. This ratio was used to calculate the total retention from the measurements of tritium content. The 2200 l/s turbomolecular pump located directly under the sample chamber maintained the total pressure in the system at 0.04 Pa. During plasma operation the ratio of hydrogen isotopes to the main impurity (water) was 1000 to 1 as determined by a mass spectrometer located on a small chamber attached to the sample chamber.

Each experiment began with turning on the plasma to a very low level with only deuterium. This was maintained until the impurities generated by the plasma initiation were removed by the pumping system. The plasma intensity was then increased at the same time that the bias on the sample was increased. Once the desired sample temperature was obtained (typically several minutes of tune-up were required), the tritium was added to the source gas. The one hour exposure began at the time the tritium was added to the gas.

After the plasma exposure, the sample was transferred to the outgassing system. Here the sample was ramped at a rate of 20 K/min from room temperature up to 1073 K. A gas mixture consisting of 99% helium and 1% hydrogen was swept over the sample and through an ionization chamber where the tritium level in the gas was continuously monitored. After passing through the ionization chamber, the gas was mixed with a small amount of oxygen and sent through a copper oxide catalyst bed where the hydrogen, tritium, and oxygen were converted into water. The water was then collected in the ethylene glycol bubblers for later tritium analysis. The total hydrogen isotope retention in the beryllium was determined by correcting for the ratio of gases in the feed gas.

The beryllium used in this experimental study was Brush Wellman S-65. This is a hot pressed material at 99.8% of theoretical density. It is 99.4% Be and approximately 0.6% BeO.

3. Experimental results

Examination of the 2 mm thick samples after removal from the plasma revealed that most of them had a dark color in the implantation zone. Auger analysis showed the dark area to be primarily beryllium oxide (almost no carbon). Part of the color change may have been due to the increased surface area in the form of sputter cones. The noted exception to the uniformly rough, dark color on the surface was the experiment performed at 973 K with a particle flux of 3×10^{18} (D + T)/cm^2 s. This sample had a thin layer exfoliating from the center of the sample. The reason for the exfoliation of the sample at 973 K is not understood. One of the samples exposed to the plasma at 573 K actually had a higher particle flux than the 973 K experiment.

In the discussion below, unless specifically stated otherwise, use of the word tritium is intended to mean both tritium and deuterium. The experimental results for the tritium retention in the S-65 beryllium are shown in Fig. 1 where the retention is plotted versus temperature. As mentioned above, each of these experiments was performed for one hour. The particle fluxes for each of the experiments are shown in the figure. The same data are repeated as Fig. 2 where the retention is plotted as a function of particle flux.

There are several points of interest in the data set. Perhaps the most important is the apparent lack of dependence of the tritium retention on the particle flux. This is seen not only at 373 K where the retention is lower for the higher flux, but also at 773 K where varying the flux by a factor of seven had no real affect on the retained tritium. This finding suggests that mechanisms other than normal diffusion and surface recombination were controlling the retention. This can be seen again in Fig. 3 where the experimental data of Fig. 1 is repeated with data from the DIFFUSE [2] computer code. The computer results examine three different particle fluxes with the assumption of $C = 0$ at the surface boundary. Almost without exception, the conservative assumption at the boundary of zero concentration still predicts more tritium inventory than was measured. The further assumption of recombination limited release would only serve to increase the difference in measured and computed values.

An indication of what is happening in the implant region is available in the report by Anderl et al. [3]. In

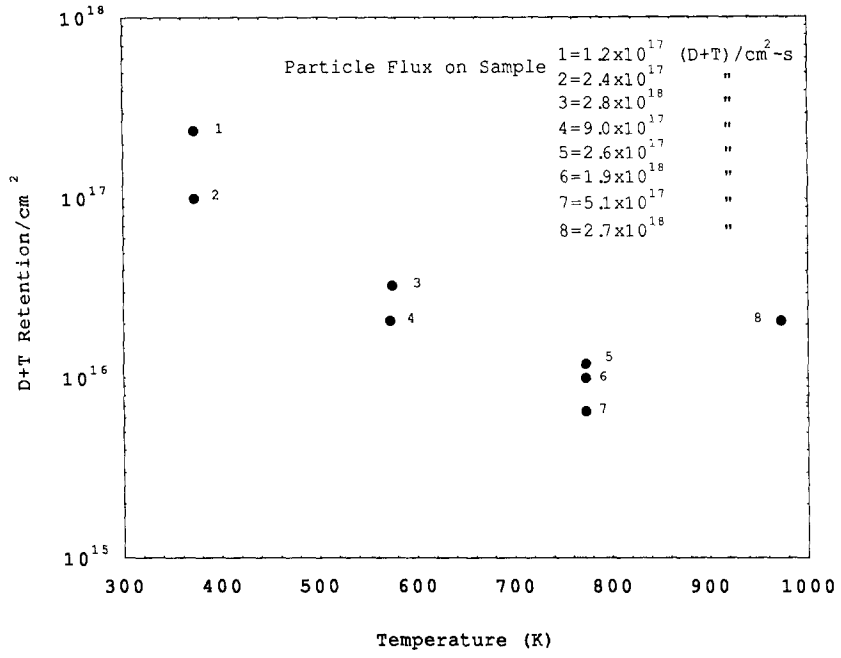

Fig. 1. Tritium retention in S-65 beryllium after plasma exposure.

their experiments using 3 keV D_3^+ on beryllium at 623 and 703 K, microscopic examination of the specimens after implantation showed a decrease in the oxide layer thickness but an increase in the near surface porosity. They postulated that the increase in porosity resulted from the formation of microscopic bubbles and voids. Guseva et al. [4] showed that a beryllium surface at 370 K exposed to 5 keV hydrogen ions progressed from pitting along grain boundaries at an ion fluence of 2.3×10^{19} H/cm^2 to severe coning (10 μm cones) at 1.5×10^{21} H/cm^2.

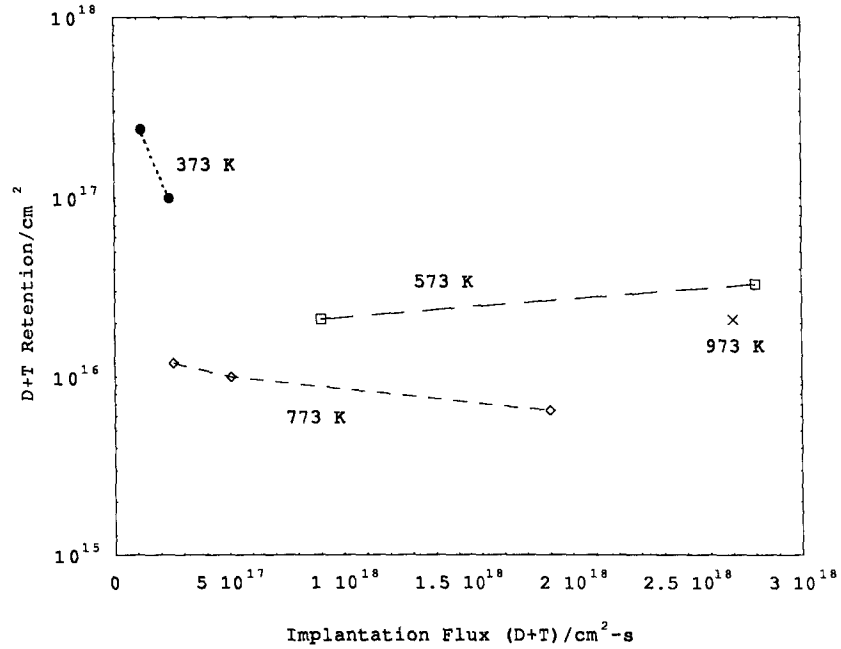

Fig. 2. Flux dependence of the tritium retention in beryllium.

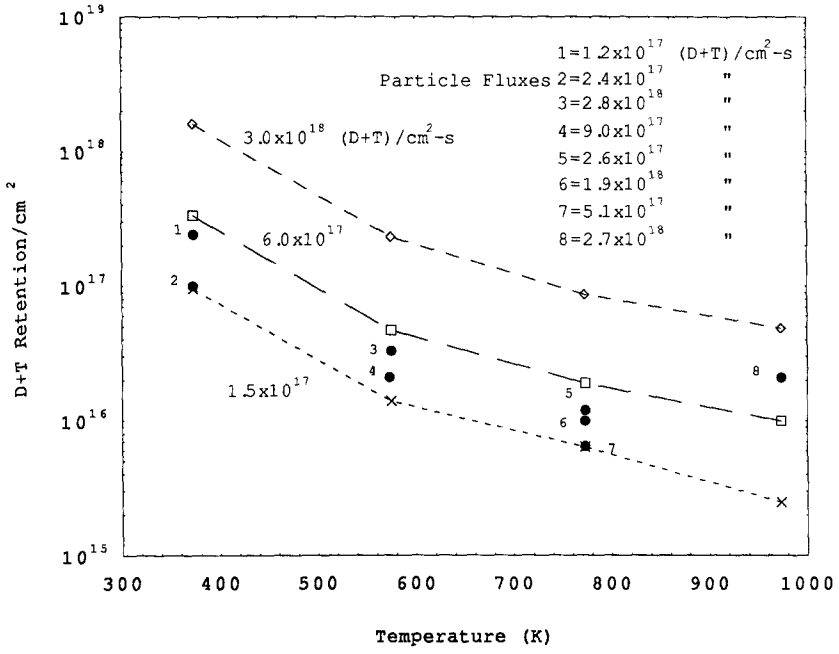

Fig. 3. Comparison of tritium retention in beryllium to that predicted by the DIFFUSE computer code assuming $C = 0$ at plasma facing surface.

Chernikov et al. [5] implanting 9 keV deuterium into beryllium at temperatures of 300, 500, and 700 K found that microscopic D_2 bubbles appeared in the metal when injected atom fractions reached about 1%. The D_2 molecule concentration peaked at 4×10^{21} cm^{-3} and backfilled from the implantation depth to the surface after which open porosity and cracks developed that let the molecular tritium escape.

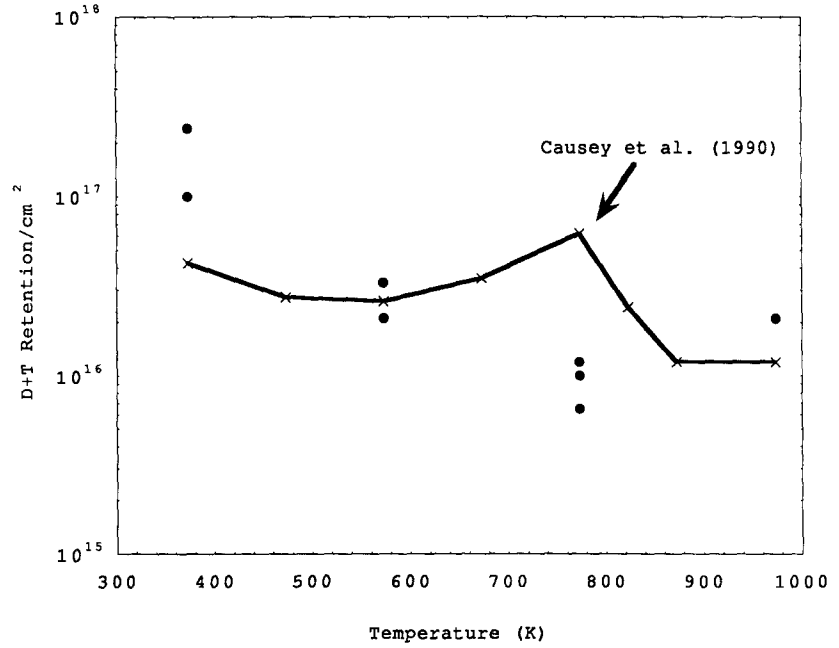

Fig. 4. Comparison of recent results of tritium retention in beryllium to earlier results of Causey et al. [7].

Based on the work of Guseva et al. [4] and Chernikov et al. [5], it appears that when the implantation layer has become saturated, essentially all of the implanting flux returns to the upstream face through the cracks and open porosity that develops. Hence the relative independence on implantation flux. The reduced inventories at higher temperatures appear to be a consequence of the reduced material strength, resulting in a lower threshold fluence for cracks to open, and a lower population of the traps in the bulk beryllium. The increase seen at the highest temperature measured occurs as the higher diffusivity at the elevated temperature carries more of the tritium deeper into the bulk. These processes, when modeled with the TMAP4 code [6], give results in good agreement with the experimental data.

The fact that the experiments were performed using 100 eV particles as apposed to keV particles does not cause any of the statements above to be incorrect. At 100 eV, the average particle depth is only a few nanometer. While the typical oxide layer on beryllium is also a few nanometer, it is believed that the oxide layer here was much thinner. Comparing the sputter rate with the arrival rate of oxygen, it is believed that the samples were very close to being free of oxide layers. This would result in the tritium ions being deposited into the metal, not the oxide.

The results from these experiments are compared to earlier results by Causey et al. [7] in Fig. 4. In the earlier experiments, the predecessor to the present day TPE was used. This experimental facility (TPX), produced an rf heated plasma that delivered a maximum flux to the sample of about 1×10^{17} (D + T)/cm^2 s. Specifically, the data shown for the earlier work was obtained using an incident 100 eV D_2^+ particle flux of 5.5×10^{16} (D + T)/cm^2 s. This is somewhat less than even the lowest particle flux used in the present experiments. With the exception of the localized peak at about 750 K reported in the earlier work, the results agree fairly well. Both data sets show the highest retention at the lowest temperature and an apparent rise in retention as the temperature of implantation in increased above 900 K. In the earlier work, the localized peak at about 750 K was postulated to be due to a surface recombination rate coefficient that decreased as temperature increased. For temperatures below about 800 K, the recombination rate coefficient was decreased even further by the oxide present on the surface. Above 800 K, the oxide and the beryllium would interdiffuse to yield a relatively clean surface with a faster recombination. Based on the new understanding of the performance of beryllium during implantation, the localized rise in the earlier data must somehow be related to the lower flux and higher oxygen potential. Even if voids and porosity were developing, there must have been sufficient oxygen present on the porosity surfaces to delay the release rate.

Other than to postulate for temperatures below 900 K that the apparent surface recombination rate coefficient of tritium in beryllium is quite rapid and that the solubility limit appears to be quite low, it is not really possible to use the results of these experiments to determine any of the parameters controlling the migration of tritium in beryllium. It is possible to partially extrapolate the results to the conditions that will exist in the ITER fusion reactor to estimate the tritium inventory in the beryllium limiter and first wall. The intermediate flux areas will likely have the least tritium inventory. This would include areas in the divertor where the temperature is in the 500–900 K range. The fluxes here would be quite intense, but the formation of open porosity will likely keep the inventory to a minimum. The two temperature extremes are more interesting. At the lowest temperatures, it appears there will be a rapid buildup of tritium in the implant zone. This is similar to the formation of the saturated layer as is well documented for carbon. Even when migration out of the saturated zone is assumed in the DIFFUSE computer code, the inventory remains low. The entire 200 m^2 first wall is only predicted to contain about 30 g of tritium after 1,000 h of continuous operation at 373 K. The beryllium that is at temperatures above 900 K is more difficult to assess. It appears from the single data point at 973 K that the tritium retention is beginning to rise. This was also the data point where the thin outer layer was seen to exfoliate. It is only possible to state that this is an unknown, and that more data at these temperatures are needed.

4. Conclusions

Tritium retention measurements in beryllium were performed over the temperature range of 373 to 973 K using 100 eV triton fluxes between 1×10^{17} and 3×10^{18} T/cm^2 s. With the exception of a single data point at 373 K, the retention is less than that predicted assuming the recombination at the surface is not rate limiting. The high release rate at the surface appears to be caused by open porosity in the near-surface area. These results can be interpreted to predict that the tritium inventory in the bulk beryllium of ITER will be quite small for temperatures below 900 K. For temperatures above this temperature, more data are required.

Note added in proof

Subsequent analysis of the beryllium samples using scanning electron microscopy and X-ray analysis has shown the black color on the sample surfaces described in Section 3 to be due to sputter cones caused by copper impurities deposited by plasma-induced arcing. A surface covered with sputter cones might be expected to have enhanced surface retention in the increased surface area, but the bulk retention could be significantly lowered due to enhanced recombination on the conical surfaces.

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