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Depth profile of tritium in plasma exposed CX-2002U

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Abstract

Depth profiles of the Ag grains density in autoradiographs, which represent tritium concentration in CX-2002U samples exposed to high flux D/T particles under various conditions, were examined and the apparent diffusion coefficients were estimated from the profiles. Plasma discharge generating D/T atomized particles with low energies increases tritium inventory in the samples by introducing high tritium concentration on the surface exposed and following diffusion process into the deep region with apparent diffusion coefficients (1.7×10^{-16} m^2/s at 293 K and 2.3×10^{-15} m^2/s at 573 K), which are much larger than the diffusion coefficients in the bulk reported. Oxygen RF-plasma exposure might be effective to remove tritium retained even at a fairly deep region in carbon fiber composite (CFC) components.

1. Introduction

Carbon fiber composite (CFC) is one of the plasma facing material candidates for deuterium (D)–tritium (T) fusion experimental machines because of its low atomic number and good thermal properties. Tritium retention and permeation are important in safety evaluation, tritium processing design and operation scenario for tritium inventory control in the vacuum vessel of a fusion reactor. Retention of hydrogen isotopes in graphite has been widely investigated using energetic hydrogen isotopes and sub-eV atomic hydrogen isotopes [1–8]. But there are fairly large discrepancies among the data reported and few data available to estimate the tritium inventory in CFC exposed to high flux D/T particles. Also there are few data to predict precisely the tritium permeation behavior through a real CFC armor tile with a thickness much larger than that used in previous experiments. This is due, in part, to difficulties in evaluation of tritium diffusion through pores in CFC.

On the other hand, tritium removal from CFC is also important from the viewpoint of tritium recovery and decontamination from the wasted CFC components. A method is needed that will remove tritium from areas of high tritium retention in the plasma facing components without heating, even with a consideration of oxygen use. Such means will make a decontamination process simple and prevent tritium from migrating deep into the bulk.

Autoradiography is a technique which can give a visual indication of real tritium behavior inside the bulk of materials when the position of tritium is well preserved through the process of charging tritium and observation [9]. Using this technique, visual examinations of tritium behavior in the CFC samples exposed to high flux D/T particles are being carried out at Tritium Engineering Laboratory (TPL) of Japan Atomic Energy Research Institute. In the present work, the tritium concentration profile in two-dimensional CFC (CX-2002U) exposed to high flux D/T atomized particles under various conditions are presented and the apparent diffusion estimated from the profile are determined. Also, the effect of oxygen plasma exposure on tritium removal from a CFC sample was examined using the autoradiography.

2. Experimental

Before plasma or gas exposure, sample disks of CX-2002U (Toyotanso Co., 10 mmϕ, 4 mm^t) were degassed at 1150 K for one hour under vacuum below 10^{-4} Pa.

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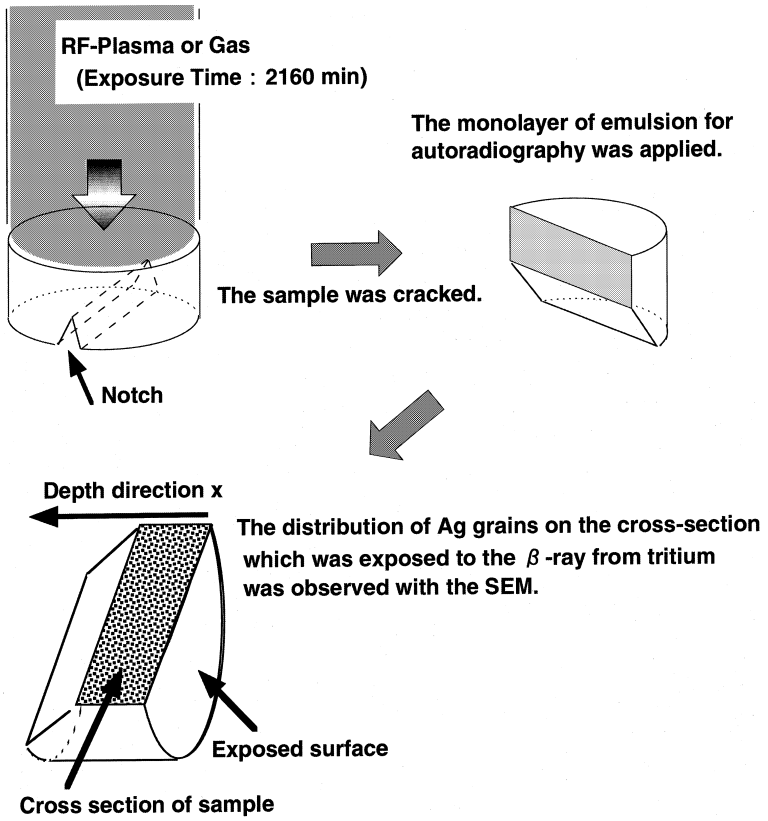

Fig. 1. Measurement procedures of autoradiography for the sample prepared in this experiment.

D/T RF (13.56 MHz, 50 W) discharge plasma was generated by introducing D_2/T_2 (D_2:$T_2 = 1000$:1) gas mixture at a flow rate of 1 cm^3/min into a quartz plasma tube [10]. The pressure in the plasma tube was maintained at 2.7 Pa and that of the main chamber at 2.7×10^{-2} Pa. Two runs of D/T RF-plasma exposure were performed for 2160 min at temperatures of 293 and 573 K. Also D/T gas exposure at 293 K for 2160 min was carried out with the same gas flow rate and tritium concentration.

Procedures of the autoradiography measurement are shown in Fig. 1. After plasma exposure, the sample was cracked along a notch, which was made on the back surface before the plasma exposure. Then a monolayer of the emulsion for autoradiography (Konica NR-H2), in which the average diameter of AgBr grains was 0.08 μm, was applied to the surface of the cracked cross-section by making the samples touch a thin film of the emulsion held in a wire loop. The sample was kept at 77 K to expose the emulsion by β-particles from tritium existing in the region close to the cross-section surface. The duration time of exposure was 14 days. The average diameter of Ag grains converted from AgBr by β-particles was 0.17 μm and the projected range of 18 keV electron in the sample is about 5 μm. Hence it is possible to observe the spatial distribution of tritium within an accuracy of 5 μm. The distribution of Ag grains on the cross-section was observed with SEM after removal of the emulsion gelatin. As an example of autoradiograph, an edge part of the cross-section of the samples exposed to D/T plasma at 293 K is shown in Fig. 2. White spots correspond to Ag grains formed by β-particles from tritium. The region in the image close to the surface of

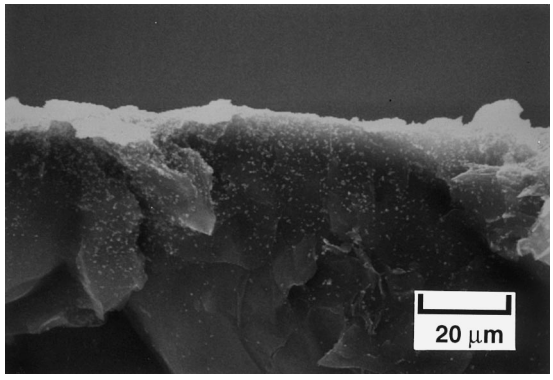

Fig. 2. An example of the autoradiograph of a cross-section of CX-2002U sample exposed to D/T RF-plasma at 293 K.

the sample was radiant due to dense electron reflection at the edge. Therefore, the density of the Ag grains, which is considered to be proportional to tritium concentrations, was measured from the area at a distance of 2.5 μm from the edge. The depth profiles shown later were taken as a function of depth from the edge.

Irradiated samples were also exposed to oxygen plasma generated by RF discharge (13.56 MHz, 50 W) for 2160 min, at a flow rate of 1 cm^3/min and a temperature of 293 K.

3. Results and discussion

Some comparisons were made for the sample exposed to gas, D/T plasma and oxygen plasma to confirm differences in tritium behavior under different treatments.

3.1. Gas exposure and plasma exposure

Fig. 3 shows the depth profiles of Ag grain density for the CX-2002U samples exposed to gas and plasma, respectively, at 293 K. The density of Ag grains which represents tritium concentration in the sample exposed to the plasma decreased rapidly from the region close to the surface, as the depth from the surface increased. On the other hand, the density in the sample exposed to gas was much smaller and changed less with the depth. This result obviously shows that the RF discharge, which generated a lot of atomized D/T particles, has an effect to increase the amount of tritium retained in the region close to the surface exposed to the plasma. The energies of atomized D/T particles were supposed to be so low that the depth profile of Ag grains densities shown in Fig. 3 could be attributed to some diffusion processes of tritium taking place beyond the very thin particle-implanted region into the deeper region of the sample. Under the condition of plasma exposure, tritium migration caused by such diffusion processes inside the sample was reached at the depth of about 120 μm or more from the plasma exposed surface.

3.2. Plasma exposure at different temperatures

Fig. 4 shows the depth profiles of Ag grain density in CX-2002U samples exposed to plasma at 293 and 573 K as a function of depth from the plasma exposed surface. The Ag grain density in the sample exposed to plasma at 293 K was almost twice as high as that of the sample exposed at 573 K at the region near the plasma exposed surface, but it decreased more rapidly with increasing depth from the surface. This result implies that at higher sample temperatures faster recycling process took place on the plasma-exposed surface.

Based on the data of the Ag grain density depth profile, which represents the tritium concentration, an approach was used to obtain apparent diffusion coefficients at 293 and 573 K. Bulk and pore diffusion processes were assumed in a one-dimensional diffusion model. It was also assumed that the tritium concentration was constant at the sample surface. The equation of calculation for the tritium concentration profile was given by

$$C(x) = C''(1 - \mathrm{erf}(x/2\sqrt{(D_t t)})),$$

where $C(x)$ is the tritium concentration at a depth x from the exposed surface (atoms/m^3), C'' the constant surface concentration (atoms/m^3), D_t the diffusion coefficient (m^2/s), t the exposed time (s) and erf( ) is the error function. This equation was applied between the

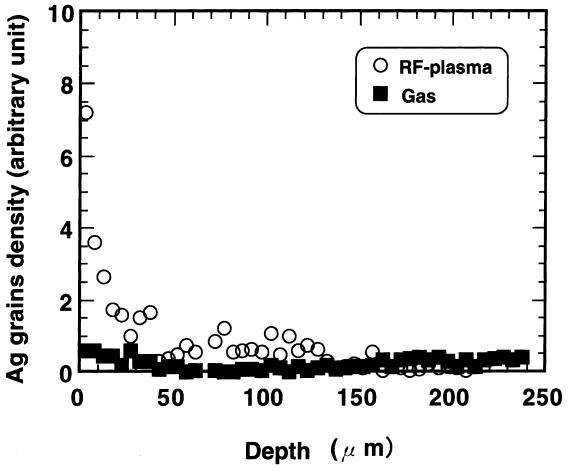

Fig. 3. Depth profiles of Ag grain density in CX-2002U exposed to D/T RF-plasma and gas at 293 K.

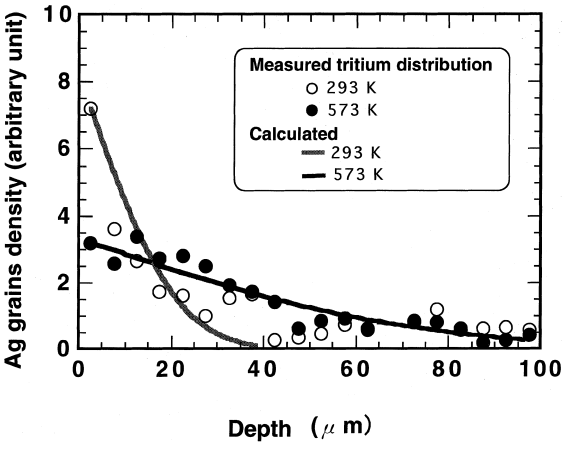

Fig. 4. Depth profiles of Ag grain density in CX-2002U exposed to D/T RF-plasma at 293 and 573 K.

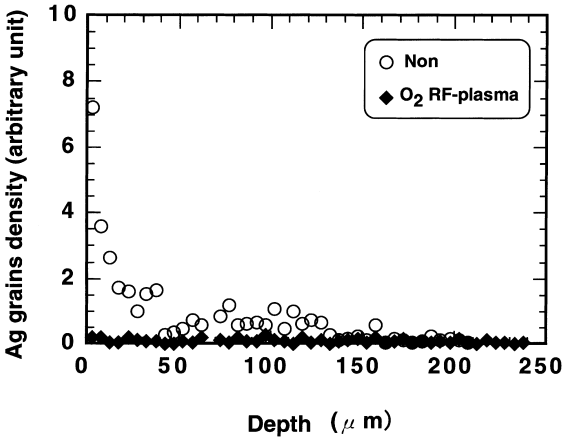

Fig. 5. Depth profiles of Ag grains density in CX-2002U irradiated by oxygen RF-plasma after exposure of D/T RF-plasma at 293 K.

surface and the point, where the amount of concentration was 1/10 of the value at the region near the surface. In this case, we fitted $C(x)$ to measured concentrations in the region of 2.5–40 μm at 293 K and 2.5–100 μm at 573 K, and derived $D_t = 1.7 \times 10^{-16}$ m^2/s at 293 K and 2.3×10^{-15} m^2/s at 573 K from the fitting curves shown in Fig. 4. The values obtained here are much larger than those reported as bulk diffusion coefficients in previous researches [11–17]. The process, to which this fact can be attributed, we suppose, is the diffusion of atomic hydrogen or hydrogen molecular radical through pores.

3.3. Oxygen plasma exposure to the tritium retained sample

Fig. 5 also shows the Ag grains density depth profiles as a function of depth in the D/T plasma exposed samples with and without the subsequent oxygen RF-plasma treatment at 293 K. It is obvious that by oxygen plasma exposure, tritium not only at the region close to the surface but also at the deep region more than 100 μm was swept out. The mechanism of the sweep out of tritium and the form of released tritium were not clear and the total decontamination factor has not been measured yet. However, it can be said that oxygen RF-plasma exposure is an effective means to remove tritium existing even at the deep region in CFC components.

4. Conclusions

Depth profiles of Ag grains density in the autoradiographs, which represent tritium concentration in the CX-2002U samples exposed to high flux D/T particles under various conditions and the apparent diffusion estimated from the profile are presented. Also an effect of oxygen plasma exposure on tritium removal for decontamination of tritium contaminated CFC components was examined. The results obtained can be summarized as follows:

1. Low energy D/T atomized particles generated by plasma discharge increases tritium inventory in CFC by introducing high tritium concentration on the surface exposed and following diffusion process into the deep region with apparent diffusion coefficient much larger than the ones in the bulk reported.
2. By increasing temperature under plasma exposure, tritium concentration at the surface was decreased, which can be attributed to the recycling processes getting faster on the irradiated surface. However, the higher temperature goes up, the deeper tritium migrates
3. According to the results of fitting the one-dimensional diffusion equation to the Ag grains density depth profiles obtained for the sample exposed to the plasma at 293 and 573 K, the apparent diffusion coefficient D_t was estimated to be 1.7×10^{-16} m^2/s and 2.3×10^{-15} m^2/s, respectively. The diffusion processes would be related to diffusion of atoms or radical molecules produced by RF-plasma discharge.
4. Oxygen RF-plasma exposure, which swept out tritium even at a fairly deep region in the sample, might be effective to remove tritium retained in wasted CFC components.

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