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Journal of Nuclear Materials 258–263 (1998) 1092–1096

journal of nuclear materials

# Tritium retention in CX-2002U and methods to reduce tritium inventory

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**Abstract**

Tritium retention in CX-2002U was studied by exposure to high flux of D/T atomized particles. The amount of tritium retained in CX-2002U appeared to be proportional to the half power of total incident fluence below 597 K and to the one third power at 897 K. No saturation was observed up to $10^{27}$ atoms/m$^2$ at 297 K. The tritium concentration profile was also measured by means of autoradiography, from which an overall diffusion coefficient of $D_t = 3.6 \times 10^{-14}$ m$^2$/s at room temperature was estimated. This value, which was much larger than the reported bulk diffusion coefficient, implies additional diffusion processes such as pore diffusion of tritium atoms or molecules. Tritium removal from the sample by exposure to air and helium plasma after irradiation of D/T atomized particles was studied. Tritium retention was reduced by about one tenth during air plasma exposure and by about a half during helium plasma exposure. © 1998 Published by Elsevier Science B.V. All rights reserved.

## 1. Introduction

Carbon Fiber Composite (CFC) is one of the candidates for an ITER plasma facing material because of its low atomic number and good thermal properties. Among the candidates, the largest amount of tritium inventory is estimated for CFCs based on the data reported, but there are fairly large discrepancies among this data [1–11]. Especially for 2- or 3-dimensional CFC materials, there are few data available to estimate the tritium inventory for a high D/T particle flux loaded on an ITER divertor. At TPL/JAERI, tritium retention in CFCs is investigated using an apparatus which can simulate the ITER divertor conditions.

In the present work we have measured tritium retention in CX-2002U exposed to atomized D/T particles with total incident fluences up to $10^{27}$ atoms/m$^2$. Tritium concentration profiles were observed by means of autoradiography, while studies of tritium removal by the exposure of irradiated samples to air and helium plasmas were also performed.

## 2. Experimental

A D/T (D:T = 1000:1) mixture gas was supplied at a flow rate of 1 cm$^3$/min and a D/T plasma was generated by a RF discharge (13.56 MHz, 50 W) [12]. The dimensions of CX-2002U sample disks were 10 mm in diameter and about 0.3 mm in thickness. The sample was degassed at 1150 K for one hour under vacuum below $10^{-4}$ Pa before plasma exposure. The pressure in the plasma tube was kept at 2.7 Pa, and in the main chamber at $2.7 \times 10^{-2}$ Pa. We assumed the flux of incident atoms on the CFC sample, which occupied the cross section of plasma tube at the end, was equal to the D/T gas flow rate, i.e. a flow rate of 1 cm$^3$/min corresponded to an atom flux of $7.4 \times 10^{23}$ at the sample surface. D/T plasma exposures were performed for 2, 10, 30 and 60 min, total fluences were $1.5 \times 10^{24}$, $7.4 \times 10^{24}$, $2.3 \times 10^{25}$ and $4.6 \times 10^{25}$ atoms/m$^2$, and sample temperatures were 293, 593 and 893 K, respectively. In case of sample temperature was 293 K, D/T plasma exposures was performed for 2160 min, total fluence was $1.7 \times 10^{27}$ atoms/m$^2$. After exposure, the sample was taken from the chamber and in a separate apparatus heated at about 1170 K in an air flow of 100 cm$^3$/min to induce combustion. The tritiated water formed was then

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PII: S0022-3115(98)00280-3

trapped with two water bubblers and a cold trap (216 K). The total tritium amount in the sample was estimated from the activity of trapped tritium measured with a liquid scintillation counter system.

The sample for autoradiography was also made by exposure to D/T plasma for 36 h (fluence: $1.7 \times 10^{27}$ atoms/m$^2$). A notch was made beforehand on the back surface and cracked there after D/T plasma exposure to observe its cross-section. The monolayer of the emulsion for autoradiography (Konica NR-H2), in which the average diameter of AgBr grains was 0.08 μm, was applied by making the samples touch a thin film of the emulsion held in a wire loop. The sample was kept in a light-tight box filled with helium gas and kept at 77 K to expose the emulsion to the $\beta$-ray from tritium decay. The duration time of exposure was 6 days. The average diameter of Ag grains converted from AgBr by a $\beta$-ray was 0.17 μm and the range of 18 keV electron in the sample is about 5 μm, so it was useful to observe the spatial distribution of tritium with an accuracy of 5 μm. The distribution of Ag grains on the cross-section was observed with SEM after removal of the emulsion gelatin.

To observe the effects on reduction of tritium retention in CFCs, the irradiated samples were exposed to air and helium plasmas. Air plasma exposure was performed for 60 and 120 min (air flow rate: 1 cm$^3$/min) to the samples which were irradiated by D/T plasma for 60 min ($4.6 \times 10^{25}$ atoms/m$^2$), and helium plasma exposure was for 60 min (gas flow rate: 1 cm$^3$/min).

## 3. Results and discussion

Thermal desorption spectra (TDS) of $D_2$ from the sample are shown in Fig. 1. Two peaks in deuterium release rate were observed at different temperatures, one at about 850 K and the other at about 1350 K. This result is similar to that of previous work on isotropic graphite [12]. The first peak can be attributed to deuterium detrapping from trap sites, and the second one should be derived from a chemical bond of tritium and carbon according to the analogy discussed in the work.

Tritium retention in CX-2002U, which is given by multiplied measured total tritium by 1001, as a function of total fluence, is shown in Fig. 2. It is obvious that retention is not saturated even at high fluences and is proportional to the half power of total incident fluences at 293 and 593 K. This implies that the increase is correlated to a diffusion process of hydrogen in CFC materials. At 893 K, retention appears to be proportional to about the one third power of the total incident fluence. This result implies that some process to suppress the increase of tritium retention such as recombinational release are effective at higher temperatures.

The autoradiograph of a cross-section of the sample is shown in Fig. 3. White spots correspond to Ag grains formed as a result of local tritium decay. The concentration of tritium should be proportional to the number density of Ag grains. The relative tritium concentration profile as a function of depth $x$ from irradiated surface is shown in Fig. 4. The concentration decreased rapidly up to a depth of $2 \times 10^{-5}$ m, and then decreased gradually. An approach to obtain an overall diffusion coefficient which may include both bulk and pore diffusion was tried by using the one dimensional diffusion equation with a condition of a constant tritium concentration at the sample surface given by

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

where $C(x)$ is the tritium concentration at a depth $x$ from the exposed surface (atoms/m$^3$), $C''$ is the constant

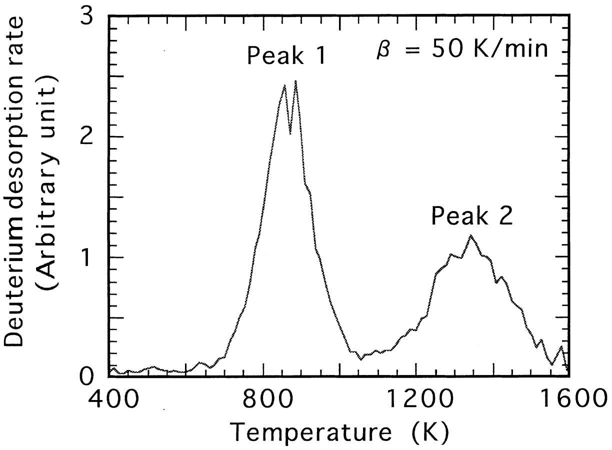

Fig. 1. Thermal desorption spectrum of $D_2$ from CX-2002U. (temperature of CX-2002U during plasma exposure: 293 K, fluence: $9.1 \times 10^{25}$ atoms/m$^2$).

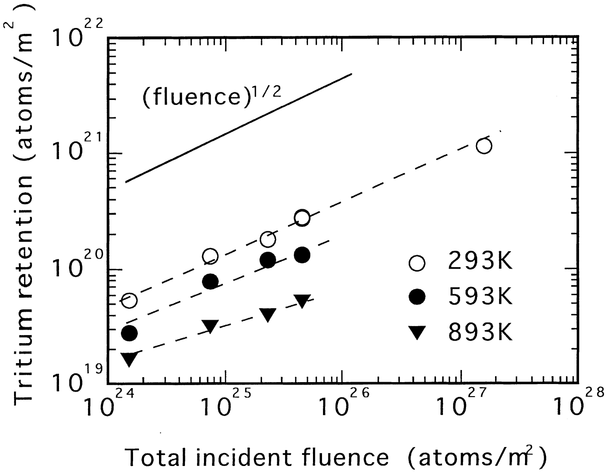

Fig. 2. Retention of tritium in CX-2002U as a function of total incident fluence.

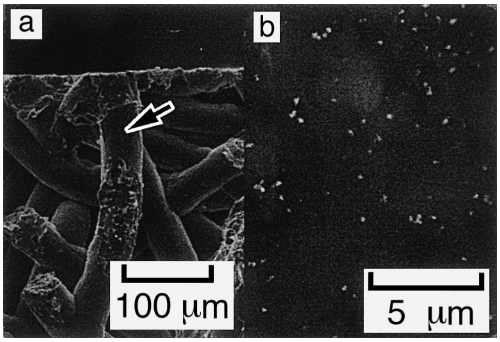

Fig. 3. Autoradiographs of a cross-section of CX-2002U. (a) A view of cross-section of CX-2002U. (b) Expanded view of the area indicated by arrow (depth is 70 μm).

concentration (atoms/m$^3$), $D_t$ represents the bulk and pore diffusion coefficients (m$^2$/s), $t$ is the exposed time (s) and erf( ) is the error function. In calculation $D_t$, we assumed that the constant concentration in contact with the sample at a depth of $1.75 \times 10^{-5}$ m did not include the saturation layer. We fitted $C(x)$ to measured concentrations and derived $D_t = 3.6 \times 10^{-14}$ m$^2$/s. In Fig. 4, fitted $C(x)$ points are also plotted. Tanabe et al. [9] suggested this diffusion process concerns the growth of a saturated layer. Langley et al. [13] reported that the total retention saturated at a lower incident fluence than in this experiment. The $D_t$ value obtained here is rather large compared with previous work, suggesting that diffusion processes such as atomic or molecular diffusion in interlinked pores should be considered.

Fig. 5 shows tritium retention in CX-2002U after exposure in air and helium plasmas. Helium plasma

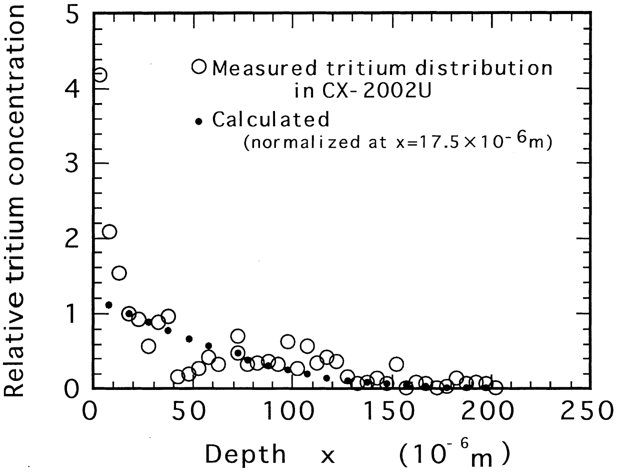

Fig. 4. Tritium concentration profile in CX-2002U.

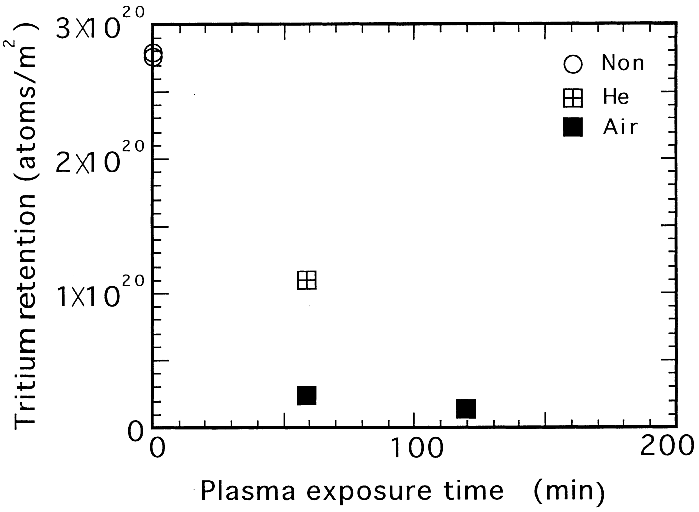

Fig. 5. Tritium retention in CX-2002U after exposure of air and He plasmas.

exposure reduced the amount of tritium retention to about half of the initial retention of the sample exposed to D/T atomized particles. On the other hand, air plasma exposure reduced the amount of tritium retention to about one tenth in both cases of exposure for 60 and 120 min. This difference can be attributed to the different reaction mechanism, i.e. the reaction mechanism between helium and carbon is only physical sputtering and that of air and carbon is mainly a chemical reaction. To reduce tritium retention in CFCs, exposure to air plasma may be effective, but oxygen will introduce disadvantages into the fusion plasma. It will be necessary to estimate oxygen levels retained in samples when air plasma exposure is employed to reduce the tritium inventory.

## 4. Conclusions

The tritium retention in CX-2002U at 293, 593 and 893 K has been studied using a mixture of deuterium and tritium atoms of low energies and high flux. A tritium concentration profile has also been observed by means of autoradiography. The results obtained here can be summarized as follows:

(1) Hydrogen isotopes retained in CX-2002U exist in two states, which correspond to two peaks in the thermal desorption spectra of $D_2$ at temperatures about 850 and 1350 K. This result was similar to that of our previous work on isotropic graphite; the first peak can be attributed to deuterium detrapping from trap sites and the second one should be derived from a chemical bond of tritium and carbon by the analogy discussed in previous work.

(2) The total hydrogen isotope retention in CX-2002U was not saturated and seemed to be proportional to the half power of the total incident fluence up to $1.7 \times 10^{27}$ atoms/m$^2$ at 293 K and $4.6 \times 10^{25}$ atoms/m$^2$ at 593 K. This implies that the increase of tritium retention is correlated with a diffusion process of hydrogen in the CFC microstructure. At 893 K, the retention appeared to be proportional to about the one third power of the total incident fluence. This result suggests that recombinational release is effective at higher temperatures.

(3) By means of autoradiography, a tritium concentration profile was measured and an overall diffusion coefficient of $D_t = 3.6 \times 10^{-14}$ m$^2$/s was estimated. This $D_t$ value was rather larger than others reported, so that diffusion processes such as diffusion of tritium atoms or molecules in interlinked pores might play an important role in tritium retention.

(4) Air plasma exposure reduced the amount of tritium to about one tenth, while one fifth of that was obtained for a helium plasma. Since oxygen is detrimental to fusion plasmas, it is necessary to estimate oxygen concentrations in CX-2002U when air plasma exposure is employed to reduce tritium inventory.

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