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Hydrogen isotope behavior in the first wall of JT-60U after deuterium plasma operation

Y. Oya [a,*], T. Tanabe [b], M. Oyaidzu [c], T. Shibahara [d], K. Sugiyama [b], A. Yoshikawa [c], Y. Onishi [c], Y. Hirohata [e], Y. Ishimoto [f], J. Yagyu [f], T. Arai [f], K. Masaki [f], K. Okuno [c], N. Miya [f], S. Tanaka [g]

[a] *Radioisotope Center, The University of Tokyo, 2-11-16, Yoyoi, Bunkyo-ku, Tokyo 113-0032, Japan*
[b] *Interdisciplinary Graduate School of Engineering Science, Kyushu University, 6-10-1, Hakozaki, Higashi-ku, Fukuoka 812-8581, Japan*
[c] *Radiochemistry Research Laboratory, Faculty of Science, Shizuoka University, 836, Oya, Suruga-ku, Shizuoka 422-8529, Japan*
[d] *Graduate School of Engineering, Nagoya University, Furo-cho, Chikusa-ku, Nagoya 464-8603, Japan*
[e] *Graduate School of Engineering, Hokkaido University, Kita-13, Nishi-8, Kita-ku, Sapporo 060-8628, Japan*
[f] *Japan Atomic Energy Agency, 801-1, Mukoyama, Naka, Ibaraki 311-0913, Japan*
[g] *Department of Quantum Engineering and Systems Science, Graduate School of Engineering, The University of Tokyo, 7-3-1, Hongo, Bunkyo-ku, Tokyo 113-8656, Japan*

Abstract

Retention of hydrogen isotopes in the carbon (isotropic graphite) first wall tiles of JT-60U was studied by secondary ion mass spectrometry and thermal desorption spectroscopy. The surface morphology and erosion/deposition profiles of the tiles were characterized using scanning electron microscope and X-ray photoelectron spectroscopy. The upper area is mainly eroded, while the bottom area of the inboard wall is dominated by deposition. In contrast to the divertor area, hydrogen isotope retention in the eroded wall area was generally larger than that in the deposition dominated area. Measured near surface concentrations of hydrogen isotopes in the wall tiles, as well as the D/H ratios, were a little higher than those in the divertor area. This indicates direct implantation of high-energy D from NBI into the first wall. The lower temperature of the first wall relative to the divertor tiles would reduce desorption and/or replacement of implanted D by subsequent D or H impingement.

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1. Introduction

The elucidation of hydrogen isotope behavior in carbon materials for fusion reactor is one of the most critical issues for fusion safety [1–5]. We have extensively studied retention characteristics of hydrogen (H), deuterium (D) and tritium (T) in the JT-60U divertor area [6–13] and found that most T produced by DD reactions was implanted

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[*] Corresponding author. Present address: Radiochemistry Research Laboratory, Faculty of Science, Shizuoka University, 836, Oya, Suruga-ku, Shizuoka 422-8529, Japan. Tel.: +81 54 238 4803; fax: +81 54 238 3989.
E-mail address: syoya@ipc.shizuoka.ac.jp (Y. Oya).

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into graphite tiles at the depth of a few μm. However, H and D were retained in the near surface area. Most of the inner divertor area except the area near the pumping slot was covered by thick redeposited layers with a maximum thickness of around 104 μm and the redeposited layers retained D uniformly throughout. On the other hand, the outer divertor area was erosion dominated and rather high hydrogen isotope (H + D) retention was found only in very near surface region less than 0.5 μm. Therefore, total hydrogen isotope retention in the divertor area of JT-60U is much less than that observed in other tokamaks like JET and TFTR [4,14,15].

However, for the evaluation of the total hydrogen isotope recycling in a fusion reactor, it is important to reveal the hydrogen isotope retention in the first wall. This motivates us to examine the first wall tiles for erosion/deposition and hydrogen isotope retention.

2. Experimental

Fig. 1 shows a cross sectional view of the vacuum vessels with (a) W-type divertor and (b) open divertor in JT-60U. All the samples were made of isotropic graphite. As shown in the figure, poloidally four tiles used in the W-type divertor configuration were investigated, along with one used in the open divertor configuration. Tiles 5Id4, 5Lb4 and 13Lg6 were located at the upper area of the first wall, and 5Md4 and 5Af4 were placed in the lower area of the inboard wall as shown in Fig. 1. Samples with size about $10 \times 10 \times 0.5\ \mathrm{mm}^3$ were cut from these tiles and named 5Md4, 5Af4, 5Id4, 5Lb4 and 13Lg6. All tiles except 13Lg6 were exposed to a total of about 21 000 DD shots and 4300 HH shots from 1991 to June 2002 [10–12]. These tiles were removed from the vacuum vessel without final HH discharge for T removal. During these experimental campaigns, 11 times of boronization using $B_{10}H_{14}$ and $B_{10}H_{14} + D_2$ were done for these tiles. Different from these tiles, 13Lg6 taken from the open divertor regime was subjected to five times of boronization using $B_{10}H_{14}$ only and was exposed to total of about 5250 DD shots and 2400 HH shots between April 1991 and October 1994 in the upper inboard position shown in Fig. 1(b). The NBI power for the open divertor configuration was slightly higher than that for the W-type divertor.

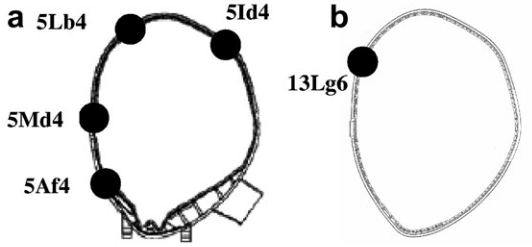

Fig. 1. Cross sectional view of the vacuum vessel with (a) W-type divertor and (b) open divertor in JT-60U and sampling positions.

Erosion and deposition profiles of the samples were investigated by a scanning electron microscope (SEM) (Hitachi type S-300H). Detection limit of the redeposition layer thickness was about 0.2 μm [16]. The depth profiles of H and D retained in the samples were analyzed by secondary ion mass spectrometer (SIMS) (ULVAC-PHI ADEPT1010 Dynamic SIMS System) [8–12]. Cesium ions (Cs^+) were used with an energy of 5 keV. The sputtering rate was nearly 1 μm/h in case of the graphite tiles. The negative signal intensities of H and D were normalized by that of $^{11}B + {}^{12}C$ for comparison. The chemical states of tile surfaces were also studied by X-ray photoelectron spectroscopy (XPS) (ULVAC-PHI ESCA5500MC system), using a Mg-Kα 400 W X-ray source [8,10,11]. The depth profile was also analyzed by Ar ion (Ar^+) sputtering with an energy of 4.0 keV. The rastered area was set to be $2 \times 2\ \mathrm{mm}^2$. The sputtering rate was estimated to be 16.3 nm/min. Thermal desorption spectroscopy (TDS) was applied to these samples with a heating rate of $0.42\ \mathrm{K\ s^{-1}}$ up to the temperature of 1200 K [9,13].

3. Results and discussion

Fig. 2 shows typical SEM images for the first wall tiles. In the samples located at the upper side of the first wall, namely 5Id4, 5Lb4 and 13Lg6, small hollow and wavy structure surfaces were found, which may correspond to the erosion trace. However, for 5Md4 and 5Af4, the surface profiles were clearly different and the tiles were covered by the deposition layer with a thickness less than a few μm, which would be the deposition dominated area.

Depth profiles of H and D were analyzed by SIMS and the integrated SIMS signal intensity ratios of $D/(^{11}B + {}^{12}C)$ and $H/(^{11}B + {}^{12}C)$ were obtained with the sputtering time of 6000 s as summarized in Fig. 3. Several sampling positions along the poloidal direction were measured. The figure shows that D retention ($D/(^{11}B + {}^{12}C)$) was much

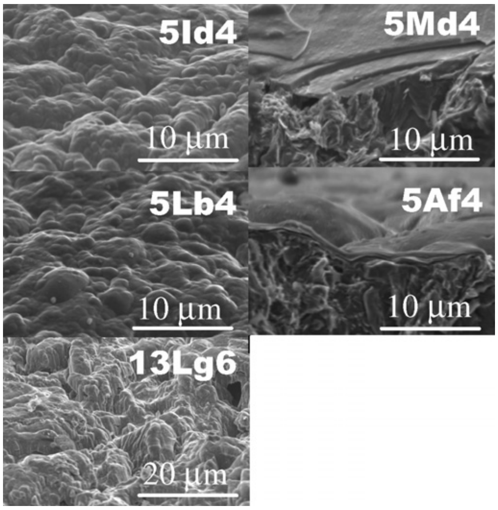

Fig. 2. Typical SEM images of the samples in JT-60U.

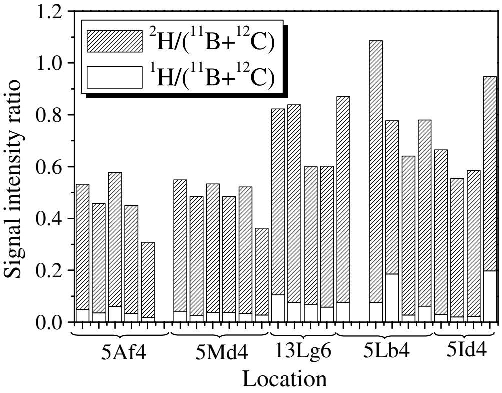

Fig. 3. Integrated signal intensity ratio of the first wall samples.

higher than H retention ($H/(^{11}B + ^{12}C)$) and the total retention (H + D) for the deposition dominated area (5Af4 and 5Md4) was relatively smaller than that for the erosion dominated area (5Id4, 5Lb4 and 13Lg6). These retention characteristics are quite opposite from the previous observation for the divertor tiles, where H retention was larger than D retention and the total retention was much large for the deposition dominated area [10–12].

To evaluate the chemical characteristics of the sample surfaces, XPS observation and Ar^{+} sputtering were sequentially performed. Fig. 4 shows the typical depth profiles of chemical composition for the deposited area (5Md4). In 5Md4, a substantial carbon deposition layer of about 80 nm in thickness was observed on boron layers of about 1.3 μm followed by the carbon matrix. The XPS peaks shifted in accordance with the concentration change with depth. For 5Md4, for example, the peak position of C 1s was shifted from 285.9 eV at the surface to 283.2 eV at the depth of 168 nm and shifted back to 284.5 eV at 1.3 μm, while B 1s, 192.8 eV at the surface, shifted to 188.5 eV at 168 nm with no further shift with depth. The depth profile for 13Lg6 (data not shown) indicated a typical pattern for eroded tiles, i.e. no boron layers observable, though a small amount of boron, less than 14%, had penetrated rather deeply. Since the solubility of boron in graphite is about 10%, this boron content might correspond to the formation of a solid solution of B and C.

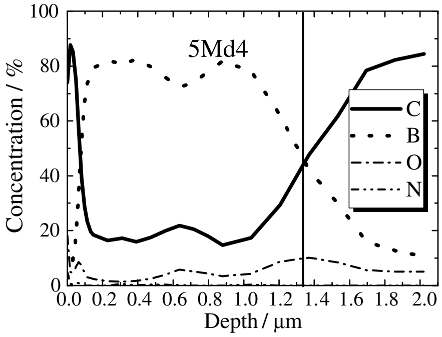

Fig. 4. Concentration profiles of 5Md4 estimated by XPS.

To determine the hydrogen isotope retention and their desorption behavior, TDS was applied to these samples. Fig. 5 shows the TDS spectra of D_2 for some samples. For 5Id4 and 5Lb4, which were erosion dominated samples, a large desorption peak was observed at about 700 K. The desorption behavior for deposition dominated 5Md4 and 5Af4 was quite different from the eroded ones. The peak was broadened and shifted to higher

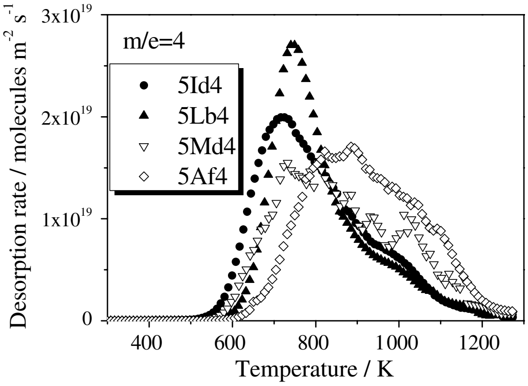

Fig. 5. TDS spectra of D_2 for typical first wall tiles.

temperature. This indicates that D was retained at deeper depth compared to that in erosion dominated area. As discussed in another paper [13], the peak shift of D_2 toward higher temperature corresponds to C–D formation with higher binding energy. This would be induced by the D implantation at higher temperature or by low D concentration over a wide area in the tile. Since hydrogen retention in boron at the wall temperature of JT-60U is very small, D injected into the boron layers would diffuse toward the surface and/or depth through open pores to allow deeper penetration in the deposition dominated area compared to that in the erosion dominated area.

The hydrogen isotope retention profiles derived from the present study are summarized in Table 1. Typical hydrogen isotope retentions in the divertor area were also shown for comparison. It should be noted that the largest hydrogen isotope retention was observed in the upper area of the first wall, i.e. erosion dominated area, and the D/H ratio reached almost 1.0, significantly high compared to that in the erosion dominated outer divertor area of only 0.08.

According to these experimental results, hydrogen isotope distribution can be discussed. Since 5Md4, 5Af4, 5Id4 and 5Lb4 were subjected to boronization using $B_{10}H_{14} + D_2$, one would expect a significant amount of D to be retained in the boron layers. Nevertheless, the hydrogen isotope profiles and retentions for these samples were very similar to those for 13Lg6, which was subjected to boronization using only $B_{10}H_{14}$ and exposed to total of 5250 DD shots and 2400 HH shots. In a separate experiment, D retained in the boron layers during boronization was found to be replaced with D from DD discharging plasmas after ~500 DD discharges [17]. Therefore, we believe most of D retained in the first wall reflects the D distribution by DD discharges, and the influence of D that remained in the boron layers during boronization must be very small.

Table 1
Comparison of H + D retention and D/H ratios in JT-60U (one column)

Location	H + D retention (10^{20} atoms/m^2)	D/H ratio
5Id4	10.4	0.80
5Lb4	10.7	0.69
13Lg6	6.2	1.02
5Md4	8.9	0.89
5Af4	9.3	0.94
Inner divertor	8.4	0.31
Outer divertor	4.1	0.08

As already mentioned, the upper area of the first wall in JT-60U was mainly eroded, while the bottom area of the inboard first wall was deposition dominated. In addition, referring to the plasma geometry or shape, 5Id4, 5Lb4 and 13Lg6 were placed nearest to the plasma. Hence plasma surface interactions on the surface of 5Id4, 5Lb4 and 13Lg6 would be stronger than those of 5Md4 and 5Af4 located relatively far from the plasma. This suggests that 5Id4, 5Lb4 and 13Lg6, on which even boron layers remained, are likely eroded by energetic particles from the plasma. By removing the boron layers, a porous structure, typical of the original tile structure, clearly appeared as seen in SEM micrographs and the large retention of hydrogen isotopes, especially deuterium, was observed in these three samples. Since the temperature of the tile surface was kept at around 573 K during discharges, the diffusion of hydrogen isotopes would be prohibited and they were kept near the surface. This causes total hydrogen isotope retention in the near surface region to be high even though they were at the eroded area. In the case of the deposition dominated area, the carbon was redeposited on the boron layer and some mixture of these layers would be formed. However, the flux of energetic hydrogen isotopes from the plasma would be low and the density of the carbon deposition layer would be also low compared to the graphite matrix. Therefore, hydrogen isotopes could easily move down and low concentration of hydrogen isotopes would be extended throughout the carbon deposition layer and the boron layer. This would cause the TDS peak to broaden.

It should be mentioned again that compared to the divertor tiles, a rather high degree of hydrogen isotope retention was observed in the near surface of the first wall tiles. We have attributed this to the direct implantation of high-energy deuterium originating from NBI (average power ~9 MW) into the first wall. In addition, the lower temperature of the first wall than that of the divertor tiles would prohibit desorption and/or replacement of the implanted D by subsequent D or H impingement.

4. Conclusions

Retention of hydrogen isotopes in five selected tiles used as the first wall of JT-60U was studied by means of SIMS and TDS. The surface morphology and erosion/deposition profiles were also examined by SEM and XPS. It was found that the upper area was mainly eroded, while the bottom area of the inboard first wall was deposition dominated. In contrast to the divertor area, where hydrogen isotopes were mainly retained in the carbon redeposited layers, the hydrogen isotope retention was larger in the erosion dominated area than in the deposition dominated area. The concentration of hydrogen isotopes in the first wall surfaces was a little higher than that in the divertor area as was the D/H ratio. This indicates that higher energy deuterium implantation into the first wall owing to the ripple/orbital loss of NBI deuterium should have a large contribution to the retention during DD discharges. Such deuterium must be implanted rather deeply. Furthermore, the lower temperature of the first wall (~573 K) compared to that of the divertor tiles could also make it hard to desorb deuterium or replace it by H.

Acknowledgement

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