

Deuterium and Hydrogen Retention Properties of the JT-60 and JT-60U Divertor Tiles

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The retention properties and depth profiles of deuterium and/or hydrogen in the graphite tiles located in the divertor region used in the W-shaped divertor of JT-60U with inner side pumping and JT-60 were investigated by thermal desorption spectroscopy (TDS) and secondary ion mass spectroscopy (SIMS). Total amount of retained hydrogen in the samples used in JT-60 and hydrogen and deuterium in JT-60U was proportional to the thickness of the re-deposited layers. Assuming homogeneous hydrogen distribution in the re-deposited layers and carbon density of 0.9 g/cm^3 , H/C in the re-deposited layers was determined to be approximately 0.03 from this slope of the linear relationships. This value is almost the same as that observed in the tiles of JT-60U and is much smaller than those observed in JET and other devices. The re-deposited layers are very likely subjected to high temperatures because of their poor thermal contact to the substrate owing to their porous structure.

Keywords: hydrogen, deuterium, retention, re-deposited layer, depth profile, JT-60, JT-60U

I. Introduction

The accumulation of tritium in in-vessel components of thermonuclear fusion devices is a major safety concern. Therefore, it is very important issue to evaluate retention characteristics of hydrogen isotopes (T, D, H) in in-vessel components of current plasma devices [1-7]. We have been studying the retention of hydrogen isotopes in plasma facing graphite tiles exposed to both deuterium and hydrogen discharges in JT-60U by using various methods under the joint research between Japanese universities and JAERI [8-11]. We have found that most of tritium produced by the D-D

reactions was implanted into depth of micrometer range, and tritium areal distribution was independent of carbon deposition profiles, while H and D seem to be very much influenced by the carbon deposition as well as the surface temperature of the tile. Although most of the tiles used in JT-60 were covered with re-deposited layer with the maximum thickness of $70 \text{ }\mu\text{m}$, for JT-60U erosion is dominant in the outer divertor region and redeposition in the inner divertor region [12].

In this study, the retention properties of deuterium and/or hydrogen in the graphite tiles located in the divertor region used in the

JT-60U and JT-60 were investigated by TDS. The depth profiles of hydrogen and/or deuterium retained in the graphite tiles were also investigated by SIMS and Elastic Recoil Detection (ERDA).

II. Experimental

II.A Samples

Fig.1 shows the cross sectional view of the vacuum vessel of JT-60 with an open divertor structure and lower X-point configuration (a), and sampling positions in three divertor tiles made by isotropic graphite (ETP-10, Ibidem) (b). All tiles were exposed to total of 1800 hydrogen discharges with the limiter configuration including 300 divertor discharges during January 1988 to October 1988. Most of the tiles were covered with re-deposited layers of the maximum thickness of 70 μm [13]. Operational temperature of the vacuum vessel and the base temperature of the tiles were kept at 573 K. However, the surface temperature of the tile rose especially at the position where frequency of the strike point hit for total of 300 divertor discharges was large [14].

For TDS analysis, twelve samples (8mm \times 8mm \times 1mm) were cut from the tiles to include the surface area; six from the inner strike region (Sample 1~6), three from the private flux region (Sample 7~9), three from the outer strike region (Sample 10~12). One sample from which re-deposited layers were completely removed (referred as a removed sample) was also analyzed by TDS to compare hydrogen retention in the re-deposited layers and that in the substrate. Two samples (Sample D and E) were analyzed by SIMS. Hydrogen depth profile in the re-deposited layer of a tile located in the private flux region (similar position to

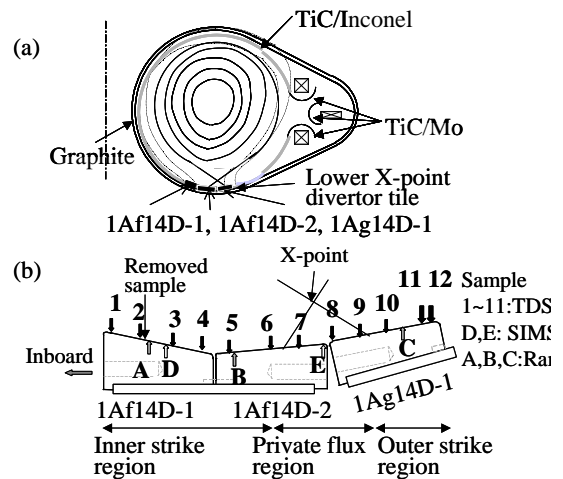


Fig.1 Cross-sectional view of the vacuum vessel of JT-60 (a) and sampling positions (b).

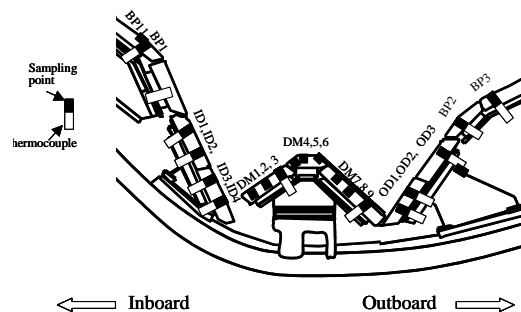


Fig.2 Cross-sectional view of the divertor region in JT-60U and sampling position

sample E) during different hydrogen discharge campaign was also determined by ERDA.

Fig.2 shows the cross sectional view of the JT-60U W-shaped divertor. The W-shaped divertor was pumped from the inner slot by cryopumps. All tiles received 3600 shots of deuterium discharges during two discharge campaigns from June 1997 to October 1998. After each of two-deuterium discharge periods, a hydrogen discharge experiment, of totally 700 shots, was carried out for detritiation before air ventilation. Among the 3600 DD discharges, about 300 shots of NBI heating were performed with the output range of 14–23 MW. In these discharges, accordingly, the surface temperature of the divertor tiles rose above 1300K [5]. The NBI power for HH discharges was about half

compared to that for the DD discharges. Hence, the temperature increase during the HH discharges would be much less than that during the DD discharges. The operational temperature of the vacuum vessel and the base temperature of the tiles were also ~573 K. Boronization was conducted twice during these campaigns. Thirteen thermocouples were embedded in the divertor tiles 6 mm beneath the tile surface, to monitor the tile temperatures during the discharge.

Samples (5mm × 10mm × 1mm) for SIMS measurements were cut from the tile surface area just above the thermocouples; four samples (ID1~ID4) from the inner divertor tiles, three (OD1~OD3) from the outer divertor tiles, four (BP1, BP11, BP2, BP3) from the baffle plates, and nine (DM1~DM9) from the dome units were prepared (see Fig.2). TDS analysis was conducted for six samples (ID1, 2, 3, OD1, 3, DM5, BP1, 3). Sample size was 8mm × 8mm × 0.5mm. For ID1, an additional measurement after removing the re-deposited layers was also conducted.

II.B Analysis of the sample

Deuterium and/or hydrogen retention properties were investigated by using a TDS apparatus. The sample was put on a sample holder and heated from room temperature to 1273 K with a constant ramp rate of 0.42 K/s, and then was held at 1273 K for 10 min. Scanning mass range was from 1 to 55 amu. For the JT-60 tiles exposed to only hydrogen discharges, the dominant mass numbers observed by a Quadrupole Mass Spectrometer (QMS) during the heating were 2(H₂), 15(CH₄), 26(C₂H₄), 30(C₂H₆) and 39(C₃H₈). For the JT-60U tiles exposed to deuterium discharges following hydrogen discharges, the dominant

mass numbers were 2(H₂), 3(HD), 4(D₂), 15(CH₄), 19(CD₃H), 20(CD₄), 26(C₂H₄), 30(C₂H₆), 32(C₂D₄), 36(C₂D₆) and 39(C₃H₈). Here, the fragment ions of CH₄, C₂H₄ and C₃H₈ were used to evaluate the amount of retained H because the parental (main) ions of these gases overlap with other fragment ions.

Depth profile of hydrogen concentration within the depth of 0.3 μm, i.e., less than the thickness of the re-deposited layer, was conducted for only one sample (JT-60 sample) by ERDA. Helium ions with energy of 1.7 MeV were used as a primary beam. Depth profiles of hydrogen, deuterium, boron and oxygen retained in the graphite tiles were analyzed by SIMS using 5 keV Cesium ions (Cs⁺) as a primary beam. The sputtering rate by Cs⁺ ion was approximately 1 μm/h. The beam of 32 μm in diameter was rastered over 400 × 400 μm² area. In order to eliminate the effect of crater edges, the negative secondary ion was collected only from the center of the rastered area. The ion intensities of H and D were normalized by that of ¹²C for comparison.

The thickness of the re-deposited layers was estimated by Scanning Electron Microscope (SEM). Since the tile have rough surface, the measured thickness of the re-deposited layer has an error of 1 μm.

III. Results

III.A. Hydrogen retention of graphite tiles used in JT-60

The most dominant species desorbed from the samples was H₂ molecule. Hydrocarbons such as CH₄, C₂H₄, C₂H₆ and C₃H₈ were also desorbed as minor desorption species. Fractions of H₂ gas to the total amount of hydrogen atoms retained in the samples are approximately

80~70 %. Desorption peaks of CH₄ and H₂ appeared around 750 K and 973 K, respectively, and desorption rates of CH₄ and H₂ increased with the thickness of the re-deposited layers. Fig.3 shows the total amounts of hydrogen desorbed from the sample as a function of the average thickness of the re-deposited layers. The total amount of retained hydrogen in the samples were $1.38\sim 10.9 \times 10^{22}$ H/m². The amount of hydrogen desorbed from the removed sample was quite small compared to those from the samples with the re-deposited layers, indicating that hydrogen was mainly retained in the re-deposited layer. In addition, one can be note that the total amount of the retained hydrogen in the samples shows good linear relationship with the thickness of the re-deposited layers except one sample. This indicates hydrogen was retained in the re-deposited layers uniformly with a constant hydrogen concentration irrespective of their thickness or poloidal position. From the slope of this straight line, we can determine hydrogen concentration in the re-deposited layers as 1.3×10^{21} H/m²/1 μ m. Assuming homogeneous hydrogen distribution in the re-deposited layers and carbon density of 0.90 g/cm³, H/C ratio in the re-deposited layers is ~ 0.03 . Here, the carbon density of the re-deposited layer was measured by using both SEM and microbalance [15]. This H/C ratio is much smaller than those observed in JET [2,4,6] and other low temperature operating machines [6,4,7] where H/C is 0.1 ~ 0.4. Fig.4 shows depth profiles of SIMS represented by signal intensity ratios of H/¹²C for the inner divertor tile (sample D) and the private flux region tile (sample E) within the depth of 1.7 μ m. Depth profiles of ERDA represented by H/C ratio of the private flux

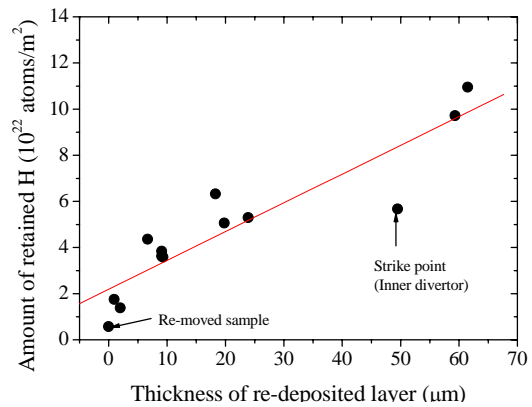


Fig.3 Total amounts of hydrogen desorbed from the samples as a function of the thickness of the re-deposited layers.

region tile were also shown for comparison. The thickness of the re-deposited layers of D and E samples were 40 and 1 μ m, respectively. The surface concentration was higher for the sample E, but it decreased exponentially with the depth, while the concentration of the sample D having the thick re-deposited layers kept rather constant with the depth. This again confirms the above discussion that the hydrogen concentration in the re-deposited layers is constant. As shown in Fig.4, in the tile located in the private flux region, where was covered with only thin re-deposited layers, H/C ratio within the depth of 300 nm was observed to be ~ 0.05 by ERDA, while H/C ratio measured by TDS was ~ 0.031 . Thus the surface hydrogen retention for the tiles which was not covered

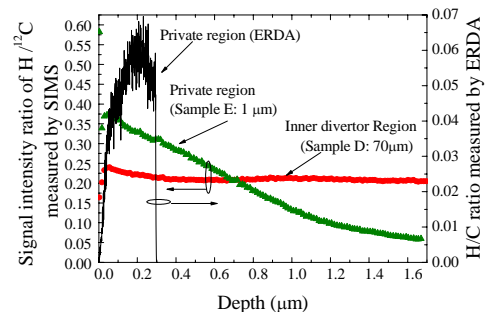


Fig.4 Depth profiles of H/¹²C of sample D and E measured by SIMS and that of H/C ratio measured by ERDA

with the thick re-deposited layers, was rather high but hydrogen did not diffuse into the depth and decreased rapidly with the depth. Assuming $H/^{12}C$ ratio measured by SIMS proportional to H/C by ERDA, the H/C of the sample D was roughly 0.036 which was almost the same as that measured by TDS.

III.B. Hydrogen and deuterium retentions of graphite tiles used in JT-60U

It was reported that the outer divertor tiles were mostly eroded (up to 20 μm) while the inner divertor tiles were covered by deposited layers with maximum thickness of 60 μm and on the dome top tile no significant deposition or erosion was observed [12]. Fig.5 compares typical depth profiles of $H/^{12}C$, $D/^{12}C$ and $(H+D)/^{12}C$ signal intensity ratios for (a) erosion dominated region of the outer divertor tile (OD3), (b) deposition dominated region of the inner divertor tiles (ID3) and (c) the plasma facing dome top tile (DM5). One can see that the $H/^{12}C$ ratio near the surface was approximately one order of magnitude larger than that of $D/^{12}C$ indicating effective replacement of deuterium retained in near surface regions. In addition, the ratio of $H/^{12}C$, $D/^{12}C$ and $(H+D)/^{12}C$ of the sample ID3 with thick re-deposited layer was one order of magnitude smaller than those of OD3 and DM5 samples and kept rather constant with the depth. Similar result was observed in the tile used in JT-60 (see Fig.4).

Fig. 6 shows thermal desorption spectra of H_2 , HD, D_2 and CH_4 for the ID3 sample covered by the re-deposited layers with the thickness of 60 μm . In addition to those dominant species, various hydrocarbons such as CD_4 , CD_3H , C_2H_4 , C_2H_6 and C_3H_8 were also desorbed as minor

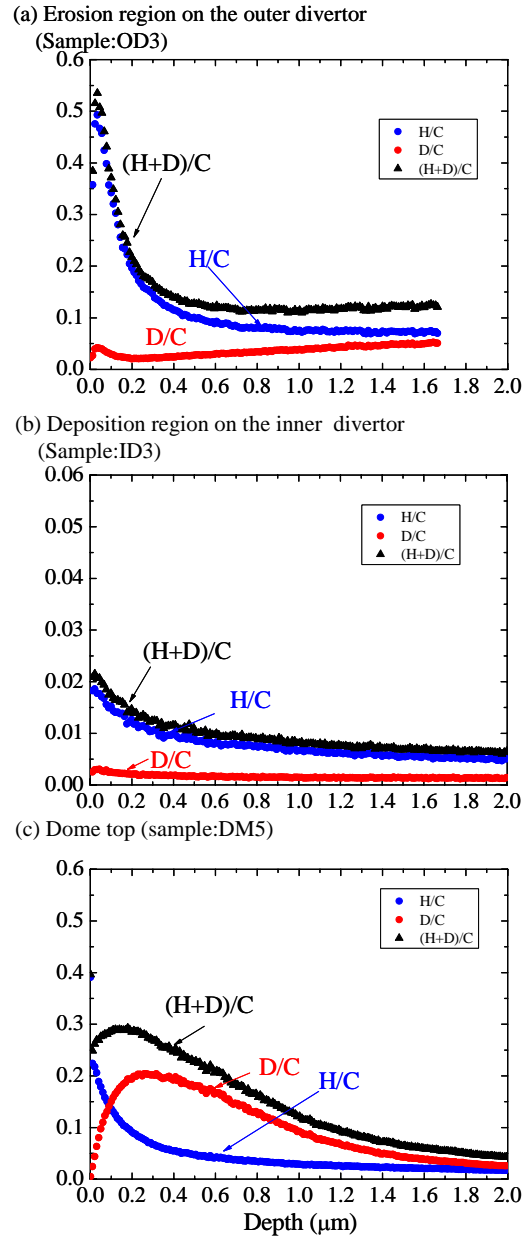


Fig.5 Typical depth profiles of signal intensity ratios of $H/^{12}C$, $D/^{12}C$ and $(H+D)/^{12}C$ for (a) erosion dominated region of the outer divertor, (b) the deposition dominated region of the inner divertor region and (c) the plasma facing dome top tile.

desorption species. Main desorption peak of H_2 appeared around 1070 K, while the desorption of HD and D_2 were peaked at higher temperatures than that of H_2 , reflecting the deeper D retention profile (see Fig.5). Desorption of CH_4 was peaked around 700 K. The total retained amount (retention) of H+D

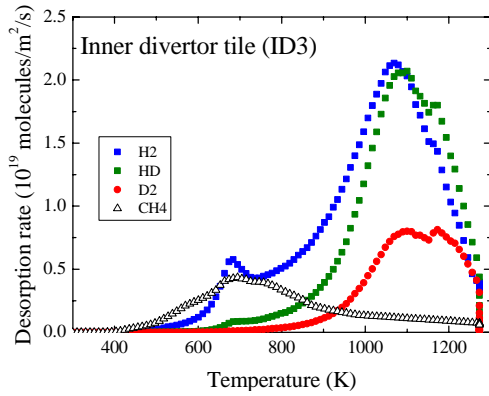


Fig.6 TDS spectra of H₂, HD, D₂ and CH₄, CHD₃ and CD₄ desorbed from the inner divertor tile covered by the re-deposited layer with the thickness of ~60µm.

was determined by integrating all mass spectra considering the number of H and D atoms included and their sensitivity. About 60% of the retained hydrogen was desorbed as hydrogen molecules (H₂, HD, D₂) and the remaining 40% as hydrocarbon molecules.

Fig. 7 compares the hydrogen retention for all samples including ID1, the sample after the removal of the re-deposited layer (named as the "removed sample"). The amount of retained hydrogen isotopes is clearly correlated with the thickness of the deposited layers (from 2.3×10^{22} to 1.3×10^{23} atoms/m²), i.e., ID3 from the inner divertor tiles covered with deposited layers of 60 µm in thickness [12] showed the largest retention, and OD1 from the outer divertor tiles with the erosion depth of about 20 µm showed the smallest. The retention of the samples from the inner baffle plate was also larger than that of the outer baffle plate. The total H+D retention in the dome top tile was comparable to those of OD3 and BP3. It is also important to note that the total H+D retention of the inner divertor tile increased with the thickness of the re-deposited layers (see Fig.8). The outer divertor tiles showed much less retention and OD1, the most

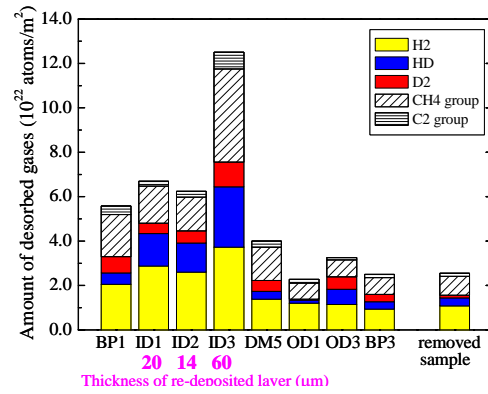


Fig.7 Amount of H₂, HD, D₂ and CH₄-group and C_{2,3} groups desorbed gases from the tiles.

eroded tile, showed the smallest retention, nearly the same as that of the removed sample. These results clearly indicate that hydrogen isotopes are mainly retained in the re-deposited layers. In Fig. 8, the total H+D retentions in the inner divertor tiles are plotted as a function of the thickness of the re-deposited layers. One can see very good linear relationship as already noted in the HH tiles (see Fig.3). From the slope of the line i.e., 1.45×10^{21} H+D/m²/1µm, (H+D)/C ratio in the re-deposited layers was determined to be 0.032 ± 0.003 . Here measured carbon density of the re-deposition layer (0.91 g/cm^3) was used. This value is the same as that

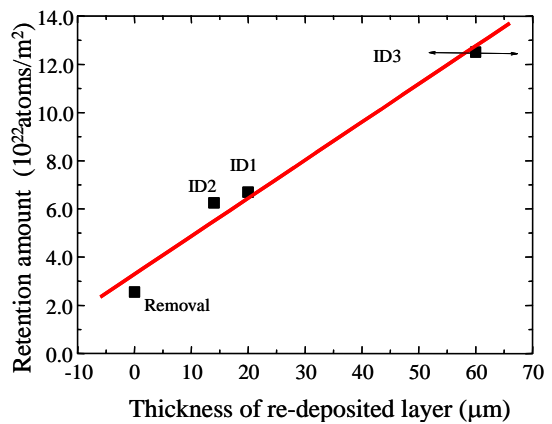


Fig.8 Total amount of retained H and D atoms in the inner divertor tiles vs. thickness of re-deposited layers

observed in the divertor tiles exposed to the HH discharges in the JT-60 (H/C \sim 0.03) and much smaller than those observed in JET and other devices [2,4,6,8,16]. Such low retention is most likely owing to the temperature increase of the divertor tiles during the discharge as discussed later. It is also important to note that the straight crosses at line crosses at $-20 \mu\text{m}$ in the thickness of the re-deposited layers when the linear relationship is extrapolated to zero concentration, in Fig. 8. This clearly indicates that the substrate also retained hydrogen isotopes, as evidenced by the desorption of D containing gases in TDS spectra of the removed sample (see Fig.7). A similar result was observed in the tiles exposed to only hydrogen discharges in JT-60 (see Fig.3).

Fig. 9 compares the atomic ratio of retained D and H (D/H) for all samples. Interestingly, D/H for most samples were nearly 0.4 except for OD1 of 0.09. In more detail, D/H seems to correlate to the plasma loading history of the samples, i.e., D/H for the inner divertor tiles increased (0.38~0.45) towards the most common position for the divertor strike point during DD discharges [12].

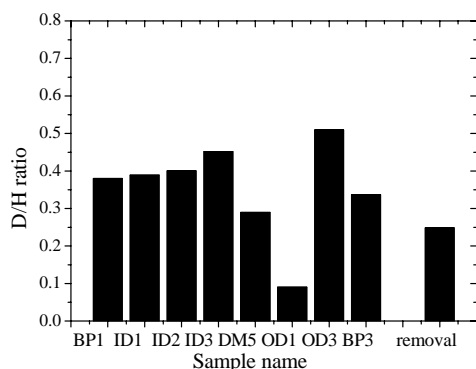


Fig.9 Atomic ratio of retained D and H (D/H)

IV. Discussion

As appeared in Fig. 7 and 8, the H+D retention was different with sample positions and clearly correlated to the thickness of the deposition layers, showing nearly constant concentration of 0.03 in (H+D)/C ratio in the re-deposited layers which was the same as that observed in the tile exposed to only HH discharge in JT-60 (see Fig.3) and is much smaller than those observed in JET and other devices. In JT-60U, the operational temperature and the base temperature of the tiles were kept at 573 K, which was higher than other devices. In addition, the poor thermal contact of the deposition layers could increase their temperature [12].

The smallest retention was observed for OD1 which was eroded with the thickness of $20 \mu\text{m}$ [13], because it was hit most frequently by the divertor strike points and experienced the highest temperature of $\sim 1373 \text{ K}$ [5] during DD discharges. This means that D dynamic retention is limited by both high heat load and erosion of the tile during DD discharges. But the H retention shows no appreciable difference among OD3, BP3 and the removed sample as shown in Fig. 3. Thus, H retention during the HH discharge plays an important role for the consideration of hydrogen isotope retention.

Interestingly, the D/H ratio was almost the same, 0.4 ± 0.1 irrespective of the tile position except the OD1. On the other hand, the ratio of the number of the DD discharges and the HH ones were around 5. If the retention characteristic is not different in both discharges, the D/H ratio is much higher (~ 5.0) than measured value (~ 0.4). If the amount of hydrogen retained in the removed sample is the same as that of substrate of all samples, D/H

ratio shall become large (see Fig.7), but still these D/H ratios are considerably smaller than 5.0. This indicates that the tile temperature might be high during DD discharges. Still the D/H of 0.4 is too low, even the DD discharges contained around 10% of H. This suggests that a part of the D retained during the DD discharges was replaced by H during the HH discharges. In addition, the HH discharges very likely add the H dominated re-deposited layers on the D dominated deposition layers on the inner divertor tiles. According to rough estimation, the HH discharges likely produced a few μm of deposition layers, though the thickness may vary in the tile position. In addition, the lower temperature during the HH discharges could result in higher H/C than D/C in the DD deposited layers. The depth profiles well reflect such retention profiles of H and D. Correspondingly, the desorption peak temperatures of HD and D₂ are higher than that of H₂. Thus the hydrogen retention in the JT-60U or JT-60 divertor tiles reflects the history of plasma exposure and/or temperatures. In this respect, one should be very careful for estimation of tritium retention from data obtained at lower temperatures.

V. Conclusions

Deuterium and/or hydrogen retention in the graphite tiles located in the divertor region of JT-60 and JT-60U were investigated by TDS and SIMS. The H+D retention in the re-deposited layers on the tiles used in JT-60U was ~ 0.03 in (H+D)/C ratio, which was the same as that observed in the tile of JT-60 and is much smaller than those observed in JET and other devices. That is because, the temperatures of the JT-60U divertor tiles during NBI heated

DD discharges rose above 700K. The poor thermal contact of the deposition layers to the substrate could increase their temperature to reduce their hydrogen retention further. The important role of temperature in reduction retention is also supported by less D retention in the sample located in the erosion-dominated region, which was clearly heated up above 1300K during the NBI heated DD discharges.

Thus, the present results indicate that tritium retention in the re-deposited carbon layers in the ITER first wall could be reduced significantly comparing the current estimation based on the JET results, if the surface temperature could be substantially higher than 573 K.

Acknowledgement

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