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# Correlation between hydrogen isotope profiles and surface structure of divertor tiles in JT-60U

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## Abstract

The present paper is devoted to depth profiles by secondary ion mass spectroscopy (SIMS) of hydrogen/deuterium in tiles taken from the dome unit area of JT-60U. This information is correlated with surface features, particularly from the aspect of erosion and deposition, determined by scanning electron microscope (SEM) and X-ray photoelectron spectroscopy (XPS). The outer divertor-facing surface was mostly covered by re-deposited layers a maximum of 10 μm thick, while the inner divertor-facing side was eroded. The deposition profile is opposite to the observation for the divertor area in most tokamaks that the outer divertor side is eroded, while the inner deposited. However, H + D retention was higher for the deposited layers than that for the eroded area. Nevertheless, hydrogen retention seems very small and showed no appreciable effects on C1s spectra of XPS compared to the constituent elements boron and oxygen.

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## 1. Introduction

The understanding of tritium and deuterium behavior in in-vessel components is critical for fusion reactor safety evaluation [1–4]. Recently, plasma-facing graphite tiles exposed to DD discharges in JT-60U at the Japan Atomic Energy Research Institute (JAERI) have been available for research studies for PMI and hydrogen and deuterium/tritium retention [5]. The hydrogen isotope behavior in the tiles during the inner side pumping have been investigated by the tritium imaging plate technique [6–9], secondary ion mass spectroscopy (SIMS), X-ray photoelectron spectroscopy (XPS) and scanning electron microscopy (SEM) [10–12]. These studies demonstrated that tritium, due to its high energy origin, was implanted rather deep by the orbital loss and ripple loss mechanisms [6–9], while hydrogen/deuterium profiles in divertor areas were complicated and very much influenced by surface temperature and erosion/deposition [10,11,13].

The present paper is devoted to depth profiles of hydrogen/deuterium in tiles taken from the dome unit area of JT-60U, determined by SIMS, and surface examination of erosion and deposition, using SEM and XPS. For tiles used in the divertor area, similar studies are reported in a separate paper in this conference [14].

## 2. Experimental

Fig. 1(a) shows the cross section of the W-shaped divertor of JT-60U. Detailed structure of the dome area is shown in Fig. 1(b). The dome units are covered by three carbon tiles. Two tiles at the dome top (5DM-1bp) and the outer wing (5DM-1cp) were made of carbon

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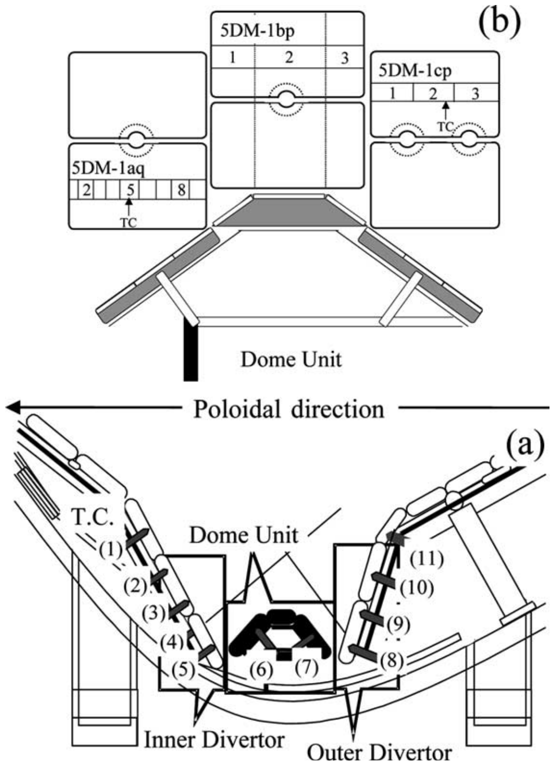

Fig. 1. (a) Cross section of the W-shaped divertor of JT-60U and (b) detailed structure of carbon tiles in the dome area and the area of samples examined.

fiber composites (CX-2002U) and the other tile of isotropic graphite (5DM-1aq). Sample disks (about $10\times10\times1$ mm$^3$) were cut from these tiles and numbered as indicated in the figure.

All tiles were subjected to total of 6700 DD discharges, 4300 for inner side pumping and the remainder for both side pumping. 350 HH discharges were also performed after the DD discharges to remove tritium accumulated in the tiles. Boronization was performed three times during this discharge period. The temperature of tiles was measured by thermocouples embedded 6 mm beneath the surface as indicated in Fig. 1(a) and was 650 K at the maximum (Ave.: 600 K). However, surface temperature of the tiles would be much higher.

The depth profile of hydrogen isotopes retained in the samples was analyzed by SIMS (ULVAC-PHI ADEPT1010 Dynamic SIMS System), using cesium ion (Cs$^+$) as the primary ion with an energy of 5 keV and a beam current of 100 nA at 45° to the surface normal. The beam size was about 32 μm and the rastering area was set at $400\times400$ μm$^2$. Depth profiling with a profilometer showed the sputtering rate to be around 1 μm h$^{-1}$.

The XPS measurements were also performed for surface characterization (ULVAC-PHI ESCA5500MC system), using a Mg-Kα 400W X-ray source and a concentric hemispherical analyzer. The depth profile was also analyzed by Ar ion (Ar$^+$) sputtering with an ion energy of 4.0 keV and a raster size $1\times1$ mm$^2$ at 30° orientation to the surface normal. For XPS, the sputtering rate was nearly 10 nm min$^{-1}$ measured by the profilometer. To remove surface contamination, 6 min (corresponding to 60–70 nm) of Ar$^+$ sputtering was required for most cases. Nevertheless, charging up the sample during XPS analysis made the estimation of true chemical shifts very difficult [13]. Therefore, the chemical shifts were estimated by referring to the O1s peak assumed to originate from the carbonyl group at 531.4 eV [15].

## 3. Results

Fig. 2 shows SEM images of sample surfaces. On the inner divertor-facing side, no re-deposition layers were observed. The surface kept the original surface structure typical of IG-430U graphite as seen in Fig. 2(a) for the sample 5DM-1aq5. Fig. 2(b) for the dome top area (5DM-1bp2), which directly faced the plasma, also shows no re-deposition. The outer divertor-facing surface (outer wing tile, 5DM-1cp2), in contrast, shows a rather smooth structure with open pores (Fig. 2(c)), indicating the existence of a re-deposited layer. A separate measurement of the thickness has shown that most of the tile surface of the outer wing tile (5DM-1cp) was covered by re-deposition layers and the maximum of 10 μm thick layers appeared near the bottom edge. On the outer divertor-facing side of the dome top tile (inner wing tile, 5DM-1bp3), the re-deposition showing lamellar layers was also found but the thickness was smaller than that of the outer dome wing (5DM-1cp).

Fig. 3 compares depth profiles of H and D determined by SIMS for (a) the inner divertor-facing surface (5DM-1aq5), (b) the dome top (5DM-1bp2) and (c) the outer divertor-facing surface (5DM-1cp3). Since SIMS analysis does not give absolute amounts, signal intensities of negative ions of H and D were normalized to that of C for comparison.

At the inner divertor-facing surface (5DM-1aq5), with the eroded area, deuterium retention was quite low and almost nothing was retained in deeper area, while in the outer divertor-facing surface (5DM-1cp3) covered by the re-deposited layers, deuterium was retained with nearly constant amount. On the dome top area (5DM-1bp2), where there was no appreciable erosion nor re-deposition, the deuterium retention was highest at the surface layer (<0.2 μm) but clearly decreased with depth.

Hydrogen profiles were nearly the same for all samples, and the highest retention appeared at or very near the surface and sharply decreased to certain levels irrespective of the samples. Considering that hydrogen

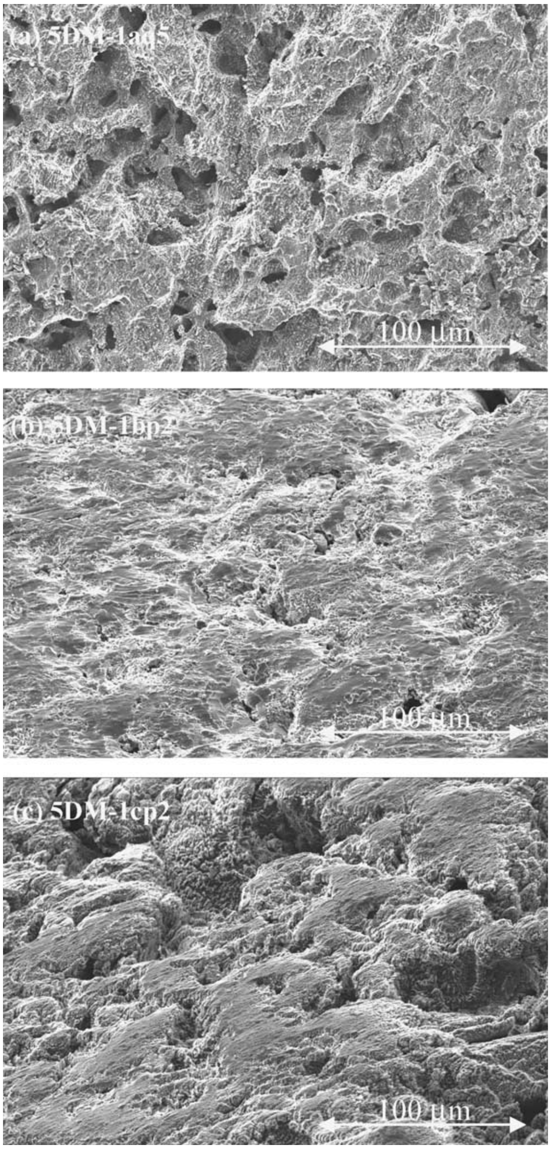

Fig. 2. SEM images observed for (a) 5DM-1aq5 (the inner divertor-facing side), (b) 5DM-1bp2 (the dome top area) and (c) 5DM-1cp2 (the outer divertor-facing side).

discharges were performed after the deuterium discharges and before ventilation to remove the accumulated tritium produced by D–D reactions, we conclude that most of deuterium retained near surfaces was replaced by hydrogen. Thus H + D profiles shown in Fig. 3 must correspond to deuterium retention during the deuterium discharges. As seen in Fig. 3, the highest retention was observed at the re-deposited area and the least at the eroded area.

Fig. 4 compares the integrated retention of H and D within the thickness of 1.7 µm obtained by integration of $H/^{12}C$ and $D/^{12}C$ SIMS signal ratios for all samples.

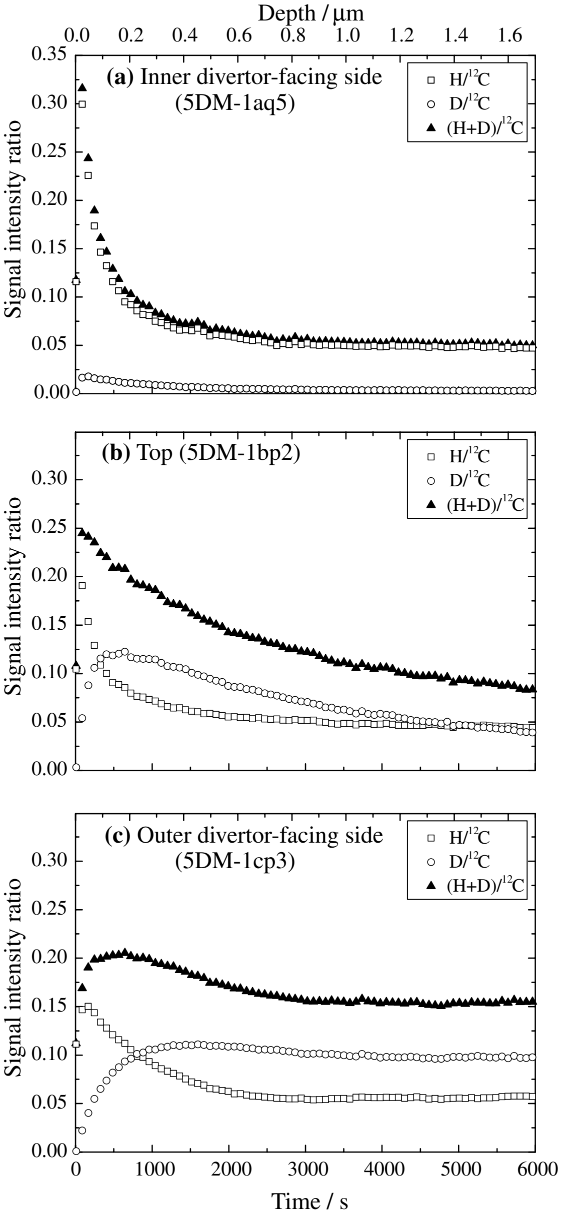

Fig. 3. Depth profiles of $H/^{12}C$, $D/^{12}C$, and $(H+D)/^{12}C$ signal intensity ratios determined by SIMS for (a) the inner divertor-facing surface (5DM-1aq5) with the eroded area, (b) the dome top (5DM-1bp2) where there was no appreciable erosion nor re-deposition and (c) the outer divertor-facing surface (5DM-1cp3) in the re-deposited area.

One can see that the H + D retention in the inner divertor-facing side was smaller than that in the outer divertor-facing side. This indicates that the retention of hydrogen isotopes on the re-deposited area is larger than that on the eroded area. The H retention in all samples was almost the same. This indicated that the hydrogen

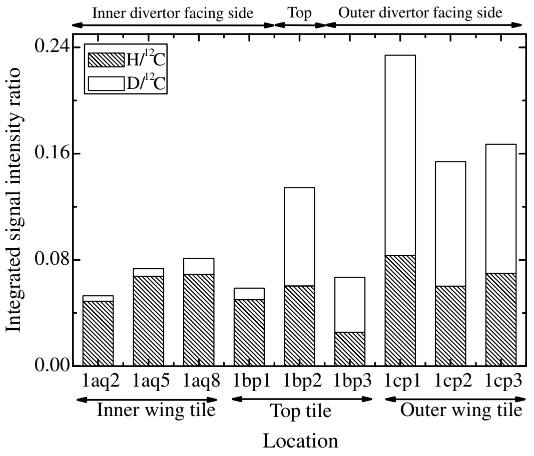

Fig. 4. Comparison of H and D retentions within 1.7 μm depth, given as the integrations of $H/^{12}C$ and $D/^{12}C$ signal ratios in the depth for all samples.

retention saturated at a certain levels irrespective of erosion or re-deposition surface.

In XPS measurements, B1s, C1s and O1s peaks were observed. Fig. 5(a) and (b) shows XPS spectra respectively for B1s and C1s observed after removal of the surface contaminants, i.e. $Ar^+$ sputtering for 6 min (corresponding to 60–70 nm). Before surface cleaning the B1s peaks for all samples showed single peaked structure, boron oxide at ~192 eV. After the surface cleaning, the B1s peaks split into two peaks as shown in Fig. 5(a) indicating the existence of a carbon-related peak at ~188 eV [16]. The atomic concentration of boron in the outer divertor-facing surface (~4%) was almost twice that in the inner divertor-facing surface. In contrast to boron, the atomic concentrations of oxygen in all samples was almost constant (~6%). Because there was little boron oxide, most of oxygen is probably bonded to carbon, forming some carbonyl groups.

The peak energy of C1s for all tiles was a little lower than that for cleaved surfaces of pyrolitic graphite (PG) and the outer divertor-facing surface shows slightly higher peak energy than that of the inner divertor-facing surface as shown in Fig. 5(b). The XPS spectra of all samples show that peak shapes were asymmetric with larger shoulder at lower energy side compared to PG. In XPS studies for hydrogen implanted graphite [17], it is known that the C1s peak energy first shifts toward the low-energy side due to damage formation induced by the implantation, and then turns upward with increasing hydrogen fluence.

## 4. Discussion

In the divertor area in most of tokamaks, the inner divertor is deposition dominated and the outer divertor erosion dominated [8]. However, observations in the dome area of JT-60U are opposite, i.e. the outer divertor-facing dome wing was re-deposition dominated and the inner divertor-facing one erosion dominated. Nevertheless H + D profiles show the higher retention on the re-deposited area, and the least on the eroded area, similar to other tokamaks. In addition, H + D profile of the eroded tile shows a sharp decay very near the surface, probably owing to the retreating surface by erosion. The H profiles are very similar for all tiles and penetrate rather deep with nearly constant concentration, suggesting pore diffusion. This indicates that mechanisms of hydrogen uptake or exchange to deuterium are similar for all dome tiles, or the re-deposition layers on the dome tile shows similar characteristics to hydrogen retention as the eroded surface.

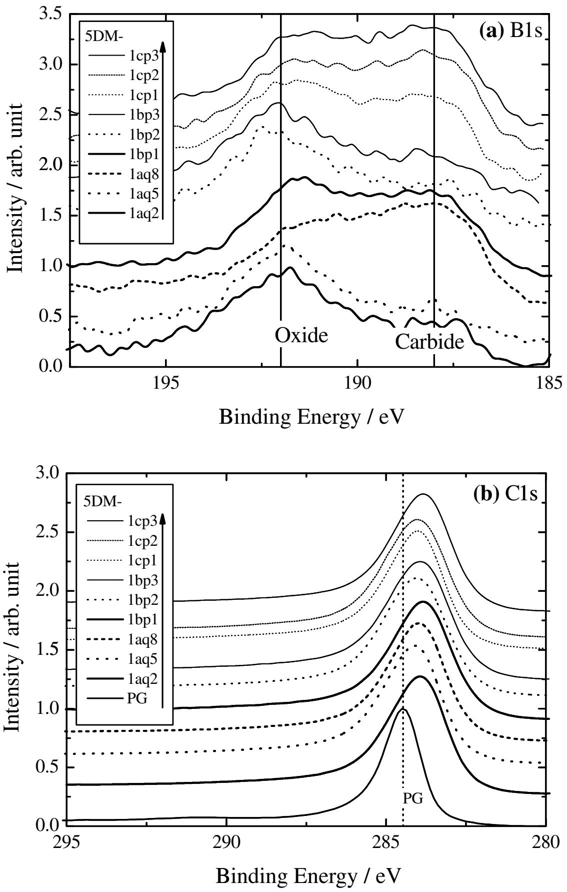

Fig. 5. Comparisons of XPS spectra of (a) B1s and (b) C1s observed on the inner divertor-facing (5DM-1aq2, 1aq5 and 1aq8), the dome top (5DM-1bp1, 1bp2 and 1bp3), the outer divertor-facing (5DM-1cp1, 1cp2 and 1cp3) tiles and Pyrolytic Graphite (PG) after $Ar^+$ sputtering for 6 min (removing 60–70 nm).

In a separate paper by Hirohata et al. [14] on H/D retention in divertor tiles, it was shown that the depth profiles of hydrogen isotopes were quite different for erosion dominated outer divertor tiles and re-deposition dominated inner divertor tiles [14]. In particular, retention of H and D in the deposited surface layers on the inner divertor tile was quite small even smaller than that in the erosion dominated outer divertor tile. Most probably, the poor adhesion or porous nature of the deposited layers on the inner divertor of JT-60U inhibited plasma heat load conduction to the substrate, resulting in temperature escalation of the deposited layers to release H/D. Actually, the depth profiles and retention of hydrogen isotopes for both divertors are inversely correlated to the heat load.

In contrast, the dome area is not likely loaded by a very high heat flux, and the temperature increment during shots must be smaller than that of the divertor tiles. Hence, hydrogen retention was higher at the re-deposited layers as observed in most tokamaks.

Recent NRA [18] and TDS [19] analysis shows the absolute (H + D)/C ratios in those tiles were less than 0.1, giving much less than those observed in other tokamaks, which is another indication that tiles were subjected to higher temperatures. In depth profiling of C1s in XPS, we found lower energy shifts with increasing depth (not shown). This seems to correspond to reduction of retained H + D in depth. However, taken such small amounts of hydrogen retention compared to boron + oxygen (about 10%) into account, these low energy shifts could be due to the removals of B and O. In other words, hydrogen retention in the samples was too small to give clear influence on XPS measurements.

## 5. Conclusion

To reveal the hydrogen isotope behavior in the graphite tiles used for the dome unit with two side pumping in JT-60U, depth profiles of hydrogen and deuterium retained in carbon tiles were analyzed by SIMS and the chemical states of C1s, B1s and O1s were evaluated by XPS. The existence of the re-deposition layers was examined by SEM.

It was found that the inner divertor-facing side of the dome unit was erosion dominated, while the outer divertor-facing side was covered by the re-deposited layers. This is opposite to the observations in divertor area of most tokamaks. Nevertheless, deuterium retention in the re-deposited layer on the outer divertor-facing side was the highest, and was the least at the eroded inner divertor-facing side as often observed for high hydrogen retention in re-deposited layers. The chemical shift of C1s XPS peak clearly correlated with hydrogen retention, but its FWHM did not, indicating the nature of tile surfaces very complex. The characteristics of the deposited layers for hydrogen seems quite depended on its temperature history, which controls the hydrogen retention.

In all tiles, deuterium on the top surface was completely replaced by hydrogen indicating that hydrogen discharges employed for detritiation seems to work well to exchange most of deuterium retained by forming C–D bonds in near-surface areas irrespective of the tile positions. However, deuterium deeply retained was not effectively replaced by hydrogen discharges, particularly for the dome top tiles which retained large amounts of deuterium.

## References

[1] G. Federici, R.A. Anderl, P. Andrew, et al., J. Nucl. Mater. 266–269 (1999) 14.
[2] G. Janeschitz, J. Nucl. Mater. 290–293 (2001) 1.
[3] J.P. Coad, N. Bekris, J.D. Elder, et al., J. Nucl. Mater. 290–293 (2001) 224.
[4] C. Stan-Sion, R. Behrisch, J.P. Coad, et al., J. Nucl. Mater. 290–293 (2001) 491.
[5] N. Miya, these Proceedings. doi:10.1016/j.jnucmat.2004.04.219.
[6] T. Tanabe, K. Miyasaka, K. Sugiyama, et al., Fusion Sci. Technol. 41 (2002) 877.
[7] K. Sugiyama, K. Miyasaka, T. Tanabe, et al., Phys. Scr. T 103 (2003) 58.
[8] T. Tanabe, N. Bekris, P. Coad, et al., J. Nucl. Mater. 313–316 (2003) 478.
[9] K. Masaki, K. Sugiyama, T. Tanabe, et al., J. Nucl. Mater. 313–316 (2003) 514.
[10] Y. Hirohata, Y. Oya, H. Yoshida, et al., Phys. Scr. T 103 (2002) 15.
[11] Y. Oya, Y. Hirohata, Y. Morimoto, et al., J. Nucl. Mater. 313–316 (2003) 211.
[12] Y. Gotoh, J. Yagyu, K. Masaki, et al., J. Nucl. Mater. 313–316 (2003) 370.
[13] Y. Oya, Y. Morimoto, M. Oyaidzu, et al., Phys. Scr. T108 (2004) 57.
[14] Y. Hirohata, Y. Oya, H. Yoshida, et al., these Proceedings. doi:10.1016/j.jnucmat.2004.04.031.
[15] S. Gardner, C. Singamsetty, G. Booth, et al., Carbon 587–595 (1995) 587.
[16] W. Cermignani, T. Paulson, C. Onneby, et al., Carbon 33 (1995) 367.
[17] Y. Gotoh, Fusion Technol. 6 (1984) 424.
[18] T. Hayashi et al., private communications.
[19] T. Shibahara et al., private communications.