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Hydrogen isotope behavior in in-vessel components used for DD plasma operation of JT-60U by SIMS and XPS technique

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Abstract

The behavior of hydrogen and deuterium retained in the graphite tiles placed in the dome top unit, the outer divertor unit and the outer baffle plate of JT-60U with W-shaped divertor were investigated by secondary ion mass spectroscopy and X-ray photoelectron spectroscopy (XPS). It was found that deuterium on the surface of the tiles was replaced by hydrogen due to hydrogen discharges at the final stage. The amount of deuterium on the dome top unit largely depends on the location of the dome unit. Especially, the amount of that on the inner side and the center of the dome top units were high compared with that on the outer side. The binding energies of C 1s and B 1s peaks on the surface of the inner side of dome top unit were largely shifted to higher energy by XPS. These results indicate that hydrocarbons and boron oxide might be formed on the surface. It is concluded that the behavior of hydrogen and deuterium and the chemical state of graphite were influenced by the temperature of the surface, the position of the strike point and the flux of the particles during the operation.

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1. Introduction

The understanding of tritium and deuterium behaviors in in-vessel components is one of the most critical issues for a future fusion reactor on a viewpoint of safety evaluation [1–4]. Quite recently, plasma facing graphite tiles used in JT-60U at Japan Atomic Energy Research Institute (JAERI) become available for various investigations. Tritium retentions on graphite tiles used in JT-60U were analyzed by the tritium imaging plate technique, and it was clear that the highest tritium level was found at the center of the dome top unit or the private region and the outer baffle plates, where the plasma did not hit but the distance from the plasma was the shortest [5]. However, the retention of the other hydrogen isotopes, namely hydrogen and deuterium, were not clarified sufficiently and it is important to understand

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the behavior of the hydrogen isotopes in plasma facing materials for the investigation of plasma wall interaction [6].

In this study, the depth profiles of hydrogen and deuterium retained in the graphite tiles placed in the dome top unit, the outer divertor unit and the outer baffle plate, which were used for ~3600 deuterium plasma (DD) discharges with deuterium neutral beam injection, followed by ~700 hydrogen plasma (HH) plasma discharges for about one year operation in JT-60U, were investigated by secondary ion mass spectroscopy (SIMS). The chemical states of graphite, boron and oxygen were also studied by X-ray photoelectron spectroscopy (XPS).

2. Experimental

Fig. 1 depicts the cross-section of the divertor region in JT-60U with W-shaped divertor. Thirteen thermocouples were installed in the divertor regions, monitoring the tile temperatures during discharge experiments. The position of the thermocouples was 6 mm under the tile surface. The graphite tiles used in this study were isotopic graphite (IG-430U) and carbon fiber composites (PCC-2S and CX-2002U), which were used for the outer baffle plates, the dome top unit and the outer divertor unit. All tiles were exposed to ~3600 shots of DD discharges for the June 1997–October 1998 experimental period. The experimental conditions during the DD plasma discharge were as follows: the W-shaped divertor was used, and the vacuum vessel was evacuated from the inner divertor slot by cryopumps. Boronizations were performed two times during this experimental period [7,8]. After the DD plasma discharges periods, 380 and 320 shots of HH discharge were carried out to remove the surface tritium.

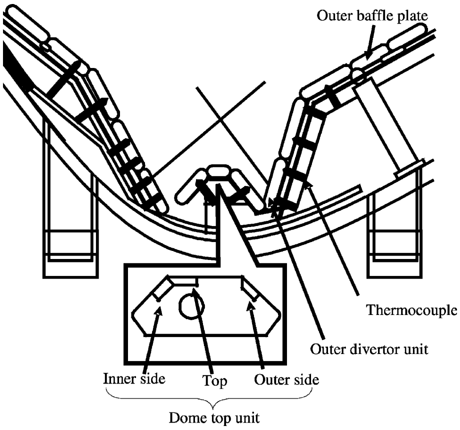

Fig. 1. Cross-sectional diagram of the divertor region in JT-60U.

The depth profiles of hydrogen and deuterium retained in graphite tiles were 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° orientation from the surface normal. The beam size was about 32 μm and the rastering area was set to be 400×400 μm^2. The sputter rate was determined nearly 1 μm/h by the calculation and the measurement of depth profiling by a profilometer (Dektak). The negative signal intensities of hydrogen isotopes were normalized by that of ^{12}C for comparison.

The chemical states of graphite, boron and oxygen were also studied by XPS (ULVAC-PHI ESCA5500MC system), using a Mg-Kα 400 W X-ray source and a hemispherical electron analyzer. The depth profile was also analyzed by Ar ion (Ar^+) sputtering with an energy of 4.0 keV and a fluence range of 2.93×10^{21}–1.54×10^{23} m^{-2}.

3. Results and discussion

Fig. 2(a)–(c) respectively show the depth profiles of $D/^{12}C$, $H/^{12}C$ and $(H + D)/^{12}C$ signal intensity ratios within the sputtering time of 6000 s measured by SIMS. The locations of these samples are illustrated in Fig. 1. It was found that the signal intensity ratio of $H/^{12}C$ in all the tiles was the highest on the surface of the tile and decreased with the increase of sputtering time. The concentration of hydrogen decreased toward the bulk of the tile, while the concentration of the deuterium increased. The largest signal intensity ratio of $D/^{12}C$, 0.20 was observed in the center of the dome top unit. The $D/^{12}C$ ratio of the inner side of the dome top unit was slightly lower, with a value of 0.15. However, the $D/^{12}C$ and $H/^{12}C$ ratios of the outer side of the dome top unit are much smaller than that of other locations. The $D/^{12}C$ ratio of the baffle plate is almost as large as that of the inner side or the center of the dome top unit and reached a value of 0.18. This might be due to the fact that the temperature of the outer baffle plate measured during the discharge was lower than that of other locations, and the distance from the plasma was the shortest. In the outer divertor unit, $H/^{12}C$ ratio on the surface is much larger than that on the dome unit and the baffle plate. However, the retention of deuterium is much smaller because of the high temperature during DD discharges and a high flux during the HH discharges. $H/^{12}C$ ratios in all the tiles decreased quickly toward the bulk and became constant between 0.02 and 0.1. In Fig. 2(c), the total amount of hydrogen and deuterium in the inner side of the dome top unit is the highest.

Fig. 3 shows the summary of the experimental results by SIMS. The integral signal intensity ratios of $H/^{12}C$ and $D/^{12}C$ within the sputtering time of 6000 s, which

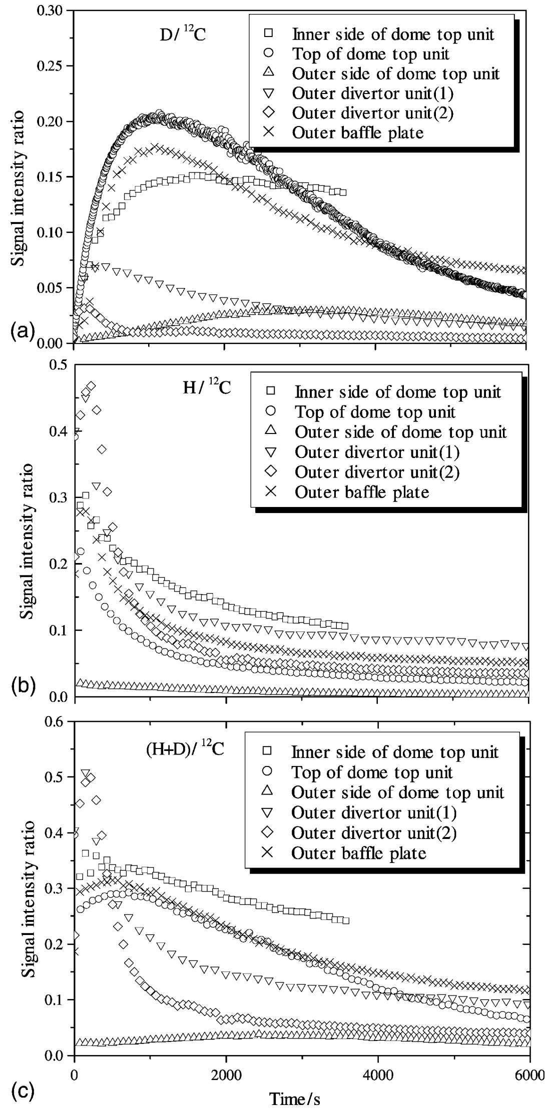

Fig. 2. Depth profiles of the signal intensity ratios of (a) D/^{12}C, (b) H/^{12}C and (c) (H + D)/^{12}C in the graphite tiles of the dome unit, the outer divertor unit and the baffle plate within the sputtering time of 6000 s by SIMS.

corresponds to the depth of about 1.7 μm, and the highest temperatures during the DD discharges were shown in this figure to illustrate the correlation between deuterium concentration retained in tiles and the temperature during the DD experiment. Here, it is assumed that the temperature of the dome top tile was the same as that of dome wings because the thermocouples were not installed in all the tiles as shown in Fig. 1. It is found that the signal intensity ratio of D/^{12}C in the tiles whose temperatures are less than 400 °C were almost higher and above 0.1. However, in the tile whose temperature is much higher and over 500 °C, the deuterium concentration is much lower. The minimum intensity position on the divertor unit also coincides with the position of the strike point, and the temperature of the unit might increase during the discharge. These results indicate that the trend of the amount of deuterium retained in the tile depends on the temperature. However, the amount of deuterium would depend not only on temperature but also the place of strike point and flux of the particles. Especially, the amount of deuterium in the dome unit would also be influenced by the flux of the particles because of the distance from the plasma was short and the temperature during the DD discharge was almost the same. These results are consistent with previous studies [5,9].

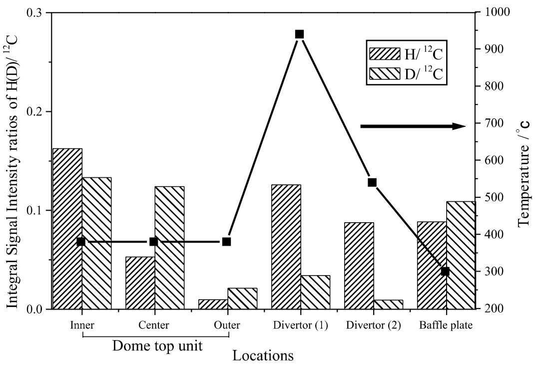

Fig. 3. Correlation between the integral signal intensity ratios of H/^{12}C and D/^{12}C within the sputtering time of 6000 s, which corresponds to the depth of about 1.7 μm, and temperature during the DD discharge in graphite tiles of the dome unit, the outer divertor unit and the baffle plate. The peak sputtering time corresponds to the depth of the maximum peak intensity.

Fig. 4 shows the photoelectron spectra of C 1s from the dome unit by XPS. The back side of the center of the dome top unit tile was used as standard, of which the binding energy was 284.57 eV. The peak top position of the inner side of the dome top unit was clearly different from the standard and shifted to a higher binding energy than 285.56 eV. In the center of the dome top unit, the shoulder peak was also confirmed in higher binding energy. After the Ar^{+} sputtering fluence of 2.93×10^{21} m^{-2}, the spectra of all the dome unit tiles were shifted to the standard C 1s position. It was reported that the binding energy of C 1s for amorphous hydrogenated carbon (a-C:H) and CO are 284.9–285.5 and 286.4–286.8 eV, respectively [10]. This indicates that hydrocarbons were formed only on the surface of the tile. This experimental result may also be caused by the divertor characteristics that impurities are easily concentrated inside the vacuum vessel and much of the dust may exist in the inner area of the W-shaped divertor region. Three following characteristics were found: (1) the peak shift of C 1s for the inner side of the dome top unit was the highest; (2) FWHM is also the largest for

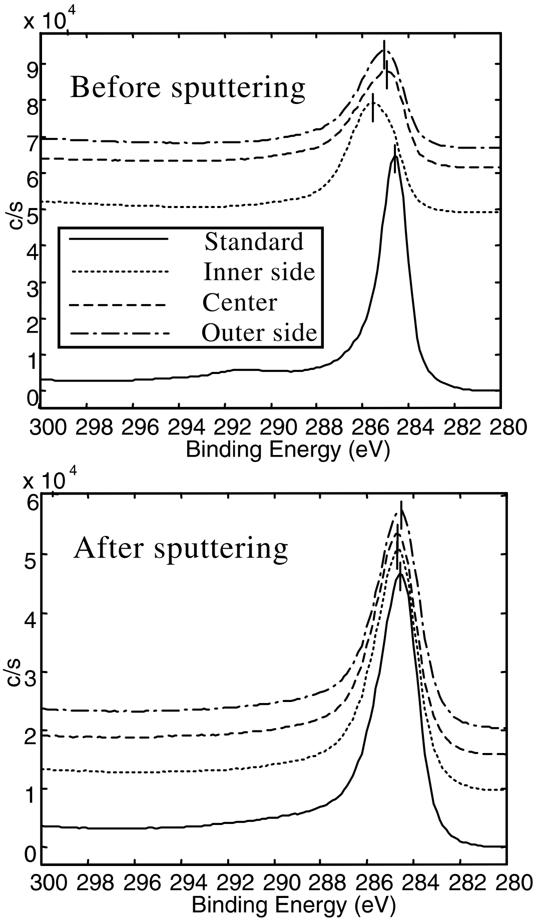

Fig. 4. Photoelectron spectra of C 1s from the inner side, the center and the outer side of the dome top unit by XPS.

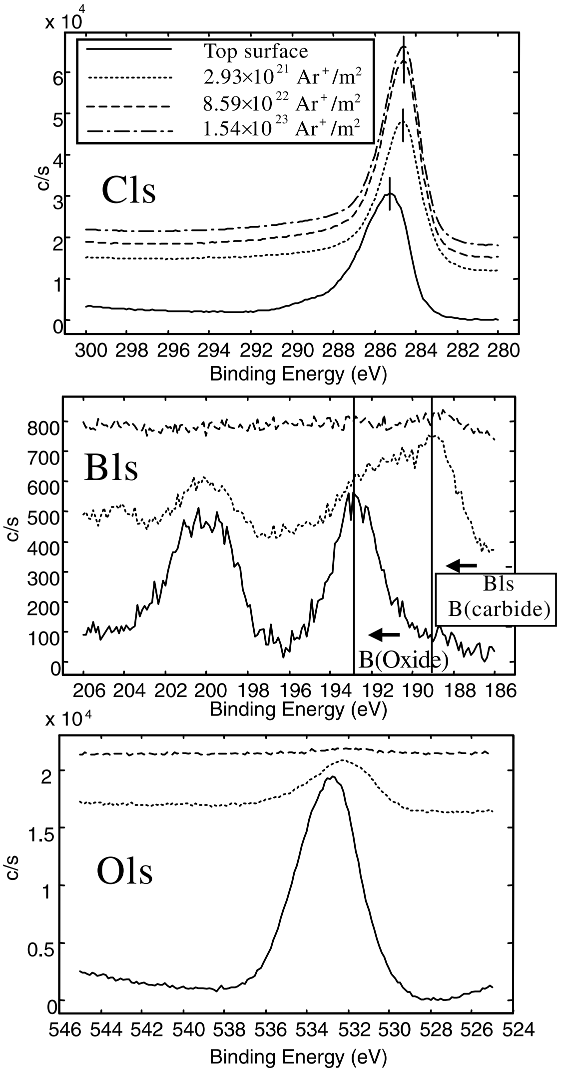

Fig. 5. Photoelectron spectra of C 1s, B 1s and O 1s from the center of the dome top unit by XPS.

the inner side; (3) FWHM for the outer side is the smallest. The hydrocarbon exists mostly on the inner side and not much hydrocarbon exists on the outer side of the dome top unit. These results are consistent with the experimental results by SIMS.

Fig. 5 also shows the photoelectron spectra of C 1s, B 1s and O 1s of the dome top unit, respectively. The chemical shift of C 1s indicates the formation of hydrocarbon as mentioned above. The peak position of B 1s was shifted to a higher binding energy of 193 eV, indicating that boron oxide was formed on the surface of the tile. After the Ar$^+$ sputtering of 2.93×10^{21} m^{-2}, the B 1s peak was shifted to a lower binding energy and divided into two peaks, B(oxide) and B(carbide) or B 1s. The O 1s peak was also weakened by the Ar$^+$ sputtering. It indicates that the oxygen is combined with boron.

From these experimental results, it is concluded that the deuterium exists mostly on the surface of the dome unit and the baffle plate and the hydrocarbon was formed on the surface of the tile. A small amount of boron also exists on the surface and forms boron oxide by gettering oxygen as impurity. The experimental results by XPS agree well with those by SIMS.

4. Conclusion

The behavior of hydrogen and deuterium retained in the graphite tiles placed in the dome unit, the divertor unit and the baffle plate of JT-60U were analyzed by SIMS; the chemical states of graphite and boron were also studied by XPS.

It is found that the hydrogen exist on the surface of the tiles mainly by the HH discharge after DD plasma discharge. The signal intensity ratio of D/^{12}C ratio in the center of the dome top unit reached a value of 0.20 and the deuterium concentrations in the inner side of the dome top unit and the outer baffle plate were also high. By comparing the experimental results and the temperatures during DD operation, the results indicate that the amount of deuterium retained in the tile depends largely on the temperature.

The XPS analysis also revealed that the binding energies of C 1s and B 1s peaks on the surface of the inner side of the dome top unit were largely shifted to higher energies compared with that of the outer side of the dome top unit. These results indicate that hydrocarbons and boron oxide might be formed on the surface of the inner side of the dome top unit.

It is concluded that the behavior of hydrogen and deuterium and the chemical state of graphite were influenced by the temperature of the surface, the position of the strike point and the flux of the particles during operation.

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