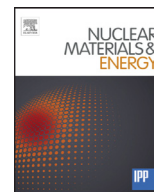




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Effects of modified surfaces produced at plasma-facing surface on hydrogen release behavior in the LHD



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ABSTRACT

In the present study, an additional deuterium (D) ion irradiation was performed against long-term samples mounted on the helical coil can and in the outer private region in the LHD during the 17th experimental campaign. Based on the release behavior of the D and hydrogen (H) retained during the experimental campaign, the difference of release behavior at the top surface and in bulk of modified surfaces is discussed. Almost all samples on the helical coil can were erosion-dominant and some samples were covered with boron or carbon, while a very thick carbon films were formed in the outer private region. In the erosion-dominant area, the D desorbed at much lower temperatures compared to that of H retained during the LHD plasma operation. For the samples covered with boron, the D tended to desorb at lower temperatures compared to H. For the carbon deposition samples, the D desorbed at much higher temperatures compared to no deposition and boron-covered samples, which was very similar to that of H. The D retention capabilities at the top surface of carbon and boron films were 2–3 times higher than no deposition area. The results indicate that the retention and release behavior at the top surface of the modified layer can be different from that of bulk substrate material.

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1. Introduction

The first wall of fusion devices receive energetic hydrogen isotopes and helium particles produced by a charge-exchange reaction at the plasma edge, which causes erosion, deposition of eroded materials and irradiation defects at the surface. This phenomenon modifies the surface conditions of the first wall and could change the hydrogen retention and release behavior of the surface [1–7]. Although the hydrogen flux to the first wall is much smaller than divertor region, the hydrogen recycling during plasma discharge in the first wall could be significant due to its large area. The hydrogen isotopes retention in the plasma-facing surface usually increases with discharge time during plasma discharge, which could

enhance hydrogen recycling. Therefore, to understand and control the hydrogen release from the modified layer becomes more important as the discharge duration grows longer. In addition, hydrogen isotopes retention is also important from the tritium safety point of view. Hydrogen retention and release behavior has been intensively investigated by performing hydrogen ion/plasma irradiation against a virgin plasma-facing material followed by thermal desorption spectroscopy (TDS). However, the hydrogen retention and release behavior of the modified layer produced in fusion devices has not been sufficiently investigated.

A long-term sample mounted on the plasma-facing surface and a small piece of sample cut from first wall panels are useful tool to grasp how the surface is modified by plasma-surface interactions (PSIs) [1–7]. And, in general, TDS analysis is used for investigation on the hydrogen isotopes release behavior in the modified layer. The TDS data includes information on release behavior of hydrogen retained both at the near surface region and at deep regions [1,2,6,7]. On the other hand, it is necessary for evaluation

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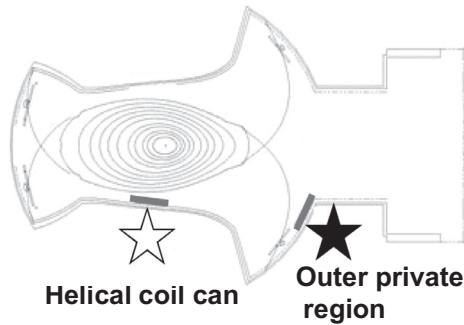


Fig. 1. Poloidal positions of long-term samples.

of the hydrogen recycling behavior during plasma discharge to understand the hydrogen release behavior, especially, at the near surface region of the modified layer. Long-term samples mounted in the vacuum vessel in the LHD during an experimental campaign contain hydrogen (H) used as a discharge gas. During the main discharge in the LHD, the average implantation depth of hydrogen particle irradiated to the first wall, which normally has incident energy of less than a few keV, is smaller than ~ 50 nm. Then, the layer from the surface to depth of ~ 50 nm is here called “top surface”. Since the implanted hydrogen during the discharge is once retained in the top surface, the release behavior of the top surface could greatly influence the hydrogen recycling. It has been reported that the H exists from the top surface to deep in bulk in the sample [8]. Therefore, comparing the release behavior of the H and an additionally irradiated hydrogen isotope, such as deuterium (D), could reveal the difference of the release behavior from the bulk and from the top surface. Although this method might be pretty simple, it would be a fresh technique focused on the top surface of modified layer.

In the present study, a deuterium (D) ion was additionally irradiated against the long-term samples mounted on the plasma-facing surface in the LHD, and the relationship of D retention and release behavior and the surface conditions was investigated. In addition, based on the release behavior of the D and hydrogen (H) retained during the experimental campaign, the difference of the release from the top surface and from deeper regions is discussed.

2. Experimental

In the 17th experimental campaign (in FY2013) of the LHD, hydrogen (H) and helium (He) was used as a working gas of main discharge. A glow discharge conditioning was conducted using He and other gases at night. In the beginning of the experimental campaign, boronization using glow discharge with a mixture of helium and diborane gas was once conducted. The locations of long-term samples in the vacuum vessel are shown in Fig. 1. The area of the first wall and graphite divertor tile is 780 m^2 and 30 m^2 , respectively [9]. The materials of the samples are stainless steel (SS), which is the same material as the first wall of the LHD, and silicon (Si). Since the element of Si wasn't contained in any plasma-facing material of the LHD, this sample is useful to detect the redeposition elements. 5–6 samples with a size of $5 \times 10 \text{ mm}^2$ were mounted at each poison. The samples on the helical coil can and in outer private region were mounted at every toroidal sector (totally ten positions).

These samples were extracted from the vacuum vessel after the experimental campaign and preserved in a vacuum at room temperature for approximately 2 years. After that, the samples were analyzed in several methods and used for an additional D ion irradiation experiments. The depth profile of atomic composition was

analyzed with Auger electron spectroscopy (AES), in which Ar^+ ion with energy of 3 keV was used for sputtering. The hydrogen retention and release behavior were investigated with thermal desorption spectroscopy (TDS) [10]. In the TDS analysis, the sample was heated from room temperature to 1273 K with heating rate of 0.5 K/s, and then held at 1273 K for 20 min. The sample temperature was monitored by a thermocouple directly attached to the sample. The desorption of gases during the heating was monitored by a quadrupole mass spectrometer. In order to clear the difference between the releases behavior of hydrogen retained during the long period of the experimental campaign and that of hydrogen retained at the top surface in the modified layer, an additional D irradiation against the samples were performed using an ECR ion source at Hokkaido University [11]. In this irradiation, D was selected as an irradiated ion species to distinguish the species retained during the additional irradiation from H retained during the LHD operation. In the irradiation, a D_3 ion with an energy of 5 keV (1.7 keV for sole D) was irradiated against the samples at the fluence of $3 \times 10^{17} \text{ D/cm}^2$. The average implantation depth of the D against stainless steel calculated by TRIM is approximately 20 nm, which is within the top surface. The sample temperature during the irradiation was room temperature, which is the same as a base temperature of the first wall in the LHD [9]. After the irradiation, the release behavior and retention of the retained D was investigated with TDS with the same condition described above. During the TDS, the D_2 desorption was monitored by a high resolution quadrupole mass spectrometer, which can separately detect D_2 from He.

3. Results and discussion

Depth profiles of atomic composition obtained with AES analysis are shown in Fig. 2. The deposition pattern on the helical coil can could be divided roughly in three types: little deposition area (Fig. 2(a)), boron deposition area (Fig. 2(b)) and carbon deposition area (Fig. 2(c)). Although a little amount of carbon was observed on the sample of little deposition area (Fig. 2(a)), this area was thought to be a net erosion area judging from early studies on erosion of the surface of the coil can [12]. The depth profile of the other toroidal sectors 3, 4, 5, 7, 8 and 9 were almost the same as the sector 2 (Fig. 2(a)). For the sample at the toroidal sector 1 (Fig. 2(b)), a boron film with a thickness of ~ 150 nm, which was evaluated with TEM observation, was observed. The thick boron film caused by boronization was due to this sector being close to the glow discharge anodes located at 10.5 sector and diborane inlet nozzle located at 1.5 sector [12,13], which were used for boronization conducted in the beginning of the experimental campaign. For the sample at the toroidal sector 10 (Fig. 2(c)), a 130 nm thick carbon film was observed. This deposition would be due to redeposition of carbon sputtered at graphite divertor tiles and/or at a neutral beam injection (NBI) armor tiles [6,14].

Since a very thick carbon film was formed on the samples in the outer private region, the depth profile of deposited elements in these samples was investigated with GDOES. Although an evaluation of atomic composition with GDOES analysis is difficult, this method can perform a qualitative analysis of depth profile of deposits including hydrogen with a very high etching rate [15]. A typical depth profile of the deposits in the outer private region at toroidal sector 5 is shown in Fig. 3. From this result and earlier studies [6,14], the main element of the deposits was thought to be carbon. This carbon deposition was owing to redeposition of carbon sputtered at the divertor tiles. The thickness of the carbon films estimated by measuring the depth of the etching crater was $1\text{--}2 \mu\text{m}$, which was similar to the samples at other toroidal sectors.

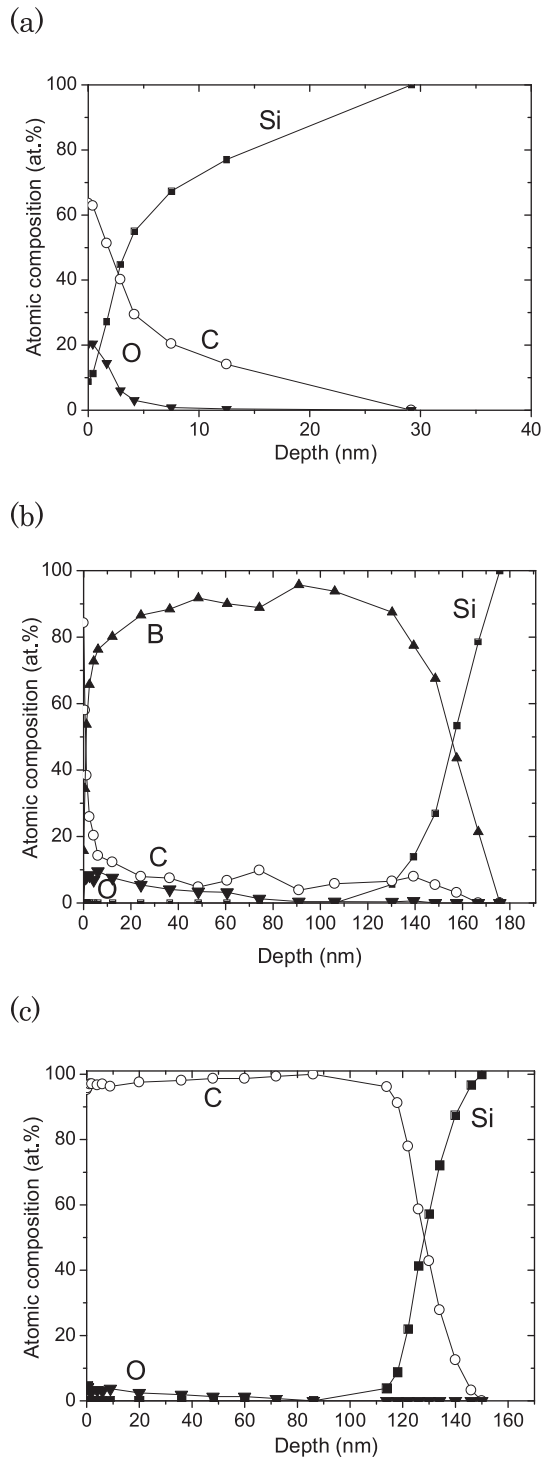


Fig. 2. Depth profiles of atomic composition for Si samples mounted on helical coil can at toroidal sectors 2 (a), 1 (b) and 10 (c).

Fig. 4 shows the TEM image of cross section of stainless steel sample mounted on the helical coil can at toroidal sector 5 (erosion area). Within the depth of ~50 nm from the top, He bubbles with a size of few nm were observed, which was also seen in other samples of erosion area. The He bubble layer was formed during He grow discharge conditioning and main discharge using He [16,17].

Fig. 5 shows thermal desorption spectra of H₂ and D₂ for stainless steel samples mounted on the helical coil can at toroidal sec-

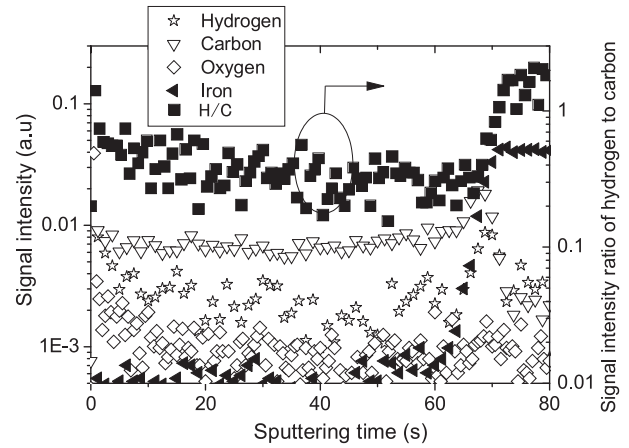


Fig. 3. Depth profile of deposits and the ratio of signal intensity of hydrogen to carbon for SS sample in the outer private region at toroidal sector 5.

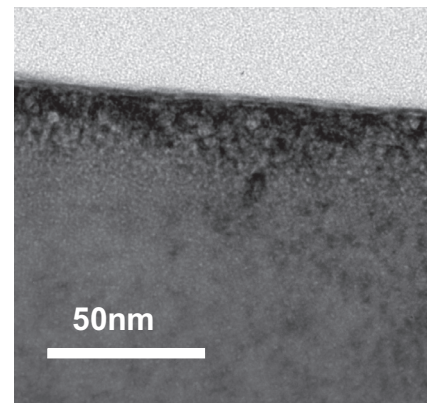


Fig. 4. TEM image of cross section of stainless steel sample mounted on helical coil can at toroidal sector 5 (erosion area).

tors 2 (erosion area) (a), 1 (boron deposition area) (b) and in the outer private region at toroidal sector 3 (carbon deposition area) (c). Note that H was retained mainly during the hydrogen discharges in the LHD plasma operation, and D was retained during the additional D ion irradiation performed against the long-term samples after the LHD experimental campaign, and that the sample used to obtain TDS spectra of H₂ was separate from that of D₂.

For the sample at erosion area (Fig. 5(a)), most of D desorbed in the temperatures below 600 K, while H₂ started to desorb at around 530 K and the desorption spectrum had major peaks at 720 K and 910 K. Possible three reasons for the fact no H₂ desorption was observed at low temperatures (< 530 K) are as follows. One is the vessel baking conducted in the experimental campaign. In the LHD, a vessel baking at 368 K was periodically conducted [9], which would lead to desorption of H₂ trapped with low trap energy. The second is H₂ desorption during a main plasma discharge. Although the surface temperature of the first wall during the main discharge, unfortunately, weren't measured, the surface temperature could rise to a certain extent due to a heat load from the plasma, which could give rise of H₂ desorption. The third one is a long-term preservation of the sample after extracting from the vacuum vessel. The sample was preserved in a vacuum for approximately 2 years since the sample had been extracted from the LHD. Therefore, some of the hydrogen retained in the sample with a small trap energy might have been released until the D irradiation was performed [18,19]. The reason for H₂ desorption at higher temperatures (> 600 K) is not clear at present. H retained in deep

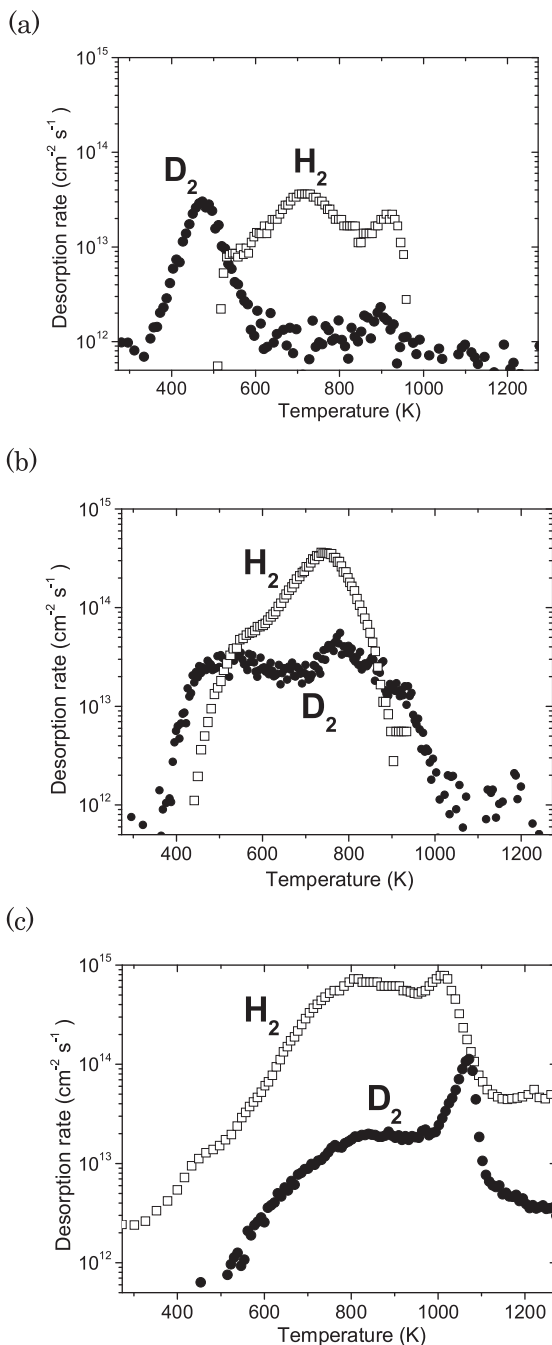


Fig. 5. Thermal desorption spectra of H_2 and D_2 for stainless steel samples mounted on the helical coil can at toroidal sectors 2 (erosion area) (a), 1 (boron deposition area) (b) and in the outer private region at toroidal sector 3 (carbon deposition area) (c).

regions due to diffusion into the bulk during long-term plasma exposure might be a reason for that.

For the sample covered with boron (Fig. 5(b)), the H_2 and D_2 desorption started at ~ 400 K and the spectra had two major peaks at around 500 K and 750 K. The lower and higher desorption peaks are attributed to the release of H (or D) forming B-H-B (or B-D-B) and B-H (or B-D) bonding, respectively [20]. Although the desorption rate of D_2 at lower temperature peak was almost the same as that at higher temperature peak, the desorption rate of H_2 at lower temperature peak was much smaller than that at higher temperature peak. This reason would be the same as the case of erosion area described above. Namely, H weakly trapped in the boron

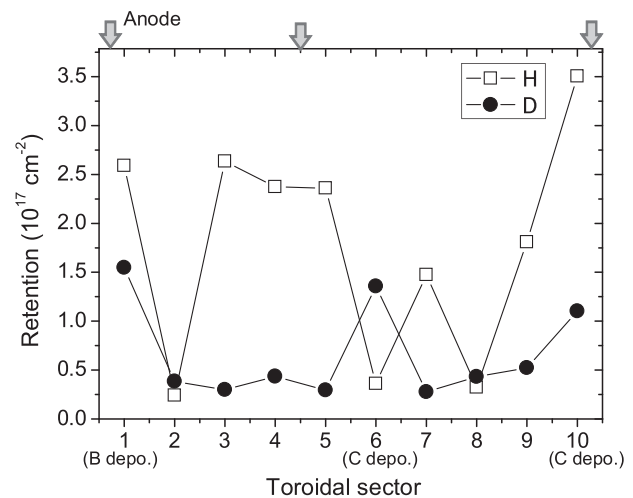


Fig. 6. Toroidal distributions of H and D retention for SS samples mounted on the helical coil can.

film might have been desorbed due to wall temperature rise during baking or plasma discharge, or long-term preservation of the sample.

For the sample covered with carbon (Fig. 5(c)), the TDS spectrum of H_2 and D_2 had major desorption peak at higher temperature, 1170 K, comparing to the erosion and boron deposition areas. This is due to a strong bonding between H (and D) and carbon atom [21]. It was found from the GDOES analysis (Fig. 3) that the H was retained almost uniformly through the carbon film with a thickness of 1–2 μm , which was much deeper than the implantation depth of D, ~ 20 nm. Then, the fact that the desorption spectra of H_2 was very similar to that of D_2 (Fig. 5(c)) indicates that the hydrogen isotopes desorption from the carbon film might be detrapping-dominant process. The results of the additional D irradiation against long-term samples indicate that the hydrogen isotopes release behavior at the top surface of the modified layer can be different from that of hydrogen isotopes retained during the long period of plasma operation.

The H and D retention for the samples mounted on the coil can are shown in Fig. 6. The toroidal distribution of H and D retentions were significantly different each other. In the D irradiation condition used in the present study, the D retention was larger for the samples with boron and carbon deposition. Hydrogen isotopes irradiations against such materials have shown that hydrogen isotopes retention in carbon materials and boron has higher capability of retaining hydrogen isotopes at the surface of deposits. On the other hand, considering the lower H retention for the sample at the sector 6, where a thick carbon film was observed, the H retention seemed to be larger near the glow discharge anodes located at toroidal sectors 4.5 and 10.5, which is a similar tendency with the former experimental campaigns [12]. This indicates that the H retention during long-term operation of the LHD could be more influenced by glow discharge rather than the surface deposits. Hydrogen isotopes can diffuse to the deeper region of metals during the long exposure to hydrogen discharges [8], which can enhance the hydrogen retention. This would be a reason for the large retention in the samples at the vicinity of the glow discharge anodes. The difference of the retention between D and H indicate that the hydrogen isotopes retention behavior of the top surface of modified layer can be different from that of bulk hydrogen retention.

4. Summary

In the present study, an additional D irradiation was performed against the long-term samples mounted on the first wall in the LHD, and the relationship between the surface conditions and the D retention and release behavior was investigated. It was found that the surface of the helical coil can was erosion-dominant and some area was covered with boron and carbon, and a very thick carbon films were observed in the outer private regions. In the erosion dominant-area, the release behavior of D was much different from that of H. The H, which was retained during several months of the LHD plasma operations, desorbed at higher temperature compared to that of D. For the samples covered with boron, the desorption spectra of both H and D had major peaks at the same temperatures, and the D tended to desorb in the lower temperatures. For the samples covered with carbon, the D desorbed at much higher temperatures compared to no deposition and boron-covered samples, which was similar to that of H. The D retention retained at the top surface of carbon and boron films was 2–3 times higher than no deposition area. The results indicate that the hydrogen isotopes retention and release behavior at the top surface of the modified layer can be different from that of bulk substrate material, and that since the HCC area is more close to the plasma and the wall temperature would easily rise up compared to the OPR, fuel hydrogen recycling at the erosion-dominant area on the HCC, where retained hydrogen desorbs at low temperatures, could be firstly enhanced in the early phase of discharge.

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