Effects of Mild Baking on Hydrogen Removal from the Modified Surface of the First Wall in the LHD

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An additional deuterium irradiation was performed for long-term samples mounted on the first wall in the large helical device (LHD) to investigate the effect of mild baking (at 368 K) on fuel hydrogen removal from the modified surface. The efficiency of baking strongly depended on the surface conditions. In the erosion dominant area, almost all the retained deuterium was removed by baking, whereas only 17% and 13% of the retained deuterium were removed from the boron and carbon deposition areas, respectively. The results suggest that the erosion dominant area in the first wall can act as a pumping wall during the main discharge, and, in contrast, the fuel hydrogen retention can continuously increase in boron and carbon deposition areas owing to the low removal efficiency of fuel hydrogen given that the deposition continues during the plasma discharge.

Keywords: baking, desorption, retention, surface modification, deposition, deuterium irradiation

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Removal of fuel hydrogen from the plasma-facing wall in fusion devices is necessary for reducing fuel hydrogen recycling and tritium inventory. Isothermal heating of the wall, which is baking, is widely applied as one of the primary techniques for fuel hydrogen removal. This technique will be used in the ITER as well [1]. In the large helical device (LHD), baking is performed during intervals between the LHD plasma discharges, and the baking temperature is set below 368 K because the wall is close to a superconducting magnet coil [2]. This mild baking is effective for removing adsobates on the surface [2]. However, it is unclear whether this baking is effective for removing the fuel hydrogen retained on the wall during the main discharge. The plasma-facing wall receives energetic particles, leading to the erosion of the wall material, redeposition of the eroded material, and irradiation damage. These phenomena can modify the fuel hydrogen retention and desorption behavior of the wall surface [3]. A long-term sample mounted on plasma-facing surface is useful for evaluating the fuel hydrogen retention and desorption behavior of the modified surface. As fuel hydrogen in the plasma-facing material exists not only near the surface region but also in the deep region owing to hydrogen diffusion into the bulk [4], the thermal desorption spectroscopy (TDS) analysis of long-term samples obtained after the plasma operation hardly provides information on fuel hydrogen desorption from the top of the surface region. In the present study, an additional deuterium (D) irradiation was performed for the long-term sample mounted on the first wall in the LHD. Then, the samples were heated at the LHD baking temperature of 368 K to investigate the effectiveness of mild baking in the removal of fuel hydrogen retained in the modified surface. These results also provide information to understand the wall-pumping capability after baking.

Positions of the long-term samples mounted on the first wall in the LHD are shown in Fig. 1. At each position, several stainless steel samples, which are made of the same material as the LHD first wall, with dimensions $10 \times 5 \times 1\,\mathrm{mm}^3$ were mounted before the beginning of the 17th experimental campaign (FY2013). After the campaign, the samples were extracted from the vacuum vessel

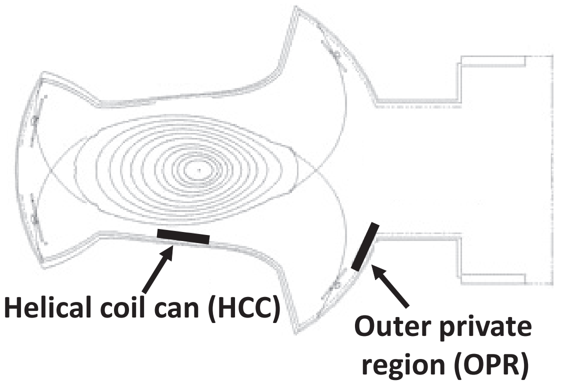

Fig. 1 Cross-sectional view of the LHD and sample positions. The HCC is close to the main plasma, and OPR is close to the graphite divertor tile.

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author's e-mail: y-nobuta@eng.hokudai.ac.jp and analyzed using several techniques. The depth profile of atomic composition of deposits on the sample was investigated using Auger electron spectroscopy (AES) and glow discharge optical emission spectroscopy. The irradiation defects at the top surface region were evaluated using transmission electron microscope (TEM). After the surface analysis, the samples were subjected to an additional D ion irradiation to evaluate the D desorption behavior at the top surface of the modified layer. D was selected as the ion species for irradiation to distinguish the species retained during the additional irradiation from the hydrogen that was used as a working gas for the LHD plasma operation. The D irradiation was performed using an electron cyclotron resonance ion source [5]. D ions with the energy of 5-keV D_3^+ (1.7 keV for single D ion) were irradiated at the fluence of 3×10^{17} D/cm^2. Using SRIM code, the implanted depth of D ions at this energy is estimated as several tens of nm, which is much smaller than the depth for hydrogen ions (~ μm) retained during the LHD plasma operation [4]. Thus, D desorption in this study can be considered as desorption from the top surface of the modified layer. During the irradiation, the substrate was at room temperature. The D-irradiated samples were used for TDS analysis wherein the samples were heated up to 1273 K with a ramp rate of 0.5 K/s. Other samples were used for a baking experiment wherein the samples were heated at a fixed temperature of 368 K, which is equal to the baking temperature of the first wall in the LHD. After baking for 6 h, the desorption behavior of the remaining D was investigated via TDS analysis under the same condition as described above. During TDS analysis and baking, the sample temperature was measured using the thermocouples directly attached to the sample, and the desorbed gases were measured using a high-resolution quadrupole mass spectrometer (QMS), which can separately detect D_2 and helium.

The deposited elements on each sample and the thickness are presented in Table 1. The deposition patterns differed greatly from position to position and were roughly divided into three areas: the erosion dominant, boron (B) deposition, and carbon (C) deposition areas. Almost no deposition was observed for HCC samples in the sectors 2 - 5 and 7 - 9. These sectors are considered as a net erosion area, judging from the early study in the wall erosion of the LHD [6]. For the HCC sample in sector 1, a 150-nm-thick B film was observed. In the 17th experimental campaign, boronization was conducted as a wall conditioning in the beginning of the campaign. This B deposition is attributed to the vicinity of this sector to the glow discharge anode and diborane inlet nozzle [7]. For the HCC sample in sectors 6 and 10 and all OPR samples, a thick C film was observed. This C deposition is attributed to the redeposition of C sputtered at the graphite divertor tile near the sample [8].

Figure 2 shows the TDS spectra of D_2 for HCC samples in toroidal sector 2 (Fig. 2 (a)) and 1 (Fig. 2 (b)) and for the OPR sample in sector 3 (Fig. 2 (c)) before and after baking. For the sample without baking in the erosion dominant area (Fig. 2 (a)), the TDS spectra shows a major peak at 480 K. Further, the desorption rate of the peak was approximately three times larger than that for the as-received sample irradiated by D ions under the same condition. In the top surface region of the HCC sample, helium bubbles formed by helium main and glow discharges during the LHD operation were observed [3]. These helium bubbles can act as trap sites for hydrogen isotopes. This bubble formation can be a reason for the higher retention capability for implanted D compared to the as-received sample. After baking, the major peak largely decreased. This indicates that a baking temperature of approximately 110 K lower than the TDS peak temperature (480 K) is effective for removing the D from the erosion dominant area. As the D desorption rate during the baking was too low to be measured by QMS, the time constant of D_2 desorption could not be estimated in this experiment. Assuming that D_2 desorption is considered as a first-order kinetic process,

Table 1 Deposited elements and their thickness at each sample position.

Poloidal position	Toroidal sector number	Deposition or erosion	Deposited elements	Deposition thickness
Helical Coil Can (HCC)	2-5, 7-9	erosion	A little amount of carbon and oxygen	a few nm
	1	deposition	boron (~90 at.%) carbon (~10 at.%) oxygen (a few at.%)	~150 nm
	6	deposition	carbon (~80 at.%) boron (~ 20 at.%) oxygen (a few at.%)	~75 nm
	10	deposition	carbon (~98 at.%) oxygen (~ 2 at.%)	~130 nm
Outer Private Region (OPR)	1-10	deposition	carbon	~1-2 μm

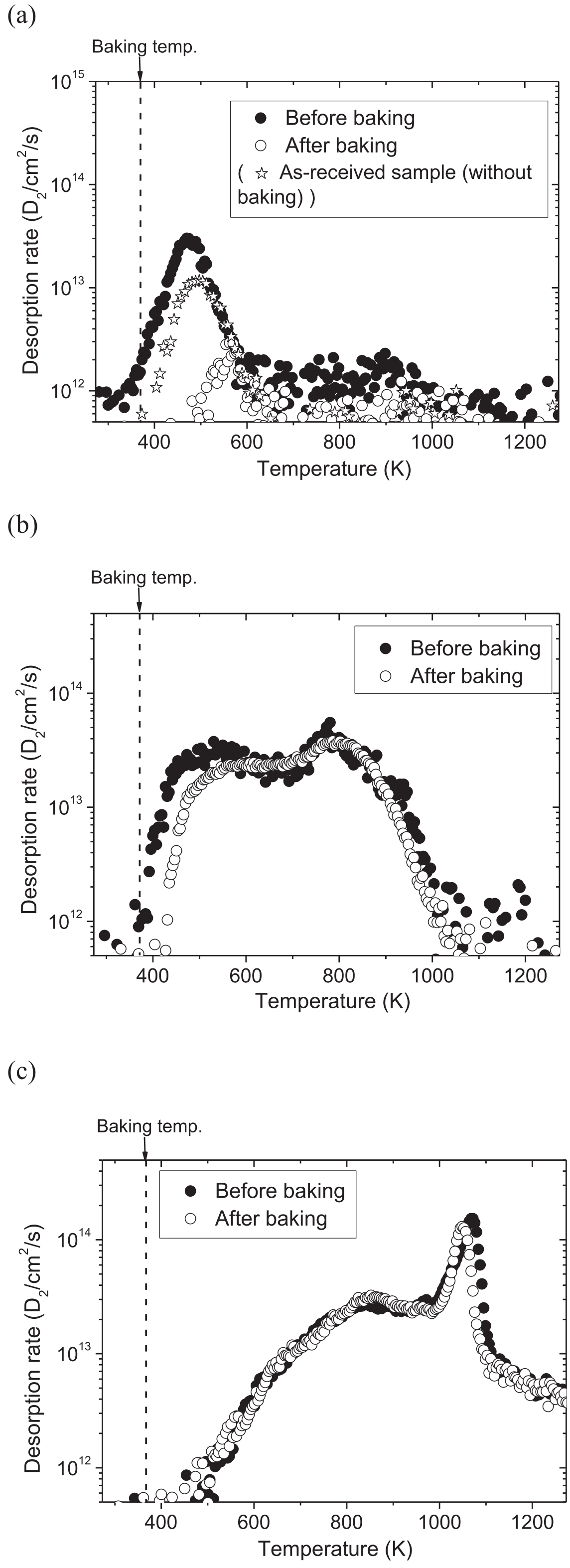

Fig. 2 Thermal desorption spectra of D_2 before and after baking at 368 K for the HCC samples in sector 2 (a) and 1 (b) and the OPR sample in sector 3 (c). Figure 2 (a) also shows the D_2 desorption spectra for the as-received sample without baking.

the time constant, τ, for D_2 desorption during baking is expressed by the following equation [9]:

$$\tau = \nu^{-1} \exp(E/k_B T),$$

where ν is the average attempt frequency, E is the activation energy of detrapping from defects, k_B is the Boltzman constant, and T is baking temperature. This equation indicates that as E becomes large, long time is required for the hydrogen isotopes to desorb. In other words, even if the baking temperature is lower than a certain temperature, at which the desorption peak is observed in TDS analysis for a sample, baking for a long time can result in continuous and slight D desorption from the sample. The time evolution of D retention during baking, $A(t)$, is expressed by the following equation:

$$A(t) = A_0 \exp(-t/\tau),$$

where A_0 is the D retention just before baking. Considering an 88% reduction in D retention for the HCC sample after 6 h of baking, τ for the sample is estimated as 2.83 h (10188 s). Using this value and $\nu = 1 \times 10^{13}$ s^{-1} [10], the activation energy of detrapping, E, is estimated as 1.24 eV; this activation energy is in the range of the energy required for hydrogen isotopes to detrap from a helium bubble, 1.2 - 1.8 eV, and from a vacancy $\leq$ 1.43 eV (depending on the number of D in a vacancy) [11]. This estimation indicates that the long-term release of D from such trap sites during baking may cause the large reduction of D in the case of the HCC sample.

For the sample covered with B (Fig. 2 (b)), the spectra exhibited two major peaks at 500 and 800 K, which are attributed to the detrapping of D from B-D-B and B-D bonding, respectively [12]. After baking, the desorption rate of the lower temperature peak decreased slightly, whereas that of the higher temperature peak did not change. This indicates that mild baking resulted only in slight removal effect for D that is weakly bonded with B (B-D-B). For the samples covered with C deposition (Fig. 2 (c)), a major desorption peak was observed as high as 1050 K. This desorption peak is caused by the strong bonding between the C atom and D in the C film. After baking, the desorption spectra for the C film showed almost no change.

D retention just after D irradiation and the reduction of the retained D after baking is shown in Fig. 3. Almost all D retained in the sample in the erosion dominant area desorbed in the form of D_2 and HD, whereas a slight desorption of methane (CD_4) was also observed for samples in the B and C deposition areas. The desorption of borane (BD_x) was negligible. The D retention shown in Fig. 3 is estimated from the desorption amount of D_2, HD, and CD_4. Deuterium retention for B and C films was 4 - 5 times larger than that for the erosion dominant area. This indicates that the top surface of the B and C films possess a higher capability of retaining implanted D. A large part of the retained D was removed by baking in the erosion dominant area, whereas only 17% and 13% were removed from the B and C deposition areas, respectively. Note that these reduction ratios of D retention after baking at 368 K could change with increasing/decreasing baking time.

In the present study, to evaluate the effects of mild

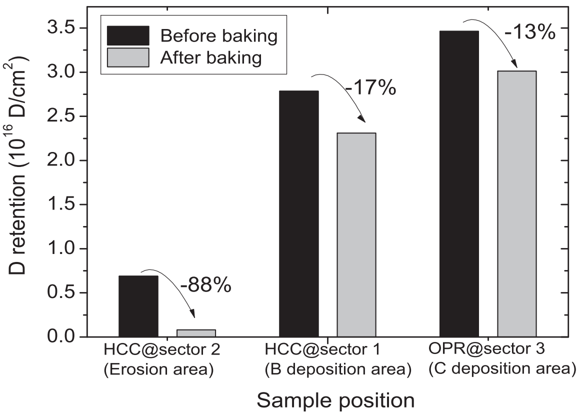

Fig. 3 D retention before and after baking for the HCC samples in sectors 2 and 1 and the OPR sample in sector 3. These results were evaluated by thermal desorption spectra.

baking (at 368 K) in the LHD on fuel hydrogen removal from the modified surface, an additional D ion irradiation was performed for long-term samples mounted on the first wall of the LHD. The effects of baking on D removal significantly depended on the surface conditions. In the erosion dominant area, the implanted D desorbed at low temperatures of approximately 480 K. From the TDS spectra of D_2 before and after baking, it was found that baking could remove approximately 90% of the implanted D in the erosion dominant area. This indicates that baking at temperatures lower than the TDS peak temperature is effective for fuel hydrogen removal in the erosion dominant area. In contrast, mild baking exhibited a small effect in the B and C deposition areas. Owing to the high efficiency of fuel hydrogen removal in the erosion dominant area, the first wall just after baking can act as a pumping wall during the main discharge. In contrast, in the B and C deposition areas, fuel hydrogen retention could continuously increase owing to its low removal efficiency by baking given that the deposition continues during plasma discharge in the LHD.

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