

# Deuterium Retention in Graphite and its Removal by Inert Gas Glow Discharge\*

Akira MATSUMOTO, Yuji YAMAUCHI, Tomoaki HINO, Kiyohiko NISHIMURA<sup>1)</sup>  
and Yoshio UEDA<sup>2)</sup>

*Hokkaido University, Kita-13, Nishi-8, Kita-ku, Sapporo 060-8628, Japan*

<sup>1)</sup>*National Institute for Fusion Science, 322-6 Oroshi-cho, Toki 509-5292, Japan*

<sup>2)</sup>*Osaka University, 2-1 Yamadaoka, Suita, Osaka 565-0871, Japan*

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The hydrogen isotope retention and its removal by inert gas glow discharge for graphite was investigated by residual gas analysis. The amount of retained hydrogen isotope in graphite during the deuterium glow discharge was about  $1.0 \times 10^{17}$  cm<sup>2</sup>, which was one order of magnitude larger than that in 316L stainless steel or tungsten. The removal ratio of hydrogen isotope by helium, neon and argon glow discharge cleaning were 49%, 22% and 6% respectively. These removal ratios were similar to those for the stainless steel, but larger than the tungsten. The removal ratio in graphite can be explained by both the depth distribution of incident deuterium in the materials and the etching depth.

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## 1. Introduction

In the fusion device, the reduction of hydrogen isotope retention in plasma facing walls is one of the significant concerns from the viewpoint of control of fuel particle density and tritium inventory. Glow discharge cleaning (GDC) using an inert gas has been widely used in many fusion devices such as the Large Helical Device [1–3] and tokamaks [4], in order to reduce the fuel hydrogen retention and remove the impurities on the walls. However, the inert gas particles are retained in the walls during GDC and then re-emitted into core plasma during main discharge. This phenomenon causes the radiation loss and the fuel dilution. Therefore, the inert gas retention in the walls also must be evaluated. On the other hand, graphite material is one of candidates for plasma facing material because of its low atomic number and high tolerance to high heat load. However, the removal efficiency of fuel hydrogen by GDC in graphite has not been investigated in detail.

In the present study, the hydrogen isotope retention in graphite by deuterium glow discharge and the reduction of retained hydrogen isotope by inert gas glow discharge was examined by using a glow discharge device with a graphite liner by residual gas analysis (RGA). The inert gas retention in graphite was also estimated.

## 2. Experimental

Figure 1 shows the schematic view of the glow dis-

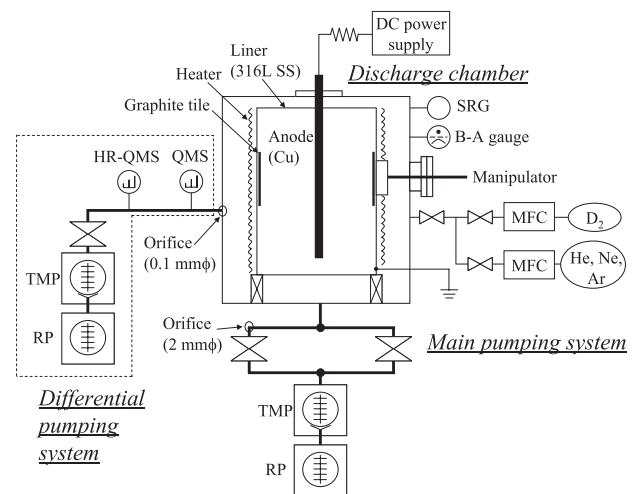


Fig. 1 Schematic view of GD device.

charge (GD) device used in the present study. The cylindrical liner made of 316L stainless steel (SS) was installed in the device, and graphite tiles of IG-430U (Toyo Tanso Co.) were set at the inside of the liner. The liner with the graphite tiles were baked at 150°C for 20 h by the heater before plasma discharge experiment. The constant-flow discharge gases were introduced into the discharge chamber through each mass flow controller (MFC), and the GD was conducted between a copper anode and the liner at room temperature. The initial discharge pressure was 8 Pa. The discharge duration was 2 h. The discharge voltage was 150–300 V. The partial pressure of retained or released gas for the graphite tiles during the discharge was quantita-

author's e-mail: akatsuki.will.1231@eng.hokudai.ac.jp

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Table 1 The amounts of hydrogen isotope retained or released during each discharge, and the removal ratio of hydrogen isotope and inert gas retention in graphite, 316L stainless steel and tungsten by the inert gas GDC.

Gas of GDC	First discharge (D <sub>2</sub> GD)		Second discharge (inert gas GDC)		Discharge gas retention [cm <sup>-2</sup> ]
	Hydrogen isotope retention [cm <sup>-2</sup> ]	Released deuterium [cm <sup>-2</sup> ]	Released hydrogen [cm <sup>-2</sup> ]	Removal ratio of hydrogen isotope [%]	
He	$6.0 \times 10^{16}$	$2.9 \times 10^{16}$	$8.6 \times 10^{14}$	49 SS: 45 W: 4.6	$7.5 \times 10^{14}$
Ne	$5.0 \times 10^{16}$	$7.1 \times 10^{15}$	$3.6 \times 10^{15}$	22 SS: 30 W: 3.4	
Ar	$5.5 \times 10^{16}$	$2.6 \times 10^{15}$	$9.6 \times 10^{14}$	6 SS: 15 W: 2.9	

SS: 316L stainless steel, W: tungsten

tively measured by a quadruple mass spectrometer (QMS) installed in the differential pumping system. In addition, the deuterium gas pressure during helium GDC was measured using the high-resolution (HR) QMS.

First deuterium (D<sub>2</sub>) GD was conducted to retain deuterium in the graphite tiles. After the D<sub>2</sub> GD the graphite tiles were subsequently exposed to helium (He), neon (Ne) or argon (Ar) GDC. The amounts of retained or released gas during the discharges were estimated from the change of partial pressure during the discharge, i.e. by RGA.

### 3. Results and Discussion

#### 3.1 Deuterium retention in graphite

Figure 2 shows time evolutions of D<sub>2</sub>, HD and H<sub>2</sub> partial pressure during the first D<sub>2</sub> GD. D<sub>2</sub> partial pressure rapidly decreased, and a part of the implanted deuterium was combined with hydrogen which led to the release of HD when the D<sub>2</sub> plasma was ignited. Furthermore, hydrogen which was initially retained in the graphite was released as H<sub>2</sub>. The release as hydrocarbon was not observed. The net amount of hydrogen isotope retention in graphite was estimated to be  $1.0 \times 10^{17}$  cm<sup>2</sup> from the data of Fig. 2. The hydrogen isotope retention in the graphite was larger than that in 316L SS [5] by a factor of about 4 and in tungsten [5] by a factor of about 2, respectively. The differences in the implantation depth of deuterium and the formation of radiation damage were responsible for the difference in the hydrogen isotope retention.

#### 3.2 Deuterium removal by inert gas GDC

After D<sub>2</sub> GD, He, Ne or Ar GDC was conducted to examine the removal of hydrogen isotope retained in the graphite. Figures 3 (a), (b) and (c) show time evolutions of D<sub>2</sub>, HD, H<sub>2</sub> and inert gas partial pressure during He, Ne and Ar GDC, respectively. Large desorptions of D<sub>2</sub>, HD and H<sub>2</sub> from the graphite tile were observed in the

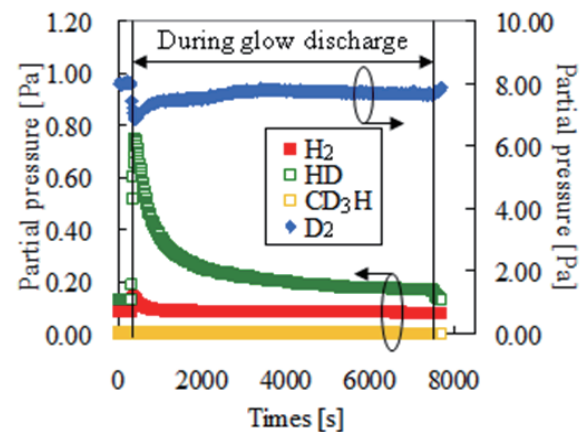


Fig. 2 Time evolutions of D<sub>2</sub>, HD and H<sub>2</sub> partial pressure during D<sub>2</sub> GD.

initial phases of all GDCs. The main desorption species during the He GDC was D<sub>2</sub>. The He partial pressure did not change during the discharge. This indicates that the helium retention in the graphite was negligible during the He discharge. The amount of desorbed hydrogen isotope during the He GDC was estimated to be  $3.0 \times 10^{16}$  cm<sup>2</sup>, which corresponded to 49% of the amount of retained hydrogen isotope during D<sub>2</sub> GD before the He GDC. The D<sub>2</sub>, HD and H<sub>2</sub> desorption were also observed for neon and argon GDC as shown in Figs. 3 (b) and 3 (c).

The amounts of hydrogen isotope retained or released during each discharge, and the removal ratio of hydrogen isotope and inert gas retention by inert gas GDC in graphite were summarized in Table 1. For reference, the results on 316L SS and tungsten were also shown. The amount of desorbed hydrogen isotope during Ne GDC corresponded to 22% of the hydrogen isotope retention by the antecedent D<sub>2</sub> GD. The amount of desorbed hydrogen isotope during Ar GDC corresponded to about 6.5% of the hydrogen isotope retention. The change in inert gas partial pressure dur-

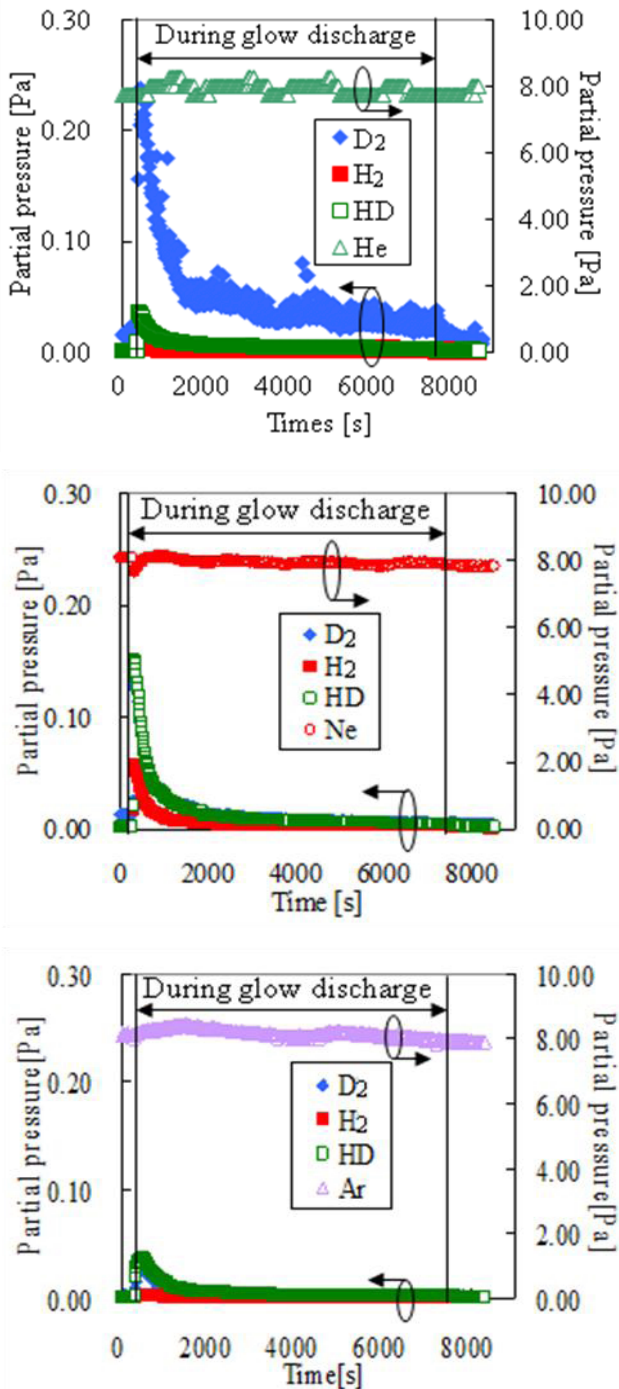


Fig. 3 Time evolutions of D<sub>2</sub>, HD, H<sub>2</sub> and inert gas partial pressure during (a) He GDC, (b) Ne GDC and (c) Ar GDC.

ing the initial period just after the ignition was observed for only Ne GDC. The neon retention in graphite estimated was three orders of magnitude smaller than the deuterium retention. It was noted that the neon retention in graphite is enough small for to ignore the possibility of the mixing of neon particle in the core plasma.

The He GDC could remove hydrogen isotope retained in the graphite tile most efficiently. This might result from that the mass of helium atom was close to that of carbon

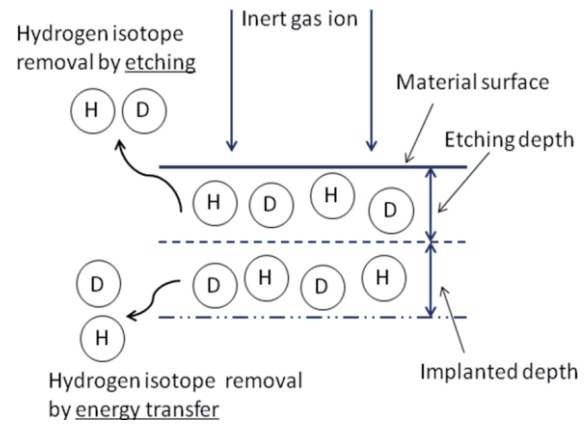


Fig. 4 Schematic view of the mechanism of hydrogen isotope removal by inert gas GDC.

and the implantation depth of helium was longer than other two discharge gases. Figure 4 shows the schematic view of the mechanism of hydrogen isotope removal by inert gas GDC. Hydrogen isotope must be released through the energy transfer from inert gas ion to carbon or hydrogen isotope atoms and sputtering of carbon. The helium ion could influence hydrogen isotope retained in deep region, and the efficiency of the energy transfer between helium and carbon/hydrogen isotope was very high compared with the cases of neon and argon. Hence, the hydrogen isotope removal ratio of He GDC became large. On the other hand, the hydrogen isotope removal ratio of Ar GDC was small. This might result from that the implantation depth of argon ion was small and sputtering yield of argon ion for graphite was also smaller than that of helium or neon [6].

Table 2 shows that the etching depth by inert gas GDC, the implantation depth of inert gas, the fraction of deuterium in etching depth and the fraction of deuterium from etching depth to implantation depth of inert gas ion in the graphite. The estimation was examined by SRIM-code 2008 [7]. Total deuterium removal ratio speculated by SRIM calculation was evaluated from the summation of the fraction of deuterium in etching depth and the fraction of deuterium from etching depth to implantation depth. Total hydrogen isotope removal ratio, which was obtained under the assumption that the hydrogen has almost same distribution with deuterium, were also shown this speculation. For Ne and Ar GDC, the hydrogen isotope removal ratios obtained by the present experiments were roughly consistent with the speculated ones. However, for He GDC, the value obtained by the experiment was smaller than the speculated one. This might result from re-deposition of sputtered carbon on the inner wall of the device and simultaneous re-incorporation of hydrogen isotope into the re-deposition layer.

The removal ratio of hydrogen isotope by inert gas GDC in graphite was almost equal to those for the stainless steel, but larger than the tungsten. The etching depth

Table 2 The etching depth by inert gas GDC and implantation depth of inert gas ion, and fraction of deuterium in graphite and total removal ratio calculated by SRIM.

Gas of GDC	Etching depth [nm]	Implantation depth [nm]	Fraction of deuterium in etching depth [%]	Fraction of deuterium in from etching depth to implantation depth [%]	Total deuterium removal ratio speculated by SRIM calculation [%]	Total hydrogen isotope removal ratio speculated by SRIM calculation [%]
He	5.3	2.1	34.9	20.1	34.9 ~ 55.0	69.8 ~ 100
Ne	1.3	1.5	3.5	8.7	3.5 ~ 12.2	7.0 ~ 24.4
Ar	0.7	1.7	1.1	9.2	1.1 ~ 10.3	2.2 ~ 20.6

of stainless steel by neon or argon GDC was extremely large compared with those of the graphite. However, thickness of re-deposition layer of sputtered particle increased with the etching depth, and re-incorporation of the hydrogen isotope in the re-deposition layer also became large. Therefore, hydrogen isotope removal ratio by neon or argon GDC in the stainless steel resulted in close value with those in the graphite in spite of large etching depth in the stainless steel. On the other hand, hydrogen isotope removal ratio in the tungsten was very small regardless of gas of GDC. This might result from that the surface structure of the tungsten. In the tungsten, oxygen or carbon impurities in the bulk were chemical etched and the voids were formed by D<sub>2</sub> GD, hence the tungsten grains at the top surface were scraped off. Thereby, the pores were formed at the top surface and the inert gas ions were difficult to be uniformly injected to surface layer. Therefore, carbon impurities, which remained in the pore and retained hydrogen isotope, were not removed efficiently by inert gas GDC, and hydrogen isotope removal ratio in the tungsten was speculated to become very small.

#### 4. Conclusion

The amount of hydrogen isotope retention and its removal by inert gas glow discharge cleaning, and the inert gas retention in graphite was examined. In this study, the

results were obtained as follow.

1. The hydrogen isotope retention in graphite was estimated to be  $1.0 \times 10^{17}$  cm<sup>2</sup>, which is approximately one order of magnitude larger than that in 316L SS and in tungsten. This result is owing to the formation of active carbon bonds.
2. The removal ratio of hydrogen isotope by helium, neon or argon glow discharge cleaning were 49.0%, 21.7% and 6.5%, respectively. These results were explained by both the ion implantation depth and the etching depth.
3. The removal ratio in graphite was similar to those for the stainless steel, but larger than the tungsten. This might result from the influence of re-incorporation of hydrogen isotope by re-deposition layer, and the surface structure.

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