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Fuel hydrogen retention of tungsten and the reduction by inert gas glow discharges

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Abstract

Polycrystalline tungsten was exposed to deuterium glow discharge followed by He, Ne or Ar glow discharge. The amount of retained deuterium in the tungsten was measured using residual gas analysis. The amount of desorbed deuterium during the inert gas glow discharge was also measured. The amount of retained deuterium was 2-3times larger compared with a case of stainless steel. The ratios of desorbed amount of deuterium by He, Ne and Ar glow discharges were 4.6, 3.1 and 2.9 %, respectively. These values were one order of magnitude smaller compared with the case of stainless steel. The inert gas glow discharge is not suitable to reduce the fuel hydrogen retention for tungsten walls. However, the wall baking with a temperature higher than 700 K is suitable to reduce the fuel hydrogen retention. It is also shown that the use of deuterium glow discharge is effective to reduce the in-vessel tritium inventory in fusion reactors through the hydrogen isotope exchange.

1. Introduction

Control of fuel hydrogen retention is quite important both for reductions of in-vessel tritium inventory and hydrogen recycling in fusion reactors. The in-vessel tritium inventory and the fuel hydrogen recycling affect safety issues of fusion reactors and the confinement of fusion plasmas, respectively. Tungsten [1] is regarded as a candidate for the plasma facing material of fusion reactors, so that the fuel hydrogen retention and the tritium inventory of tungsten have to be investigated. The performance of the control

method on the fuel hydrogen retention such as inert gas glow discharge has to be clarified. In this study, gas retention of polycrystalline tungsten is measured by a residual gas analysis (RGA) in a glow discharge apparatus [2-6] with liner walls of polycrystalline tungsten. The liner wall is taken cathode, so that the ion species is implanted into the tungsten. First, the deuterium retention is measured by using D₂ glow discharge. The hydrogen retention is also measured by using H₂ glow discharge to find the effect of hydrogen isotopes on the retention. The deuterium glow discharge followed by the inert gas (He, Ne or Ar) glow discharge is conducted to measure the amount of desorbed deuterium during the inert gas glow discharge [3-6]. The effect of the inert gas glow discharge on the reduction of deuterium retention is investigated. Similar experiments were previously conducted for the case of stainless steel (SS) wall [2]. The present results are compared with the case of SS. The experiment on the hydrogen isotope exchange is also conducted. From this experiment, the exchange ratio of tritium by deuterium during the deuterium glow discharge is evaluated. The small tungsten sample irradiated by the deuterium glow discharge is prepared. The desorption spectrum of deuterium is obtained by using a thermal desorption spectroscopy (TDS). The wall temperature required to reduce the fuel hydrogen retention as well as the tritium retention is obtained.

2. Experiments

In the glow discharge apparatus shown in Fig.1, the discharge pressure and the discharge time were taken 8 Pa and 2 h, respectively, for every type of the discharge. The anode voltage was in the range from 300 - 400 V. The anode is made by copper and the chamber made by SS is grounded (cathode). Inside of the chamber wall is fully covered by sheets of polycrystalline tungsten with a purity of 99.9% (Nilaco). The average grain size is approximately 100 nm. The ion fluence is obtained by measuring the ion current from the chamber to the earth. The ultimate pressure of the vacuum chamber is 10⁻⁶ Pa. Numerous gas such as H₂, D₂, He, Ne or Ar is driven to the chamber with a constant flow rate through a mass flow controller (MFC). The gas retention is obtained by a residual gas analysis (RGA). Before the start of the glow discharge, the gas is supplied with a constant flow rate. Just after the glow discharge turns on, the ion species is implanted into the tungsten, so that the gas pressure drops. The gas pressure then recovers to the previous value after the ion implantation saturates. The change of the partial pressure is monitored by a quadrupole mass spectrometer (QMS). The amount of the decreased gas pressure corresponds to the amount of retained gas.

In the glow discharge apparatus, a sample holder is placed at the center of the chamber as shown in Fig.1. The sample holder is also grounded. Small tungsten samples are placed on the holder. After the glow discharge, these samples are extracted from the chamber. Thermal desorption spectroscopy (TDS) is conducted in order to look the gas desorption behavior. The depth profile of the atomic composition is also analyzed by using Auger electron spectroscopy (AES). The surface morphology is observed both by atomic force microscopy (AFM) and scanning electron spectroscopy (SEM).

3. Results

The hydrogen glow discharge was conducted as shown in Fig. 2. The implantation saturated approximately within 60 min. The partial pressure drop δp was integrated with the time t to obtain $\int \delta p dt$ [Pa·s]. The amount retained at the wall surface δQ was obtained as $\delta Q = \int \delta p dt / S$ [Pa·m³], where S [m³/s] is the pumping speed. Then, the amount of retained hydrogen can be obtained. The amount of retained hydrogen was (5 – 6) $\times 10^{16}$ H/cm². The deuterium glow discharge was similarly conducted as shown in Fig. 3. The partial pressure of D₂ dropped but the partial pressures of HD and H₂ increased. The implanted D and the background H in W produce HD and H₂. The net reduced amount of deuterium gas pressure corresponds to the amount of retained deuterium, the deuterium retention. This value was $\sim 5.0 \times 10^{16}$ H/cm², which is very close to the amount of retained hydrogen. We repeated the measurements for both the hydrogen retention and the deuterium retention. The significant difference between hydrogen and deuterium retentions was not found. Thus, the tritium retention is regarded as the same as the hydrogen or deuterium retention. This result was same in the case of SS. However, the amount of retained deuterium or hydrogen in tungsten was (2-3) times larger than that in SS. This reason is explained later.

The tungsten wall was exposed to the H₂ glow discharge until the hydrogen retention saturated. After that, the deuterium glow discharge was conducted to replace the hydrogen into the deuterium. The result was similar with Fig. 3. Namely, the desorption of H in forms of HD and H₂ occurred due to the isotope exchange between H and D. The ratio of exchanged H by D was ~ 60 % in the 2 h deuterium glow discharge. This result indicates that the ratio of tritium exchanged by deuterium during the deuterium glow discharge becomes ~ 60 % when the deuterium glow discharge is carried out for the wall containing the tritium in fusion reactors. The exchange of hydrogen isotopes is quite effective to reduce the tritium retention at the surface of tungsten wall.

The depth profiles of atomic composition before and after the deuterium glow

discharge were shown as Fig. 4. Even after the deuterium discharge, both carbon and oxygen contents slightly increased. Namely, the carbon content that well traps the deuterium was not decreased. In the case of SS, the carbon concentration almost disappeared after the deuterium glow discharge. Figure 5 shows the desorption spectra of gases containing deuterium. The large desorption peak of hydrocarbon was observed. This peak temperature is relatively low, 700 K. It is noted that desorption of water ($m/e=18$) was little observed. The tungsten surface became rough after the deuterium glow discharge (Fig.9 (b)). These results suggest that most of deuterium is trapped by the carbon content, and the deuterium retention of the tungsten became larger compared with the case of SS since the carbon content is hardly removed.

The inert gas glow discharges using He, Ne and Ar were conducted after the deuterium discharge (Figs. 5-7). The deuterium discharge was done until the deuterium retention saturated. During the inert gas discharge, the partial pressure of deuterium increased. The ratio of desorbed deuterium was obtained from this pressure rise. The removal ratios of the deuterium retention in He, Ne and Ar discharges were 4.6 %, 3.1 % and 2.9 %, respectively. This ranking is proportional to the implantation depth of inert gas ion calculated from SRIM Code as shown in Fig.8. This ranking is also inversely proportional to the deposition rate owing to the sputtered particles. The amounts of retained He, Ne and Ar were 1/10, 1/100 and 1/1000 of the deuterium retention, respectively. This ranking is also proportional to the implantation depth. The helium retention is relatively large, so that the helium may emit into the plasma during the plasma discharge. However, the emission of Ne or Ar may be ignored since the retention of Ne or Ar is very small. The removal ratio for the deuterium retention in tungsten was one order of magnitude smaller than that in SS. In the present experiment, polycrystalline tungsten with a grain size of 100 nm was employed. As shown in Fig.9, the surface became quite rough after the plasma irradiation, perhaps owing to the drops of grains weakly bonded by the ion bombardment. The deuterium trapped in the carbon at the bottom region is little removed by the bombardment of inert gas ions, so that the removal ratio becomes small. On the other hand, the surface of SS remained flat even after the glow plasma discharge. This is the reason why the removal ratio of deuterium retention in tungsten was significantly smaller, compared with the case of SS. If the surface of tungsten is kept flat, the carbon content retaining a large amount of deuterium is removed by the inert gas discharge and then the removal ratio shall be significantly increased.

4. Summary

Polycrystalline tungsten was exposed to deuterium glow discharge. The amount of retained deuterium in the tungsten was measured using residual gas analysis. The amount of retained deuterium was several times larger compared with a case of stainless steel. This reason is due to the growth of rough surface and the existence of carbon content. He, Ne and Ar glow discharges were conducted after the deuterium glow discharge to reduce the deuterium retention. The ratio of removed deuterium amount during the inert gas glow discharge was measured. The ratios of desorbed deuterium amount by He, Ne and Ar glow discharges were 4.6, 3.1 and 2.9 %, respectively. These values were one order of magnitude smaller compared with the case of stainless steel. Namely, the deuterium trapped at the rough surface was little removed by the inert gas ion bombardment. Thus, the inert gas glow discharge is not suitable to reduce the fuel hydrogen retention.

However, if the wall baking with a temperature higher than 700 K is carried out, most of retained deuterium can be desorbed. The in-vessel tritium inventory is similarly reduced significantly if the wall baking is done.

The present systematic results on fuel hydrogen retention, the reduction of deuterium retention by inert gas discharge, the hydrogen isotope exchange and the deuterium desorption spectrum are quite useful to control the in-vessel tritium inventory and fuel hydrogen recycling in fusion plasma.

Acknowledgements

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Figure captions

Fig.1 Glow discharge apparatus.

Fig.2 Change of H_2 partial pressure during H_2 glow discharge.

Fig.3 Changes of D_2 , HD and H_2 partial pressures during D_2 glow discharge.

Fig.4 Depth profile of atomic composition before and after D_2 glow discharge.

Fig.5 Thermal desorption spectra of gases containing deuterium in the sample irradiated by D_2 glow plasma.

Fig.6 Changes of He, D_2 , HD and H_2 partial pressures during He glow discharge.

Fig.7 Changes of He, D_2 , HD and H_2 partial pressures during Ne glow discharge.

Fig.8 Changes of He, D_2 , HD and H_2 partial pressures during Ar glow discharge.

Fig. 9 Change of surface morphology after plasma irradiations, (a)before irradiation, (b)after D_2 plasma irradiation, and (c)after D_2 plasma irradiation followed by He plasma irradiation.

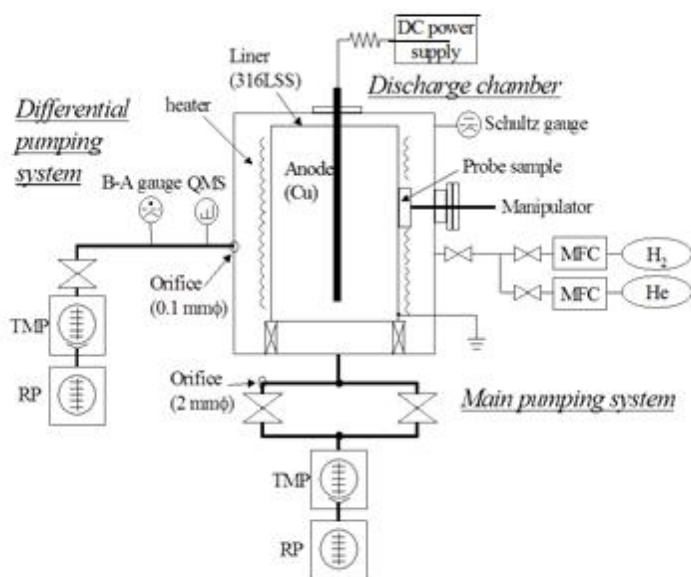


Fig.1 Glow discharge apparatus.

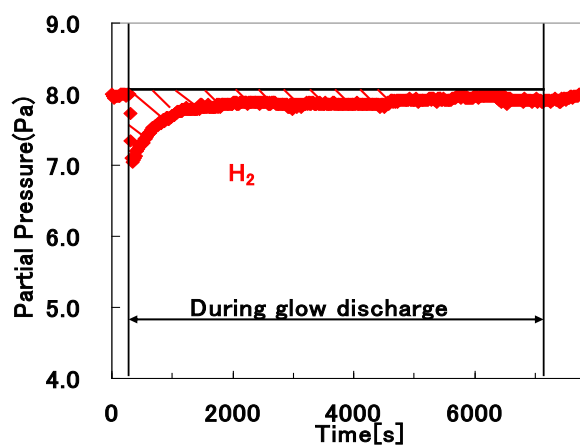


Fig.2 Change of H_2 partial pressure during H_2 glow discharge.

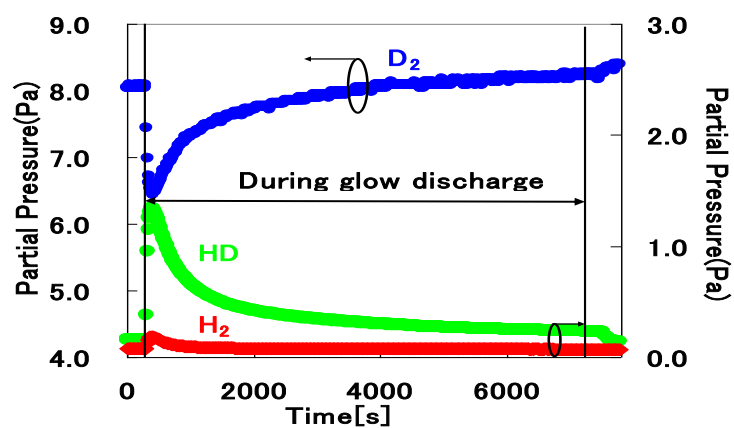


Fig. 3 Changes of D₂, HD and H₂ partial pressures during D₂ glow discharge.

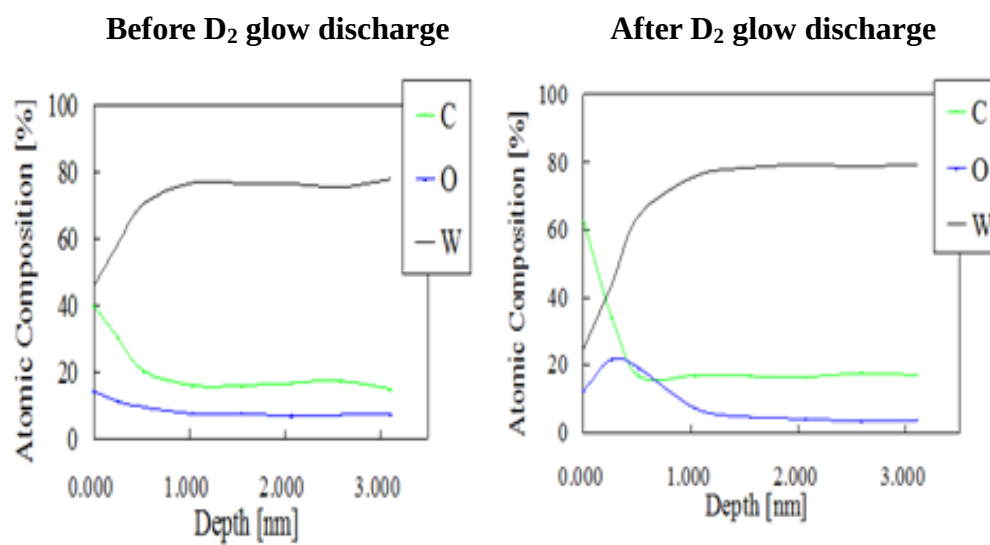


Fig.4 Depth profile of atomic composition before and after D₂ glow discharge.

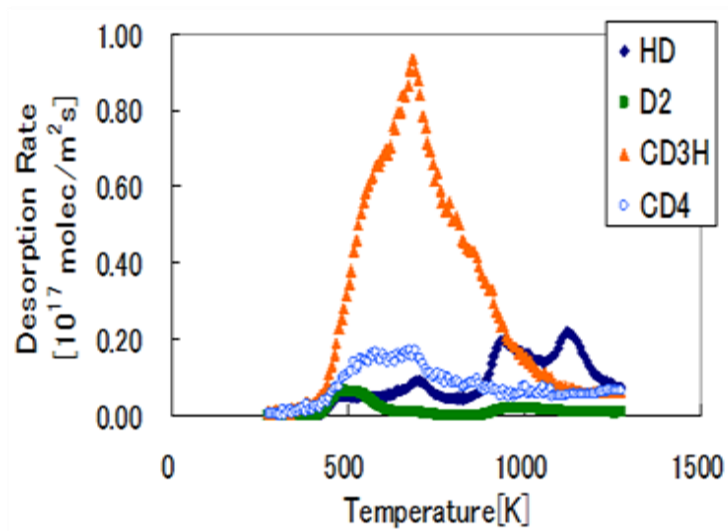


Fig.5 Thermal desorption spectra of gases containing deuterium in the sample irradiated by D_2 glow plasma.

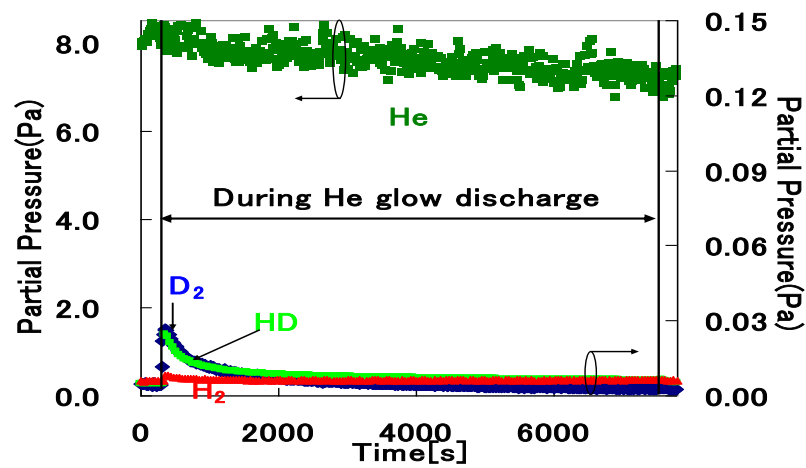


Fig.6 Changes of He, D_2 , HD and H_2 partial pressures during He glow discharge.

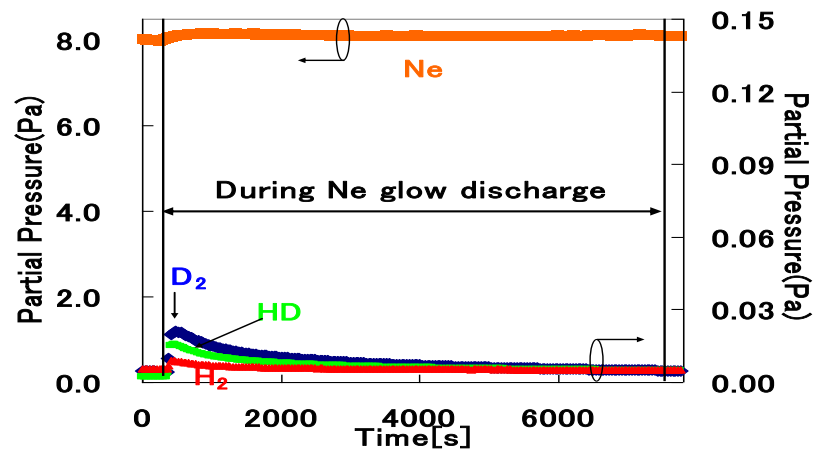


Fig.7 Changes of He, D₂, HD and H₂ partial pressures during Ne glow discharge.

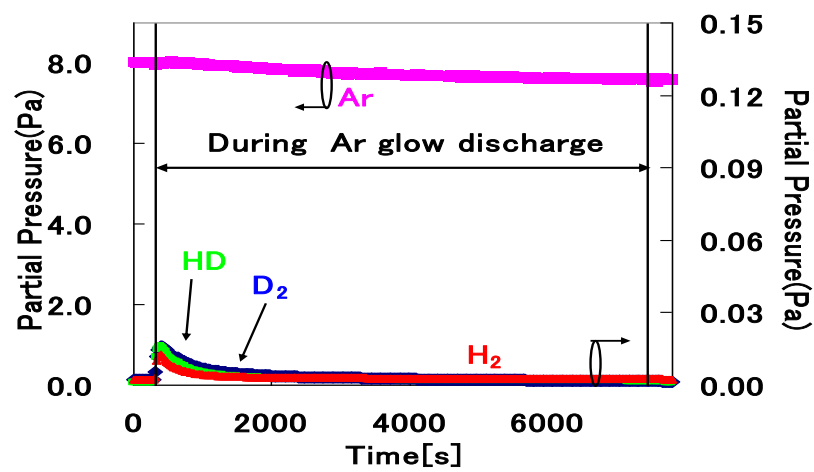
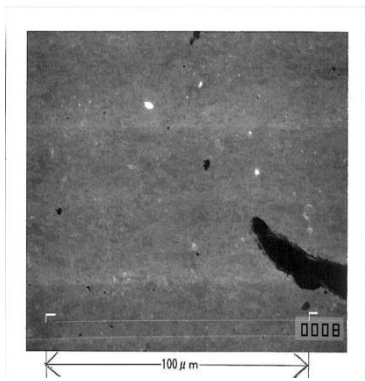
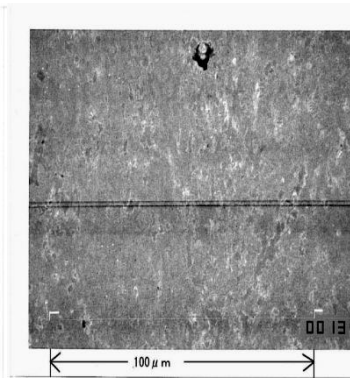


Fig.8 Changes of He, D₂, HD and H₂ partial pressures during Ar glow discharge.

(a) Before Irradiation



(b) After D_2 Irradiation



(c) After D_2 and He Irradiations

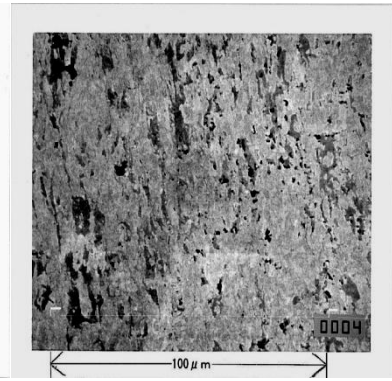


Fig. 9 Change of surface morphology after plasma irradiations, (a) before irradiation, (b) after D_2 plasma irradiation, and (c) after D_2 plasma irradiation followed by He plasma irradiation.