Study on plasma – tungsten surface interactions using the new experimental device EXPRESS

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An experimental apparatus named as EXPRESS (Effluence and Exchange Probe for Recycling Estimation at NIFS and Shizuoka University) has been designed and established in Shizuoka University to acquire laboratory data (well-conditioned data) needed for the design of an actual reactor. The behaviors of deuterium retention and the surface chemical state after EXPRESS plasma operation were evaluated and compared with that for LHD (Large Helical Device, NIFS) hydrogen plasma exposed sample. It was found that a thin impurity deposition layer with the thickness less than 0.3 nm was formed on the surface, which was one thirteenth thinner than that on LHD plasma exposed tungsten. The TDS experiment showed that major deuterium desorption stage for EXPRESS plasma exposed tungsten was located at the lower temperature less than 550 K, although that for LHD plasma exposed one was found at the temperature between 550 K – 1000 K. These facts suggest that the impurity concentration in the deposition layers have a large influence on the deuterium retention behavior in tungsten.

Keywords: deuterium plasma exposure, TDS, XPS, impurity deposition layer, tungsten

1. Introduction

Tungsten will be used as a plasma facing material in future fusion devices. Many studies have been devoted to elucidate fundamental tritium retention and transport behaviors in tungsten under well-controlled condition [1-3]. However, it is quite difficult to apply these data obtained under well-controlled condition to actual and larger-scale plasma devices or fusion devices where conditions are much more complex. For example, a wall temperature gradient will arise when the plasma is controlled. Therefore, the key parameters should be acquired to complete laboratory data (well-controlled condition data) needed for the design of an actual reactor design. The design of a new plasma experimental device, namely EXPRESS (Effluence and Exchange Probe for Recycling Estimation at NIFS and Shizuoka University) was designed to clarify the hydrogen isotope recycling dynamics. This devise has these following features: i) many samples can be placed on the sample stage for plasma exposure and the sample temperature gradient can be controlled, and ii) 1 GBq / day of tritium can be handled.

As a preliminary experiment, the deuterium plasma exposure to pure tungsten was performed in this study to characterize and evaluate the behaviors of the deuterium retention and the surface chemical state after EXPRESS plasma operation at R.T. These experimental results obtained at EXPRESS were compared with those in LHD (Large Helical Device, NIFS) plasma exposed tungsten and only D_2^+ implanted tungsten for the evaluation of fuel recycling during plasma-surface interactions in EXPRESS.

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2. EXPRESS Apparatus

The recently designed plasma experimental device to investigate hydrogen isotope recycling behavior and plasma surface interaction, EXPRESS consists of three parts: i) a plasma generator and plasma exposure system, ii) a fuel storage and purification system, and iii) a fuel recovery-supply systems. Figure 1 shows the schematic view and picture of the plasma generator and plasma exposure system. The plasma generator and plasma

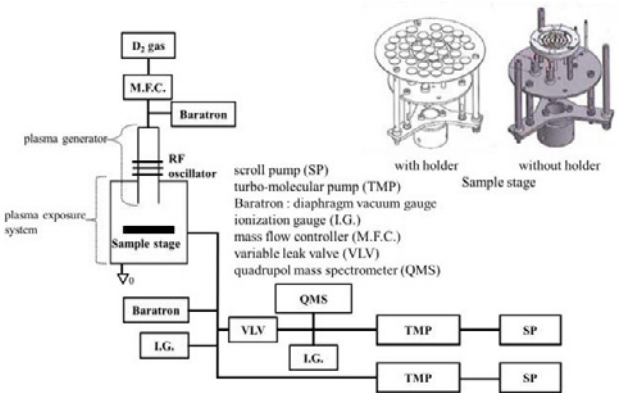

Fig. 1 Schematic view and picture of the plasma generator and plasma exposure system in EXPRESS.

exposure system can be evacuated to 1×10^{-5} Pa by a scroll pump (SP) and a turbo-molecular pump (TMP) combination. The pressure in the system is measured by Baratron and ionization gauge (I.G.). The source gas is supplied through a mass flow controller (M.F.C.) at a flow rate of 0.1-5.0 sccm into the plasma generation tube made by quartz with a size of 150 mm in length and 42 mm in diameter. The excitation frequency is fixed at 13.56 MHz by a RF matching box. The bias voltage can be changed from 0- 200 V between the chamber wall and the sample stage to control implantation energy similarly larger-scale plasma devices. The size of sample stage is 100 mm in diameter, which is made of tungsten. A heating unit is installed beneath the center of sample stage to generate a temperature gradient up to 200 K between the center and the edge of the sample stage as shown in Fig. 2. This apparatus is used to investigate the study of plasma-surface interactions on the first wall of actual reactor. The heating unit can heat up to 873 K at the center of stage. The ion flux can be measured by a faraday cup installed in the plasma exposure system.

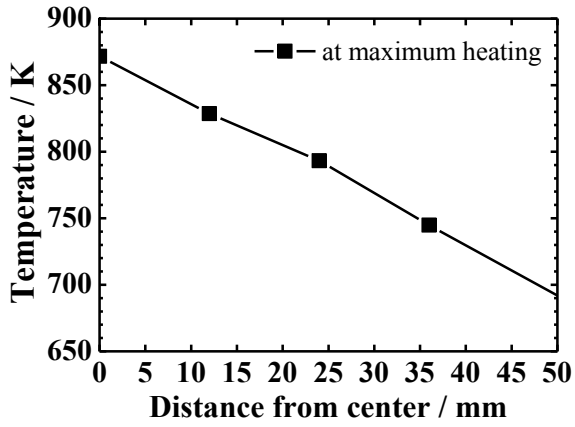

Fig. 2 The maximum temperature profile on the sample stage in EXPRESS.

3. Experimental

A disk-type polycrystalline tungsten with stress-relieved conditions (heated at 1173 K for 1 hour), which was purchased by Allied Material Co. Ltd, was used for all experiments. The sample size was 10 mm diameter and 0.5 mm thickness. The samples were polished mechanically to the roughness of less than 1 μm by SiC abrasive papers and 1-3 μm diamond suspensions, and preheated at 1173 K for 30 minutes under ultrahigh vacuum (< 10^{-6} Pa) to remove the impurities and damages introduced during the polishing processes.

After preheating, the sample was introduced into the plasma exposure system of EXPRESS. The deuterium gas was supplied at a flow rate of 3.0 sccm and the pressure of 3.5 Pa. The D plasma was exposed to the samples with the D_2^+ energy of 63 eV to implant at the lowest possible energy for the stable plasma operation. The ion flux was 3.4×10^{19} D^+ m^{-2} $s^{-1}$and the fluence was 1.0×10^{24} D^+ m^{-2} at R.T. This sample was named as EXPRESS sample.

After plasma exposure, TDS (Thermal Desorption Spectroscopy) were performed from R.T. to 1173 K with a heating rate of 0.5 K s^{-1} to investigate the deuterium desorption and retention behaviors. The chemical states and the thickness of the impurity layer on tungsten were evaluated by XPS (X-ray Photoelectron Spectroscopy) (ESCA1600 system, ULVAC-PHI Inc.) using a Mg - Kα X-ray source (1253.6 eV) and 1 keV Ar^+ sputtering technique [4]. The thickness of impurity deposition layer was defined as the depth where impurity concentration was reached to be half compared to that on the surface. The sputtering rate by Ar^+ implantation was estimated by AFM (Atomic force microscope).

Two types of tungsten samples, namely D_2^+ imp. sample and LHD sample, were prepared to compare the deuterium retention behavior. In the D_2^+ imp. sample, 3.0 keV D_2^+ implantation was performed for tungsten with flux of 1.0×10^{18} D^+ m^{-2} s^{-1} and fluence of 1.0×10^{22} D^+ m^{-2} at R.T. In the case of LHD sample, the sample was exposed to 134 shots (exposure time 271 seconds) of hydrogen plasma in LHD, with the ion energy of about 10-100 eV and the temperature of less than 373 K, at a first wall position in the plasma operation campaign of 2011.

4. Results and discussion

Figure 3 shows the TDS spectra for EXPRESS and LHD and D_2^+ imp. samples. The TDS spectrum for the D_2^+ imp. sample consists of two desorption stages at 400 K and 550 K, attributing to the desorption stages of deuterium adsorbed on the surface and/or trapped by dislocation loops (Peak 1), [5,6] and deuterium trapped by vacancies (Peak 2) [7] respectively. In the D_2 desorption stages for the EXPRESS sample, Peak 1 was the dominant stage which is similar for the D_2^+ imp. sample, but the ratio of Peak 1 to

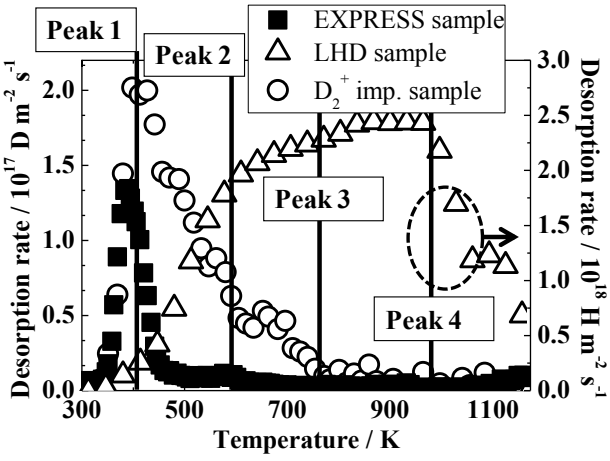

Fig. 3 TDS spectra for the EXPRESS, LHD and D_2^+ imp. samples

Peak 2 was quite different among these samples, It can be said that the plasma exposure hardly produces the irradiation defects in tungsten due to lower D^+ implantation energy according to SRIM code calculation [8], indicating that the formation of irradiation defects was limited for EXPRESS sample, and most of deuterium would be accumulated on the sample surface. In the LHD sample, additional desorption stages were observed at around 750 K (Peak 3) and 900 K (Peak 4). It was reported that the W-O bond in WO_3 was one of the stable deuterium trapping sites with a desorption temperature of around 750 K [9]. In our previous study, the deuterium implanted into HOPG was trapped as C-D bond after dissociation of C-C bond, which was desorbed at 900 K [10]. Therefore, the desorption stages of deuterium at these higher temperatures in the LHD sample may be assigned to that bound to oxygen as O-H (Peak 3) and carbon as C-H bonds (Peak 4), respectively.

Figure 4 shows W-4f XPS spectra for the top surfaces of the EXPRESS, LHD and D_2^+ imp. samples. The W- $4f_{7/2}$ XPS spectra consisted of two chemical states located at 30.8 eV for metallic W and 35.8 eV for WO_3. [11,12]. The abundance of WO_3 for total W-4f XPS peak area was 23% for the EXPRESS sample, and 63% for the LHD sample as shown in Table 1. In the EXPRESS sample, the major chemical state of tungsten was the metallic state which was different from the LHD sample, indicating that most of deuterium for the EXPRESS sample would be interacted with metallic W rather than WO_3. Figure 5 shows the depth profiles of W, WC, WO_3 and C for the EXPRESS sample. The impurities were accumulated within the depth of ~ 0.3 nm for the EXPRESS sample. Table 2 summarizes atomic concentrations on the top surface and thickness of impurity deposition layers on the EXPRESS and LHD samples. Large amount of oxygen and carbon were accumulated on the surface for both samples and its impurity concentrations were almost the same. However, the thickness of the impurity deposition layer for the EXPRESS sample was about one thirteenth thinner than that for the LHD sample. It can be said that the thin impurity deposition layer was achieved by EXPRESS device and the formation of impurity deposition layer hardly affected on deuterium retention as O-D bonds for the EXPRESS sample, resulting in decrease of Peak 3 in the TDS spectrum of the EXPRESS sample compared to the LHD sample.

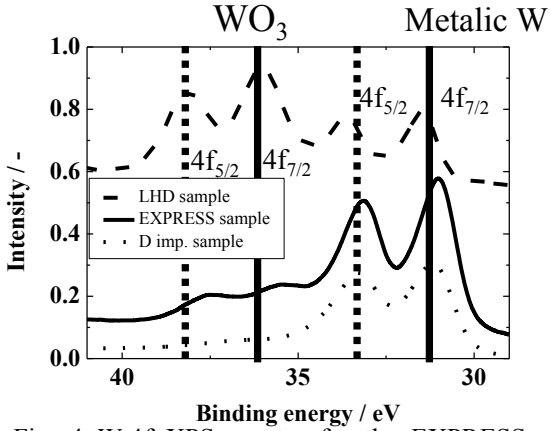

Fig. 4 W-4f XPS spectra for the EXPRESS, LHD and D_2^+ imp. samples

Table 1 Ratios of W and WO_3 on the top surface of W

	W	WO_3
EXPRESS	77%	23%
LHD	37%	63%
D imp.	86%	14%

Figure 6 shows C-1s XPS spectra of EXPRESS, LHD and D_2^+ imp. samples. Three chemical states were found at the binding energies of 286.1 eV, 284.8 eV and 283.3 eV, attributing to C-O, C-C and C-W bonds, respectively [13-15]. C-O and C-C bonds were major chemical states in the LHD sample, although C-C bond was formed by plasma exposure for the EXPRESS sample. However, implantation depth of carbon was limited within the surface region and the total amount of carbon for the EXPRESS sample was quite small compared to that for the LHD sample indicating that carbon would hardly affect the behavior of hydrogen isotope retention in the EXPRESS sample.

It was concluded that the EXPRESS sample had the thinner impurity deposition layer compared with LHD sample and hydrogen isotope retention is affected by carbon and oxygen chemical states and the amounts of impurity in deposition layer. In the EXPRESS sample, most of the impurities were adsorbed on the top surface

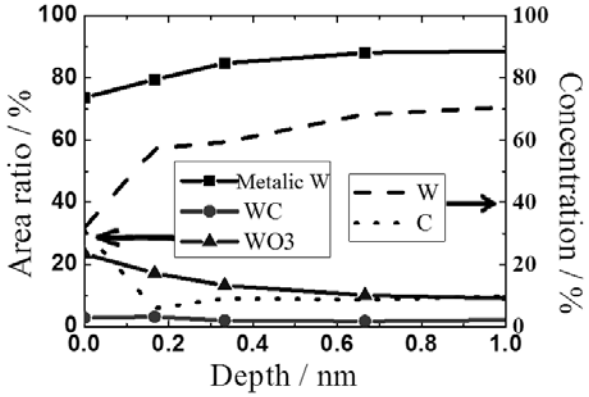

Fig. 5 Depth profiles of W and C for the EXPRESS sample

Table 2 Atomic concentrations for the surface of the EXPRESS and LHD samples and the thickness of the impurity deposition layers.

	W	C	O	Thickness
EXPRESS	34%	30%	36%	0.3 nm
LHD	18%	34%	48%	4 nm

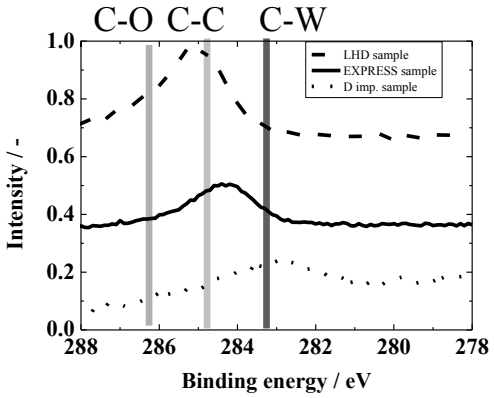

Fig. 6 C-1s XPS spectra for the EXPRESS, LHD and D_2^+ imp. samples.

after deuterium plasma exposure. However, the amounts of impurities beneath the surface were too small to affect the deuterium retention behavior. In addition, the deuterium was hardly trapped by irradiation defects because the formation of defects was limited due to low energy ion irradiation. Therefore, the deuterium was only retained by surface adsorption during deuterium plasma exposure in EXPRESS.

In further experiments, longer time (higher fluence) plasma exposure and oxygen and carbon contaminated plasma exposure will be needed to make impurity deposition layers that have a variety of thickness and chemical state in order to clarify the influence of impurity deposition layer on deuterium retention behavior.

5. Conclusions

A new equipment to acquire laboratory data (well-conditioned data) needed for the design of an actual reactor named EXPRESS has been designed and is establishing. The deuterium plasma exposure was performed to characterize and evaluate the behaviors of deuterium retention and surface chemical states after the EXPRESS plasma operation. The deuterium retention behaviors for tungsten after EXPRESS plasma exposure was studied and these results were compared to those for the LHD and D_2^+ imp. samples. It was found that most of deuterium was desorbed at the lower temperature less than 550 K for the EXPRESS sample, which was almost the same with the D_2^+ imp. sample. However, the TDS spectrum for the LHD sample was quite different from that for the EXPRESS and D_2^+ imp. samples, which would be caused by the formation of thick impurity deposition layer on the surface for the LHD sample. These facts indicate that the amounts of impurity in the deposition layer could affect on the deuterium retention behavior in tungsten used in actual plasma devices and fusion reactors.

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