

# Tritium Retention on Stainless Steel Surface Exposed to Plasmas in LHD (II)\*)

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Effects of plasma exposure for tritium retention have been studied using both samples of a plasma-exposed stainless steel and a bare stainless steel. It was seen that the plasma-exposed surface was covered with thin deposition layers. Constituent elements of the deposition layers mainly formed metallic oxides except for C and O, but a part of Fe and Mo was a metallic state. Both samples were simultaneously exposed to tritium gas at room temperature with no heat treatment, and a considerable increase in tritium retention was observed for the plasma-exposed sample. In addition, it was clarified that distribution of tritium retention on the plasma-exposed surface was non-uniform even exposure at room temperature. Increase in tritium retention and non-uniformity of tritium distribution were discussed from viewpoints of distribution and chemical states of constituent elements of the deposition layers.

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## 1. Introduction

It is recognized that reduction of tritium retention in the fusion reactor materials as well as inhibition of tritium permeation is one of great important issues from viewpoints of safety and economy of tritium in the fusion system, since a huge amount of tritium is used in a future fusion reactor.

Retention and permeation of tritium in/through the fusion materials will be changed by surface states of materials, e.g., surface modification of materials by exposure to plasmas. This is due to a reason that adsorption and/or absorption process of tritium is very sensitive for the chemical and physical properties of a material surface.

As a model, retention behavior of tritium for stainless steel samples exposed to plasmas in the Large Helical Device (LHD) has been studied so far by present authors [1, 2]. In this study, stainless steel samples were exposed to plasmas at locations of 1.5U, 5.5U, 6.5L and 9.5L in LHD for one cycle. Surfaces of them were covered with carbon, boron and metallic oxides, although the amount of them was different in each sample. After plasma exposure, they were exposed to tritium gas in the specially designed tritium exposure device. Prior to the tritium exposure, the samples were pre-heated in vacuum under the given tem-

perature condition.

It was seen that plasma exposure resulted in a great increase in the tritium retention when the samples were pre-heated at high temperatures before tritium exposure. In addition, remarkably non-uniform tritium distribution appeared in surface of the 6.5L sample, while surfaces of the other samples were almost uniform. Namely, it is important to study the effects of plasma exposure for evaluation of tritium retention.

In the present study, therefore, effects of pre-heating temperature in vacuum and surface analyses of the deposition layers were intensively studied using a 6.5L sample exposed to plasmas.

## 2. Experimental

The samples used in this study are stainless steel type 316, and size of the sample plate was  $10 \times 10 \times 0.5$  mm. After the sample plate was finally polished with a buff, it was fixed at the location of 6.5L in LHD before beginning of plasma experiments. Then it was exposed to the plasmas for one cycle of the 13th cycle, which was operated for 13 weeks from October 2009. After the plasma experiment, the sample plate (plasma-exposed SS316) was removed from the wall of LHD and then it was provided with surface analyses and tritium exposure experiments.

As a first step, surface of the plasma-exposed SS316

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was observed by a digital microscope. This sample was also used to analyze surface state of deposition layers by means of Laser Raman spectroscopy (JASCO NRS-7100) and X-ray photoelectron spectroscopy (XPS: Thermo Fisher Scientific ESCALAB250X).

Procedures of tritium exposure are as follows. As received sample removed from the 6.5L location was loaded into the specially designed tritium exposure device and it was evacuated below  $6 \times 10^{-7}$  Pa at room temperature. After the evacuation, the sample was exposed to tritium gas diluted with deuterium for 4 hours at room temperature. The pressure of tritium gas was 2.66 kPa. The amount of tritium retention in surface layers of the sample was evaluated by  $\beta$ -ray-induced X-ray spectrometry (BIXS), which was employed argon as a working gas [3], and tritium distribution of a sample surface was examined by the imaging plate technique. Two-dimensional mapping of  $\beta$ -ray dose was obtained by measuring the intensity of photo-stimulated luminescence with the imaging analyzer (Fujifilm FLA-7000). The spatial resolution was  $25 \times 25 \mu\text{m}$ .

A bare stainless steel sample (bare SS316) was also used to compare with tritium retention for the plasma-exposed SS316. Evacuation and tritium exposure conditions were the same as examinations for the plasma-exposed SS316.

### 3. Results and Discussion

#### 3.1 Surface observation

Figure 1 shows a photograph of the surface exposed to plasmas for one cycle at the location of 6.5L in LHD. Although it is not clear from the photograph, color change of the sample surface was observed. Remarkable change of color appeared in the plasma-exposed surface: the surface was looked like black. Such color change is due to deposition of materials on the surface. Thickness of a deposition layer has not been measured yet. However, as an example, photograph of the cross section of deposition layers (*ca.*  $2 \mu\text{m}$ ) is described in elsewhere [1]. The surface was covered with thin layers which consisted of B, C, O, Ti, Cr, Fe, Ni and Mo as described later.

Figure 2 shows a Raman spectrum observed for plasma-exposed SS316. The spectrum for Papyex, which is highly oriented graphite, is also described in the figure as a reference spectrum. Two broad peaks appeared in the region of interesting for the plasma-exposed SS316: one is  $1579 \text{ cm}^{-1}$  (Peak I) and the other is  $1356 \text{ cm}^{-1}$  (Peak II). Peak I was assigned to the vibration mode of  $E_{2g}$ , and Peak II was assigned to the vibration mode of  $A_{1g}$ . Raman shifts of these two peaks observed for the plasma-exposed SS316 agreed well with those observed for Papyex, although peak width and the peak ratio of (Peak II/Peak I) was largely different. Namely, these results indicate that a part of carbon deposited on the plasma-exposed SS316 is graphitization and the grain size of graphite structure is very small.

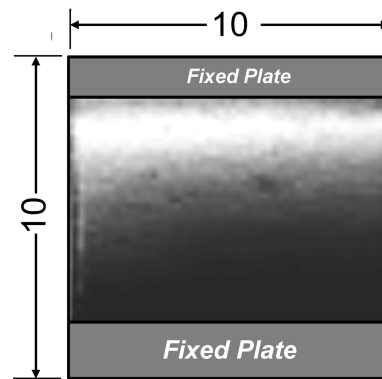


Fig. 1 Photo of the SS316 sample surface exposed to plasmas. The sample was kept with a fixed plate on the wall.

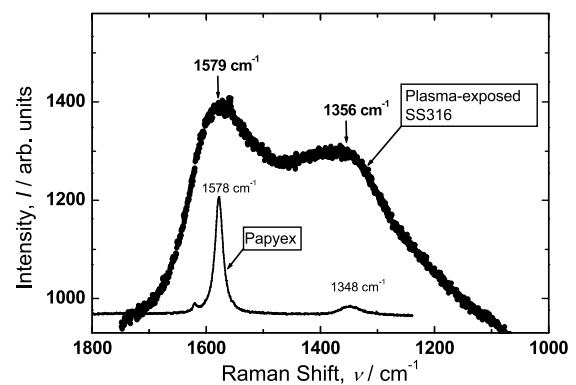


Fig. 2 Raman spectrum for the plasma-exposed surface.

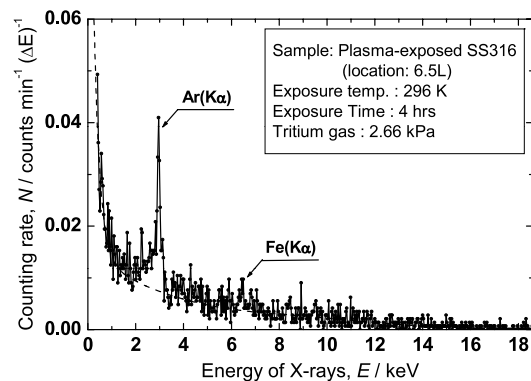


Fig. 3 X-ray spectrum observed by BIXS after exposure to tritium gas at room temperature for the plasma-exposed SS316.

#### 3.2 Tritium retention

Figure 3 shows an X-ray spectrum observed for plasma-exposed sample. The X-ray spectrum was observed by means of BIXS. The dashed line in the figure shows the background level, which was obtained by measuring for 100 hours by the present X-ray detector. A sharp peak appeared clearly in the spectrum, which is the characteristic X-rays of  $\text{Ar}(K\alpha)$ . It is known that intensity of  $\text{Ar}(K\alpha)$  X-rays is proportional to the amount of tritium re-

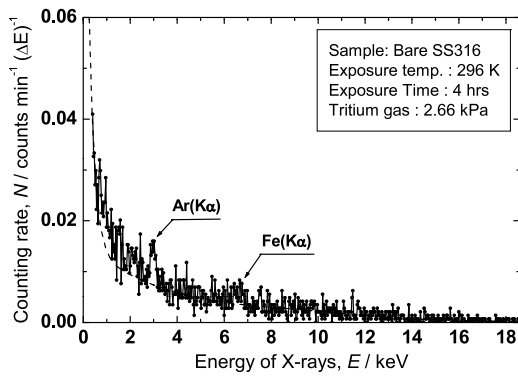


Fig. 4 X-ray spectrum observed by BIXS after exposure to tritium gas at room temperature for the bare SS316.

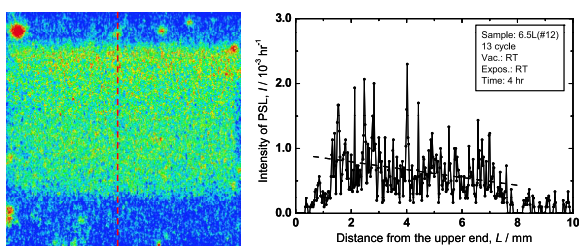


Fig. 5 Tritium distribution observed by a technique of imaging plate for the plasma-exposed SS316 surface.

tained in surface layers of a sample [4]. In addition to the Ar(Kα) peak, a very weak peak can be seen around 6.4 keV. This was assigned to Fe(Kα). This indicates that iron atoms exist near surface of deposition layers, although chemical states of iron atoms are not clear.

In comparison with the plasma-exposed SS316, tritium retention for a bare SS316 sample was examined in a similar manner. The measurement result is shown in Fig. 4. Although two peaks were similarly observed in the X-ray spectrum, their intensities were considerably smaller than those observed in Fig. 3. From these results, it was seen that tritium retention largely increases by exposure to plasmas. This is due to the impurity layers deposited on the surface of SS316.

Figure 5 shows distribution of tritium retained in deposition layers of the plasma-exposed surface, which was observed by the IP technique. The result of line analysis (right) at the red broken line in the IP image (left) is also shown. Tritium concentration is described by a color measure, and tritium concentration increases with color changes from blue to red. Since blue regions in both upper and lower part of the IP image had been covered by fixed plates in LHD, those regions did not expose to plasmas. As clearly seen from the IP image, it was seen that much tritium was adsorbed on the plasma-exposed surface and the tritium retention gradually decreases toward the lower part from the upper part as described by the broken line in the figure of line analysis. Namely, it was clarified that non-

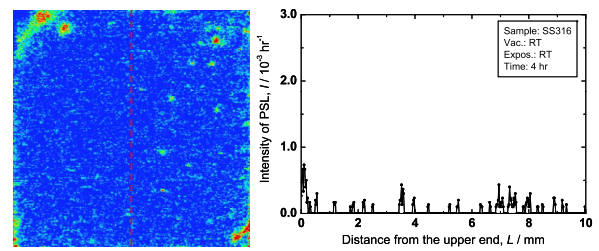


Fig. 6 Tritium distribution observed by a technique of imaging plate for the bare SS316 surface.

Table 1 Summary of BIXS and IP data.

Sample	Intensity	BIXS/cpm			IP/ GBq m <sup>-2</sup>
		Ar(Kα)	Fe(Kα)	Fe(Kα)/Ar(Kα)	
Plasma-exposed SS316		0.135	0.030	0.22	0.105
Bare SS316		0.026	0.014	0.53	0.016
Ratio		5.2	2.1	-----	6.5

uniform distribution of tritium appeared even exposure at room temperature.

To confirm increase in tritium retention in the deposition layers, tritium gas was exposed to the bare SS316 at room temperature, and an IP image was measured with the same manner. The result is shown in Fig. 6. Most part of the IP image was blue, and it is seen that the tritium retention for the bare SS316 surface is very lower than that for the plasma-exposed surface. These results agree with measurement results of BIXS.

Table 1 shows a summary of peak intensities of characteristic X-rays observed by BIXS and the surface tritium concentration estimated from IP measurements. Tritium retention by the plasma-exposed SS316 is 5-6 times greater than that by the bare SS316. In addition, the ratio of Fe(Kα)/Ar(Kα) for the plasma-exposed surface was below 1/2 in comparison with that for the bare SS316 surface. This indicates that the concentration of iron atoms in the deposition layers is lower than that in surface layers of the bare SS316.

### 3.3 Surface analysis

To examine the difference in tritium retention of both samples, distribution of all constituent elements on the surface of deposition layers and chemical states of the elements were analyzed by XPS. The results are shown in Fig. 7. The analyses were carried out continuously from the upper end to the lower end of the plasma-exposed SS316 surface shown in Fig. 1. Width of analyzing part was 200 μm. Intensity of each peak observed for a given element is described by a color measure which is shown in the right side of each analysis data.

Eight elements were observed in the deposition layers on the plasma-exposed surface: B, C, O, Ti, Cr, Fe, Ni and Mo. Titanium is due to usage in the LHD as a titanium sublimation pump. Among these elements, all el-

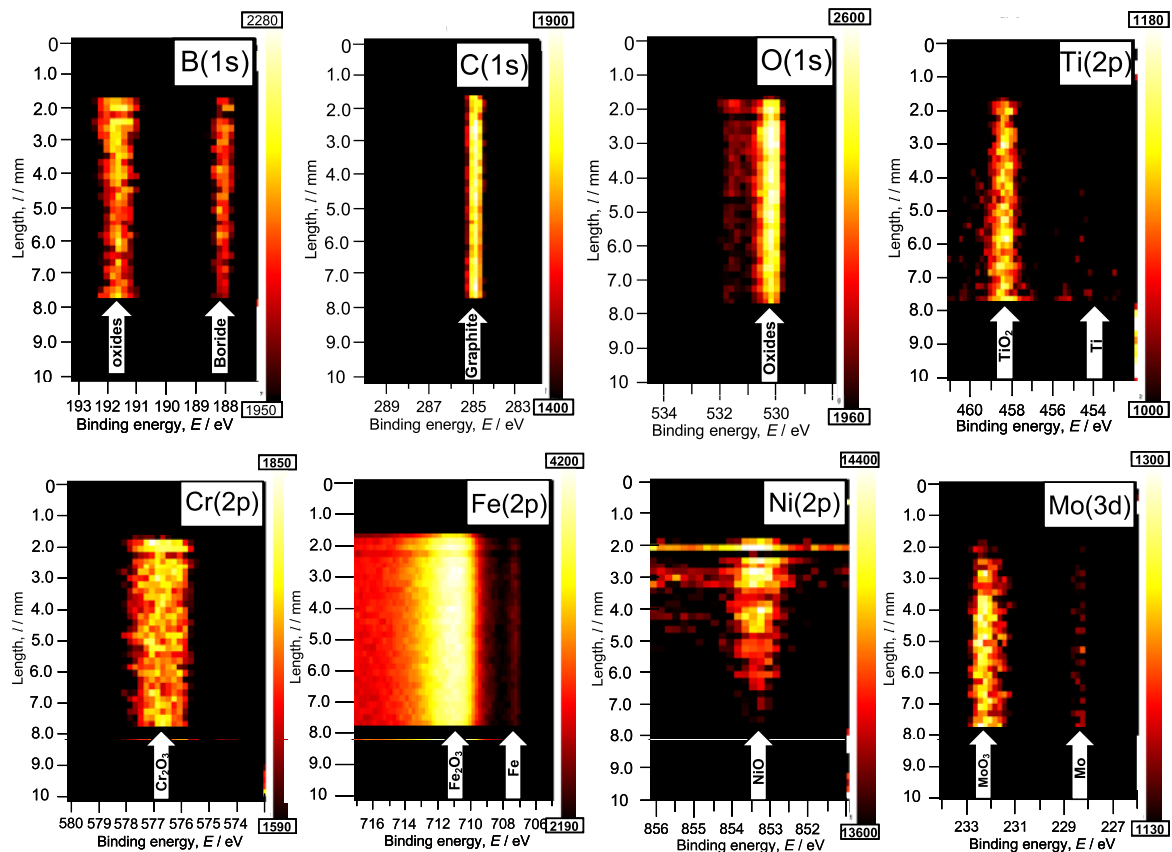


Fig. 7 Vertical distribution of constituent elements on the deposition layer analyzed by XPS.

elements other than carbon and oxygen were basically oxidized, but a part of iron and molybdenum was the metallic states. Furthermore, it should be noticed that only nickel distributed non-uniformly on the surface of deposition layers. It is suggested, therefore, that nickel oxide as well as metallic iron and molybdenum may play an important role for non-uniform distribution of tritium.

#### 4. Summary

Effects of plasma exposure for tritium retention behavior were studied using a plasma-exposed stainless steel, which was prepared by exposing to plasmas for one cycle in the Large Helical Device. Surface of the plasma-exposed sample was analyzed by laser Raman spectroscopy and X-ray photoelectron spectroscopy, and it was seen that the sample was covered with carbon, oxygen, metallic oxides (a part of metal species was metallic state), and so on. Tritium retention for the plasma-exposed sam-

ple was above 5 times larger than that for the bare stainless steel sample, and also non-uniformity of tritium distribution was observed for the plasma-exposed surface even exposure at room temperature. These phenomena were correlated with the distribution of metallic elements and chemical states of them.

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