Tritium Retention on Stainless Steel Surface Exposed to Plasmas in LHD[*]

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Tritium retention of samples exposed to plasmas in the Large Helical Device (LHD) during each campaign in 12th, 13th and 14th cycles has been studied. Small sample plates made of stainless steel type 316L were fixed in advance at four different walls in LHD: location of a sample plate was 1.5U, 5.5U, 6.5L and 9.5L. After plasma exposure in each cycle, these samples were exposed to tritium gas at a temperature of 300 or 623 K. Retention behavior of tritium in surface layers of each sample was mainly examined using β-ray-induced X-ray spectrometry (BIXS) and X-ray photoelectron spectroscopy (XPS). The energy spectra observed by BIXS and XPS showed the depositions of boron, carbon, titanium, chromium, iron, nickel and molybdenum with oxygen. Tritium retention of the samples exposed to plasma increased than that of a bare SS316L sample, although it was largely different in the location of a sample. When the samples were exposed to tritium gas at 300 K, the order of magnitude of tritium retention was as follows: 9.5L≫5.5U>6.5L>1.5U for 12th cycle, 6.5L>9.5>1.5U>5.5U for 13th cycle, and 6.5L>1.5U~5.5U>9.5L for 14th cycle.

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1. Introduction

Since a huge amount of tritium is used in a future thermonuclear fusion reactor, it is well recognized that the reduction of tritium retention in the plasma-facing materials (PFM's) of the fusion reactor is one of great important issues from viewpoints of safety and economy of tritium in addition to controlling fuel particle balance in the reactor core. However, trapping and release behavior of tritium in/from the PFM's will change when they are attacked by high energy particles, because PFM's are always bombarded by neutrons, fuel particles, and the impurity particles. In addition, surface of PFM's is eroded by chemical and physical sputtering, and components of PFM's redeposit on the surface. Namely, it is indispensable to study in advance the effects of exposure to plasmas for tritium retention.

It is also important to examine preliminarily the trapping and release behavior of tritium in the PFM's and construction materials of the Large Helical Device (LHD), because the fusion experiments using deuterium gas have been planned as a next step experiments in LHD [1]. The deuterium experiments are accompanied by the production of neutron and tritium. The maximum quantity of tritium production is estimated to be 55 GBq/year (1.5 Ci/year). A part of tritium generated in the device is exhausted by the vacuum pumps, while the others are mainly retained in the materials of the first wall and diverter. Tritium retention and its removal in/from PFM's and structural materials have been recognized as one of key issues from viewpoints of safety operation and preserve environment.

A part of tritium trapped in the surface layers of PFM's will be chronically released from the materials depending on the temperature and concentration gradient in the materials. Such release behavior is of a serious problem when an operation of the device is stopped and an atmospheric gas is introduced into the device for maintenance of the device. This is due to a reason that a chemical form of tritium released from the materials is mainly the oxidized form of tritium.

From these viewpoints, changes in the tritium retention by plasma exposure were studied using samples exposed to various plasma experiments in LHD during one campaign from the 12th to the 14th cycle.

2. Experimental

The samples used in this study are stainless steel type 316L (SS316L), and size of the sample plates was $10 \times 10 \times 0.5$ mm^3. After these sample plates were finally

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polished with a buff, 12 plates were fixed at different locations in LHD (1.5U, 5.5U, 6.5L and 9.5L) [2] before beginning of a plasma campaign, and exposed to the plasmas during one campaign: namely, 12th cycle was continued for 13 weeks from September 2008, 13th cycle was for 13 weeks from October 2009, and 14th cycle was for 11 weeks from October 2010. After each campaign of plasma experiments, the sample plates were removed from the wall of LHD and then three plates of each location were provided for surface analyses and tritium exposure experiments.

As a first step, surfaces of the plasma exposed samples were observed by a digital microscope. These samples were also used to analyze the depositions and chemical state of surface elements by X-ray photo- electron spectroscopy (XPS) and Raman spectroscopy.

Tritium exposure procedures are as follows. Four kind samples removed from each location were loaded in the tritium exposure device, and those were evacuated below 5×10^{-6} Pa at room temperature or 673 K. After evacuation, the samples were exposed to tritium gas diluted with deuterium at room temperature or 623 K for 4 hours under the given pressure. The amount of tritium retention in the surface layers of a sample was evaluated by β-ray-induced X-ray spectrometry (BIXS) and tritium distribution of a sample surface was examined by the imaging plate technique. The details of BIXS are described elsewhere [3].

3. Results and Discussion
3.1 Surface observation

Figure 1 shows photographs of surfaces exposed to plasmas in the 13th cycle as an example. As clearly seen form the photographs, color of the sample surfaces was different among samples. Remarkable change of color appeared in the 6.5L and 9.5L samples: the surfaces were looked like black. Such color change is due to the materials deposited on the surface. Similar color changes were also observed for samples of the 12th and 14th cycles. Thickness of the deposited materials was different in four samples, although thickness of deposited materials on each sample has not been measured. However, as an example, photograph of the cross section of a deposition layer (*ca.* 2 μm) is described in elsewhere [4].

3.2 Tritium retention

In comparison with the plasma exposed samples, at first, the retention behavior of tritium was examined for a bare SS316L sample which was not exposed to any plasma experiments. This sample was exposed to tritium gas for 4 hr at 623 K. The tritium concentration and total pressure of tritium gas applied to this experiment were 0.6%-T and 1.3 kPa, respectively. Figure 2 shows the X-ray spectrum observed by BIXS after tritium exposure. Three peaks appeared in the X-ray spectrum. These peaks were assigned to characteristic X-rays of Ar(Kα), Cr(Kα) and Fe(Kα) as
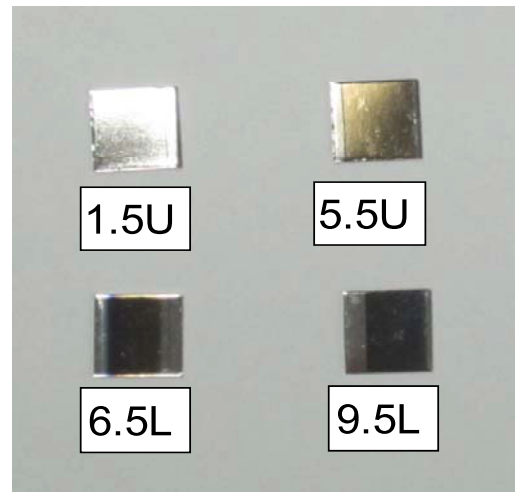

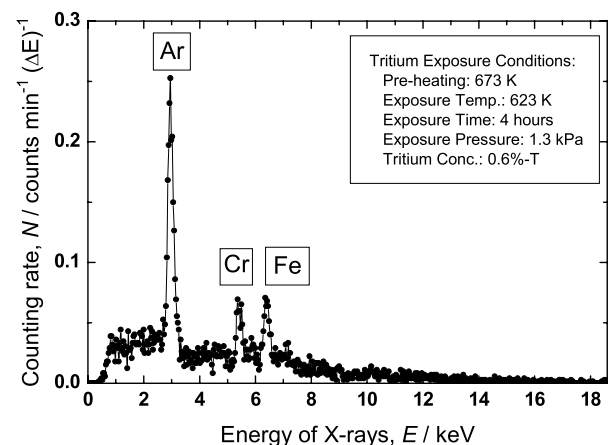Fig. 1 Photograph of the sample surfaces observed by the digital microscope.

Fig. 2 β-ray-induced X-ray spectrum observed for bare SS316L sample.

is shown in the figure. It is known that intensity of Ar(Kα) X-rays is proportional to the amount of tritium retained in surface layers of a sample. Since average energy of β-rays emitted from tritium nuclei is as low as 5.7 keV, an escape depth of β-rays in stainless steel is smaller than 1 μm.

The intensity of Ar(Kα) X-rays observed was 2.7×10^{-2} counts/s. From this intensity, we can estimate the amount of tritium retained in surface layers [5]. The amount of surface tritium was estimated to be 11.1 kBq. Taking the tritium concentration and surface area of the sample into account, the amount of tritium was as much as 8.2 GBq/m^2.

Figure 3 shows an example of X-ray spectrum observed by BIXS for 9.5L sample exposed to plasmas in the 13th cycle. Six characteristic X-ray peaks appeared in the spectrum, which were assigned to the elements of Mo, Ar, Ti, Cr, Fe, and Ni from the lower energy side. From this spectrum, it can be seen that components of stainless steel deposited on the sample surface by erosion of first wall
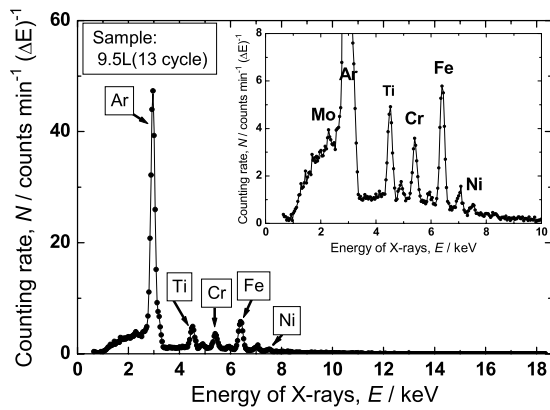
Fig. 3　X-ray spectrum observed for the 9.5L sample exposed to plasmas in the 13th cycle. Tritium concentration; 17.2%-T, total pressure of tritium gas; 0.41 kPa, exposure temperature; 623 K, and exposure time; 4 hr.
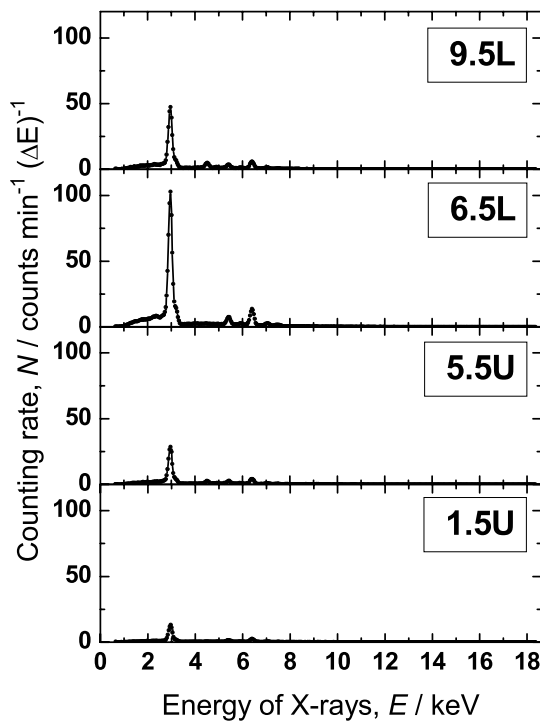

Fig. 5　Changes in the intensity of Ar(Kα) X-rays at room temperature with time.

Fig. 6　Tritium distribution on the surfaces of 6.5L and 5.5U samples measured by imaging plate technique.

623 K for 4 hr. Plural X-ray peaks appeared in every spectrum. However, X-ray intensity was different in each sample. Maximum tritium retention appeared in the 6.5L sample.

To examine the release behavior of tritium retained in these samples, changes in the intensity of Ar(Kα) X-rays at room temperature were tracked for about one month. The tracking result was shown in Fig. 5. It is clearly seen from the figure that only X-ray intensity of the 6.5L sample gradually decreased with time. This indicates that the amount of tritium retention decreased with time by release from the plasma-exposed surface.

After measurements by BIXS, these samples were kept at ambient temperature. About 300 days later, tritium distribution of both surfaces of the 6.5L and 5.5U samples were measured by the imaging plate technique as shown in Fig. 6. Remarkable nonuniformity of tritium distribution was observed for 6.5L, whereas the surface of 5.5U was almost uniform. As was seen from Fig. 5, difference in initial tritium retention between both samples was 3.3 times, and after 300 days the red area in the 6.5L sample

Fig. 4　Comparison of X-ray spectra observed for the samples which were exposed to plasmas at four locations.

of LHD. Titanium was also founded, which was used in the LHD as a sublimation pump. Furthermore, it was seen from XPS analyses that boron, carbon and oxygen were also deposited. The intensity of Ar(Kα) X-rays observed for the 9.5L sample was about 150 times larger than that for the bare SS316L sample shown in Fig. 2, although tritium conditions differed between both samples. Namely, it is suggested that tritium retention of the present sample remarkably increased owing to exposure to plasmas.

Comparison of X-ray spectra observed for four samples is shown in Fig. 4. These samples were exposed to plasmas in the 13th cycle, and exposed to tritium gas at
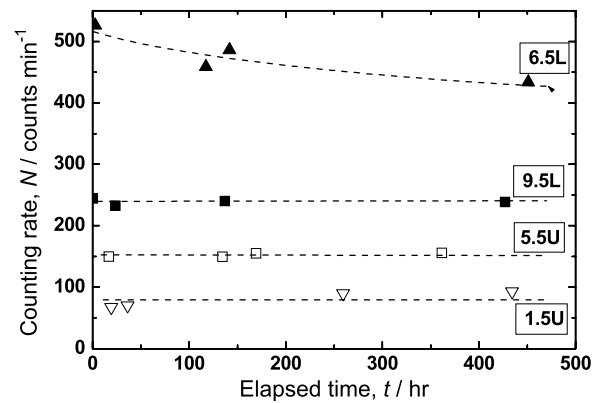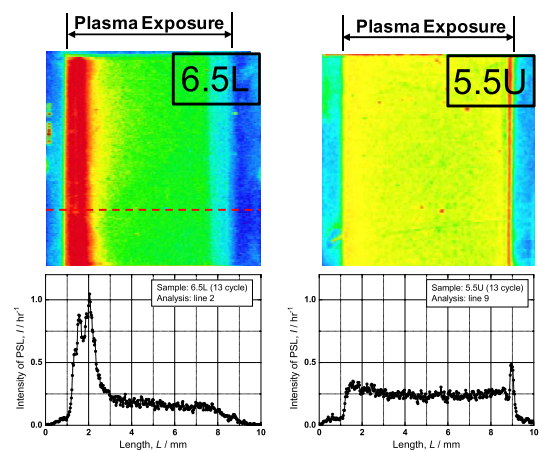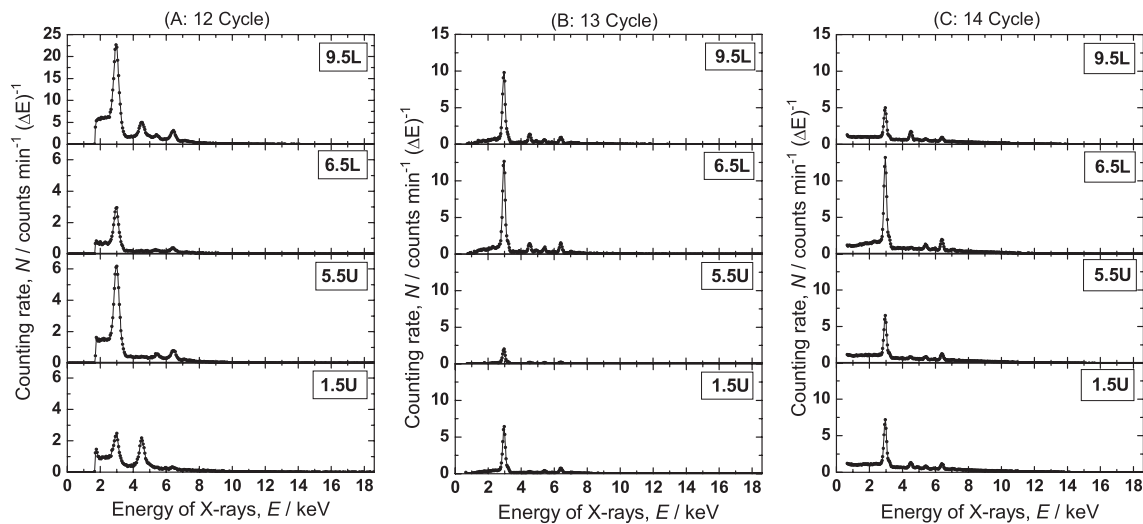
Fig. 7   Comparison of X-ray intensities observed for the samples exposed to plasmas in 12th, 13th and 14th cycle. Exposure temperature to tritium gas was around 300 K for the samples.

was 3.5 times higher than the orange area in the 5.5U sample. Namely, it is considered that nonuniformity of tritium distribution in the 6.5L sample is due to main desorption of tritium retained in the part of green area. This nonuniformity might be caused by different depositions on the surface. Therefore, it is necessary to examine in detail the deposited species on the surface in the near future.

All data mentioned above were obtained by exposing to tritium gas at 623 K. To study the effects of radiation damages by bombardment of high energy particles on tritium retention, the tritium exposure and evacuation was conducted at about 300 K. Results of BIXS measurements are shown in Fig. 7. The order of magnitude of tritium retention was 9.5L≫5.5U>6.5L>1.5U for 12th cycle, 6.5L>9.5L>1.5U>5.5U for 13th cycle, and 6.5L>1.5U~5.5U>9.5L for 14th cycle. The amount of tritium retention remarkably decreased with lowering exposure temperature. In addition, changes in the order of tritium retention were also caused by exposure temperature of tritium. These results suggest that deposition layers have different trapping sites and their trapping activities appear in different temperatures.

4. Summary

Retention behavior of tritium for samples exposed to plasmas (12th, 13th, and 14th cycle) in the Large Helical Device (LHD) was studied using mainly β-ray-induced X-ray spectrometer. Type 316L stainless steel samples were fixed at locations of 1.5U, 5.5U, 6.5L, and 9.5L in the LHD before beginning of plasma experiments. After plasma exposure, the samples were exposed to tritium gas. As a result, the following results were obtained:

(1) Surface of the samples was covered with carbon, boron and metallic oxides, although the amount of them was different in each sample.

(2) Tritium retention remarkably increased by plasma exposure in comparison with a bare SS316L sample, and it differed in the sample locations in LHD.

(3) When the samples were exposed to tritium gas at 300 K. the order of magnitude of tritium retention was as follows: 9.5L≫5.5U>6.5L>1.5U for 12th cycle, 6.5L>9.5L>1.5U>5.5U for 13th cycle, and 6.5L>1.5U~5.5U>9.5L for 14th cycle.

(4) Remarkably nonuniform distribution of tritium appeared in the surface of 6.5L sample exposed to plasmas at 13th cycle, while the surface of 5.5U sample was uniform.

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