

Tritium Retention on the Surface of Stainless Steel Samples Fixed on the Plasma-Facing Wall in LHD^{*)}

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Effects of pre-heating for retention and distribution of tritium have been studied using samples fixed on the wall of the Large Helical Device during a plasma campaign. The samples were fixed at four different locations. The plasma-facing surface of the samples was covered with deposition layers of different thickness in each sample. Retention behavior in deposition layers was observed using β -ray-induced X-ray spectrometry and imaging plate technique. Pre-heating of the samples in vacuum was changed in a temperature range from 300 to 623 K, and subsequent tritium exposure was carried out at 300 K in every runs. Non-uniformity of tritium distribution clearly appeared even in the as-received samples which was not pre-heated. It is considered, therefore, that non-uniform adsorption sites of tritium have been produced during a formation process of deposition layers. In addition, it was seen that the amount of tritium retention increased with an increase in the pre-heating temperature, indicating that adsorption sites of tritium were newly formed in the deposition layers by heating in vacuum.

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1. Introduction

From viewpoint of the safety and economy of tritium in a fusion system, it is of a great importance to reduce tritium retention in the reactor core as well as inhibition of tritium permeation through fusion reactor materials, since a huge amount of tritium is used in a fusion reactor as a fuel. Retention and permeation behavior of tritium are strongly affected by the surface states of the fusion reactor materials, e.g., erosion, deposition, modification, and so on. This is due to a reason that adsorption and/or absorption process of tritium are very sensitive for material surface states.

Present authors have studied so far effects of deposition layers for tritium retention behavior by using stainless steel samples fixed on the plasma-facing wall in the Large Helical Device (LHD) for one cycle as a model sample [1-3]. Surfaces of the samples were analyzed in detail by laser Raman spectroscopy and X-ray photoelectron spectroscopy. As a result, it was seen that the plasma-facing surface of samples was covered with carbon, oxygen, metallic oxides (a part was metallic state), and so on. Tritium retention for the plasma-facing surface was above 5 times larger than that for the rear surface (wall side). Furthermore, significant non-uniformity of tritium distribution was observed, when a plasma-facing sample was

pre-heated in vacuum and then it was exposed to tritium gas at higher temperatures. Although such a phenomenon was correlated with the distribution of metallic elements and chemical states of them, it is important to study systematically the effects of pre-heating in vacuum for evaluation of tritium retention.

Prior to the tritium exposure at 300 K, the plasma-facing samples were pre-heated in vacuum under the given temperature conditions, and effects of pre-heating temperature for the tritium retention and distribution have been studied.

2. Experimental

Samples used in this study are stainless steel type 316, and sample size was $10 \times 10 \times 0.5$ mm. After the sample was finally polished with a buff, it was fixed at four different positions (1.5U, 5.5U, 6.5L and 9.5L) of the plasma-facing wall in LHD before beginning of plasma experiments. Four sample positions placed along a toroidal direction are shown in Fig. 1. Then it was exposed to various plasma experiments during one cycle of the 15th cycle, which was operated for 16 weeks from June 2011. After ending of the plasma experiments, the samples were removed from the wall and then it was provided to tritium exposure runs.

Procedures of tritium exposure are as follows. As-

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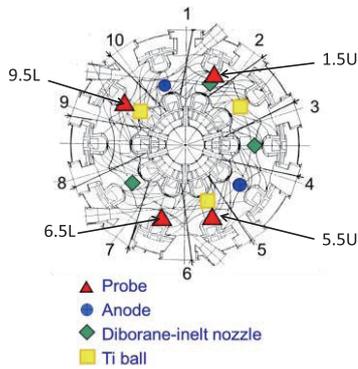


Fig. 1 Sample positions placed along a toroidal direction.

received samples removed from four locations were set in the specially designed tritium exposure device, and it was evacuated below 6×10^{-7} Pa at room temperature. After the evacuation, the samples were 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 retained in surface layers of a sample was evaluated by β -ray-induced X-ray spectrometry (BIXS) [4, 5], which was employed argon as a working gas. Tritium distribution of a sample surface was examined by imaging plate (IP) technique. Two-dimensional mapping of β -ray dose was obtained by measuring the intensity of photo-stimulated luminescence with the imaging analyzer (Fujifilm FLA-7000). The spatial resolution was $25 \times 25 \mu\text{m}$.

3. Results and Discussion

3.1 Surface of the samples

The plasma-facing surfaces were observed by an optical microscope prior to tritium exposure experiments. Surfaces of the SS316 samples were changed after plasma experiments as shown in Fig. 2. All of the sample surfaces were covered with thin deposition layers. Different colors appeared in each sample, which is similar to phenomenon observed so far [2]. As seen from the photo, colors of 1.5U and 5.5U samples were brown and blue, while those of 6.5L and 9.5L samples were gray and dark gray. It is considered that the difference in color is due to difference in thickness and constituents of the deposition layers.

3.2 Retention of tritium

The adsorption behavior of tritium for four samples was examined at first without additional heat treatment except for vacuum pumping. Figure 3 shows X-ray spectra measured by means of BIXS. As seen clearly from the figure, characteristic X-ray peak of Ar($K\alpha$: 2.96 keV) appeared in all samples, indicating that a given amount of tritium is retained in the surface layers of samples. Intensity of the Ar($K\alpha$) peak of 1.5U sample was largest among four samples, and that of other samples was almost similar intensity. Deposition of Fe (6.40 keV) and Ti (4.51 keV)

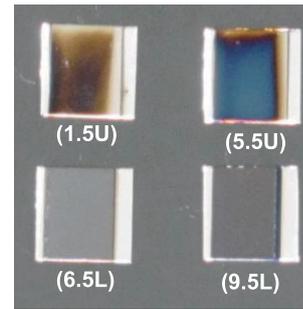


Fig. 2 Photographs of the sample surfaces used in this study.

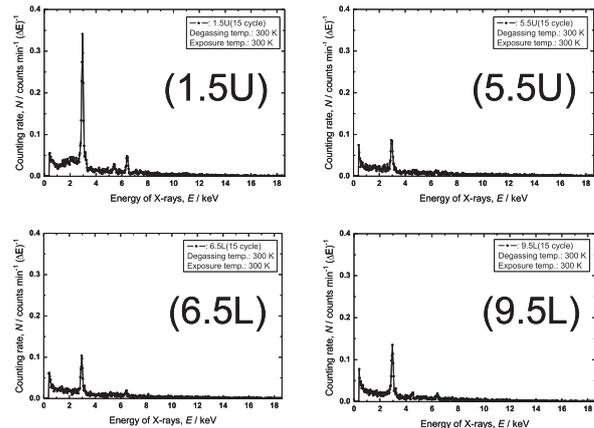


Fig. 3 BIXS spectra observed for four samples located at different positions. Tritium gas was exposed to the samples at 300 K.

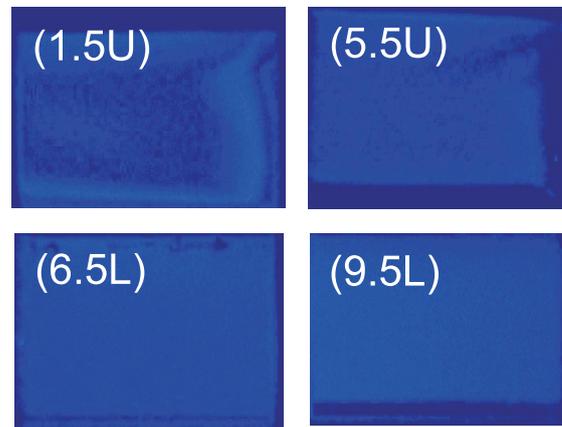


Fig. 4 Tritium distribution observed by IP technique.

was confirmed in 1.5U, although peak intensities were very weak.

Distribution of tritium retained in surface layers was measured by IP technique, which is shown in the Fig. 4. White part indicates tritium adsorption. Although a small amount of tritium adsorption was observed for all samples, tritium distribution on the 1.5U sample surface was significantly non-uniform even though tritium exposure at room temperature without pre-heating in vacuum. Such a

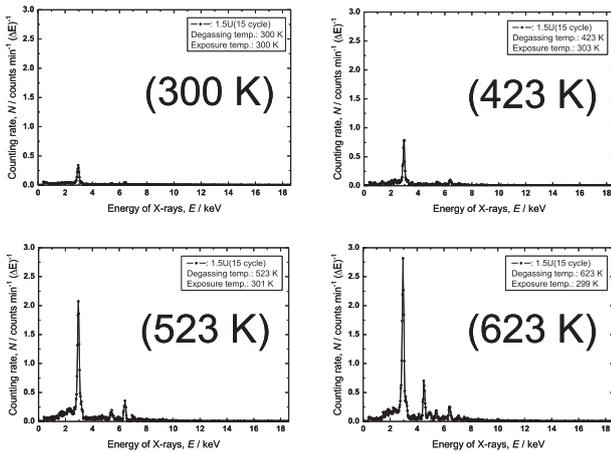


Fig. 5 Effects of pre-heating temperature in vacuum on tritium retention. The sample used was 1.5U.

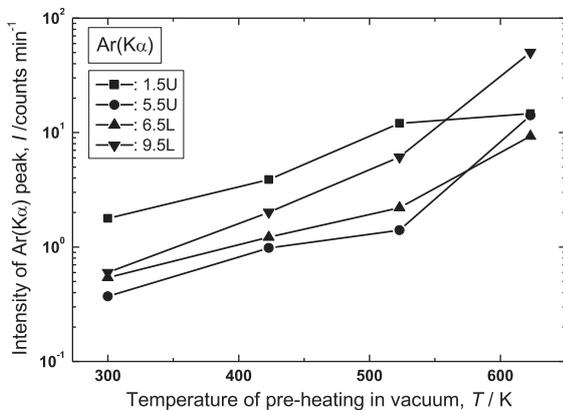


Fig. 6 Changes in the intensity of Ar(Kα) peak with pre-heating.

non-uniform distribution of tritium has been observed for the samples which were pre-heated in vacuum and then exposed to tritium gas at high temperatures [2, 3]. This indicates, therefore, that non-uniform tritium distribution is originally formed on the plasma-facing surface of 1.5U sample during plasma experiments.

To examine the effects of pre-heating temperature in vacuum on tritium retention, the same samples were repeatedly exposed to tritium gas after pre-heating in vacuum at a given temperature. The pre-heating temperature was changed from 300 to 623 K, and exposure temperature to tritium gas was kept at 300 K in every run.

As an example of effects of pre-heating, BIXS spectra observed for the 1.5U sample are shown in Fig. 5. In this sample, two major changes were observed by increase in the pre-heating temperature. One is that the intensity of Ar(Kα) peak grows up with a temperature rise, and the other is a drastic increase of the Ti(Kα) peak at 623 K. Changes in the intensity of an Ar(Kα) peak for all samples are summarized in Fig. 6. The intensity increased exponentially in all samples up to 523 K, but at 623 K different increase behavior appeared.

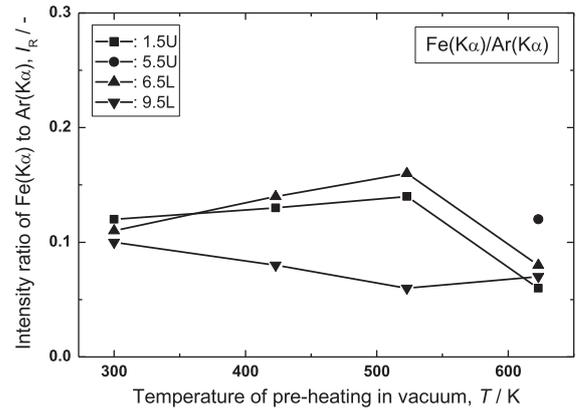


Fig. 7 Changes in the intensity ratio of Ar(Kα) peak to Fe(Kα) one.

It is expected that intensity of an Fe(Kα) peak is proportional to that of an Ar(Kα) peak, if a retention site of tritium is mainly metallic iron and/or iron oxides in surface layers. This is due to a reason that intensity of an Ar(Kα) peak depends on the intensity of β-rays escaped from deposition layers of the sample surface. Changes in the intensity ratio of Fe(Kα)/Ar(Kα) with pre-heating temperature are shown in Fig. 7. As seen clearly from the figure, the ratio is not constant. Similar increase tendency was observed for 1.5U and 6.5L sample below 523 K, while the opposite tendency appeared above this temperature. In the 9.5L sample, on the other hand, quite different behavior was observed.

The increase tendency will be caused by the increase in adsorption sites of iron and other species of which activation characteristics are similar to iron species, while the decrease tendency is due to increase in the different adsorption sites in addition to iron species. For example, the candidates are boron, carbon, titanium and nickel. It is already known from the surface analyses that these elements exist in deposition layers on the sample surface [3]. It is hard to observe the characteristic X-rays of boron and carbon by the present X-ray detector owing to low energy X-rays, and the deposition amounts of nickel and titanium are very small in comparison with those of iron and chromium.

The peak intensity of Cr(Kα) was almost proportional to that of Fe(Kα) as shown in Fig. 8. Namely, it is estimated that retention behavior of chromium is similar to that of iron. In addition, changes in the intensity ratio of Ti(Kα) to Ar(Kα) is shown in Fig. 9. In the 9.5L sample, a Ti(Kα) peak was observed for all the samples, and significant increase in the ratio appeared at 623 K, although the ratio was constant below 523 K. This indicates that titanium species was significantly activated by pre-heating at 623 K.

These indicate that not only iron and chromium but also other adsorption/absorption sites contribute as retention sites, which existed in a deeper region than the escape depth of β-rays. It is considered that activation temperature

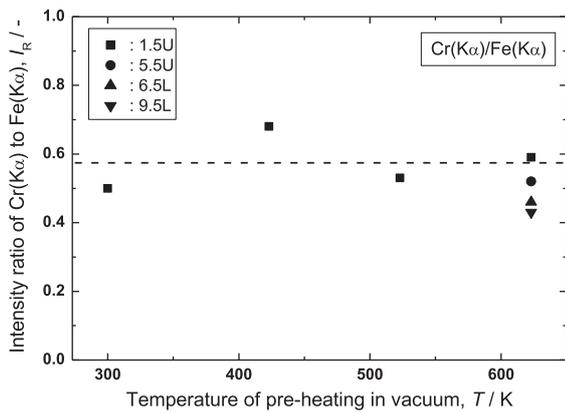


Fig. 8 Changes in the intensity ratio of Fe(Kα) peak to Cr(Kα) one.

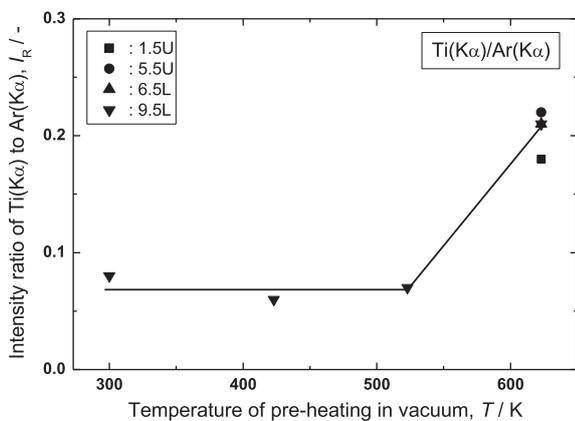


Fig. 9 Changes in the intensity ratio of Ti (Kα) peak to Ar (Kα) one.

of these retention sites is strongly dependent on chemical properties of each retention site. Namely, tritium retention will be controlled by not only the amount of metallic species but also their chemical states.

3.3 Desorption behavior of tritium

All samples were finally exposed to tritium gas at room temperature after pre-heating at 623 K. After these samples were provided to measurements by BIXS and IP technique, they were kept in ambient atmosphere. IP measurements of these samples were again carried out 100 days later. The result is shown in Fig. 10.

Large difference in tritium retention and distribution appeared among four samples. In addition, drastic changes appeared in both samples of 1.5U and 5.5U, whereas there were not apparent changes in both samples of 6.5L and 9.5L. In particular, non-uniform desorption from the surface layers is observed for both samples of 1.5U and 5.5U. Further studies are required to clarify a mechanism of non-uniform desorption of tritium.

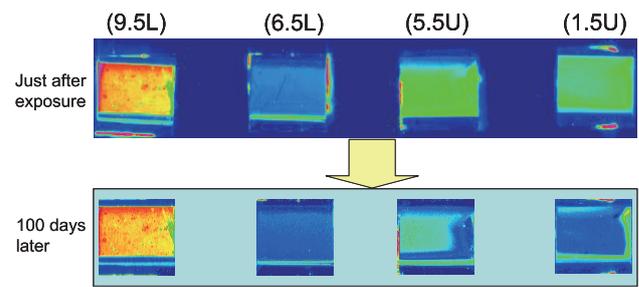


Fig. 10 Desorption behavior of tritium at ambient atmosphere.

4. Summary

To examine the effects of pre-heating in vacuum of the plasma-facing samples for retention and distribution of tritium, four kinds of stainless steel samples which were fixed at locations of 1.5U, 5.5U, 6.5L and 9.5L during one cycle in the Large Helical Device were exposed to tritium gas at 300 K after pre-heating at a temperature range from 300 to 623 K in vacuum. The amount and the distribution of tritium retained in surface layers were measured by BIXS and IP technique, respectively.

Although the amount of tritium retained in surface layers was very small for all samples, non-uniform distribution of tritium in 1.5U and 5.5U samples was significant. In this test, all of samples were exposed to tritium gas at 300 K without pre-heating in vacuum.

It was seen from changes in the peak intensity of Ar(Kα) measured by BIXS that the tritium retention increased with increase in the pre-heating temperature. Intensities of Fe(Kα) and Cr(Kα) peak increased with an increase in pre-heating temperature, but those were not proportional to intensity of Ar(Kα) in every samples. In addition, drastic increase in Ti(Kα) peak appeared in all samples by pre-heating at 623 K.

After the final exposure to tritium gas at 623 K, desorption behavior of tritium was examined under the condition of an ambient atmosphere. Although almost uniform desorption from the surface layers appeared in 6.5L and 9.5L, non-uniform desorption was observed for both samples of 1.5U and 5.5U.

Acknowledgements

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