富山大学水素同位体科学研究センター研究報告31：49－57，2011.

論　文

高密度・低エネルギー重水素プラズマに曝露したタングステンの表面トリチウム濃度分布

磯部　兼嗣[1)]、V. Kh Alimov[1)]、山西　敏彦[1)]、鳥養　祐二[2)]

1）日本原子力研究開発機構 核融合研究開発部門トリチウム工学研究グループ
〒319-1195　茨城県那珂郡東海村白方白根 2-4
2）富山大学 水素同位体科学研究センター
〒930-8555　富山市五福 3190

Tritium concentration in tungsten surface exposed to low-energy, high-flux D plasma

K. Isobe[1)], V. Kh Alimov[1)], T. Yamanishi[1)] and Y. Torikai[2)]

1) Tritium Technology group, Japan Atomic Energy Agency,
Tokai-mura Naka- gun, Ibaraki, 319-1195, Japan
2) Hydrogen Isotope Research Center, University of Toyama
Gofuku 3190, Toyama 930-8555, Japan

(Received December 13, 2011; accepted March 9, 2012)

Abstract

The limits on tritium inventory in the vacuum vessel and the need for prevention of impurity ingress into plasma make plasma-surface interaction on tungsten an important issue. It is well known that plasma exposure on tungsten makes some kinds of blisters on the surface and increases the hydrogen inventory. On the other hands, there is a possibility that plasma exposure would change the characteristic of surface and surface region in tungsten and cause the increase of tritium inventory. To understand the change of tritium inventory in the surface region and surface in tungsten by plasma exposure, tritium concentration in tungsten exposure by low-energy (38eV), high-flux D plasma with was examined with

BIXS after thermal exposure of tritium gas. D plasma exposure was carried out at specimen temperature around 495 and 550K. After that, specimens were exposed to gaseous tritium diluted with deuterium at 473K for 3 hours. The tritium concentration was measured with BIXS. The tritium concentration in surface and surface region was found to be increased by plasma exposure. And its concentration of tungsten exposed at 495 K was estimated to be twice higher than that of as-received tungsten.

1. Introduction

Tungsten is one of the candidate materials for plasma-facing components in fusion reactor. The limits on tritium inventory in the vacuum vessel and the need for prevention of impurity ingress into plasma, make plasma-surface interaction on tungsten an important issue. Surface morphology and hydrogen inventory of tungsten after plasma exposure have been studied with many kinds of plasma condition. Most of the results on hydrogen isotope retention and recycling for W have been reviewed by Causey and Venhaus,[1,2] and Skinner et al.[3] It has been found that deuterium plasma exposure to tungsten makes some kinds of blister on the surface and increases the hydrogen inventory.[4-10] On the other hands, there is a possibility that plasma exposure would change the characteristic of surface and surface region in tungsten and cause the increase of tritium inventory. The authors have studied the deuterium depth distribution of tungsten with Nuclear reaction analysis and clarified deuterium depth distribution after D plasma exposure.[10] Especially, deuterium was increased with approaching surface and its concentration was two orders of magnitude higher than that of inner region at 330 K of the fluence of 10^{26} D/m^2. This result indicated that tritium concentration in the surface and the surface region that was exposed to plasma is important with respect to not only the plasma surface interaction, but also the inventory and the decontamination of tritium in fusion reactor. In case that the hydrogen inventory of tungsten surface and surface region would be increased by plasma exposure, tritium inventory in the vacuum vessel should be taken into account not only hydrogen implantation but also the

increase of hydrogen concentration in surface and surface region by plasma exposure. Beta ray-induced-X ray spectrometry (BIXS) can measure the tritium concentration of surface region in nm order due to the short path of beta-rays released from tritium. On the other hand, this method required tritium and tritium has to be introduced into specimen by using some methods. We have tried to introduce tritium to specimens with thermal exposure of tritium gas and to apply BIXS method to measure the tritium concentration of recrystallized tungsten exposed to D plasma. In this study, tritium concentration of tungsten exposed with low-energy (38eV), high-flux D plasma was examined with BIXS. Tritium concentration measurements were carried out not only in the recrystallized tungsten but also in the ITER grade tungsten that was not exposed to D plasma.

2. Experimental

The tungsten specimens exposed to plasma were the recrystallized tungsten (A.L.M.T. Corp., Japan) with 99.99 wt% purity and major impurities (in weight ppm) of Mo and Fe around 10, C and O less than 30. The specimens were finally recrystallized at 2073 K after cut into 10 × 10 × 2 mm and polishing. Fig. 1 shows the schematic diagram of liner plasma generator used in this study. The plasma was ignited by an electron-emitting filament made of LaB_6 with a shape of a hollow dual-spiral and maintained by an arc discharge power supply applied between the filament (cathode) and grounded chamber wall (anode), being assisted by the confinement coil. The water cooled specimen holder is isolated from the grounded chamber wall so that the specimen can be negatively biased to adjust the energy of ions impinging onto the specimen. The specimen is passively

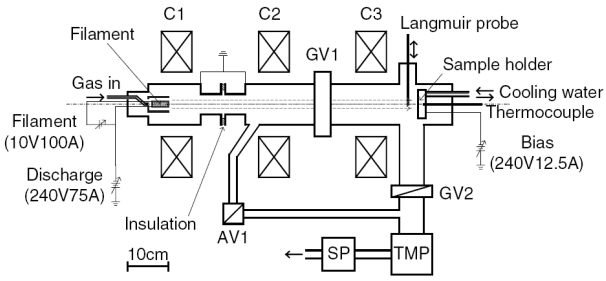

Fig. 1. Schematic view of a linear plasma generator that can produce low-energy and high flux plasmas.

heated by the plasma itself and the temperature rise was adjusted by inserting several mica plates of different thickness between the specimen and the holder. The temperature is monitored using a type K thermocouple tightly pressed to the backside of specimens. The ions species can be controlled by adjusting the operational parameters of plasma generator. In this study, plasma beams highly enriched with a single species of D_2^+ with a ratio of over 80% were obtained, and the main impurity in the plasma was oxygen with a concentration less than 1 ppm. The area of the specimen exposed to the plasma was 9 mm in a diameter, and incident flux and energy were fixed at 10^{22} $D^+/m^2/s$ and 38 eV/D, respectively. D plasma exposure to recrystallized tungsten was carried out at around 495 and 550 K of specimen to the same fluence of 10^{26} D/m^2. The temperature dependence of deuterium inventory of tungsten exposed such conditions have been reported.[10] After plasma exposure, the surface of tungsten specimen was observed with scanning electron microscope (KEYENCE VE-9800) at a tile angle of 45 ^{o}.

Tritium was introduced into the specimens thermally by exposure to hydrogen gas containing tritium. Fig. 2 shows the schematic view of tritium exposure apparatus. The tritium exposure apparatus consisted of a vacuum pumping system, a quartz tube for specimen heating and a tritium source. The tungsten specimens, the tungsten after plasma exposure, as received recrystallized tungsten (non-exposure) and ITER grade tungsten, were placed in quartz tube of specimen heating. ITER grade tungsten (A.L.M.T. Corp., Japan) used in this study were with over 99.96 wt% purity and major impurities (in weight ppm) of Mo and Fe around 3, C and O less than 10. The specimens were finally annealed for

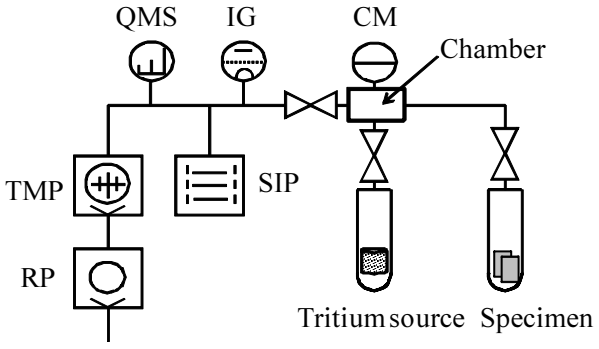

Fig. 2. Schematic view of tritium exposure apparatus.

stress relief after cut into 10 × 10 × 2 mm and polishing. Glass wool were inserted between each specimen to avoid the contact of each specimen. After the installation of specimen in the heating tube to the apparatus, specimen tube and the apparatus except the tritium source were evacuated by the vacuum pumping system to less than 10^{-4} Pa. Before the tritium exposure, specimens were heated at 473 K for degassing the quartz tube, the glass wool and specimens. At this temperature, we have already confirmed that deuterium in tungsten was not released to the vacuum by thermal desorption spectroscopy. After the high vacuum was obtained, a valve above the specimen heating tube was closed and the tritium source was also heated up after opening the valve above tritium source. Tritium in the source was diluted with deuterium and the tritium concentration is 7.8 %. During the heating of tritium source, the tritium was released into the chamber equipped with CM. The pressure of hydrogen containing tritium reached around 1 kPa, the valve above tritium source was closed. And tritium released into the chamber was introduced into the specimen heating tube by opening the valve above specimen. The exposure of tritium was kept 5 hours. After tritium was re-stored into the tritium source sufficiently, specimens were removed from the tritium exposure apparatus.

BIXS measurement was carried out in the chamber in which Ar gas flowed continuously. Details of the BIXS measurement have been reported.[11] Thin Be plate was used as the window of X-ray detector (CANBERA SSI 50150 Model), because of the energy of X-ray induced by tritium is very low. BIXS measurement of each specimen was continued until the peaks appeared clearly.

3. Results

Fig. 3 shows the results of surface observations with SEM after D plasma exposure. On the surface of tungsten exposed at 495 K, two kinds of blister appeared. One type had

larger diameter over 10 μm, and the other had small diameter under 1μm. On the other hand, there was only one kind of blisters, its diameter was under 1 μm, on the surface of tungsten exposed at 550 K. This temperature dependence of blister morphology has been already reported in [10] and these observations suggested the deuterium inventory of tungsten in this study was the same as the previous study. According to previous results, a maximum deuterium inventory at the 10^{26} D/m^2 fluence is 7×10^{21} D/m^2 around 500 K. Over 500 K, the deuterium inventory decreased drastically. And deuterium inventory at around 550 K was one order of magnitude smaller than that for 500 K. Therefore, the deuterium inventory at the exposure temperature of 495 K was considered to be higher than that of 550 K.

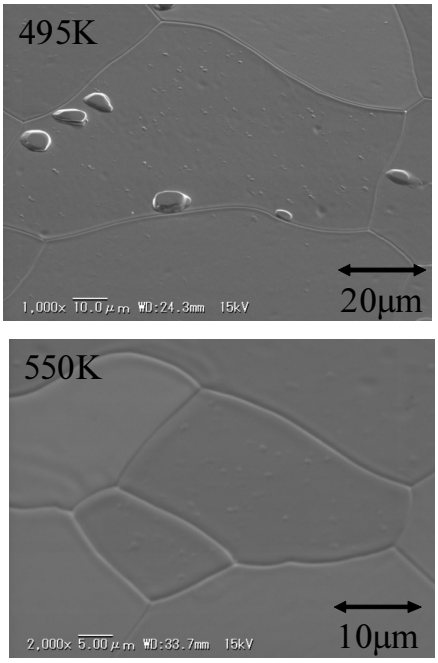

Fig. 3. Surface observations result after D plasma exposure

BIXS measurement results of tungsten exposed to D plasma, as-received recrystallized tungsten and the ITER grade tungsten were shown in Fig. 4. The horizontal axis is the energy of X-rays and the vertical axis is X-ray count rate. Since the BIXS measurement time was wdifferent from each other, X-ray count was normalized into counts per second. Two peaks appeared at the energies of 1.8 keV and 3.2 keV in X-ray spectrum. The peaks at 1.8 keV and 3.2 keV are characteristic X-rays of W (Mα) and Ar (Kα) respectively. Beta-ray released from tritium induced characteristic X-rays depending on its

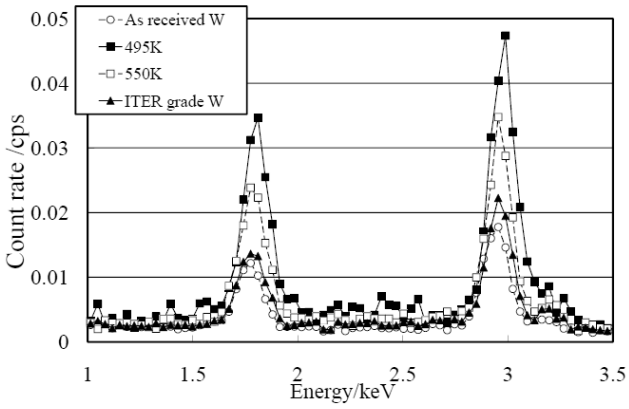

Fig. 4. BIXS measurement results of each specimen

depth. Beta-ray released from tritium on the surface can induce Ar atom in the chamber. Therefore, Ar(Kα) depends on tritium concentration from surface to beta-ray escape depth in tungsten. On the other hand, beta-ray released from tritium in tungsten can excite W atom. Therefore, peak intensity of W (Mα) x-rays depends on tritium concentration in tungsten from surface to tungsten bulk. Here the maximum depth of tritium inducing Ar(Kα) and W(Mα) x-rays was estimated to be in 20 nm and 500 nm, respectively.[11] So that two peaks as shown in Fig. 4 were obtained by BIXS measurement, tritium existed in tungsten surface to 20 nm depths and to 500 nm depths were obtained successfully by the method of this study. Especially, tritium concentration of surface region of tritium existed in 20 nm depths and of tritium in surface could be clarified by this measurement. According to the count rate of each peak, the count rate of the tungsten exposed at 495 K was the highest. And the count rate of the tungsten exposed at 550 K was the second one. The count rate of ITER grade tungsten was slightly higher than that of as-received recrystallized tungsten. This result agrees with the inventory and NRA results of previous paper.[10] Therefore, BIXS measurement was shown its performance to obtain the tritium concentration on surface and surface region. From the result of fig. 4, tritium concentration in surface region and surface was found to be increased by plasma exposure. In other words, plasma exposure increased the amount of tritium trapping site in tungsten surface region and surface. Therefore, the result of fig. 4 suggested that plasma exposure would increase the tritium inventory of tungsten by not only hydrogen implantation but also the increase of the amount of hydrogen trapping site. The reason of the increase of trapping site could not be clarified in this study. To understand reasons of the increase, the surface condition after plasma exposure and tritium distribution should be clarified in detail.

Here, hydrogen concentration in surface region and surface can be estimated from the count rate of Ar(Kα) x-rays. The count rates from 2.746 keV to 3.404 keV were chosen

for the start of the Ar(Kα) peak and the end of the peak respectively. The count rate subtracted background was summed up between start of the peak and end of the peak. After that, tritium concentration was estimated by radioactivity of Ar(Kα) x-rays. Tritium concentration of as-received tungsten is 3×10^{14} T/cm^2, tungsten exposed at 495 K is 7×10^{14} T/cm^2 and tungsten exposed at 550 K is 4×10^{14} T/cm^2, respectively. From this estimation, it was found that the tritium concentration of tungsten exposed at 495 K is about twice higher than that of as-received tungsten and the tritium concentration of tungsten exposed at 550 K is slightly higher than that of as-received tungsten. This estimation of tritium concentration also indicated that plasma exposure increased hydrogen trapping site in surface and surface region and suggested that the amount of trapping site by plasma exposure was reached twice higher than that of as- received tungsten .

4. Conclusions

To understand the change of the surface region and surface in tungsten by plasma exposure, we have introduced tritium in tungsten specimens by thermal exposure to tritium gas and applied BIXS method to measure the tritium concentration of recrystallized tungsten exposed to D plasma. Two peaks were observed in BIXS measurement and tritium concentration in the surface region and surface in tungsten were successfully obtained. Tritium concentration in the surface region and surface was found to be increased by plasma exposure. And its concentration of tungsten exposed at 495 K was estimated to be twice higher than that of as-received tungsten.

Acknowledgement

A part of this work is performed with the support and under the auspices of the Hydrogen Isotope Research center, University of Toyama Collaborative research program.

References

(1) R. A. Causey and T. J. Venhaus, Phys. Scripta, **T94** (2001) 9.
(2) R. A. Causey, J. Nucl. Mater., **300** (2002) 91.
(3) C. H. Skinner, A. A. Haasz, V. Kh. Alimov, et al., Fusion Sci. Technol., **54** (2008) 891.
(4) G. -N. Luo, W. M. Shu, M. Nishi, J. Nucl. Mater., **347** (2005) 111.
(5) G. -N. Luo, W. M. Shu, M. Nishi, Fusion Eng. Design, **81** (2006) 957.
(6) W. M. Shu, A. Kawasuso, Y. Miwa, E. Wakai, G.-N. Luo, T. Yamanishi, Physica Scripta, **T128** (2007) 96.
(7) V. Kh. Alimov, J. Roth, Physica Scripta, **T128** (2007) 6.
(8) W. M. Shu, G. -N. Luo, T. Yamanishi, J. Nucl. Mater., **367-370** (2007) 1463.
(9) W. M. Shu, E. Wakai, T. Yamanishi, Nucl. Fusion, **47** (2007) 201.
(10) V. Kh. Alimov, W. M. Shu, J. Roth and et al., Physica Scripta, **T138** (2009) 014048.
(11) M. Matsuyama, S. Nakagawa, M. Enyama and et al., J. Nucl. Mater., **329-333** (2004) 752-756.