# Tritium distribution on the tungsten surface exposed to deuterium plasma and then to tritium gas

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Tungsten (W) samples were exposed to Low-energy (38 eV/D), high flux ($10^{22}$ $D^+/m^2/s$) deuterium (D) plasma at 495, 545 and 550 K to a fluence of $10^{26}$ $D/m^2$. After the plasma exposure, tritium was introduced into the samples by exposure to deuterium-tritium gas mixture at 473K. Tritium distribution on the W surface was examined by the techniques of imaging plate and autoradiography. The results of the imaging plate revealed that tritium was concentrated mainly within the area exposed to the D plasma, and the concentration of tritium is slightly varied within the area. Autoradiography of the W surface showed that tritium was concentrated on the grain boundary and blisters.

Keywords: Tungsten, Plasma-surface interaction, Tritium, Imaging plate, Tritium autoradiography.

## 1. Introduction

Since tungsten (W) is one of the candidate materials for plasma-facing components of fusion reactor, many investigations relating to the plasma-W interaction have been carried out. From a view point of safety, the tritium inventory in plasma-facing materials is a one of key issues due to limitation of tritium inventory in the vacuum vessel. Most of the results on hydrogen isotope retention and recycling in W materials have been reviewed by Causey and Venhaus [1,2], and Skinner et al [3]. It has been reported that during exposure to D plasmas, blisters with various shapes and sizes depending on the exposure temperature are formed on the W surface [4-14]. Blister formation is accompanied by deformation of the near-surface layers [12, 13], and this deformation is thought to create additional traps for hydrogen isotopes. There is also a possibility that amount of hydrogen isotope which could be solved or trapped in the deformed W near-surface layer increases. In this study, W samples with D-plasma-induced blisters on the surface were exposed to $D_2$-$T_2$ gas mixture, and the distribution of tritium on the W surface was examined by the techniques of imaging plate and autoradiography.

## 2. Experimental

The W specimens exposed to D plasma were prepared from polycrystalline tungsten (A.L.M.T. Corp., Japan) with a purity of 99.99 wt%. The samples were fully recrystallized at 2070 K for 1 hour after cutting into $10 \times 10 \times 2$ $mm^3$ and polishing. The grain size was 20-200 μm.

The linear plasma generator used for delivering a plasma beam is described elsewhere [4]. A plasma beam highly enriched with a species of $D_2^+$ to over 80% was obtained. The bias voltage of -80 V was applied to the W sample resulting in a incident of 38 eV/D, taking into account the plasma potential of about -4 V measured by a Langmuir probe. The incident deuterium ion flux was fixed at $10^{22}$ $D^+/m^2/s$, and the samples were exposed to ion fluence of $10^{26}$ $D/m^2$. The sample was passively heated by the plasma itself and exposure temperature was set by the thermal contact between the sample and the cooled holder. D plasma exposure to recrystallized W was carried out at around 495, 545 and 550K. At the fluence of $10^{26}$ $D/m^2$, the deuterium inventory of tungsten exposed at temperature over the range of 500 - 550 K become higher than that at other temperature range [15]. The surface area of the W samples exposed to the plasma was 9 mm in diameter. For a comparison, as-received recrystallized W sample was also used for tritium loading. Note that the as-received sample was not exposed to the D plasma.

4 months after the D plasma exposure, the samples were loaded with tritium from gaseous phase. Fig. 1 shows a schematic view of a tritium exposure apparatus. The tritium exposure apparatus consisted of a vacuum pumping system, a quartz tube for gas exposure equipped with an external heater, quadrupole mass spectrometer

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(QMS), and a tritium source. The W samples were placed in the quartz tube, in doing so pieces of glass wool were inserted between samples to avoid a contact between them. The quartz tube with samples was heated in vacuum at 473K for degassing until termination of gas release that was controlled by QMS. At this temperature, we have already confirmed that deuterium in tungsten is not released to the vacuum by thermal desorption spectroscopy [15]. When high vacuum was obtained, tritium diluted with deuterium was introduced into the quartz tube from the tritium source. W samples were exposed at 473 K to D (92.2%)-T (7.8%) gas mixture at a total gas pressure of 1 kPa for 5 hours.

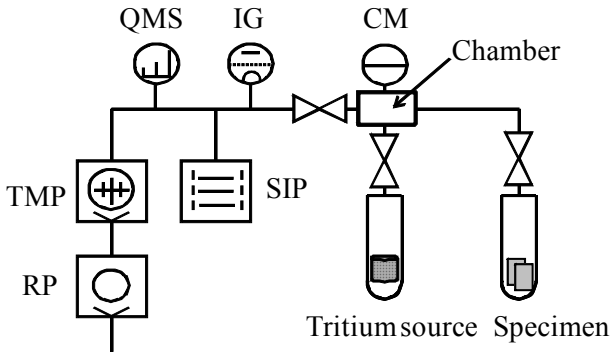


Fig. 1 Schematic view of tritium exposure apparatus.
QMS:Quadropole mass spectrometer,
IG:Ionization gauge, CM:Capacitance manometer
SIP:Suptter ion pump, RP:Rotary pump
TMP:Turbo molecular pump

The amount of tritium retained in the W near-surface layers was evaluated by the imaging plate technique. Imaging plate (IP) is a 2-dimensional radiation detector with high sensitivity. IP used in our experiments was BAS-TR2025 (Fuji Photo Film Co. Ltd.) which consists of 50 μm thickness photostimulable phosphor mounted on a plastic support. The IP surface was contacted to the W surface and exposed to tritium beta electrons in 1 week. After the exposure to tritium beta electrons, the IP was processed by an IP reader FUJIFILM BAS2500 (Fuji Photo Film Co. Ltd.) in order to obtain the digital image. Tritium activity is expressed as the photo-stimulate luminescence (PSL) intensity, which is proportional to the absorbed radiation energy.

Tritium autoradiography technique was applied to two specimens; recrystallized W exposed to the D plasma at 495K and as-received, non-exposed recrystallized W. At first, to avoid direct contact of the W samples with AgBr grains in radiographic emulsion, thin collodion film was deposited on the W surface by dipping the samples in collodion diluted by ethanol. The monolayer of the radiographic emulsion (ILFORD L4) was placed onto the collodion film by a wire-loop method. The specimens were kept in a light-tight box at around 273 K for 12 days to expose the emulsion to the tritium beta electrons. Then, specimens were dipped in a developer solution and then in a fixer solution. Under exposure to beta electrons, the AgBr grains were transformed into Ag grains. The distribution of Ag grains was observed by the scanning electron microscope (SEM).

## 3. Results and Discussion

Surface morphology of the recrystallized W after D plasma exposure at 495K is shown in Fig. 2. Grain boundary could be clearly observed in Fig. 2. Large blisters with sizes of a few tens of microns and small blisters with size of less than a few microns are observed. The large blisters demonstrate a multi-layered structure like steps. The small blisters are formed on the flat surface of crystal grains.

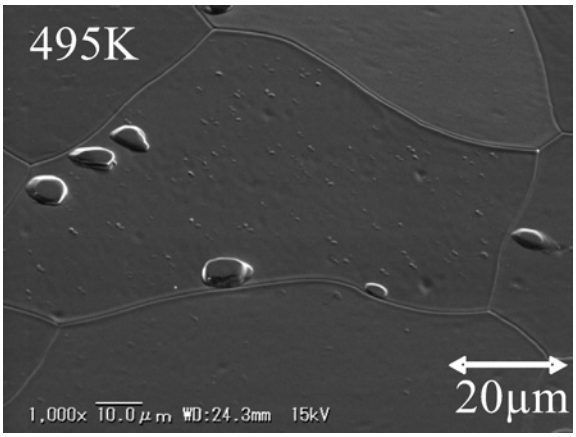


Fig. 2. SEM image of recrystallized W exposed at 495 K to low-energy high flux D plasma with fluence of $10^{26}$ D/m$^2$. The surface was tilted at an angle of 45ºto the electron beam.

Fig. 3 shows the results of IP measurements. IP measurement can easily observe tritium distribution of the whole of specimen as shown in Fig. 3. A level of darkness corresponds to a concentration of tritium in near-surface layer. As seen Fig. 3 (a, b, c), tritium is concentrated on the surface area exposed to the D plasma. It is known that under exposure to high flux D plasmas, vacancy-type defects, voids and cracks are generates in W, and these defects are responsible for accumulation deuterium [11-14, 16]. Obviously, subsequent exposure to tritium leads to isotope exchange in the near-surface layer [17] and accumulation of tritium at the defects created under previous D plasma exposure.

The average tritium concentration of the W surface exposed at 550 K is significantly higher than that for the W surface exposed at 545 K, although the difference in the exposure temperature is only 5 K. Obviously, a small variation in the exposure temperature within this temperature range has a significant influence on a

concentration of the D-plasma-induced traps generated in near-surface layer. This suggestion is supported by sharp temperature dependence of the D retention in the recrystallized W exposed to low-energy, high-flux D plasma [16].

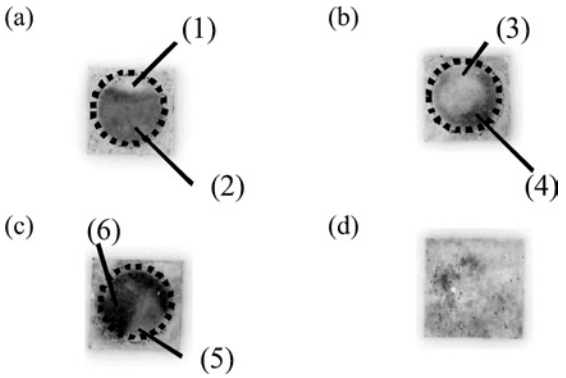


Fig. 3. IP images for recrystallized W exposed to low-energy, high flux D plasma at 495 K (a), 545 K (b), 550 K (c), and for as-received recrystallized W (d). Dashed circles (a-c) indicate the surface areas exposed to D plasma. Tritium was loaded into all samples at 473 K, 1 kPa D-T gas mixture for 5 hours.

However, within the exposed surface area the tritium distribution is not uniform. Intensities of PSL at points ((1)-(6)) in Fig. 3 are shown in Table I. The ratio between the maximum and minimum PSL intensities on the same exposed sample surface ((2)/(1), (4)/(3), (6)/(5)) varied from 4:1 to 6:1. It was reported recently that hydrogen isotopes can be retained by tungsten oxide layers [18]. Probably, the surfaces of the W samples were nonuniformly oxidized before tritium gas exposure.

Table I Intensity of photo-stimulate luminescence at some points on the plasma-exposed W surface

| Point | (1) | (2) | (3) | (4) | (5) | (6) |
|---|---|---|---|---|---|---|
| PSL/mm$^2$ | 320 | 1800 | 500 | 2100 | 560 | 3200 |

Figure 4 demonstrates autoradiographic patterns, in which white spots correspond to Ag grains tracing the tritium beta electrons. The Ag grains, as white lines, were observed along grain boundary (Fig. 4 (a)). The Ag grains decorate also blisters and are distributed uniformly on the grain surface (Fig. 4 (b, c)). However, the Ag grains are not observed on the surface of as-received recrystallized W (Fig. 4 (d)). As was mentioned above, various types of trapping sites for hydrogen isotopes are generated in W under exposure to high-flux D plasma. Under subsequent exposure to tritium gas at elevated temperature, tritium is thought to be occupied these trapping sites. It should be noted that techniques of imaging plate and autoradiography allow detecting tritium retention in a shallow, near-surface layer because the escape depth of tritium beta electrons in

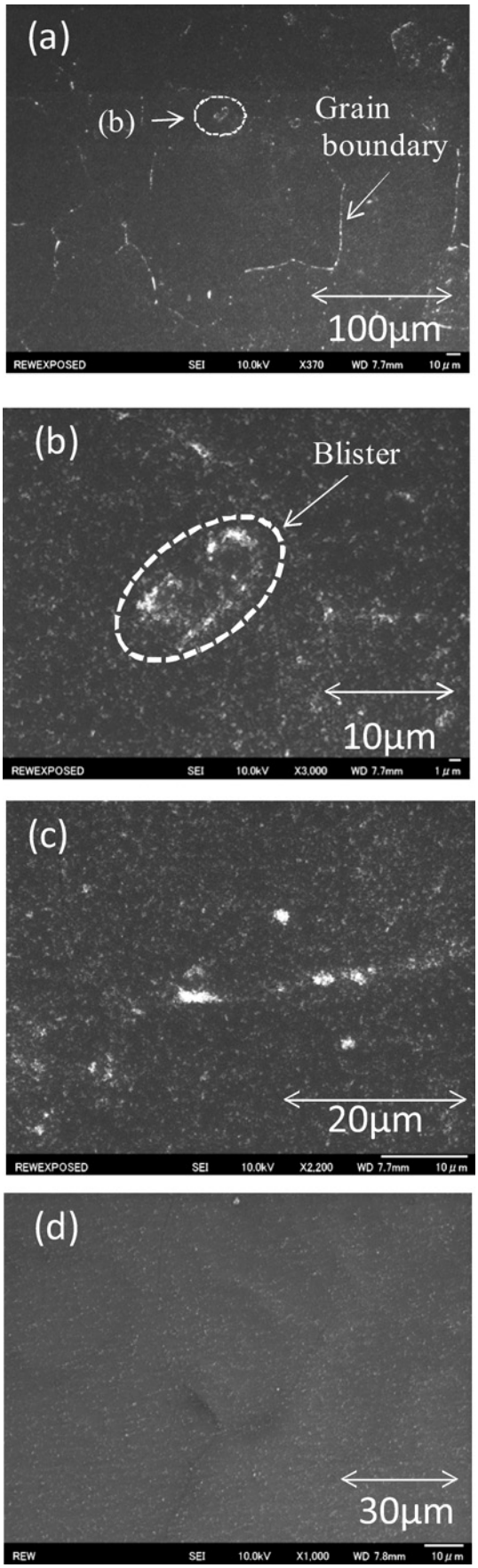


Fig. 4. Autoradiographic patterns of recrystallized W exposed with D plasma at 495K (a, b, c) and as-received recrystallized W (d). Tritium was loaded into these samples at 473 K, 1 kPa D-T gas mixture for 5 hours.

W does not exceed one micrometer [19, 20]. Thus, results of this study shows that under exposure to the D plasma at elevated temperatures, traps in the near-surface layer are generated mainly along grain boundaries and in layers subjected to tensile stress due to material migration above the surface during formation of blisters [12].

## 4. Conclusion

Techniques of imaging plate (IP) and autoradiography were applied to observe a distribution of tritium on the W surface preliminary exposed to low-energy, high-flux D plasma at elevated temperatures (495-550 K) and then to tritium gas at 473 K. According to IP images, tritium was highly concentrated within D-plasma-exposed surface area; however, the tritium concentration was varied within this area. With the use of autoradiography, it was found that tritium was concentrated on the grain boundary and along/on blisters formed under D plasma exposure. These results suggest that D plasma exposure generates traps for hydrogen isotopes in the W near-surface layer.

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