On the possibility of low-temperature removal of hydrogen isotopes from tungsten in conditions of thermonuclear and plasma facilities

L B Begrambekov, S S Dovganyuk, A E Evsin and A S Kaplevskiy.

National Research Nuclear University MEPhI (Moscow Engineering Physics Institute), 115409 Kashirskoe shosse 31, Moscow, Russia Federation

e-mail: ssdovganyuk@plasma.mephi.ru

Abstract. Acceleration of hydrogen isotope trapping and desorption has been observed in the experiments with stainless steel (Activated Surface Penetration - ASP) under irradiation by hydrogen atoms and hydrogen plasma ions with oxygen impurity. ASP is considered in this paper for a method of low-temperature hydrogen isotope removal from tungsten and aluminum-coated tungsten under irradiation by hydrogen plasma ions with oxygen impurity. It is shown that periodic irradiation by hydrogen plasma ions with oxygen impurity allows preventing accumulation of implanted deuterium ions in tungsten.

1. Introduction

Beryllium and tungsten are chosen as the materials of the first wall and the divertor of the ITER tokamak, respectively. Due to the possible presence of oxygen impurity in ITER plasma, existence of oxides on their surfaces, as well as the surface of beryllium layer deposited on tungsten. The development of the low-temperature hydrogen isotope removal method from tungsten and tungsten with beryllium layers is relevant for safety reasons as tritium trapping in the first wall and the divertor of the tokamak is limited as 700g.

Barrier properties of oxide layers relative to hydrogen diffusion are well known [1-4]. It was shown in work [1] that barrier properties of the oxide layer on stainless steel relative to hydrogen isotope diffusion change under ion and plasma irradiation. It was found that irradiation of 12X18H10T-grade steel (closest analogue is AISI 321) by hydrogen atoms and hydrogen plasma ions with oxygen impurity drastically improve hydrogen isotope diffusion through the surface of stainless steel (Active Surface Penetration – ASP), accelerating hydrogen isotope trapping and desorption at low temperatures (≤ 500 K).

The goal of this work was to study the possibility of using ASP phenomenon for developing the method of low-temperature removal of hydrogen isotopes from tungsten and aluminum-coated tungsten under the irradiation by hydrogen plasma with oxygen impurity. As beryllium is toxic, tungsten samples with aluminum coating were used in the experiments due to the simplicity of aluminum’s usage in laboratory conditions, not imposing additional safety measures, and its physical and chemical properties closely resembling those of beryllium.
2. Sample preparation and experimental setup

Tungsten samples of 7x7x1 mm$^3$ were mechanically polished, washed in an ultrasonic ethanol bath and then annealed at pressure of $\leq 5\times10^{-6}$ Pa and temperature of 1600 K to remove “technological” hydrogen. For a part of these samples, 200±10 nm thick aluminum coating was deposited on one side in argon plasma with hydrogen impurity. Residual gas pressure did not exceed $5\times10^{-4}$ Pa. Deposition rate was 0.055 nm/s. Temperature of the samples was 450-500 K. EDS analysis showed formation of an oxide layer on aluminum coating exposed to the atmosphere.

Experiments were conducted in the «MIKMA» stand [6]. Deuterium plasma ions were implanted into tungsten samples and tungsten side of aluminum-coated samples. The samples were then irradiated by hydrogen plasma ions with oxygen impurity. The cycles of irradiation repeated and deuterium content was measured using thermal desorption spectrometry (TDS) analysis after each cycle.

Residual gas pressure in the plasma chamber of the stand did not exceed $3\times10^{-3}$ Pa. Deuterium, and then hydrogen plasma was initiated between a glow cathode and an anode. Plasma ion flow irradiating the sample included mainly molecular ions of the respective hydrogen isotope (82-85%), with the rest being mono- and triatomic ions. During the analysis of the results it was assumes that the sample was irradiated exclusively by H$_2^+$ or D$_2^+$ ions. The temperature of the sample was regulated by radiation from a flat tungsten spiral behind the back of the sample. The energy of irradiating ions was $E=650$ eV/at., the ion flow on the sample was $j=3.7\times10^{19}$ at/m$^2$s, the irradiation dose was $\Phi=1.3\times10^{23}$ at/m$^2$, the temperature of the samples during deuterium implantation and hydrogen ion irradiation was $T=500$ K.

Residual gas pressure in the TDS camera was $1.3\times10^{-5}$ Pa. During the TDS measurements, samples were heated at a rate of 5 K/s, the desorption of H$_2$, HD, D$_2$, H$_2$O, HDO and D$_2$O molecules was registered. Desorption of other hydrogen- and deuterium containing molecules was negligible. The amount of deuterium and hydrogen in the samples after each stage of the experiment was calculated based on these results.

The first sample implanted with deuterium was used for a TDS measurement of hydrogen and deuterium content after the implantation. The second sample was exposed to vacuum for 1 hour at 500 K i.e. for the duration of hydrogen plasma with oxygen impurity irradiation. Hydrogen and deuterium content was then measured in the sample. This allowed deducing whether particles trapped would desorb during exposure to vacuum without plasma irradiation. After deuterium implantation, the third sample was irradiated by hydrogen plasma with 1% oxygen impurity; in case of aluminum-coated sample, the film was irradiated. The parameters of irradiation by H$_2$+1% O$_2$ plasma were as follows: $E=50$ eV/at., $j=3.7\times10^{19}$ at/m$^2$s, $\Phi=1.3\times10^{23}$ at/m$^2$, $T=500$ K. TDS was then used to measure the change of hydrogen and deuterium content after irradiation by H$_2$+1%O$_2$ plasma ions. Experiments were conducted with one, two, three and five repetitions of deuterium implantation and H$_2$+1%O$_2$ plasma irradiation.

3. Results of the experiments for tungsten samples

Deuterium and hydrogen atom content in tungsten samples before and after the experiments are shown in Table 1. Interaction of deuterium ions during implantation (Table 1 row 2) with surface oxide layer initiated hydrogen trapping from residual gas molecules on the surface in the quantities roughly equal to the quantity of trapped deuterium. Exposure of tungsten samples to vacuum (Table 1 row 2, 3) did not lead to desorption of implanted deuterium atoms. Irradiation by H$_2$+1%O$_2$ plasma (Table 1 row 2, 4) initiated desorption of ≈40% implanted deuterium atoms, and the hydrogen in the ample increased to about 2.8 of the initial value. The results of the experiment showed that ASP develops on the surface of tungsten and reduces the quantity of previously implanted deuterium. However, to make a conclusion on the possibility of using ASP to remove hydrogen isotopes from tungsten, it was necessary to make sure that the effect is repeated for multiple cycles of deuterium implantation and H$_2$+1%O$_2$ plasma irradiation.
Table 1. Hydrogen and deuterium content in tungsten samples after deuterium implantation, exposition to vacuum, exposition to atmosphere and H$_2$+1%O$_2$ plasma irradiation of the side deuterium was implanted in.

| № | Experiment                                      | Deuterium content, $\times10^{20}$ at/m$^2$ | Hydrogen content, $\times10^{21}$ at/m$^2$ |
|---|------------------------------------------------|---------------------------------------------|--------------------------------------------|
| 1 | Before deuterium implantation                   | -                                           | -                                          |
| 2 | After deuterium implantation                    | 2.2                                         | 2.3                                        |
| 3 | After implantation and exposition to vacuum     | 2.2                                         | 2.3                                        |
| 4 | After irradiation by H$_2$+1%O$_2$ plasma       | 1.3                                         | 4.8                                        |

4. Results of the experiments for aluminum-coated tungsten samples.
Deuterium and hydrogen content in aluminum-coated tungsten before and after deuterium implantation into tungsten, exposition to vacuum and H$_2$+1%O$_2$ plasma irradiation are shown in Table 2.

Table 2. Hydrogen and deuterium content in aluminum-coated tungsten samples after deuterium implantation, exposition to vacuum, exposition to atmosphere and H$_2$+1%O$_2$ plasma irradiation of aluminum coating.

| № | Experiment                                      | Deuterium content, $\times10^{20}$ at/m$^2$ | Hydrogen content, $\times10^{21}$ at/m$^2$ |
|---|------------------------------------------------|---------------------------------------------|--------------------------------------------|
| 1 | Before deuterium implantation                   | -                                           | 3.6                                        |
| 2 | After deuterium implantation into tungsten      | 2.9                                         | 3.1                                        |
| 3 | After implantation and exposition to vacuum     | 3.1                                         | 3.3                                        |
| 4 | After irradiation of Al by H$_2$+1%O$_2$ plasma | 1.5                                         | 4.0                                        |

It can be seen that a significant amount of aluminum was absorbed into the sample from the residual gas (Table 2 row 1). Its decrease in content during deuterium implantation (Table 2 row 2) can be explained by sputtering of the surface layer by deuterium ions and isotope exchange. Exposition of the sample implanted from tungsten side to vacuum did not lead to any changes in deuterium content (Table 2 row 2, 3), which correlates with the results obtained for a non-coated tungsten sample (Table 1 row 2, 3). Irradiation by H$_2$+1%O$_2$ plasma ions of the aluminum coating initiated the desorption of approx. half (48%) of trapped deuterium (Table 2, row 2, 4), as with tungsten sample (Table 1, row 2, 4); hydrogen content increased by 11%. The conclusion can be made that ASP develops on the surface of the aluminum coating on tungsten.

5. Multiple tungsten irradiations by deuterium and hydrogen ions.
The results of the experiments with multiple iterations of tungsten irradiation by deuterium and hydrogen ions are shown on Fig. 1. It can be seen that after the second cycle deuterium concentration
inside the sample was set to $3.1 \times 10^{20}$ at/m$^2$, which remained almost unchanged until the fifth cycle. It can be seen that hydrogen is trapped at a much higher rate than deuterium, however, the increase of total hydrogen isotope content inside the sample slows down after the second iteration of the cycle, which means that periodic irradiation by $\text{H}_2 + 1\% \text{O}_2$ plasma ions prevents deuterium accumulation in tungsten.

As the first wall and the divertor of a thermonuclear reactor will be irradiated not only by fast neutrals from the center of plasma, but also slow ions and atoms from the periphery, it can be assumed that oxygen impurity sorbed on the surface of elements will assist ASP. For that reason, it can be assumed that tritium removal from tungsten and beryllium plasma-facing elements will occur at a constant rate during the operation of the reactor.

6. Conclusion
It was shown in this work that ASP develops on the surfaces of both tungsten and aluminum-coated tungsten. The features of the development of ASP on tungsten and aluminum film on tungsten allow concluding that the growth of tritium trapping in the ITER tungsten divertor tiles during the operation of the reactor can be halted by periodic irradiation by low-ionized deuterium plasma with small oxygen impurity.

Similarity of beryllium and aluminum parameters permits halting tritium trapping in beryllium-coated divertor tiles, as well as beryllium first wall tiles, in a similar fashion.

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