IMPLANTATION OF DEUTERIUM AND HELIUM IONS INTO COMPOSITE STRUCTURE WITH TANTALUM COATING

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Processes of retention and thermal release of implanted deuterium and helium from tantalum coatings of multilayer composite structure by thermodesorption spectrometry were investigated. The dependences of amount of accumulated deuterium and helium, and the shape of thermal desorption spectra as functions of the D^+ and He^+ ions fluencies and of the temperature of irradiation were shown. Possible mechanisms of these processes are proposed.

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INTRODUCTION

Tantalum and tungsten are considered [1-4] as the materials used for protective coatings of structures facing the plasma in CTF facilities. One of the advantages of their application is the insignificant accumulation of hydrogen isotopes in them under the effect of plasma flows. Being actual, this problem is studied intensively [1-7] to increase the radiation resistance of CTF facilities and to obtain new materials with improved parameters while using the ion implantation technique. In [5-8] we carried out comprehensive analysis of the radiation defects formation in matrix, of capture, retention, and thermal desorption of ion-implanted deuterium and helium in tungsten coatings of multilayer composite systems. In this paper the processes accompanying the implantation of deuterium and helium ions into the composite structures with tantalum coating: capture, retention and release of the implanted gases from the coating were studied. The effects of the irradiation dose of D^+ and He^+ ions and the temperature of the sample on the above processes under irradiation were shown. The processes of accumulation and retention of deuterium and helium under the ion irradiation and of the thermal desorption of these gases from Ta and W coatings at the subsequent heating were compared. The types of the matrix radiation damage were determined; their influence on the coatings structural features was shown.

1. EXPERIMENTS

The studies were carried out on tantalum coatings obtained by magnetron sputtering of Ta target in the Ar atmosphere at the pressure of 1.0 Pa. The deposition was carried out at the rate of 0.6 nm/s at the temperature $T = 600$ K on a 0.8 mm thick stainless steel substrate with a Ti interlayer of less than 10 nm thickness predeposited on it. The thickness of the tantalum coatings under study was 1.5 μm. The composite structure with a Ta coating was marked as (SSt + Ta). The samples were irradiated by ion beams of 20 keV He^+ or 20 keV D_2^+ (10 keV D^+), at the current density of ~ 5 μA/cm^2 to the doses Φ within the interval of $(0.1…1.1) \times 10^{18}$ cm^{-2}. Deuterium ions were implanted into the tantalum coatings at the sample temperatures T_o: 290, 370, 470, 570, and 670 K. According to the calculations in [9] for the ions D^+ (10 keV) and He^+ (20 keV) the mean projective ranges of the ion in the tantalum coatings were about 60 nm, and total ones were about 160 nm. They were comparable for the ions under study and much shorter than the coating thickness.

In the studies, the method of thermodesorption spectrometry (TDS) was used. Thermodesorption spectra of helium and deuterium were obtained using PTI-7A gas mass spectrometer calibrated with a helium leak valve GELIT-1, applying the data of [10] on the ionization cross section of helium and deuterium particles in the ion source of this mass spectrometer. The spectra of thermal desorption of helium and deuterium were obtained, and the values of the concentration C and of the capture coefficient $\eta = C/\Phi$ of the implanted gases were determined. The studies of helium and deuterium thermal desorption were carried out by heating the irradiated samples at the constant rate $\alpha = 0.8$ K/s in the temperature range of 290…1800 K. The spectra represent the dependences of the number of S particles of the implanted gas released at the given heating temperature T on this temperature. The error of the temperature measuring is ± 25 K. The sensitivity of the technique used to determine S is not worse than 2×10^{12} cm^{-2}. When the unirradiated samples were heated in the same temperature range, the maximum of background intensity for ions with $m = 4$ a.m.u. did not exceed $S = 0.003 \times 10^{16}$ cm^{-2}.

2. RESULTS AND DISCUSSION

When a composite structure with the tantalum coating irradiated by D^+ or He^+ ions is heated, thermal release of the gases implanted into the vacuum is observed. Fig. 1 shows the spectra of deuterium thermal desorption from the tantalum coating of the composition (SSt + Ta) (curves 1-5). For comparison, the data on thermal desorption from the tungsten coating of the composition (SSt + W) (curves 1'-6') taken from [8] are given. The compared samples were irradiated at the room temperature by D^+ ions with the same energy to different doses.

The spectra of the deuterium thermal desorption from the tantalum coating of the same composite structure, irradiated by D^+ ions with the same dose ($\Phi_{D^+} = 2.0 \times 10^{17}$ cm^{-2}) but at different temperatures of the samples during deuterium implantation, are shown in Fig. 2.

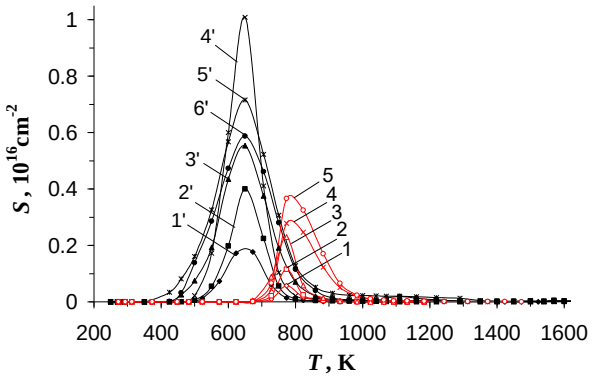

Fig. 1. The spectra of the deuterium thermal desorption from the Ta coating irradiated with D^+ ions (Φ_{D^+}, 10^{17} m^{-2}: 1 – 1.0; 2 – 2.0; 3 – 4.0; 4 – 8.0; 5 – 11.0) and, according to [8], from the W coating irradiated with D^+ ions (Φ_{D^+}, 10^{17} cm^{-2}: 1′ – 1.0; 2′ – 2.0; 3′ – 3.0; 4′ – 4.0; 5′ – 5.0; 6′ – 7.0); α = 0.8 K/s

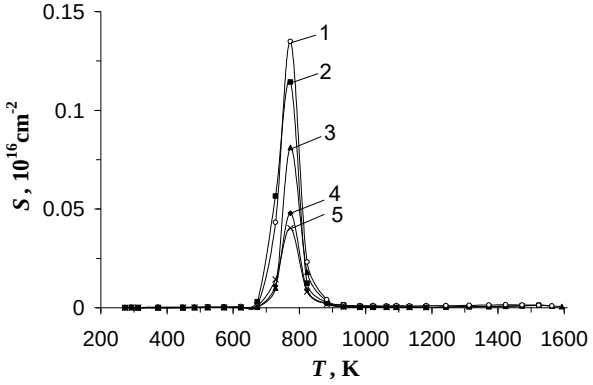

Fig. 2. The spectra of the deuterium thermal desorption from the Ta coating irradiated with D^+ ions (Φ_{D^+}, 2.0×10^{17} cm^{-2}) at different temperatures of irradiation T_o, K: 1 – 290; 2 – 370; 3 – 470; 4 – 570; 5 – 670; α = 0.8 K/s

Figs. 1 and 2 show, that a noticeable release of deuterium from the tantalum coating starts at temperatures $T \geq 620$ K and finished at $T \approx 1200$ K (curves 1-5). The maximal thermal desorption of deuterium is observed at the temperature $T_m \approx 770$ K. According to [8], a noticeable deuterium release from the tungsten coating starts at temperatures $T \geq 320$ K and finished at $T \approx 1000$ K (see Fig. 1, curves 1′-6'). The peak of thermal desorption of deuterium from the W coating has its maximum at $T_m \approx 640$ K.

The thermal desorption spectra of the deuterium, whose ions are implanted into Ta or W coatings at the irradiation doses not exceeding 11.0×10^{17} cm^{-2} for Ta and 7.0×10^{17} cm^{-2} for W, have the form of a single-peak dependence. For the peak of deuterium release from the Ta coating T_m shifts to a higher temperature, and the peak intensity is about a half of the thermal desorption peak from tungsten.

Fig. 3 shows the spectra of helium thermal desorption from the tantalum coating of the composition (*SSt* + Ta) (curves 1-4) and, for comparison, the analogous results of thermal desorption from the tungsten coating of the composition (*SSt* + W) (curves 1'-4'), taken from [8]. The compared samples were irradiated at the room temperature by He^+ ions of the same energy to different similar doses.

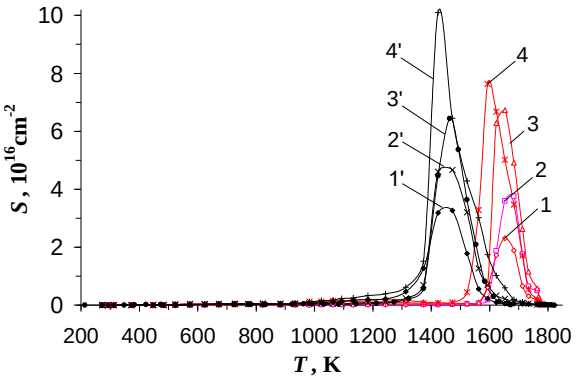

Fig. 3. The spectra of helium thermal desorption from the Ta coating irradiated with He^+ ions (Φ_{He^+}, 10^{17} cm^{-2}: 1 – 1.0; 2 – 1.9; 3 – 4.2; 4 – 6.0) and, according to [8], from the W coating irradiated with He^+ ions (Φ_{He^+}, 10^{17} cm^{-2}: 1′ – 1.0; 2′ – 2.0; 3′ – 3.1; 4′ – 4.0); α = 0.8 K/s

Fig. 3 (curves 1-4) shows, that helium released from the tantalum coating within the temperature range of $1000 \leq \Delta T \leq 1800$ K. A single peak of helium thermal desorption with maximum temperature $T_m \approx 1650$ K is observed. A noticeable release of helium from the tungsten coating, according to [8], starts at temperatures $T \geq 800$ K and finished at $T \approx 1800$ K (see Fig. 3, curves 1'-4'). The maximum helium thermal desorption from the W coating occurs at $T_m \approx 1500$ K. The comparison of the spectra of He thermal desorption from Ta and W coatings shows, that at the irradiation doses, not exceeding 6.0×10^{17} cm^{-2} for Ta and 4.0×10^{17} cm^{-2} for W, the spectra of helium thermal desorption have the form of a single-peak dependence. In this case, T_m of the peak of the helium release from the Ta coating shifts to the higher temperatures, as compared with the W coating. Any significant decrease in the peak height is not observed. It was also determined that the deuterium release from the coatings of both types occurs at lower temperatures, as compared to that of helium, (see Figs. 1, 3).

Fig. 4 shows the dependences of the concentrations *C* for deuterium (1, 1') and helium (2, 2'), implanted at the room temperature, on the irradiation dose of D^+ and He^+ ions for the tantalum coating (curves 1, 2), and for the tungsten coating data taken from [8] (curves 1', 2').

Fig. 5 shows the dependences of the capture coefficient for deuterium η_D (1, 1') and helium η_{He} (2, 2') on the dose of irradiation by D^+ and He^+ ions for Ta coating (curves 1, 2) and, according to [8], for the W coating (curves 1', 2'). As Figs. 4 and 5 show, the values of the concentration *C* and the helium capture coefficient η_{He} are much higher for both tantalum and tungsten coatings than the analogous values of *C* and η_D for deuterium. As compared to helium, deuterium accumu-

lates in the coatings of both materials to a lower concentration with the capture coefficient of about an order of magnitude lower. With increase of the dose Φ of irradiation by He^+ ions, the value of η_{He} decreases for both tantalum and tungsten coatings (see Fig. 5, curves 2 and 2'). Some decrease in η_D with an increase in Φ of D^+ ions is observed only for the tungsten coating (see Fig. 5, curve 1').

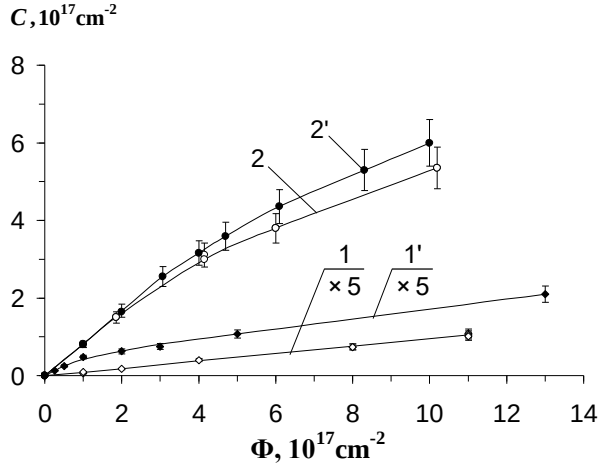

Fig. 4. Dependences of the concentrations of the implanted deuterium (1, 1') and helium (2, 2') on the dose of irradiation by D^+ and He^+ ions for the Ta coating – curves 1, 2, and, according to [8], for the W coating – curves 1', 2'; T_o = 290 K

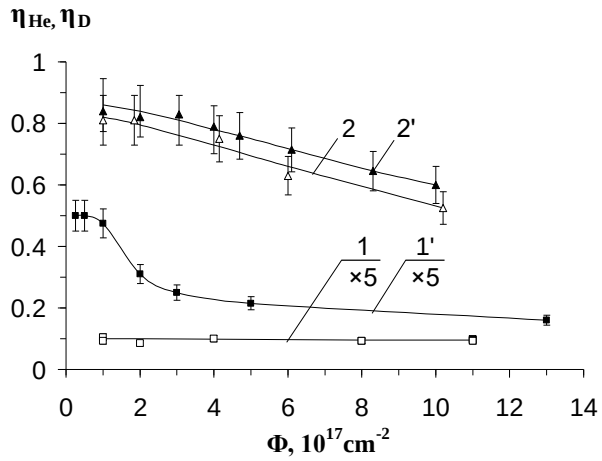

Fig. 5. The dependences of the capture coefficient of deuterium (1, 1') and helium (2, 2') on the dose of irradiation by D^+ and He^+ ions for the Ta – curves 1, 2 and, according to [8], for the W coating – curves 1', 2'; T_o = 290 K

The dependences of the capture coefficient of deuterium η_D on the temperature T_o of the sample irradiated by D^+ ions for the Ta coating are shown in Fig. 6 by curve 1, and for the W coating by curve 2. With increase of T_o some decrease in η_D is observed for both Ta and W coatings. In the analyzed interval ΔT_o some more significant (approximately tenfold) decrease in the η_D value for the tungsten coating is observed.

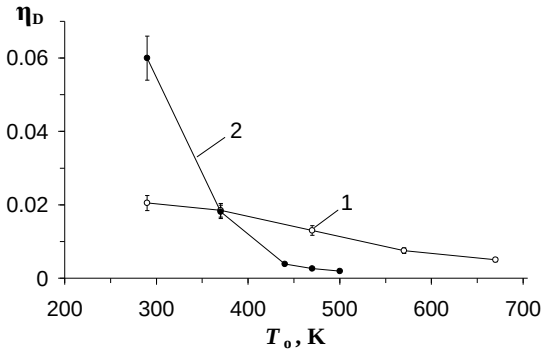

Fig. 6. The dependence of the capture coefficient of deuterium on the temperature T_o of the sample irradiated by D^+ ions for the Ta coating – curve 1, and for the W coating – curve 2; $\Phi_{D^+} = 2.0 \times 10^{17}$ cm^{-2}

From the results (see Figs. 4 and 5, curves 2 and 2') it follows, that at T_o = 290 K for Ta and W coatings, the process of He^+ ions implantation into them to various doses in the interval of $1.0 \times 10^{17} \le \Phi \le 1.1 \times 10^{18}$ cm^{-2} can be described by the same dependences $C = f(\Phi)$ or $\eta_{He} = f(\Phi)$ within the measurement error. But D^+ ions implantation into the same coatings and at the same temperature T_o up to different doses in the interval of $1.0 \times 10^{17} \le \Phi \le 1.3 \times 10^{18}$ cm^{-2} is not described by the same dependences $C = f(\Phi)$ or $\eta_D = f(\Phi)$. A particularly large difference is observed in the dependence $\eta_D = f(\Phi)$. If for the W coating the coefficient of deuterium capture decreases with the increase of the irradiation dose of D^+ ions, then for the Ta coating it does not change. This may be due to different types of the deuterium solubility: endothermic (W) or exothermic (Ta), and due to the ability to form hydrides in tantalum.

The authors of this work early carried out electron microscopic studies of changes in the microstructure of W coatings containing deuterium or helium implanted at room temperature (see [11]). The studies showed the formation of interstitial dislocation loops and dislocation nets with a mean diameter $d \ge 5$ nm and the density exceeding 3.2×10^{12} cm^{-2} in the coatings irradiated by D^+ ions ($\Phi_{D^+} \le 6 \times 10^{18}$ cm^{-2}) or He^+ ($\Phi_{He^+} < 7.0 \times 10^{17}$ cm^{-2}) while the formation of deuterium and helium bubbles was not observed. The helium bubbles were observed at $\Phi_{He^+} \ge 7 \times 10^{17}$ cm^{-2}; their mean diameter and density were 2.5 nm and 5×10^{12} cm^{-2}, respectively, at $\Phi_{He^+} = 7 \times 10^{17}$ cm^{-2}.

On the bases of the obtained results, it can be assumed that in both Ta and W coatings [8, 11], irradiated by D^+ or He^+ ions, some radiation defects are formed such as vacancy-type defects and gas-vacancy complexes, which are traps for the implanted gases, as well as the dislocation interstitial loops.

CONCLUSIONS

The radiation resistance of tantalum coatings of the composite structure (*SSt* + Ta) to the irradiation by He^+ and D^+ ions of medium energies at the doses of $1.0 \times 10^{17} \le \Phi \le 1.1 \times 10^{18}$ cm^{-2} and at different temperatures of the sample under bombardment was studied. The spectra of thermal desorption of deuterium and helium from Ta coatings into the vacuum

were studied, and the capture coefficients of these gases in the coating were determined. When implanting D^+ and He^+ ions into the tantalum coatings it was found, that deuterium accumulates in the coating with the capture coefficient of approximately an order of magnitude lower than helium, and its release from the samples into the vacuum as compared to helium occurs at lower temperatures.

It is shown that at T_o= 290 K, the accumulation of He^+ ions ($1.0 \times 10^{17} \le \Phi \le 1.1 \times 10^{18}$ cm^{-2}) in the tantalum and tungsten coatings is described by the same dependences $C = f(\Phi)$ and $\eta_{He} = f(\Phi)$ within the measurement error. The thermal desorption spectra of helium implanted at doses not exceeding 6.0×10^{17} cm^{-2} (Ta) or 4.0×10^{17} cm^{-2} (W) are described by single-peak dependences. D^+ ion implantation into the same coatings and at the same temperature T_o up to different doses $1.0 \times 10^{17} \le \Phi \le 1.3 \times 10^{18}$ cm^{-2} is not described by the same dependences $C = f(\Phi)$ and $\eta_D = f(\Phi)$.

In the tantalum coatings irradiated by D^+ or He^+ ions is supposed the formation of the following radiation defects: vacancy-type defects, dislocation interstitial loops, gas-vacancy complexes.

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ИМПЛАНТАЦИЯ ИОНОВ ДЕЙТЕРИЯ И ГЕЛИЯ В КОМПОЗИЦИОННУЮ СТРУКТУРУ С ТАНТАЛОВЫМ ПОКРЫТИЕМ

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Методом термодесорбционной спектрометрии исследованы процессы накопления и выделения дейтерия и гелия в вакуум из танталовых покрытий композиционной структуры. Количество накопленного дейтерия и гелия и вид спектров термической десорбции показаны в зависимости от дозы облучения ионами D^+ или He^+ и температуры образца при облучении. Предложены возможные механизмы этих процессов.

ІМПЛАНТАЦІЯ ІОНІВ ДЕЙТЕРІЮ ТА ГЕЛІЮ В КОМПОЗИЦІЙНУ СТРУКТУРУ З ТАНТАЛОВИМ ПОКРИТТЯМ

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Методом термодесорбційної спектрометрії досліджено процеси накопичення і виділення дейтерію та гелію у вакуум із танталових покриттів композиційної структури. Кількість накопиченого дейтерію та гелію і вигляд спектрів термічної десорбції показано в залежності від дози опромінення іонами D^+ або He^+ та температури зразка при опроміненні. Запропоновано можливі механізми цих процесів.