

COMPARISON OF STRUCTURAL DISRUPTIONS ON THE TUNGSTEN SURFACES WHICH INDUCED BY IRRADIATION OF HELIUM IONS BEAMS WITH ENERGIES 0.12 AND 4 MeV

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Helium ion beams with energies $E_{\text{He}^+} = 0.12$ and 4 MeV were used to irradiate samples of tungsten with a purity of 99.7%. Graphs of coefficient of the ionization, coefficient of production of phonon and vacancies are obtained. These processes occurred as a result of irradiation. The differential and integral curves of the sputtering coefficient from the surface of samples of tungsten were constructed, that allowed to determine the number of recoil atoms that reached the surface. The total coefficients of displacements on the surface of the samples were calculated. These characteristics are compared with similar characteristics obtained by irradiating of samples of tungsten (tungsten samples) with a purity of 99.5% with helium ions (0.12, 4 MeV). Areas in which the characteristics are similar and areas where these characteristics differ are marked.

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INTRODUCTION

The use of beams of accelerated particles allows deciding a few tasks. One of them is the simulation of the effect of ionization radiation on various materials. For these purposes, beams of accelerated ions and the beams of accelerated electrons are widely used. In our work, we considered the effect of accelerated beams of helium ions with different energies on the surface of material. To obtain helium ions beams, a set of equipment was made [1, 2].

Irradiation of materials with helium ion beams makes it possible to simulate processes and structural disturbances. These violations occur when materials are exposed to various types of ionizing radiation. Heavy ions have a scattering cross section five times larger than neutrons. Therefore, they can create 10^6 – 10^7 times more radiation damage than with neutron irradiation. In a few hours of irradiation, it is possible to get radiation damage that takes several years to develop in reactors. The beam of heavy ions must have an intensity of several microamperes.

At creation of thermonuclear reactors it is necessary to decide the problem of change of structure of material, because material is exposed to rays by an ionizing radiation. It is necessary to decide a question, what material to use for creation of the first wall of thermonuclear reactor. The importance of this issue is determined by the conditions of the location of the first wall in the active area of reactor.

The first wall can be affected by several extreme factors. The first wall of the reactor is in the region of high temperature (>1200 K). The products of nuclear fusion act on the first wall of the reactor. The streams of particles can have intensity: $\sim 10^{22} \dots 10^{24} \text{ H}^{2+} \cdot \text{m}^{-2} \cdot \text{s}^{-1}$ (<500 eV) helium ions and $\sim 10^{24} \text{ H}^+ \cdot \text{m}^{-2} \cdot \text{s}^{-1}$ (1...10 eV) protons. Also, fast neutrons influence on the first wall of the reactor (14.2 MeV, ~ 2 dpa) [3, 4].

As a result of influence of high temperature and radiation factors, different effects appear on the surface the material of the first wall and inside the material of the first wall. There is a melting of individual points on the surface, bubble boiling, evaporation, sputtering of the material of the first wall. At the same time, the effects of blistering and flecking work. Cavities, “hills” appear on the surface, and material is pulled out. Thin microscopic outgrowths (“fuzz”) appear. The microscopic outgrowths (“fuzz”) grow in height and width. The speed of growth in different directions is different.

Microscopic characteristics also change. As a result of thermal and radiation exposure, the appearance and increase of defects in the crystal lattice of the materials of the first wall occurs. All these effects lead to deterioration of characteristics and destruction of the material of the first wall.

The main candidates that are proposed for the first wall may be refractory materials. Tungsten is a promising material [5–9]. Tungsten has a number of specific characteristics that meet the requirements of material of the first wall. The temperature of melting of tungsten is high. He has a high residual strength, low coefficient of erosive sputtering.

Also, tungsten has high radiation-protective characteristics. Composite materials were created on its basis. Thermal conductivity values have been obtained for these composite materials. Changes in micro hardness were studied for various composites. All these changes occurred as a result of irradiation and accumulation of structural defects [10–12]. Tungsten is used to create hard balls in ball mills, which are used in uranium mining [13].

Tungsten retains almost no tritium and has minimal swelling. At present, technologies for applying coatings of tungsten on ceramics have been developed. Tungsten

is also used to create containers for the storage and transportation of RAW [14].

PURPOSE OF WORK

Study of changes in the characteristics of the surface of tungsten with a purity of 99.7%, which occurred as a result of irradiation the beams of ions of helium with energies of 0.12 and 4 MeV. Calculation of coefficients of ionization, appearance of photons, appearance of vacancies for these cases. Determination of coefficient of sputtering from the surface of tungsten taking into account the thickness of superficial layer. Comparison of all these values, which were obtained for a sample of tungsten with a purity of 99.7%, with the results that were obtained for a sample of tungsten with a purity of 99.5%.

THE MAIN PART

All experimental work were conducted with the use a helium ion beams accelerator [1, 2, 15, 16]. The helium ion beam had the following parameters: pulse current 700 μA , pulse length 500 μs , average current 0.7 μA , repetition frequency 2...5 imp/s, current density $(0.15...0.44) \cdot 10^{13}$ part/ cm^2 . The studies were performed using helium ion beams with energies of 4 (acceleration mode) and 1.2 MeV (injection mode). Diagnostics of beam of ions are carried out using gamma-radiation detectors [17, 18]. The control of thermal characteristics and the vacuum state of the equipment was carried out using methods IR radiometric [19].

In the process of irradiation of tungsten with helium ion beams, there is implantation of ions and change of structure of material [7, 9, 20–24]. The defects of grate of tungsten appear. There is an accumulation of defects of grate of tungsten. There is diffusion of defects of grate. These defects lead to a change in the characteristics of tungsten.

The use of helium ion beams makes it possible to simulate the processes of irradiation of materials that are used as materials for the first wall of a thermonuclear reactor. In this case, it becomes possible to avoid working with radioactive substances, to work in “cold” and “hot” modes, and to reduce the time of research.

In our case, the experiment was performed in the “cold” mode. The irradiation temperature was 380 K. That is, in the “cold” mode, we study changes in of structure of material of tungsten that appeared as a result of radiation exposure. There was no thermal effect.

In the experiments, tungsten with a purity of 99.7% (W – n) was studied. The samples of tungsten were made in the form of circles, which had a thickness of 2 and a diameter of 10 mm. Irradiation was carried out perpendicular to the end face of the circles. The ends of the circles were polished. Such a choice of the shape of the circles is due to the possibility of detecting and studying surface damage, both from the irradiation side and from the opposite side.

In previous experiments, tungsten was studied with a purity of 99.5% (W – 1) [7, 9, 21]. The composition of tungsten was determined using X-ray fluorescence analysis. Experimental data on the composition are given in [7, 9, 21]. In this work, we will compare the

results obtained for a sample of tungsten (W – n) with previously obtained results for the samples of tungsten (W – 1) [7, 9, 21].

The implementation of experimental work is a laborious process; therefore, methods of mathematical modeling were used. Numerical calculations were carried out using the SRIM code [25]. The values of ionization radiation doses were found using the Geant4 v 4.9.6p02 software package [26].

The SRIM software package makes it possible to calculate the mean free paths of ions, the distribution of vacancies, and the distribution of recoil atoms in the target material. The characteristics of damage of target depend on the loss of ions along the trajectory of run. We find the following characteristics: damage of target; sputtering of atoms from the surface of target; ionization of the material in the volume of the target; phonon production in the volume of the target.

The coefficients of ionization, coefficients of damage, and coefficients of production of phonon were obtained for a sample of tungsten (99.7%) (Table 1).

Table 1
The values of energy loss and damage of tungsten samples (W(99.7)) when irradiated with helium ion beams with energies of 0.12 (a) or 4 MeV(b)

Energy loss, $E_{\text{He}^+} = 0.12 \text{ MeV}$. Formation of vacancies = 146 vac./ion Damageability = 166 displacements/ion free run (max) = 0.5 μm			
Parameters	Ionization, %	Displacements, %	Phonons, %
Ions He^+	93.98	0.08	0.74
Cascade of Displacement	1.38	0.28	3.57

b

Energy loss, $E_{\text{He}^+} = 4 \text{ MeV}$. Formation of vacancies = 339 vac./ion Damageability – 386 displacements/ion free run (max) = 7 μm			
Parameters	Ionization, %	Displacements, %	Phonons, %
Ions He^+	99.59	0.00	0.04
Cascade of Displacement	0.10	0.02	0.24

It follows from the Table 1, that the main losses of the beam go to the ionization of tungsten. The energy spent on ionization in the volume of sample is 95% (1.2 MeV (see Table 1,a)) and 99.6% (4 MeV (see Table 1,b)). Ionization of tungsten occurs due to the ions of helium.

The processes of cascades of displacements have little effect on the change in the radiation characteristics of material of target. Also, a very small part of the beam energy is spent on the production of phonons. Graphs of changes in the processes of ionization, the formation of phonons, and the processes of segregation (redistribution) of atoms were obtained.

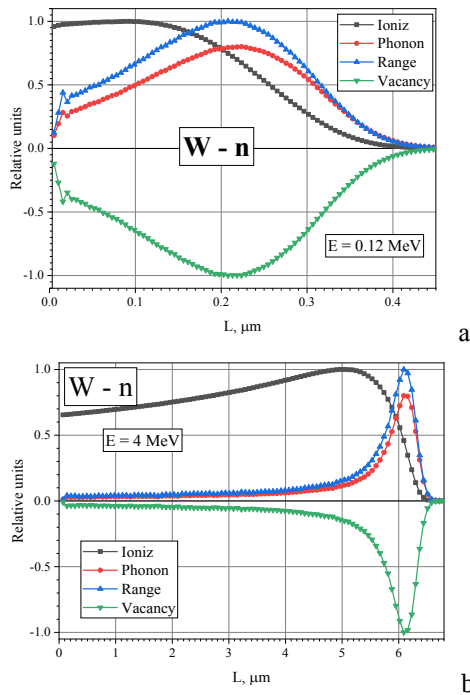


Fig. 1. Curves which show of changes in the number of photons, vacancies, ionization in the sample of tungsten (W – n (99.7%)). Irradiation with helium ions with energy: a – 1.2 MeV; b – 4 MeV

It follows from the graphs that all processes (ionization, formation of phonons and vacancies) for the ions of helium with energies of 4 MeV occur much deeper than for ions with energies of 1.2 MeV. Thus, ionization processes occur to depths of 0.4 μm (1.2 MeV) and to depths of 6.4 μm (4 MeV). The half-width of peaks of production of phonon is in the range of 0.1...0.3 μm (1.2 MeV) and 5.8...6.3 μm (4 MeV). In the case of energies of 4 MeV, the half-width is wider than for the case of 1.2 MeV.

These relations are also valid for the cases of the appearance of vacancies and the segregation of atoms. When of samples of tungsten (W – n) are irradiated with helium ions with an energy of 1.2 MeV, all processes occur up to the surface layer depth of 0.5 μm. For the case when the energy of helium ions is 4 MeV, all processes occur to depths of 7 μm. These differences depend on the mean free path of helium ions. Obviously, at low energies, the mean free path is shorter than at high energies.

A comparison was made of the results presented in this work and the results presented in the works (W – 1 (purity 99.5%)) [7, 9, 21]. The results differ slightly. All maximal values of peaks of ionization, productions of phonons and vacancies are deeper on 0.01 μm for the case of tungsten of type of W – 1 (99.5%), what for the case of tungsten of W – n (99.7%). Such results in the case of helium ion energy $E_{He^+} = 0.12$ MeV.

When irradiated with helium ions with an energy of $E_{He^+} = 4$ MeV, the maxima of the curves of ionization, production of phonons and vacancies are deeper by 0.1 μm for the case of tungsten of type of W – 1 (99.5%) than for the of tungsten of W – n (99 – 7). The shallower depth of the ionization peaks was due to the larger amount of tungsten in the sample. This does not

contradict the general laws of absorption and scattering of ions on atoms.

The quantitative values of the quantities that are presented in Fig. 1 are needed when calculating the displacement coefficient (dpa is the number of displacements per atom). Graphs have been obtained that show the change in the coefficient of displacement. To calculate the coefficient of displacement (dpa), we use the expression:

$$dpa = \frac{D_{ion} \cdot dpi}{N_A \cdot \rho_0 \cdot SL} \cdot \frac{1}{100} \sum_i \mu_i n_i, \quad (1)$$

where D_{ion} – is the radiation dose, ion/cm². The D_{ion} parameter is found from the experimental conditions. The dpi parameter is the number of displacements per ion. The dpi parameter is found from the total number of displacements per ion (see Table 1,a,b). N_A – is Avogadro's number. In this equation: ρ_0 – is the density of the sample, which was measured experimentally, $S=1$ cm² – is the unit area of the sample, L – is the maximal run of ions in a sample. The value $M = 1/100 \times \sum \mu_i n_i$ – is the molar mass of the sample. The value of μ_i – is the molecular weight of the i -th element of the sample, n_i – is the percentage of the i -th element of the sample.

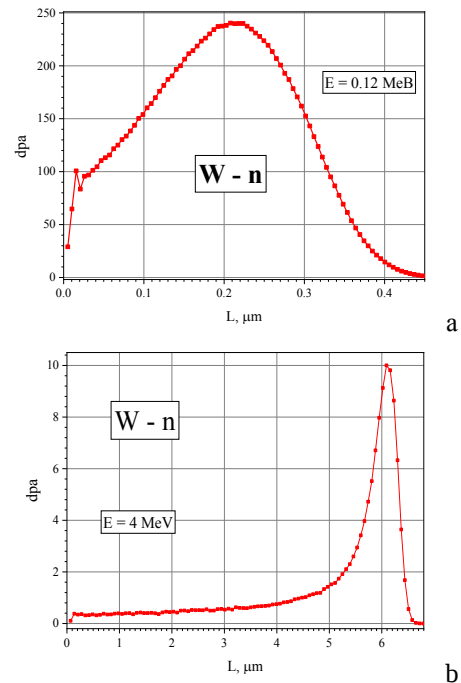


Fig. 2. Displacement formation profile in a sample of tungsten (W – n (99.7%)). Irradiation with helium ions with energy: a – $E_{He^+} = 0.12$ MeV; b – $E_{He^+} = 4$ MeV

When irradiated with ions of helium with an energy of $E_{He^+} = 0.12$ MeV (Fig. 2,a), the maximum of peak of displacement is located at a depth of 0.21 μm. The half-width of the peak of displacement was in the sample depth range of 0.08...0.31 μm. In this region, ions can produce displacements up to 200 dpa. At depths from 0 to 0.08 μm and from 0.31 μm, ions can produce displacements of no more than 130 dpa. The total coefficient of displacement was $dpa \approx 59$ at an irradiation dose of $1 \cdot 10^{18}$ ion/cm².

When irradiated with ions of helium with an energy of $E_{\text{He}^+} = 4 \text{ MeV}$ (see Fig. 2,b), the maximum of peak of displacement is located at a depth of $6.1 \mu\text{m}$. The half-width of the displacement profile peak was in the depth range of $5.8 \dots 6.3 \mu\text{m}$. In this region, ions can produce displacements up to 10 dpa. At depths from 0 to $5.8 \mu\text{m}$ and from $6.3 \mu\text{m}$, ions can produce displacements of no more than 2 dpa. The maximum contribution to displacements is made by ions with path lengths from 5.8 to $6.3 \mu\text{m}$. The total coefficient of displacement was $\text{dpa} \approx 8.7$ at an irradiation dose of $1 \cdot 10^{18} \text{ ion/cm}^2$.

Similar work was also carried out for samples of tungsten of the W – 1 type (99.5%), but in the case of irradiation with ions of helium with energy of 4 MeV [21].

An important parameter which helps to define the degree of damage of surface of material is a coefficient of sputtering of surface. The paper [7] presents the results of numerical calculations of coefficients of sputtering for separate impurities of materials which are in our samples of tungsten. These data are used to calculate the total of coefficients of sputtering from the surface of the samples. The paper [9] presents the results of numerical calculations of the total coefficients of sputtering for tungsten of W – 1 (99.5%). The samples were irradiated with ions of helium ions with energy of 0.12 MeV.

Differential characteristics of sputtering of atoms from the surface of samples of tungsten (W – n) were obtained.

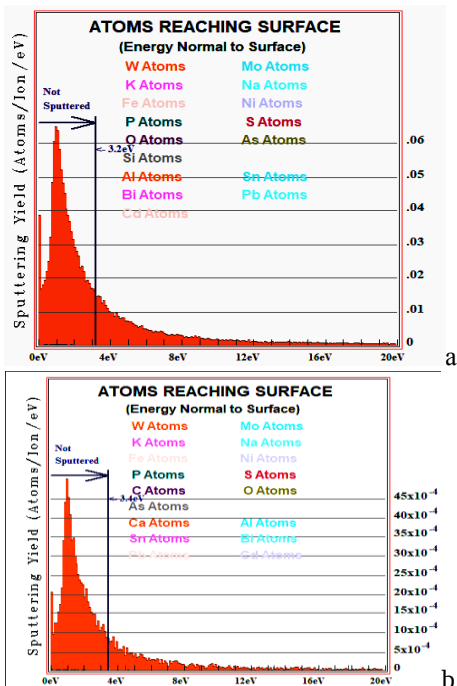


Fig. 3. Differential characteristics of sputtering of tungsten (type of W – n (99.7%)). Irradiation with helium ions with energy:
a – $E_{\text{He}^+} = 0,12 \text{ MeV}$; b – $E_{\text{He}^+} = 4 \text{ MeV}$

There are two regions on the graphs, which are separated by a straight line 3.2 (Fig. 3,a) and 3.4 eV (see Fig. 3,b). These straight lines correspond to middle energy of connection for the sample of tungsten (W – n (99.7%)) that was irradiated ions with energies of 0.12 (see Fig. 3,a) and 4 MeV (see Fig. 3,b). The

regions to the left of lines of the 3.2 (see Fig. 3,a) and 3.4 eV (see Fig. 3,b) contain atoms with energies below than energy of connection. Consequently, these atoms are not sputtered from the surface of the samples.

To the right of these lines are atoms with energies greater than energy of connection. These atoms are sputtered from the surface and contribute to the coefficient of sputtering.

Integral characteristics of sputtering were obtained. They are shown in Fig. 4.

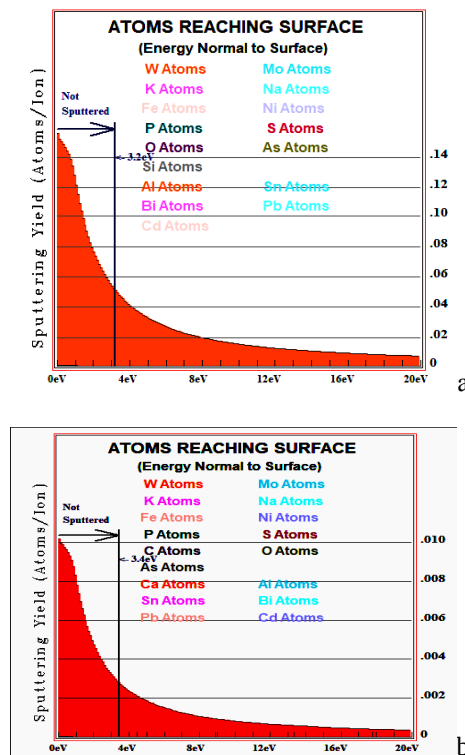


Fig. 4. Integral characteristics of sputtering of sample of tungsten (W – n (99.7%)). Irradiation with helium ions with energy: a – $E_{\text{He}^+} = 0.12 \text{ MeV}$;
b – $E_{\text{He}^+} = 4 \text{ MeV}$

The total energy of recoil atoms that reach the surface of the samples is given along the horizontal axis of the graphs. The number of recoil atoms that have reached the surface is given on the vertical axis. The number of recoil atoms is given in terms of one ion of helium. From the graphs (see Fig. 4) we can determine the number of atoms that are sputtered from the surface of sample of tungsten (type of W – n (99.7%)).

We observe a significant difference in the number of recoil atoms that appear after irradiation with ions of helium with energies of 0.12 or 4 MeV. In the case of energies of 0.12 MeV, the maximum number of atoms that are sputtered from the surface is 0.045 atom/ion. In the case of energies of 4 MeV, the maximum number of atoms that are sputtered from the surface is 0.0025 atom/ion. Amount of atoms which are sputtered from a surface, with energies of 8 eV is 0.005 atom/ion ($E_{\text{He}^+} = 0.12 \text{ MeV}$) and 0.001 atom/ion ($E_{\text{He}^+} = 4 \text{ MeV}$). For an energy of 20 eV, with which atoms of recoil reach the surface of sample, these values are 0.001 atom/ion (0.12 MeV) and 0.0005 atom/ion (4 MeV).

It can be seen from the graphs that the number of atoms that are sputtered on the surface, in terms on one ion of helium, is almost 20 times less for the case of ions of helium with energies (4 MeV) than for the case of ions of helium ions with energies (0.12 MeV).

The surface of the samples, after irradiation, differs significantly when irradiated the ions of helium with different energies (Fig. 5).

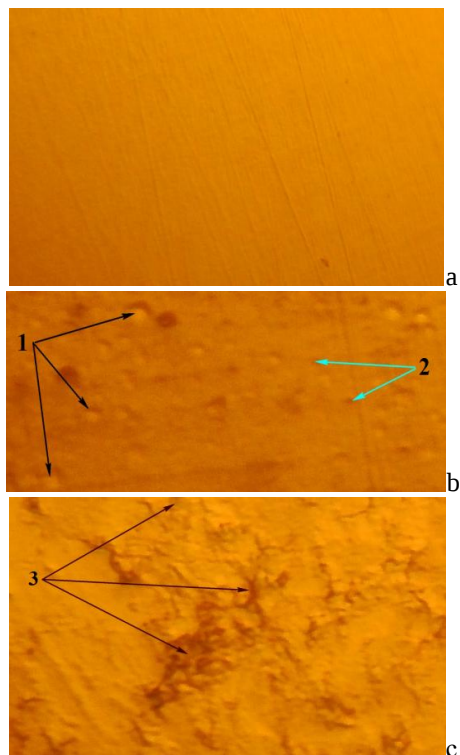


Fig. 5. Photographs of the surface of sample of tungsten (W – n (99.7%)): a – before irradiation, %; b – irradiation with helium ions with energy $E_{\text{He}^+} = 0,12 \text{ MeV}$; c – irradiation with helium ions with energy $E_{\text{He}^+} = 4 \text{ MeV}$

The non-irradiated sample of tungsten (W – n (99.7%)) has a smooth surface (see Fig. 5,a). To accurately perform the experiment, the surface of the sample was polished. This avoids the appearance of effects associated with inhomogeneities

In the case of irradiation of sample of a tungsten (W – n (99.7%)) with beams of ions of helium with an energy of 0.12 MeV, the total coefficient of displacement was $\text{dpa} \approx 59$, at an irradiation dose of $1 \cdot 10^{18} \text{ ion/cm}^2$. In the case of irradiation of sample of a tungsten (W – 1 (99.5%)) with beams of ion of helium with an energy of 0.12 MeV, the total coefficient of displacement was $\text{dpa} \approx 50$, at an irradiation dose of $1 \cdot 10^{18} \text{ ion/cm}^2$ [9, 21]. On the surface of the sample of tungsten, we observe a significant number of pits (craters) (see points similar to 1 Fig. 5,b).

Also, there are protrusions (bulges) on the surface (see points similar to 2 Fig. 5,b). The protrusions are much smaller than the depressions. As in the case of the sample of tungsten (W – 1 (99.5%)) [9, 21], there are five times more pits than protrusions. The reason for this is the inhomogeneity of the processes of flecking and blistering [24].

In the case of irradiation of sample of a tungsten (W – n (99.7%)) with beams of ions of helium with an energy of 4 MeV, the total coefficient of displacement was $\text{dpa} \approx 8.7$, at an irradiation dose of $1 \cdot 10^{18} \text{ ion/cm}^2$. In the case of irradiation of sample of a tungsten (W – 1 (99.5%)) with beams of ion of helium with an energy of 4 MeV, the total coefficient of displacement was $\text{dpa} \approx 4$, at an irradiation dose of $1 \cdot 10^{18} \text{ ion/cm}^2$ [9, 19, 21].

On the surface of the sample, we observe the presence of small tendrils of “fluff” of tungsten (see points similar to 3 Fig. 5,c). These violations are formed as a result of the diffusion of ions of helium to the surface of the material and extrusion of the material into thin tendrils. In our studies, the length of the “fluff” was three times its thickness. In experimental works [21, 23, 24] a height of fuzz was in one and a half or two times more than thickness. This may be due to the effects of annealing. A “fuzz” grows slower, when effects of annealing are. This assumption is consistent with our results. In our case, the temperature of the experiment did not exceed 380 K.

Because processes of formation and development of inhomogeneities and violations of structure of material of the first wall are in the stage of study, our experimental data are useful and necessary.

The problem of radiation resistance of tungsten under the action of beams of charged particles is also relevant in view of the current trends in the development of the latest nuclear reactors. For example, a bimetal based on tungsten and titanium is proposed for the manufacture of a neutron converter for a subcritical assembly reactor [27]. Such a coating can be formed both by rolling and by using charge particles beams. [28–30]. The observed loosening of the tungsten structure must be taken into account when creating such converters, since their operation is expected under conditions of cooling and intense irradiation, i.e., an additional factor here will be the thermoelastic effect, both due to macrocooling and due to the formation of microtracks along the passage of particles [31]. The formation of local temperature peaks in the contact zone of materials under irradiation, in turn, can lead to the formation of a liquid phase at grain boundaries [32] and create conditions for facilitated deformation by the mechanism of grain boundary sliding. Another factor that can affect the mechanism of destruction is the formation of intermetallic phases [33, 34], which causes a change in the value of the crystal lattice parameter.

CONCLUSIONS

1. The samples of tungsten (purity 99.7%) were irradiated beams of ions of helium with energies of 0.12 and 4 MeV.

2. The features of the change of processes of ionization, the processes of formation of phonons and vacancies, which go (pass) to the depths of superficial (of the surface) layer of tungsten 0.45 ($E_{\text{He}^+} = 0.12 \text{ MeV}$) and $6.8 \mu\text{m}$ ($E_{\text{He}^+} = 4 \text{ MeV}$), are determined.

3. Integral and differential characteristics of sputtering of atoms from the surface of tungsten (99.7%) are obtained, depending on the energy of ions of helium. It was found that as a result of the work of

ions with energy of 0.12 MeV, twenty times more recoil atoms appear on the surface than for ions with an energy of 4 MeV. The calculation of recoil atoms is given for one ion.

4. The total coefficients of displacement on the surface of samples of tungsten were calculated: a) dpa $\approx$ 8.7, when ions with energy of 4 MeV; b) dpa $\approx$ 59 when the energy of ions of helium is 0.12 MeV. Dose of irradiation $1 \cdot 10^{18}$ ion/cm². The profiles of the change in the coefficient of displacement with respect to the depth of the surface layer of tungsten are found.

5. Supposition is outspoken, that the increase of temperatures (annealing) on the surface of sample, prevents the growth of the tendrils (fuzz) in height.

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ПОРІВНЯННЯ СТРУКТУРНИХ ПОШКОДЖЕНЬ ПОВЕРХНІ ВОЛЬФРАМУ, ЯКІ ЗУМОВЛЕНІ ОПРОМІНЕННЯМ ПУЧКАМИ ІОНІВ ГЕЛІЮ З ЕНЕРГІЯМИ 0,12 АБО 4 MeV

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Пучками іонів гелію з енергіями $E_{\text{He}^+} = 0,12$ і 4 MeV виконано опромінення зразків вольфраму з чистотою 99,7%. Отримані графіки значень коефіцієнта іонізації, коефіцієнта генерації фононів і вакансій при опроміненні зразків вольфраму іонами гелію. Побудовані диференціальні і інтегральні криві коефіцієнта розпилення з поверхні зразків вольфраму, що дозволило визначити кількість атомів віддачі, які досягли поверхні. Пораховані сумарні коефіцієнти зсувів на поверхні зразків. Виконано порівняння цих характеристик з аналогічними характеристиками, які були отримані при опроміненні іонами гелію (0,12; 4 MeV) зразків вольфраму з чистотою 99,5%. Відзначені області, в яких характеристики схожі і області, де ці характеристики відрізняються.