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FEATURES OF STRUCTURAL DAMAGES OF SURFACE OF TUNGSTEN AS A RESULT OF IRRADIATION WITH HELIUM ION BEAMS WITH ENERGY 4 MeV

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The beams of ions of helium (E_{He}^{+} = 4 MeV) affected samples of tungsten with purity 99.5%. The radiation characteristics of tungsten have been studied. The change in characteristics was caused by irradiation with helium ion beams. Numerical values of the number of vacancies and phonons were obtained depending on the depth of the layer in tungsten (maximum at a depth of 6.1 μm). The sputtering coefficients of atoms from the surface of tungsten are found. The mean free path of helium ions was up to 70 μm. It is determined that the maximum destruction of the surface occurs in the region of the pits. Pits appear in five times more than bubbles.

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INTRODUCTION

Beams of accelerated particles are widely used in radiation science of materials and in technologies of modification of surface. Helium ion beams are of particular interest for research works and for technological works. One of settings (One of the facilities) for obtaining such beams is the accelerator complex of the NSC KIPT [1, 2].

The beams of ions of helium were applied for scientific researches. Using the charged particles it is possible to design many processes which take place at radiation damage. The processes take place in the active zones of nuclear and thermonuclear reactors. The reactors operate under stationary and transitional modes of operation. Heavy ions have a scattering cross section six times larger than neutrons. Heavy ions cause radiation damage $10^6…10^7$ times more than with neutron irradiation (Fig. 1).

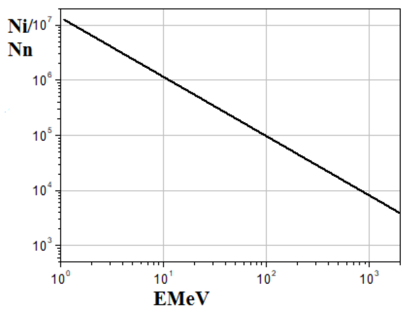


Fig. 1. Dependence of damage on energies (ionic and neutron)

At the same time, the radiation effect, which is achieved in the most powerful operating nuclear reactors for several years, can be reproduced in just a few hours, using a heavy ion beam with an intensity of several microamperes.

Researches were conducted with materials which are used in thermonuclear reactors. At creation of thermonuclear reactors it is required to decide the row of different tasks. Among them it is necessary to decide the task of choice of material of the first wall of reactor. It is related to that materials of the first wall are exposed to a few types of ionization influence [3, 4].

The material is exposed to high temperatures (>1200 K). Materials of the first wall are exposed to influence of plasma. The surface of material is exposed to influence of products of nuclear fusion. Particle flows can have such an intensity: ~ $10^{22}…10^{24}$ $H^{2+}m^{-2}s^{-1}$ (<500 eV) helium ions and ~ 10^{24} $H^{+}m^{-2}s^{-1}$ (1…10 eV) protons [5]. There is an output of ions of helium, as with low energy (in the interval of 100…200 keV, ~ 10^{15} m^{-2}) so with high energy (in the interval of 200 keV…10 MeV). During exploitation of thermonuclear reactor the first wall rapid neutrons affect (14.2 MeV, ~ 2 dpa).

Under the influence of all these factors, various effects appear on the surface of the material and inside the material. Among them are the following effects: the effect of vacation of material, and the effect of softening of the surface layer. Also, the microscopic lakes of molten metal appear on the surface of the material.

The areas of the bubble boiling appear. It causes sputtering of material of the first wall. The effects of Blistering and flecking show up intensively. Speed of growth and height of "fuzz" change. In addition to macroscopic characteristics, microscopic characteristics must also be investigated. The defects of grate in the material of the first wall appear as a result of thermal and radiation exposure. The defects of grate of material of the first wall grow also. The structure of the crystal grate changes. The grain orientation changes. The study and calculation of these effects will allow you to determine how the resistance of the material changes.

Different materials can be used as the material of the first wall. Among them are various composite materials based on ceramics. All refractory materials are considered as a candidate material for the first wall.

One of the perspective materials that can be used in the manufacture of thermonuclear reactors, there is a tungsten [6 - 8]. The choice of tungsten was determined by its specific properties.

Tungsten melts at high temperature. Tungsten has high strength characteristics at high temperatures. Tungsten has a high residual strength after cooling. Tungsten has a low erosive spray coefficient.

Its stopping power with respect to gamma radiation fluxes also led to the use of its powders in radiation protective materials and raised the question of developing new methods for obtaining dispersed tungsten-containing materials [9 - 11].

Also it should be noted that technologies for applying tungsten coatings on ceramics have been developed. Tungsten does not almost retain tritium. The value of microhardness changes as a result of irradiation and accumulation of defects [10, 12]. Tungsten is also examined as a component of protective pencil-cases at transporting and storage of RAW [13]. Since in a thermonuclear reactor, the surface of the first wall is subjected to intense action of helium ions, it is necessary to study what structural changes occur in tungsten.

PURPOSE OF WORK

Analysis of changes in the surface structure of tungsten (purity 99.5%), which occurred as a result of irradiation with an ion beam with energies of 4 MeV. Calculation of coefficients of sputtering on the depth of superficial layer. Calculation of ionization coefficients, phonon yield, appearance of vacancies. Comparison of these coefficients with the coefficients for an ion beam with energies of 0.12 MeV.

THE MAIN PART

The simulation method was used in the study of changes in the surface structure of the material of the first wall. The simulation method allows you to work in different modes (mode "cold", mode "hot", mode without radioactivity). In our case, helium ion beams with energies of 4 MeV were used.

Helium ion beams are an effective tool for irradiating tungsten. This is due to several reasons. Helium is almost insoluble in tungsten. The impact of helium ions leads to changes in the structure of grate of tungsten. Macroscopic characteristics change of tungsten as a result of violations of grate. Because solubility of helium in tungsten is low, and his influence result in considerable violations of grate of tungsten, then it is possible to get such results which in the real terms are accumulated for a few years.

The helium ion accelerator was used as a machine for irradiating the surface of tungsten [1, 2, 14, 15]. The operating parameters of the accelerator are such: 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 MeV (in the acceleration mode).

Diagnostics of the secondary radiation background was carried out using gamma-radiation detectors based on mercury compounds [16, 17]. Monitoring of the state of the vacuum equipment of the accelerator was carried out by IR radiometric methods [18]. Previously, the results of experiments were presented when helium ion beams with energies of 0.12 MeV were used (in the injection mode) [19].

As a result of irradiation of tungsten with helium ions, the possible development of several processes which have nonlinear characteristics. These processes are difficult to predict, since the irradiated object is a non-equilibrium system. The state of this system changes over time. Examples of irradiation of materials with ion beams show a gradual change in mechanical characteristics. In this case, the formation of dislocations occurs [8, 20, 21]. Implanted ions also accumulate. During this process, a number of qualitative structural transformations take place. One of these processes is the formation of subsurface defects [22].

Defects have a different appearance: pores, "hills", cavities, "fuzz". Defects change the characteristics of surface of tungsten. It is necessary to study the dynamics of development of all defects that appear as a result of irradiation. On the basis of these results, we can determine the resistance of tungsten as the material of the first wall.

The experiment was carried out in cold mode. The temperature of the experiment was 380ºC. When working in a cold mode, we can separately study structural damage to the surface of tungsten that arose as a result of radiation influence. And separately study the damage to the surface of tungsten, which arose as a result of thermal influence.

Tungsten, which was investigated in our experiments, had a purity of 99.5% (W-1). Using X-ray fluorescence analysis, the concentration of impurities was found. The composition of samples of tungsten is given in [18]. The samples had the form of pucks with a diameter of 10 mm and a thickness of 2 mm. Samples of this shape allow you to detect all effects.

Effects appear as a result of irradiation. The effects appear on the surface of the sample from the irradiation side and from the opposite side. When irradiated with helium ions with energy of 4 MeV, the following characteristics were obtained. The radiation dose was 1.62×10^{18} ion/cm^2. The radiation damage dose was 8.19 dpa. A fusion reactor must operate for several years to receive such radiation damage.

Irradiation of samples with helium ions is a laborious and costly process. Therefore, we can make a prognosis of changes in the structure of the tungsten surface. The prognosis is carried out using numerical calculations. The SRIM code [23] was used for numerical calculations.

The SRIM package makes it possible to obtain the following characteristics: ion path lengths in materials; distribution of vacancies in the volume of target; distribution of recoil atoms in the volume of target; distribution of ions in the volume of target. Also, such characteristics damage of target can be found by numerical methods: sputtering atoms of target, ionization volume of the target, phonon production in volume the target.

These characteristics depend on the ion energy losses in the material of sample. The attenuation of ionizing radiation was calculated using the Geant4 v 4.9.6p02 software package [24].

In our work, much attention is paid to the sputtering coefficient. Based on its values, we can calculate how the structure of the surface of tungsten changes. When calculating the sputtering coefficient, a thin surface layer is considered. The sputtering coefficient depends on the amount of ions that return to the surface. These ions return as a result of cascades of scattering and reflection.

The main parameters that are necessary for calculating the sputtering coefficient are: the values of superficial energy of connection and the displacement energy. These quantities must be known both for tungsten (the main element) and for all impurity elements that are included in tungsten. The sputtering coefficient is found from this value. The value is calculated as the average number of target atoms which are sputtering, calculating on one falling ion. This value is called "sputtering yield". When carrying out numerical calculations, it was considered that the irradiation dose had a value of 5×10^6 helium ions.

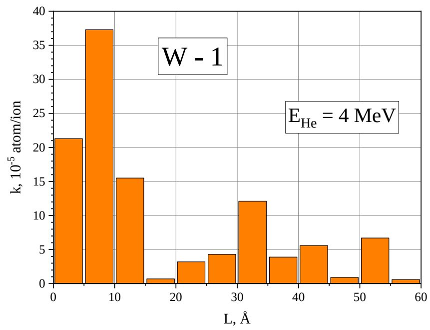
The coefficients of sputtering of atoms were got for elements which are included in the sample of tungsten of type of W-1. The coefficients of sputtering are presented depending on the thickness of layer of sample. Ions of helium had energy 4 MeV. These coefficients are resulted in Table.

Sputter coefficients (sc).
Sample of tungsten (type of W-1),
density (ρ) = 19.275 g/cm³
($k\cdot10^{-5}$ atom/ion, E_{He}^{+} = 4 MeV)

Additives	Thickness targets Å			
	5	20	40	60
(W-1)	21.1	74.3	97.0	111
(Fe)	0	0	0.4	0.1
(Al)	0	0.1	0.1	0.2
(Cr)	0	0	0.2	0.1
(Ni)	0	0	0	0.2
(Co)	0	0	0.1	0.1
(Mo)	0.1	0	0	0
(Mn)	0	0.1	0.2	0
(Ca)	0	0	0	0.1
(Si)	0.1	0	0	0
(Pb)	0	0	0.2	0.1
(O)	0	0.3	0.1	0.2
(Mg)	0	0	0	0
(S)	0	0	0	0
(P)	0	0	0	0
(∑)	**21.3**	**74.8**	**98.3**	**112.1**

The total sputtering ratio is SK ≈ 112.1×10^{-5} atom/ion. The sputtering coefficient is given taking into account the accumulation on the depth of the sample. The overall sputtering ratio is given for W-1 tungsten. The sputtering coefficient is given at helium ion energies of 4 MeV. Tungsten brings in a basic contribution to the coefficient of sputtering.

The contribution of other impurities is insignificant. Previously [18], the values of all coefficients were obtained for a W-1 tungsten sample, but in the case of irradiation with a beam of helium ions with energies of 0.12 MeV. The total sputtering coefficient, at ion energies of 0.12 MeV, is SK ≈ 1958.4×10^{-5} atom/ion (W-1) [18]. Coefficient of sputtering in 20 times greater for helium ions with energies of 0.12 MeV than for ions with energies of 4 MeV. Admixtures render greater influence on the coefficient of sputtering (case 4 MeV). The calculated data are confirmed by experimental data. In photographs of the surface of samples of tungsten, we observe a different surface structure for different radiation doses [18]. The total coefficient of sputtering strongly depends on the depth of location of layers of sample.



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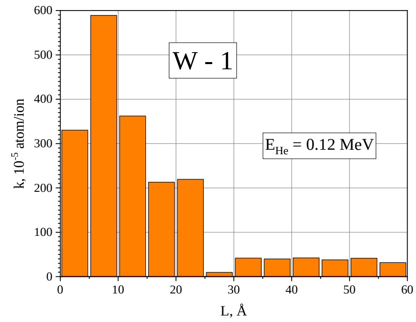


b

Fig. 2. Change of total coefficient of sputtering (∑) on the depth of sample of tungsten of type of W-1: irradiation with helium ions with energy 4 MeV (a); irradiation with helium ions with energy 0.12 MeV (b)

The distribution of the total coefficient of sputtering over the depth of the samples has interesting features. The maximum contribution is made by interactions in surface layers up to 15 Å (4 MeV). Whereas for ions with energies of 0.12 MeV, the main contribution to the total coefficient of sputtering is made by interactions in layers up to 25 Å.

A feature is that another layer appears, which affects the coefficient of sputtering (4 MeV). This layer is at a depth of 30…35 Å. Energy transfer is obtained by reflecting ions from atoms that are located in deeper layers. The number of interacting atoms per helium ion also differs significantly. For a case 0.12 MeV it is an amount anymore almost in 20 times. Accordingly, the number of surface defects is also greater.

To accurately calculate the coefficient of sputtering, it is necessary to know the energy of the recoil atoms that reach the surface. For this purpose, the integral spray characteristics were calculated (Fig. 3).

The graph shows the energy of the recoil atoms that reach the surface of the sample. The vertical axis gives the number of recoil atoms that have reached the surface. The number of recoil atoms is given in terms of one helium ion.

A straight line on graphic arts (3.8 eV) represents middle superficial energy of connection for this sample (W-1). In an area which is on the left of this line (area of Not Sputtered), there are atoms with energy below 3.8 eV. Therefore, they cannot be sputtered from the surface of sample. In the region to the right of line the 3.8 eV, there are atoms with energy more than energy of connection. These atoms are sputtered from the surface and contribute to the coefficient of sputtering.

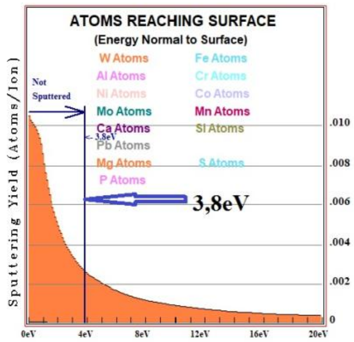


Fig. 3. Integral characteristics of sputtering of sample of tungsten (W-1). Irradiation with helium ions with energy 4 MeV

The differential characteristics of sputtering of atoms on the surface of tungsten were calculated (Fig. 4).

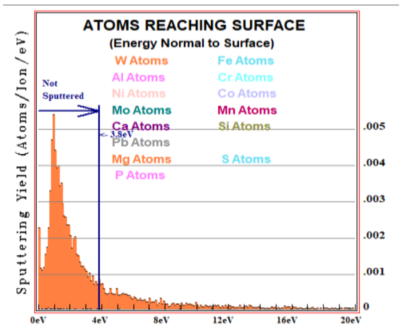


Fig. 4. Differential characteristics of sputtering of tungsten (type of W-1). Irradiation with helium ions with energy 4 MeV

From the graph (see Fig. 4), we can find the energy of atoms on the surface of the sample (tungsten of W-1). Evidently, that no more than 0.003 atoms on every ion, are sputtered from the surface.

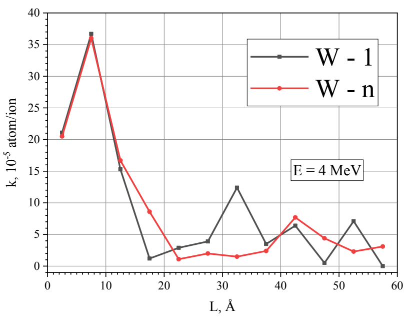


Fig. 5. Change of coefficient of sputtering depending on length of run of ions of helium (W-1)

The coefficient of sputtering depends on run-length of ions of helium (Fig. 5). It was found that the maximum number of atoms is sputtered in the range of mean free paths from 4to 12 Å. Graphs for the samples of tungsten W-1 and W-n differ little. The difference in the region is from 28 to 38 Å. We see a peak in this area. Probably, the reason is the additional interaction of atoms that interacted at a great depth. The second reason may be the processes of interaction with atoms of oxygen in the W-1 sample.

In the process of irradiation of target there is a change of its structure along the trajectory of run of ions of helium. The ionization changes of tungsten, phonons are formed, vacancies appear, and segregation (redistribution) of atoms occurs (Fig. 6).

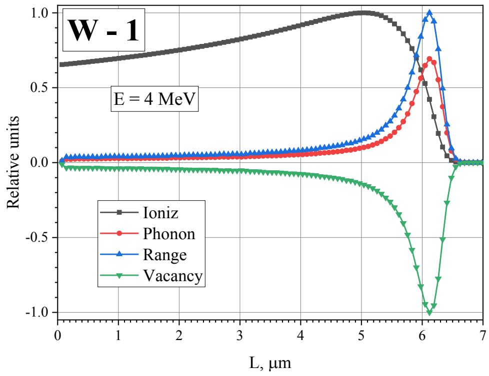


Fig. 6. Curves which show of changes in the number of photons, vacancies, ionization in the sample of tungsten (W-1). Irradiation with helium ions with energy 4 MeV

It can be seen from the graphs (see Fig. 6) that ionization processes occur in the surface layer up to 6 µm deep. Maximum ionization occurs in a layer up to 5 µm. These values are significantly larger than for the case with energies of 0.12 MeV. The processes of formation of photons and vacancies occur in the surface layer at a depth of 5 to 6.6 µm.

Half-width of the peak of output of photon and formation of vacancy is in the region from 5.7 to 6.3 µm. Thus, all structural changes occur only in the surface layer to a depth of 6.6 µm, when of samples of tungsten (W-1) are irradiated with a helium ion beam with energies of 4 MeV.

Graphs (see Fig. 6) are used to find of the coefficient of damages (*dpa* – the number of displacements per atom) and curves along which of the coefficient of damages changes. The value of the coefficient of damages (*dpa*) is found from the expression:

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

where D_{ion} is the radiation dose [ion/cm^2]. 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). N_A is Avogadro's number. In this equation: ρ_0 is the density of the sample, which was measured experimentally, S =1 cm^2 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.

The displacement profile had a maximum at a path length of 6.1 μm (E_{He}^+ = 4 MeV, dpa ≈ 60). The radiation dose was 1.62×10^{18} ion/cm^2. Ions with path lengths from 0.1 to 4 μm create no more than 2 dpa. The half-width of the displacement profile peak was in the range of mean free paths from 5.8 to 6.4 μm (Fig. 7).

The peak of displacement had sharp boundaries. Consequently, the maximum contribution to the appearance of displacements is made by ions with path lengths from 4 to 6.5 μm. Point of peak of displacements coincides with the point of peaks in the formation of photons, vacancies.

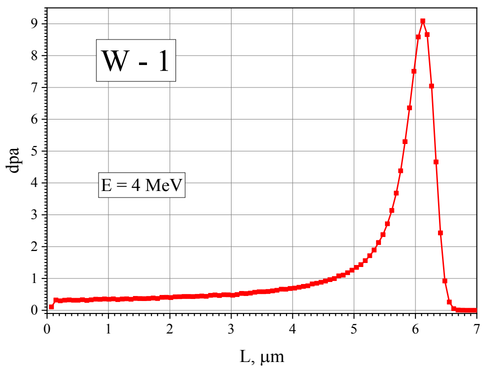
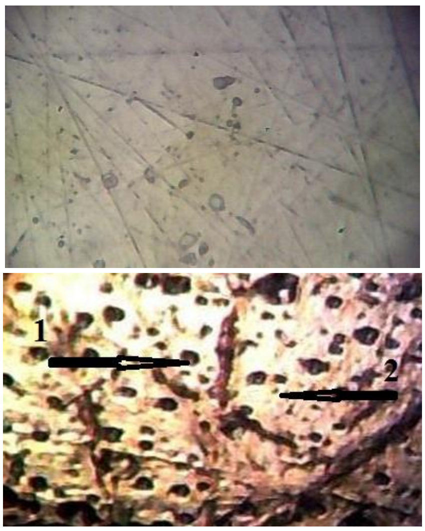


Fig. 7. Displacement formation profile in a sample of tungsten (W-1)

The surface of sample of tungsten (W-1) is presented on Fig. 8.



a

b

Fig. 8. Photographs of the sample surface (W-1); before irradiation (a); after irradiation (irradiation dose 10^{17} ion/cm^2) (E_{He}^+ = 4 MeV) (b)

On a photo (see Fig. 8,a) see the negligible quantity of spots on the surface of the sample. These are traces of technological processing. In the photo (see Fig. 8,b) a large number of pits (1) were found, which were formed as a result of effect of flecking. Also, we observe a small amount of swellings (2), which were formed as a result of the blistering effect. The amount of pits is five times amount of bubbles.

The conducted researches can serve as a supplement to systematic studies of the behavior of tungsten. Tungsten is under act of extreme electro physics and radiation factors. Including the study of how arise residual stresses and pulsed temperature fields [25, 26].

CONCLUSIONS

1. By the beams of Helium ions, with energies of 4 MeV, were irradiated the samples of tungsten with purity 99.5%.
2. Curves of changes in ionization processes along the length of the ion path are found. Also, the curves of changes in the processes of formation of vacancies and phonons along the length of the ion path are found. It was found that the processes of formation of phonons and vacancies have a maximum at a depth of 6.1 μm.
3. Integral and differential characteristics of sputtering of atoms from the surface of tungsten (W-1), depending on the energy, are obtained. The displacement and segregation profiles of atoms of tungsten are calculated.
4. The coefficients of sputtering of tungsten were obtained, along the depth of the sample (up to depths of 70 μm). Coefficient of dispersion is given depending on length of free run of ions of helium.
5. It is found that pits on-the-spot tungsten in five times more than bubbles. Supposition is outspoken, that the tendrils of tungsten (fuzz) appear due to spreading of bubbles of helium on the surface of pits, and accordingly squeezing out of tungsten.

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ОСОБЛИВОСТІ СТРУКТУРНИХ ПОШКОДЖЕНЬ ПОВЕРХНІ ВОЛЬФРАМУ В РЕЗУЛЬТАТІ ОПРОМІНЕННЯ ПУЧКАМИ ІОНІВ ГЕЛІЮ З ЕНЕРГІЄЮ 4 МеВ

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Виконано опромінення зразків вольфраму з чистотою 99,5% пучками іонів гелію (E_{He}^{+} = 4 МеВ). Розглянуто зміни окремих радіаційних характеристик вольфраму, які були ініційовані обробкою зразка вольфраму пучками іонів гелію. Чисельними методами отримані значення кількості вакансій і фононів. Кількість вакансій, фононів визначалась у залежності від глибини шару вольфраму (максимум на глибині 6,1 мкм). Знайдено коефіцієнти розпилення атомів з поверхні вольфраму для іонів гелію з довжиною вільного пробігу до 70 мкм. Максимальне руйнування поверхні знаходиться в області ямок, яких у п'ять разів більш, ніж бульбашок.