Effects of Plasma Interaction with Radiation-Damaged Tungsten

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Abstract. 14 MeV-neutrons from DT fusion reaction present a serious concern for materials of the first wall and divertor in a tokamak fusion reactor operating in steady state because of the damage produced in material structure. However, there is lack of information on radiation damage of plasma facing materials and their behavior under plasma impact. This paper is devoted to experimental investigation of tungsten at high level of radiation damage under steady state deuterium plasma. Tungsten is one of the important candidates as plasma facing material for application in ITER and, probably, beyond. The displacement damage of 1-80 dpa was produced in tungsten samples W (99,95% wt) by high-energy alpha particles $^4He^{2+}$ (3-4 MeV) from accelerator at Kurchatov Institute (cyclotron). The irradiated samples were then studied in deuterium plasma on the LENTA linear divertor simulator. Plasma exposures were made at 250 eV of ion energy and fluence 10^{21}-10^{22} ion/cm^2. Erosion dynamics of the damaged surface layer (~6 μm) as well as deuterium retention were studied. Surface modifications have been observed in the damaged material. Increased deuterium retention was detected on damaged tungsten by nuclear analysis methods; implanted helium accumulation was also measured.

1. Introduction

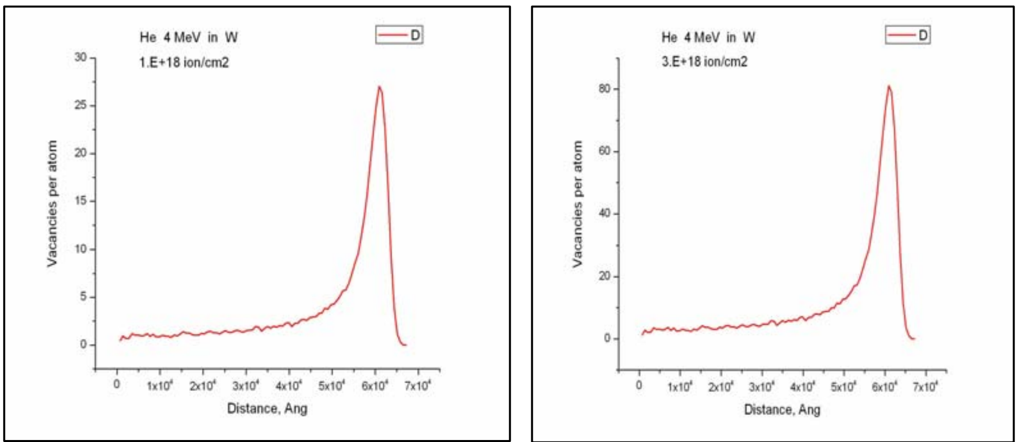
The consequences of radiation damage produced by fast fusion neutrons in plasma-facing materials and corresponding limitations of their service life in a fusion reactor should be evaluated prior to the construction of the reactor itself. No neutron sources of the needed intensity are actually available to achieve the damage level predicted for long-term operation of fusion tokamak reactor. On the other hand, charged particles accelerated to high energy present an efficient means for production of displacement damage to the levels being of interest for fusion research for a reasonable time period. Estimations of radiation damage for power fusion reactor fall in the range from a few tens to one-two hundred dpa. This paper describes experimental results obtained in the development of the research based on the modeling of neutron-induced damage by irradiation with high energy accelerated particles and study of plasma effect on the damaged materials in steady state plasma. The start of these researches was announced in [1], and the results on carbon materials at high level of radiation damage were given in our previous reports [2-6].

An experimental investigation of relation between the plasma effect on PFMs and the level of radiation defects in material is being conducted on facilities at Kurchatov Institute. Experiments are performed using the Kurchatov cyclotron providing high-energy particles to produce damage in material and the linear plasma machine LENTA simulating tokamak divertor plasma conditions. Plasma-facing materials actually considered as candidates for application in tokamak reactors have been included in the study: carbon based materials have been examined at the initial stage [2-5] and tungsten at the ongoing stage of the work [4-6]. Our results on erosion of carbon materials and its enhancements on irradiated materials are given elsewhere [4,5]. New results of the experiments with tungsten W (99,95% wt) of the composition similar to the ITER candidate grade are presented in this paper. The work is focused on the material having accumulated radiation defects at the level usually considered as a high level of radiation damage. This paper describes experiments with damaged tungsten in deuterium plasma. Accelerated high-energy ions were used to produce damage in tungsten providing high-level displacement ranging from ~1 dpa to 80 dpa. Plasma experiments have been aimed at evaluation of erosion under high plasma flux and deuterium trapping.

2. Experimental procedure

Experimental procedure included two stages: irradiations with high-energy ions were carried out at the first stage to produce damage in the material; then irradiated samples were exposed to steady-state plasma at the second stage. After each experimental operation the samples were studied by the methods described below.
Tungsten grade under the study contained 99.95% wt. of tungsten the composition corresponding to that being considered for the ITER application. Tungsten samples 10×12 mm were cut from tungsten sheet 1 mm thick, they were polished and cleaned.
Prior to the plasma exposure, tungsten samples were irradiated on the cyclotron by α-particles ($^4He^{+2}$ ions) at the energy of 3.5-4 MeV to produce damage in the material. The modeling of neutron effect relevant to fluence of 10^{22} neutron/cm^2 becomes possible because the damage produced by charged particles can reach an appropriate level at a reasonable operating time due to much higher damage rates than that from neutrons. Four high-energy ion fluences were reached in those irradiations: $\Phi = 5{\cdot}10^{17}$, 10^{18} and $3{\cdot}10^{18}$ He^{++}/cm^2, and, lately, the fourth irradiation was performed on the sample prepared for the plasma exposure to even higher dose of energetic alpha particles on tungsten surface $\Phi = 10^{19}$ He^{++}/cm^2.
The damages produced so far are concentrated in the surface layer, the depth of the damaged layer being about ion range. In our case, the thickness of the damaged layer was about 6 μm corresponding to accelerated alpha-particles stopping range in tungsten. Distribution of the defects was highly non-uniform. Fig. 1 shows the result of calculation by SRIM program [7] of primary defects distribution in tungsten irradiated by alpha particles at 4 MэB to doses $\Phi = 10^{18}$ He^{++}/cm^2 and $\Phi = 3{\cdot}10^{18}$ He^{++}/cm^2. In the latter case, the average concentration of the produced defects in the damaged layer exceeded 10 dpa.

The following primary defect values were characteristic of the three radiation doses given above $\Phi = 5{\cdot}10^{17}$, 10^{18} and $3{\cdot}10^{18}$ He^{++}/cm^2. The maximal values were 13.5 dpa, 27 dpa, 81 dpa, correspondingly, while average levels over damaged layers were 2 dpa, 4 dpa and 12 dpa. The damage minima were near the surface and these were 0.5 dpa, 0.9 dpa and 2.8 dpa.



a b

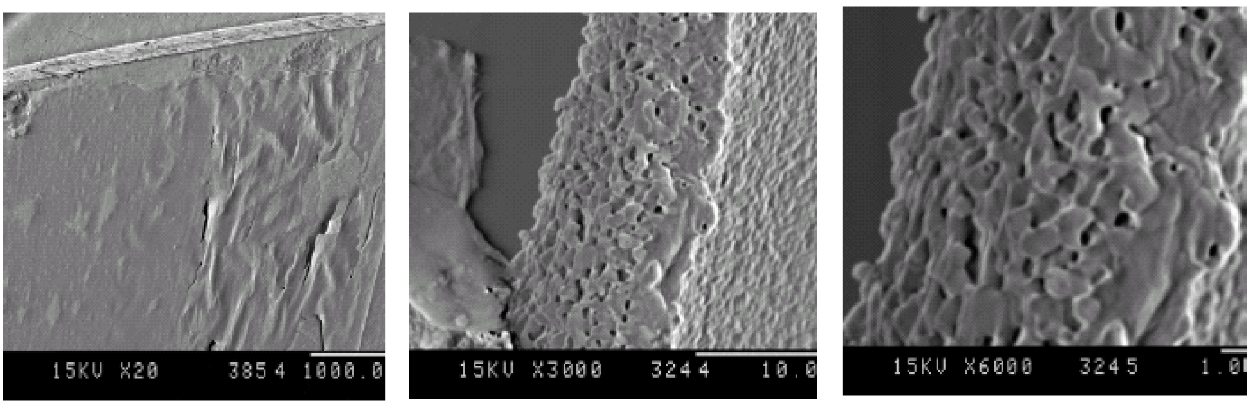
FIG. 1. Primary radiation defects in tungsten produced by 4 MeV alpha particles ($^4He^{+2}$) at fluence $\Phi = 10^{18}$ He^{++}/cm^2 (a) and $3{\cdot}10^{18}$ He^{++}/cm^2(b).

The irradiated tungsten samples were subjected to multiple exposures to steady-state deuterium plasma on the LENTA linear plasma divertor simulator. The plasma ion energy on the surface was 250 eV to simulate the edge tokamak plasma. Plasma parameters were N_e= $2{\cdot}10^{12}$ cm^{-3}, T_e= 6 eV, ion flux on the target j = 20 mA/cm^2. Successive exposures of the samples resulted in total ~10^{22} ion/cm^2 of deuterium ions on the W surface, giving about 10^{21} ion/cm^2 of plasma fluence at each exposure. The material temperature during plasma exposures as well as during irradiations with fast particles was kept below 100 C.

Evaluation of erosion yield of tungsten under deuterium plasma was made basing on the data of plasma parameters and weight loss measurements. Changes of the surface microstructure were analyzed with Scanning Electron Microscope. Swelling effect and surface profiles were detected by profilometer. Nuclear analysis methods were applied to study accumulation of gases - deuterium and helium; their content and distribution in depth were measured on accelerators at Moscow University (Van de Graaf generator and cyclotron). Elastic Nuclear Backscattering of α-particles (2.4 MeV) and protons (7.5 MeV) was used for measurements of helium content, and Elastic Recoil Detection Analysis by 1.9 MeV helium ions on Van de Graaf generator was applied to obtain deuterium distributions in depth.

3. Tungsten erosion and surface modification

Changes of the tungsten surface at different stages of the study were analyzed with SEM technique. While minor changes were found after irradiations on the cyclotron by fast alpha particles to smaller fluence $\Phi = 5{\cdot}10^{17}$ He^{++}/cm^2 [4], important changes were observed on the samples at larger fluences. Formation of large bubbles on the surface and partial splitting of the damaged layer from the bulk were found, ruptures of the layer also occurred. Fig. 2a shows a view of the surface of tungsten sample after irradiation to $\Phi = 3{\cdot}10^{18}$ He^{++}/cm^2 with the mentioned features exhibited on the surface. The structure of the splitted layer is well seen in Fig. 2b, the photo was taken in an open crack at inclined incident position of the SEM beam. Surface structure does not show important changes while formation of pores is observed in the damaged layer.



a b c

FIG. 2. Surface of the irradiated tungsten at $\Phi = 3{\cdot}10^{18}$ He^{++c}/cm^2 (a) and structure of the damaged layer as shown in crack (b) and (c). Pores are seen in the layer splitted from the bulk. Scale bars are given in microns.

Swelling effect was found to occur and it was evaluated with profilometer. The measurements gave the level of swelling at about 0.1-0.3 μm or at ≤ 5% of the total damaged layer depth.

The plasma part of the experimental procedure consisted in series of the irradiated samples successive exposures to deuterium plasma with analysis of the samples after each exposure run, and that made it possible to observe the erosion process dynamics. Multiple exposures of the samples were executed so that duration of plasma operation at each step was sufficient to get deuterium ion fluence about $\sim 10^{21}$ ion/cm^2 giving erosion depth of ~ 0.5-1 micron. Total deuterium fluence needed to erode 5-7 μm of tungsten was $\leq 2\cdot 10^{22}$ ion/cm^2.

The average erosion rate was found after each exposure by the measured weight loss, and it was estimated at about G ≅ 0.3 μm/hour. Erosion yields of the tungsten in the study under deuterium plasma ions at 250 eV in the studied condition was found by this erosion rate with the account for the measured ion current to the samples. The obtained value of the average erosion yield was $Y_{d\text{-}w} \cong (2\text{-}4)\cdot 10^{-3}$ at/ion. If we refer it to ion composition of the plasma with important content of different ion species (D^+, D^+_2, D^+_3) then we find good correspondence of the result with known data. No correlation was found in this study of the yield values with the damage level for different erosion depths of the damaged layer for the samples with different doses; also, no difference was found in erosion rate on the damaged samples as compared to undamaged tungsten.

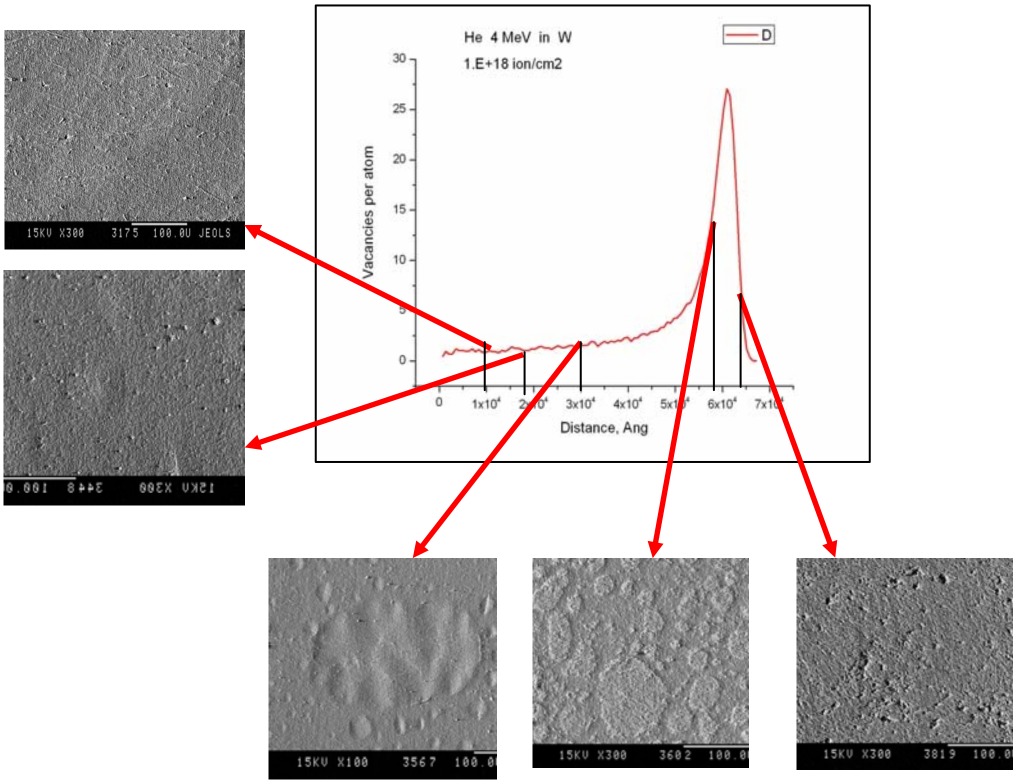


FIG. 3. Surface structure of the irradiated tungsten ($\Phi = 10^{18}$ He^{++}/cm^2) at different stages of erosion in deuterium plasma in correspondence with primary defects distribution. Scale bars given in microns.

At the same time, significant surface microstructure modification after plasma exposure has been observed on the damaged tungsten in comparison with the non-irradiated one. The SEM analysis of the surface and measurements of the profile at each step of exposure series revealed the changes of the surface structure. Dynamics of the erosion process is illustrated in Fig. 3. The figure gives SEM photos of the tungsten sample irradiated to $\Phi = 10^{18}$ He^{++}/cm^2 for five successive plasma exposures relating to different erosion depths obtained in the step-wise process. Correlation of the structure with the level of damage is seen in connection with the distribution of the primary defects taken from the calculated curve given in the figure. Erosion goes from initially smooth surface via appearance of bubbles of different sizes (from tens and hundred microns to mm range) at 2 and 3 step to form a particular spotted structure at the 4 step corresponding to the region of the maximal damage concentration (here about 27 dpa) and to come, finally, to the surface with rare residual spot roughness that is presumably formed at a depth where the whole damaged layer was eliminated by erosion.

These observations were further clarified by the measurements of the surface profiles. An example of the profiles taken with profilometer is presented in Fig. 4. These profiles were registered on the sample described above ($1\cdot10^{18}$ He^{++}/cm^2) on the fourth step of plasma exposure, i.e. approximately at the position of damage maximum when the plasma has eroded about 5-6 µm. A rough structure of spot groups is developed on the surface when erosion comes to the depth of the peak damage (ion range) (Fig. 4a, the part of the curve at 500-2000 µm; the edge part 0-500 µm was shadowed from irradiation and from the plasma). The plasma exposed part shows the developed profile with the structure elements of about $\Delta h \sim 2$ µm in height. Fig. 4b shows the same case around the border of irradiated area after 4th exposure to the plasma. Irradiated part of the surface is to the right; undamaged one is to the left. The development of the structure of $\Delta h \sim 2$ µm high is detected only on the irradiated side of the tungsten.

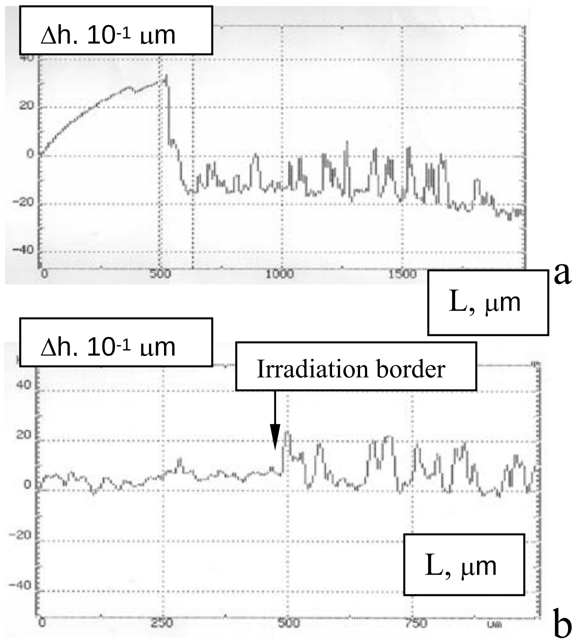


FIG. 4. Surface profile of irradiated tungsten (10^{18} α/cm^2) after plasma erosion to depth of the peak damage (~5 µm): a – profile at the sample edge (0-500 µm interval was masked from the plasma; b – the region around the border of irradiation; left - undamaged part, right - damaged part.

4. Deuterium and helium in damaged tungsten

Concentrations of hydrogen isotopes retained after deuterium plasma in the damaged tungsten have been measured by Elastic Recoil Detection Analysis. By this technique, a significant increase of the retained deuterium in the damaged tungsten structure (by an order of magnitude) has been detected. Elastic Nuclear Backscattering Method has been also applied to search for the implanted helium. Helium was detected as accumulated in tungsten to depth of ~6 micron and that corresponded to the calculated He distribution profiles [6].

Two irradiated samples have been analyzed: the sample irradiated to 10^{18} He^{++}/cm^2 and that of $3{\cdot}10^{18}$ He^{++}/cm^2 at different stages of plasma exposure steps. Comparison was made with the plasma exposed ($3.8{\cdot}10^{21}$ D/cm^2) undamaged sample (not subjected to irradiation), plasma condition being the same for all three samples. The results are presented in the Table 1.

TABLE 1: DEUTERIUM AND HELIUM IN TUNGSTEN.

Elastic nuclear backscattering, α-particles. E=2,4 MeV						ERDA, α-particles E=1,9 MeV	
Sample Nr	3,5-4 MeV α-fluence, cm^{-2}	dpa, max	D-plasma fluence, cm^{-2}	Erosion depth, μm;	He, ат.%; at/cm^2;	D, cm^{-2}	H, cm^{-2}
W3	$3{\cdot}10^{18}$	81	$2,7{\cdot}10^{21}$	0,8	-	$2{\cdot}10^{16}$; $2,4{\cdot}10^{16}$	$1,6{\cdot}10^{17}$
W2	$1{\cdot}10^{18}$	27	$1,3{\cdot}10^{22}$	4,9	10%; $4,1{\cdot}10^{17}$(0,7 μm wide)	$1.7{\cdot}10^{17}$	$7{\cdot}10^{16}$
W0	0	0	$2,8{\cdot}10^{21}$	1,17	-	$1.6{\cdot}10^{16}$	$1.5{\cdot}10^{17}$

The table shows deuterium, hydrogen and helium concentrations in the damaged (W-2 and W-3) and undamaged (W-0) tungsten samples. The data is given for the indicated alpha irradiation fluences, D-plasma fluences and plasma erosion depths.

The presented data shows that undamaged tungsten has the minimal areal density of the trapped deuterium ($1.6{\cdot}10^{16}$ D/cm^2). At the same time two other damaged samples have higher deuterium content. The largest integral deuterium retention $1.7{\cdot}10^{17}$ D/cm^2 was detected on the sample W-2 ($1{\cdot}10^{18}$ He^{++}/cm^2), and this value exceeded the retention for the undamaged case by an order. This measurement was made at the erosion depth of about 5 μm, i.e. close to or in the peak of displacement distribution. The W3 sample had also higher area D content ($(2\text{-}2.4){\cdot}10^{16}$ D/cm^2) than the non-irradiated W1 but lower than W2. The measurement on W3 was made close to the initial surface of the sample after erosion was stopped at the region where the displacement damage was at a few dpa level (see Fig. 1).

Depth distributions of the trapped deuterium in four damaged tungsten samples are shown in Fig. 5. The fourth sample labeled as W01A was taken for comparison at the state after alpha irradiation to $1.1{\cdot}10^{18}$ He^{++}/cm^2 without plasma processing. The three other samples were subjected to irradiations mentioned in the first section and to the plasma processing. It follows from this graph that deuterium penetration from the plasma into tungsten is about 100 nm in all cases for different damages of the material. On the other hand, the quantity of the retained deuterium depends on the damage level and on the erosion progress stage in plasma exposure series (erosion depth stage is given in Fig. 5 in brackets in microns). Maximal deuterium uptake was detected on the W2 sample at the stage when erosion came close to the zone of maximal damage (27 dpa) and it was about 8% at. More deep penetration of deuterium was registered around the maximum of damage [6].

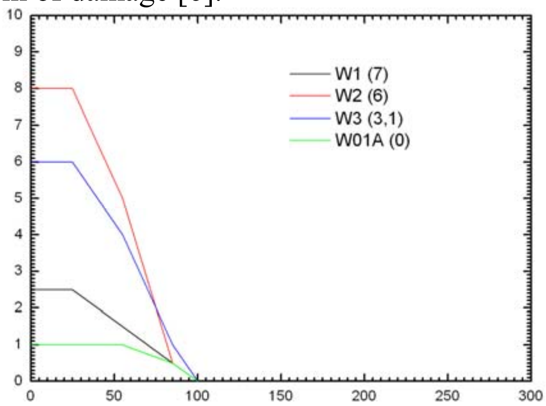


FIG.5. Deuterium concentration in tungsten in four samples: W1 - $\Phi = 5{\cdot}10^{17}He^{++}/cm^2$, α+plasma; W2 - $\Phi = 1{\cdot}10^{18}He^{++}/cm^2$, α+plasma; W3 - $\Phi = 3{\cdot}10^{18}He^{++}/cm^2$, α+plasma; W01A - $\Phi = 1.1{\cdot}10^{18}He^{++}/cm^2$, no plasma.

Helium content was measured by reaction $^4He(p,p)^4He$. Helium was found in W2 and W3 samples at depth of about 5 μm. Its total concentration was registered at $(Nt)_{He} = 4{\cdot}10^{17}$ and $(Nt)_{He} = 9{,}6{\cdot}10^{17}$ at/cm^2 correspondingly.

5. Discussion

Experimental methods and procedure adopted in this study are based on simulation of fusion neutrons producing damage of plasma facing materials by fast ions. Namely, α-particles ($^4He^{+2}$ ions) at the energy of 3.5-4 MeV were used in this work to produce damage in tungsten. Note that these very particles are generated as a product of the reaction D + T → He^4 (3,5 MeV) + n (14,1 MeV), and though the charged particles are magnetized and confined in the plasma vessel their penetration to the boundary layer and escape to the wall may not be completely ignored. It is certain, that the flux of the energetic helium ions to the wall would not be so high that their result as of a source of damage for the plasma facing materials could be comparable with neutrons. Therefore, the high level of radiation damage in tungsten of tens dpa reached in this work by alpha particles should be considered as simulation of neutron displacement damage being far higher over the effect of the helium component.

The plasma effect on the damaged tungsten was studied in the progress of the surface erosion, the dynamics of this process was revealed by the stepwise plasma exposures. The continuous erosion of the surface is important as a factor excluding the influence of surface initial condition effects on the process, if static, and this being more relevant to reactor situation. The structure of the surface was observed to change from step to step till the last stage where the damaged layer of 6 μm was totally eroded by deuterium. Formation of the blister-like structures was found in the intermediate stages (tens – hundred microns in size) and even large bubbles (mm size) and partial splitting of the damaged layer from the bulk. Large amount of helium (up to 10% in maximum) may explain these effects as well as the formation of the spots of column structures with the elements of about $\Delta h \sim 2$ μm in height at the depths about the alpha's end-of range. Helium may be accumulated in the pores observed in the damaged layer (micron size) and cavities, and they may develop under plasma bombardment

and have influence on the changes in the structure with helium outcome to the material surface. The presence of helium not only in the vicinity of the initial surface after irradiation but also in the bulk in very large quantity may be particularly important for deuterium retention in the material. Here we observed considerable increase of deuterium uptake in the damaged layer at depth of maximal damage (5-6 μm).
All these experiments were performed at temperatures below 100 C so that annealing effect on the produced defects would be avoided. Tungsten components are presumed to work in ITER in the range ~300-1300 C, the further studies should cover this range to reveal the influence of temperature on erosion and hydrogen retention.

6. Summary

Effect of neutron damage impact on characteristics of material under deuterium plasma has been studied. Tungsten W 99.95% at. similar to ITER divertor candidate was subjected to irradiation of energetic alpha particles (3.5 - 4.0 MeV), the material samples has been obtained at the level of displacement damage relevant to future fusion reactor ITER and beyond (1-80 dpa).
Plasma effect on the damaged tungsten has been examined. Erosion under steady-state deuterium plasma was studied at 250 eV of ion energy at the wall, evaluation of the erosion yield was found to be $Y_{d\text{-}w} \sim (2\text{-}4)\cdot 10^{-3}$ at/ion. Development of the surface structure has been observed on the irradiated tungsten with formation of blister-like features, cavities, and column structures. Correlation of erosion yield with the presence and the level of damage were not registered.
Large amounts of the implanted helium were detected by nuclear reactions techniques in the damaged tungsten surface layer with maximum of 10% at. Increase of deuterium retention was found on the damaged tungsten with the largest integral deuterium uptake measured $1.7\cdot 10^{17}$ D/cm^2.
The obtained results show the importance and necessity of the further investigations of hydrogen isotope retention in damaged tungsten to evaluate correctly effects of tritium accumulation in fusion reactor vessel.

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