Nuclear Instruments and Methods in Physics Research B 340 (2014) 27–33

Contents lists available at ScienceDirect

Nuclear Instruments and Methods in Physics Research B

journal homepage: www.elsevier.com/locate/nimb

Electron microscopy observations of radiation damage in irradiated and annealed tungsten

J. Grzonka [a,*], Ł. Ciupiński [a], J. Smalc-Koziorowska [b], O.V. Ogorodnikova [c,d], M. Mayer [c], K.J. Kurzydłowski [a]

[a] Warsaw University of Technology, Wołoska 141 St., PL-02507 Warsaw, Poland
[b] Institute of High Pressure Physics PAS, Sokołowska 29/37, PL-01142 Warsaw, Poland
[c] Max Planck Institute of Plasma Physics, Boltzmannstr. 2, D-85748 Garching, Germany
[d] National Research Nuclear University "MEPHI", Kashirskoe sh.31, Moscow, Russia

ARTICLE INFO

Article history:
Received 23 March 2014
Received in revised form 14 July 2014
Accepted 22 July 2014
Available online 2 September 2014

Keywords:
Tungsten
Radiation damage
Radiation-induced defect annealing
STEM observations

ABSTRACT

In the present work tungsten samples were irradiated with W^{6+} ions with a kinetic energy of 20 MeV in order to simulate radiation damage by fast neutrons. Two samples with cumulative damage of 2.3 and 6.36 displacements per atom were produced. The scanning transmission electron microscopy investigations were carried out in order to determine structure changes resulting from the irradiation. The evolution of the damage with post implantation annealing in the temperature range 673–1100 K was also assessed. Damage profiles were studied at *cross-sections*.

Scanning transmission electron microscopy studies of the lamellae after annealing revealed aggregation of defects and rearrangement as well as partial healing of dislocations at higher temperatures. The results confirm the higher density of radiation-induced dislocations in the near surface area of the sample ($1.8 * 10^{14}\ m^{-2}$) in comparison with a deeper damage area ($1.5 * 10^{14}\ m^{-2}$). Significant decrease of dislocation density was observed after annealing with a concurrent growth of dislocation loops. Transmission electron microscopy analyses show that the dislocation loops are perfect dislocations with the Burgers vectors of $\boldsymbol{b} = ½[1 1 \underline{1}]$.

1. Introduction

Tungsten (W) is one of the main candidates for plasma-facing materials (PFMs) in fusion reactors. It is a reference material in the high-flux region of the divertor in ITER and a candidate plasma-facing material for DEMO. This is due to its favourable physical properties under high heat and particle fluxes such as high melting temperature (3695 K), good thermal conductivity (175 W/mK at room temperature) and low erosion rate [1]. The low sputtering rate is beneficial in application in fusion reactors due to minimisation of plasma impurities generation [2].

The PFMs and structural components of fusion reactors are subjected to fast neutron (n) fluxes produced by the nuclear reactions of the fusion fuel. Neutron irradiation results in crystal structure modification due to the introduction and accumulation of radiation induced defects [3]. Those defects can act as trapping sites for deuterium, therefore creation of crystal lattice defects can lead to a significant increase of deuterium retention in W [4–7]. This in turn can seriously jeopardise a safe operation of a fusion reactor. Thus the prediction of hydrogen isotopes retention in n-irradiated W is an important issue for its application as PFM in fusion reactors.

The irradiation by fast heavy ions is often used to simulate the influence of fast neutron irradiation on the structure and pertinent properties of W [4,8–13], as the intensive sources of fusion neutrons allowing building up relevant damage levels in irradiated materials are still not available. Also irradiation in fast nuclear reactors is disadvantageous as a long time (about two years) is required for achieving relevant damage levels and due to different neutron energy spectra as compared to fusion neutrons. Moreover, samples irradiated by fast neutrons become activated, which renders their analysis much more complicated as compared to the samples irradiated with fast heavy ions.

The hydrogen isotope retention in solids depends on trap density and the rate at which the defect associated traps are filled by deuterium [5]. It was reported in [14] that there are at least two types of ion-induced defects that are responsible for trapping of

* Corresponding author. Tel.: +48 222348455.
E-mail addresses: j.grzonka@inmat.pw.edu.pl (J. Grzonka), lciupinski@gmail.com (Ł. Ciupiński), julita@unipress.waw.pl (J. Smalc-Koziorowska), igra32@rambler.ru (O.V. Ogorodnikova), Matej.Mayer@ipp.mpg.de (M. Mayer), kjk@inmat.pw.edu.pl (K.J. Kurzydłowski).

http://dx.doi.org/10.1016/j.nimb.2014.07.043
0168-583X/

deuterium: (1) vacancy-type defects (vacancy and vacancy clusters) which can trap D atoms and D_2 molecules and (2) dislocation-type defects, which capture D atoms. The density of radiation-induced defects that can serve as trapping sites for deuterium was found to decrease with increasing temperature. The D retention in W decreased upon annealing in the temperature range from 300 to 673 K and showed minor changes after annealing in the temperature range from 700 to 1100 K. Furthermore, a peak of the D retention was found at around 1000 K [5].

Although, transmission electron microscopy (TEM) investigations of ion induced defects in tungsten after annealing have been already reported elsewhere [15,16], the reported results were obtained on *plane-view* samples with the TEM images taken parallel to the implanted surface. Thus only a "shallow" near-surface region of the implanted target was investigated in that research. In the present work the microstructural investigations are extended into TEM samples perpendicular to the implantation surface.

2. Material and methods

The polycrystalline European ITER W samples produced by Plansee AG were cut into $10 \times 10\ mm^2$ plates with a thickness of about 0.5 mm. These plates were mechanically polished to mirror like finish and cleaned in an ultrasonic bath. Prior to implantation with tungsten ions they were recrystallized at 2200 K for 10 min.

The radiation damage was produced by implantation with 20 MeV W^{6+} ions (angle of incidence equal to 90°) at room temperature. Two doses equal to $3.7 * 10^{18}$ and $1 * 10^{19}\ W^{6+}/m^2$ were applied, which corresponds to 2.3 and 6.36 displacements per atom (dpa). The damage level was calculated using the program SRIM 2008.03 full cascade option with a threshold displacement energy of 90 eV. The implantation was carried out at IPP Garching in a chamber connected to the 3 MV tandem accelerator [4].

The Scanning Transmission Electron Microscope (STEM) observations were performed using STEM Hitachi HD2700 with an accelerating voltage of 200 kV. The examination of dislocation loops was supported by dark field (DF) observations with transmission electron microscope TEM FEI Tecnai G2 TF S-TWIN at accelerating voltage of 200 kV. The dislocation densities were calculated using line-intercept method [17]. Five randomly placed lines of different angular orientation were drawn over STEM images facilitated using DigitalMicrograph program. The number of points N, intersections of each defect image with the random lines, was counted. The dislocation density ρ was the number of points N divided by the total line length of the random lines L_r multiplied by the lamellae thickness t. The lamellae thickness was estimated to be about 80 nm.

The observations were performed at lamellae cut as cross-sections perpendicular to the implanted W target surfaces. These lamellae were produced using a Focused Ion Beam system – FIB/SEM Hitachi NB5000. During FIB preparation the implanted surface of the tungsten plates has been covered with a protective tungsten layer. The final thinning was carried out with low energy argon ions polishing using a Linda Gentle Mill device aiming at the reduction of damage introduced during FIB processing. The accelerating voltage during that process was in the range of 0.5–1 kV.

The TEM lamellae taken from implanted samples were annealed using the experimental chamber of Thermogravimetric Analyzer Netzsch STE 449 F3. During annealing the vacuum was kept at 10^{-4} mbar. The lamellae were annealed for 1 h at 673, 773, 950, 1000 and 1100 K. The heating rate was 10 K/min, whereas the cooling rate was 20 K/min. The annealing temperatures were selected based upon the results of D desorption experiments carried out by Ogorodnikova et al. [5].

3. Results

The TEM images of the samples after W^{6+} ion implantation with 20 MeV are presented in Fig. 1. The bright top tungsten layer is used as a protective coating during the FIB preparation process. The area below represents the external tungsten target surface. The tungsten targets, recrystallized before implantation, used in the experiment, reveal a relatively coarse grains structure with three (Fig. 1a) and two (Fig. 1b) tungsten grains differing in orientation visible in the FIB processed lamellae. The damage profile can be observed. The STEM observations reveal that the depth of the damaged zone depends on the dose and is equal to 2.5 and 2.8 μm for 2.3 and 6.36 dpa, respectively (Fig. 1). As already reported in [18], the damaged zone can be divided into 3 subregions: (1) near surface with high density of tangled dislocations, (2) intermediate with lower density of "long" dislocations and (3) deep with high density of "short" dislocations. A uniform image contrast is observed for the area below the damaged zone. One can observe that the depth of damage zone in the two grains of lamellae extracted from the 2.3 dpa damaged target is different and likely depends on their crystallographic orientation. The damaged zone depth inside the left [1 1 0] grain is higher than in the right [0 1 2] one (Fig. 1a).

The modification of radiation-induced defects in tungsten induced by annealing in the temperature range from 673 to 1000 K is presented in Figs. 2 and 3. The images in Fig. 2 show the near surface region and partially the intermediate area of the damaged samples, approximately 600 nm from the target surface. It can be noted that the annealing of W lamellae at lower temperatures (673 K and 773 K) leads to the rearrangement and aggregation of defects. The growth of dislocation loops can be followed at the magnified image inserts. At 950 K recrystallization of the tungsten protective layer applied during FIB preparation occurred, and the contrasts from crystallites is now observable in the top most part of the lamellae. Further rearrangement and annihilation of dislocations is observed leading to the appreciably lower defects density at the TEM image. A "decoration" of radiation-induced dislocations by dislocation loops also takes place (indicated by the arrow in Fig. 2). Annealing also considerably reduces the dislocation loops density, in the region far from implantation surface. Though a much higher amount of dislocation loops remains deep in the material in comparison to the near-surface and intermediate zones (Fig. 3).

The contrast below the damaged zone revealed in Fig. 4 is due to the damage induced by the gallium ions during FIB sample preparation. This unwanted feature is present at the entire cross section of the investigated samples. This means that the damage zone contains dislocation loops formed by agglomeration of point defects both from the W-ions damaging and from FIB processing. During annealing the enlargement of dislocation loops in the whole area of the sample takes place. The increase in size, however seems to be higher for the W damaged area (Fig. 4). This enhanced kinetics of dislocation loops growth is most probably due to a higher driving force caused by the higher defect density at the damage area.

An unwanted contamination of the surface of TEM lamellae occurred during post-irradiation annealing and introduced artefacts to the TEM image (marked with arrow in Fig. 3) for the temperatures of 950 and 1000 K. Their proliferation was observed in the sample annealed at 1100 K, leading to the entire surface of lamellae degradation, which is observable in the images presented in Fig. 5b and d. Nonetheless, the damaged zone is still observable. The images of the near surface and deeper damage regions before and after annealing in 1100 K (Fig. 5) show further process of coalescence of defects and material recovery leading to a considerable reduction of defects number.