Journal of Nuclear Materials xxx (2017) 1–7

Contents lists available at ScienceDirect

Journal of Nuclear Materials

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

Annealing effect on the microstructure of tungsten irradiated in SINQ target

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HIGHLIGHTS

- Tungsten was irradiated in a SINQ target at two doses of 1.4 and 3.5 dpa with 37 and 140 appm He at 80 and 110 °C.
- Irradiation induced defect structures were investigated.
- Due to annealing the number of dislocations and defect clusters reduced to less than half after annealing at 900 °C.
- Bubbles observed in the 1.4 dpa sample after annealing at 800 °C.

ARTICLE INFO

Article history:
Received 9 February 2017
Received in revised form
26 September 2017
Accepted 11 December 2017
Available online xxx

Keywords:
Tungsten
Microstructure
Irradiation effect
Annealing effect

ABSTRACT

In this work, the microstructure of pure tungsten irradiated in a target of the Swiss spallation neutron source is studied. The tested tungsten specimens were irradiated to two doses of 1.4 and 3.5 ddpa with 37 and 140 appm He at 80 and 110 °C, respectively. The specimen of 1.4 dpa was consecutively annealed at temperatures of 500 °C, 600 °C, 800 °C and 900 °C, for 1 h, and the post-irradiation annealing effect on the microstructure was investigated. Microstructural features such as dislocations, defect clusters, dislocation loops and bubbles were observed by means of transmission electron microscopy (TEM). TEM images were obtained in different areas of the samples, to obtain quantitative information of the dislocations and defect clusters. There was a significant change in the microstructure of the tungsten after irradiation and post-irradiation annealing. The average dislocation density and defect cluster density were evaluated.

1. Introduction

The European Spallation Source (ESS) under construction in Lund, Sweden [1] will be the world's brightest neutron source facility. A 5 MW proton beam with a duty factor of 4% impinges on a tungsten target. The neutrons are produced through a spallation process in tungsten [2], which has a high neutron production density, thanks to its high atomic number and high mass density. In the ESS target, approximately 3 MW will be deposited in the tungsten volume, which is removed by actively circulating helium gas flow to avoid the corrosion issues related to water cooling. Helium cooled tungsten should pose lower hazard risks to the environment compared to liquid metal target materials which are used at other MW class spallation sources. However, though tungsten is a metal with a high strength and the highest melting point, its low ductility and a high ductile-to-brittle transition temperature [3,4] are of a concern, particularly in irradiation conditions.

Neutron irradiation in tungsten has been examined so far by a few other researchers. Initially, Rau et al. [5,6] conducted experiments in the 1960's and found that neutron irradiation produces defect clusters and voids in the microstructure. Matolich et al. [7] determined the swelling properties of tungsten and tungsten alloys. In the recent years, Hasegawa et al. [8,9] observed a hardening effect induced by voids and dislocation loops at damage levels greater than 0.15 dpa. They also determined the defect types in tungsten depending on irradiation temperature and displacement damage [10]. Hu et al. [11] examined single crystalline tungsten after neutron irradiation at low temperatures and low doses, and the effect of annealing on the irradiated material. Klimenkov et al. [12] irradiated tungsten at 900 °C to an average damage level of

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https://doi.org/10.1016/j.jnucmat.2017.12.020
0022-3115/

Please cite this article in press as: B. Horvath, et al., Annealing effect on the microstructure of tungsten irradiated in SINQ target, Journal of Nuclear Materials (2017), https://doi.org/10.1016/j.jnucmat.2017.12.020

1.6 dpa. In the irradiated samples they observed cavity formation and transmutation product, rhenium. Fukuda et al. [13] reported dislocation loops and voids in pure tungsten and tungsten alloys in irradiation temperature and dose ranges of 531–756 °C and 0.42–0.47 dpa. Additionally, they analyzed the effects of Re content and fabrication process on microstructural changes and hardening in neutron irradiated tungsten, tungsten-rhenium alloy [14,15], and other tungsten alloys [16]. Gilbert et al. [17] simulated the primary transmutation products for tungsten under nuclear power-plant conditions, stating that rhenium can reach a concentration of 3.8 at% after five years irradiation.

Tungsten is the most critical non-structural material in the ESS target, and its mechanical integrity during operation is essential for maintaining helium circulation and retaining transmutation elements, for a reliable operation of the target during its design lifetime of 5 years. Operating at 5 MW, the tungsten in the ESS target is exposed to approximately a maximum dose of about 2 dpa per year, due to the impinging proton beam and spallation neutrons [18]. To understand the radiation induced damage in tungsten, reliable data is needed on the mechanical properties of proton irradiated tungsten. Dedicated experiments were established at the Paul Scherrer Institute to examine the behavior of tungsten irradiated by high energy protons and spallation neutrons in the SINQ targets. Specimens in the dose range of 1.3 and 25 dpa were extracted from a large piece of pure tungsten irradiated in the fifth SINQ Target Irradiation Program (STIP-V) during 2007 and 2008. Mechanical testing and microstructural examination are being conducted, starting with specimens of low doses due to the high activity of the specimens. Bend testing on specimens irradiated to doses up to 3.5 dpa at temperatures below 140 °C demonstrated strong irradiation-induced embrittlement effect. The specimens are brittle at 1.3 dpa already, even at a test temperature of 450 °C [19], which is close to the maximum temperature in the ESS target. In this study, the microstructural changes in the irradiated tungsten specimens of low doses 1.4 and 3.5 dpa were investigated to understand the embrittlement effect. Furthermore, consecutively post-irradiation annealing was performed on the 1.4 dpa specimen to study the evolution of the defect microstructure at high temperatures, which is useful to improve the understanding of radiation effect of tungsten.

2. Experimental

The material investigated is 99.9 wt% tungsten in rolled condition with average grain sizes approximately 28/17 μm (length/width). The samples were extracted from a large piece of pure tungsten irradiated in STIP-V during 2007 and 2008 covered a dose range of 1.3–25 dpa [19]. In the present work two samples with relatively lower dose were investigated. Sample 1 was irradiated to 1.4 dpa, 37 appm He, at 80 °C. Sample 2 was irradiated to 3.5 dpa, 140 appm He, at 110 °C. In the initial bulk stage, the dose rate of the approx. 4 × 2 × 1 mm large sample was about 3 mSv/h at 10 cm. TEM sample preparation was performed with the use of the focused ion beam (FIB) technique in order to examine the highly activated samples safely. With FIB, the sample size was reduced to about 2 × 20 × 20 μm. Therefore, the activity of a TEM sample was reduced to negligible level. Out of this small piece, an 8 × 8 μm^2 and 200 nm thick lamella was finally obtained. Then, by flash electropolishing (Cu TEM grid, 0.5% NaOH, in 2 °C for unirradiated, 9 °C for irradiated W samples) this lamella was further thinned down to a thickness of about 60 nm. The flash electropolishing is necessary to remove the FIB-induced damage on the surface of the thin foil. Unirradiated tungsten samples were produced with twin-jet electropolishing by using Struers TenuPol-V. Electron microscopy observation was performed with a JEOL 2010 type transmission electron microscope operated at 200 keV and equipped with energy-dispersive X-ray spectroscopy (EDS). Two-beam bright field (BF) and weak beam dark field (WBDF) imaging conditions at (g, 5 g), (g, 6 g) and g = 110 were used. In order to obtain quantitative information of dislocations, they were counted on several low magnification pictures taken from different areas of the sample. Annealing of the irradiated bulk tungsten sample was conducted at temperatures: 500, 600, 800, and 900 °C. This was performed consecutively on the 1.4 dpa specimen. In each temperature step, the bulk specimen was annealed for 1 h in vacuum (1×10^{-5} bar) in an envelope of pure tantalum foil to reduce possible oxidation effect. In each condition, 3-4 lamella samples were prepared and 1 or 2 samples were observed depending on the quality of the results obtained.

3. Results and discussion

3.1. Microstructure of unirradiated tungsten

The microstructure of the unirradiated tungsten specimens was investigated to use as a reference to compare to. By using both flash polished and jet electropolished samples, a total of 33 μm^2 area was observed to measure dislocation density. Fig. 1 shows a representative BF TEM micrograph of an unirradiated tungsten sample, thinned by twin-jet electropolishing. Dislocations up to tens of microns long were observed throughout the samples. The dislocation density was determined by counting the number of dislocation lines crossing a unit of area in the samples. The dislocations were counted in the areas where the average thickness was about 50 nm. The dislocation density was measured to be 1.6×10^{13} m^{-2} (Table 1).

3.2. Microstructure of tungsten irradiated at 1.4 and 3.5 dpa

Eight TEM samples have been fabricated from the 1.4 dpa bulk material on different grains up to 800 μm distance from each other. Six of them were observed. While the dislocation density of the *un*irradiated samples is quite different in different areas or samples, the defect cluster structure of the 1.4 dpa samples remained very similar in all the six TEM samples. Defect clusters were counted from seven areas of two best samples to evaluate the size distribution and volume density of defect clusters. The difference in the size distribution is negligible and about 25% in the defect cluster density of the two samples. For this reason, just one TEM sample was made from the 3.5 dpa specimen. The microstructure of both 1.4 and 3.5 dpa specimens showed large differences compared to the unirradiated tungsten. The BF images of these two specimens can be seen in Fig. 2. The images of these two specimens show high similarities. The long dislocations observed in the unirradiated samples were replaced by short dislocation debris of the length of a few tens of nanometers. When evaluating the defect cluster and dislocation densities developed under the different irradiation conditions, similar results are found. The dislocation densities are 2.3×10^{13} and 1.8×10^{13} m^{-2} for the 1.4 and 3.5 dpa samples, respectively. These density values are similar to that of the unirradiated tungsten. Additionally, defect clusters were also found in great amounts homogeneously distributed throughout the samples. Quantitative analysis of the size and density of defect clusters was done from WBDF images. The density of defect clusters was evaluated from the micrographs with the following three imaging conditions: WBDF (g, 6 g), (g, 5 g) and two-beam BF, while g = 110, which detect the majority of the defects and loops as observed in Fe based alloys [20]. Defect clusters were counted only in the 15–30 nm thick regions of the samples. The thickness was measured by the thickness fringes in the WBDF condition. The

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