Journal of Nuclear Materials 509 (2018) 152–157

Contents lists available at ScienceDirect

Journal of Nuclear Materials

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

Thermal diffusivity of tungsten irradiated with protons up to 5.8 dpa

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ARTICLE INFO

Article history:
Received 24 April 2018
Received in revised form
26 June 2018
Accepted 26 June 2018
Available online 28 June 2018

Keywords:
Tungsten
Irradiation effects
Spallation target
Thermal diffusivity
Thermal conductivity

ABSTRACT

Thermal properties of pure tungsten, irradiated in the Swiss neutron spallation source at the Paul Scherrer Institut, have been studied in the temperature range 25–500 °C. Disk-shaped specimens were prepared from a tungsten sheet which was irradiated with high energy protons and spallation neutrons. The specimens tested in this work received total damages of maximum 3.9 and 5.8 dpa at average irradiation temperatures of 115 and 140 °C, respectively. The thermal diffusivity of the irradiated tungsten was measured using the conventional flash method. For both specimens, the results show a significant decrease in thermal diffusivity after irradiation. Relative to unirradiated tungsten, the irradiated samples show thermal diffusivity values which are 28–51% lower, depending on temperature. Annealing of the irradiated specimen of 3.9 dpa at 1000 °C for 1 h resulted in a slight recovery of thermal diffusivity. In addition, thermal conductivity values were calculated from the thermal diffusivity data.

1. Introduction

Pure tungsten has been selected as the target material at the European Spallation Source (ESS) facility, which is currently under construction in Lund, Sweden [1]. The decision is based on the high neutron yield of tungsten, which stems from its high atomic mass and high density. Despite having been used as target material at other spallation facilities for many years, the data on the properties of high energy proton and spallation neutron irradiated tungsten are scarce. At ESS, the target will be subjected to a high power and high energy proton beam. The pulsed proton beam with a duty cycle of 4% and a repetition rate of 14 Hz delivers 357 kJ per pulse to the tungsten. The target consists of about seven thousand bricks of pure tungsten with dimensions $80 \times 30 \times 10\,mm^3$, separated by 2 mm wide channels for the gaseous helium coolant. The bricks are placed on a 2.5 m diameter, rotating wheel. The rotation of the target wheel distributes the beam energy and the radiation damage over a larger volume of the spallation material.

During the 5-year lifetime of the target, the maximum displacement damage in tungsten would be about 10 dpa. With this displacement damage dose rate, the ductile-to-brittle transition temperature (DBTT) of tungsten will increase to above the target operating temperature after only a few weeks of full 5 MW operation. The radiation induced brittle behavior of pure tungsten and potential loss of structural integrity during target operation is a point of concern. Three-point bending tests performed on tungsten specimens irradiated in a target of the Swiss Spallation Neutron Source (SINQ) to doses in the range 1.3–3.5 dpa, have shown virtually zero ductility at temperatures up to 500 °C [2]. The radiation induced embrittlement of tungsten, in combination with the high power pulsed beam is considered as one of the main contributions to potential target failure. The proton beam, pulsed at 14 Hz with a duty factor of 4%, is expected to give rise to cyclic thermo-mechanical stresses in the tungsten, possibly leading to fatigue failure at stresses far below the yield stress [3].

The calculated maximum temperature and equivalent stress in the ESS target during operation are 445 °C and 110 MPa, respectively [4]. These calculations are based on thermo-mechanical data of unirradiated pure tungsten. The temperature and secondary thermal stress in tungsten is largely determined by its thermal conductivity. However, the radiation induced degradation of thermal properties of tungsten in spallation target environment is not known. Nevertheless, there is an indication of degradation of thermal conductivity of tungsten under neutron radiation in reactor environments. The neutron radiation induced decrease of thermal conductivity is caused by rhenium transmutation due to

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https://doi.org/10.1016/j.jnucmat.2018.06.041
0022-3115/

low energy neutron capture and displacement damage. While the spallation material is bombarded by high energy protons and fast neutrons, it is also exposed to thermal neutrons moderated by hydrogen-rich moderators and water coolants. Therefore, it is expected that the thermal conductivity of tungsten in the ESS target will decrease due to the formation of irradiation induced transmutation products such as rhenium, and due to displacement damage, although the extent of degradation is still not fully understood.

There are few existing studies on neutron irradiation effects on thermal properties of tungsten-rhenium alloys. Tanabe et al. [5] studied the effect of different rhenium content in tungsten-rhenium alloys on the thermal diffusivity in the temperature range 300–1000 K. It was observed that thermal diffusivity of pure tungsten decreases with increasing temperature, whereas the alloys, having 5–25 wt% rhenium, do not show a sharp change in thermal diffusivity as a function of temperature. Alloys with relatively higher rhenium content show a marginal increase in thermal diffusivity, beyond a certain temperature. The calculated thermal conductivity from the experimentally determined thermal diffusivity was compared to the one derived from electrical resistivity data using Wiedemann-Franz law. This law assumes that thermal conductivity of a metal is determined by the kinetics of electron transport, excluding phonon transport effects. The analysis of the difference between the two values indicated that the degradation of thermal conductivity with higher contents of rhenium is mainly due to the electron contribution.

In a study on electrical resistivity of neutron irradiated tungsten and tungsten-rhenium alloys, Hasegawa et al. [6] irradiated specimens in a fast reactor (JOYO) and in a mixed spectrum reactor (HFIR) to obtain different amounts of rhenium as the transmutation product. In both reactors the displacement damage was 1 dpa, and the irradiation temperatures were similar: 500 °C at HFIR and 600 °C at JOYO. Because of the high flux of thermal neutrons at HFIR, the rhenium production rate is higher due to neutron capture followed by beta decay. At HFIR, pure tungsten is expected to change to W-5Re-0.5Os after 1 dpa, and at JOYO the rhenium content is expected to be less than 0.03%. The resulting difference in the increase of electrical resistivity between the two equivalent specimens, after irradiation, is almost 50%, which should be mainly due to different radiation induced rhenium alloying. The observed loss of electrical conductivity can be related to the loss of thermal conductivity, in part, by Wiedemann-Franz law.

The effect of neutron irradiation on thermal diffusivity of pure tungsten and some tungsten-rhenium alloys has been studied by Fujitsuka et al. [7]. The specimens were irradiated in JMTR at 60 °C to thermal and fast neutron fluences of 1.03×10^{20} and 3.37×10^{19} n/cm^2 ($E > 1$ MeV), respectively. The thermal diffusivities were measured at temperatures up to 700 °C. Pure tungsten showed a decrease in thermal diffusivity with increasing temperature, both before and after irradiation. At room temperature, thermal diffusivity of the irradiated pure tungsten specimen is approximately 15% lower. The addition of rhenium (5–25 wt%) significantly lowers the diffusivity of all the alloys compared to that of pure tungsten, also after irradiation. With an addition of only 5 wt% rhenium, thermal diffusivity at room temperature drops by nearly 50% compared to that of the pure tungsten. After irradiation thermal diffusivity of the W-5% Re alloy is only marginally changed.

To summarize, the data from the reviewed neutron irradiation studies of tungsten indicate a decrease of thermal diffusivity in the range of 15–50%, showing a clear dependence on the amount of rhenium present. The rhenium content makes the electron effect more dominant, while the displacement damage accounts for the phonon effect. However, the radiation environment of tungsten as a spallation target material is different from the reactor environments. The high energy protons interact with materials in different ways than neutrons and the tungsten is also exposed to fast secondary neutrons with a kinetic energy of up to an order of 1 GeV. In this work, we present results from thermal diffusivity measurements of pure tungsten irradiated at the Swiss Spallation Neutron Source (SINQ). The specimens are irradiated by a mixed flux of high energy primary protons and fast secondary neutrons up to a displacement damage dose of 5.8 dpa.

2. Experimental

2.1. Material and specimens

Two slices of 1 × 10 × 100 mm were cut from the cross-section of a 1 cm thick cross-rolled tungsten plate, with a purity of 99.9 wt% tungsten. The average grain sizes were in the range 17–25 μm. From these two slices, several small plates of 1 × 8 × 30–50 mm were cut and irradiated in Target-7 of SINQ at the Paul Scherrer Institut (PSI). These plates were irradiated with high energy protons and spallation neutrons up to doses of about 28 dpa. This irradiation campaign took place during the years 2007–2008, as a part of the fifth SINQ Target Irradiation Program (STIP-V). After the irradiation, several disk-shaped specimens, with a diameter of 6 mm, were cut from the plates using electro-discharge machining (EDM). Due to the high activity of the irradiated tungsten, only two of the lowest doses could be used for measurements in this study. The specimens received doses of 3.9 dpa with 158 appm He at an average irradiation temperature of 115 °C, and 5.8 dpa with 245 appm He at an average irradiation temperature of 140 °C. The irradiation temperature is nearly linearly dependent on the proton beam current received by the target. The variation of the current, and thus the temperature, during the irradiation of the STIP V specimens is estimated to ±15% around the average values.

The main transmutation product in the irradiated specimens is expected to be rhenium, which is produced in tungsten by thermal neutron capturing (n, γ) followed by beta decay (β^-). Natural tungsten consists largely of the isotopes W184 (30.7%) and W186 (28.6%). The (n, γ) cross-section of thermal neutrons with a typical kinetic energy of 0.025 eV is 1.62 barn for W184 and 37.9 barn for W186. The calculated maximum thermal neutron flux in the SINQ target is 1.5×10^{13} n/cm^2/s/mA [8] and the accumulated beam charge on the SINQ Target 7 is 9.83 Ah. This leads to an estimation of the rhenium fraction in this tungsten material to be approximately 2%.

The irradiated tungsten specimens were mechanically polished on both sides in several steps, finishing with standard colloidal silica suspension. In addition, two unirradiated tungsten reference specimens, polished and unpolished, were also used. The unpolished specimen was used to study the effect of surface roughness and emissivity differences on the signal-to-noise ratio during measurement. The average thicknesses were measured using a point-type micrometer. All the specimens were around 1 mm thick and 6 mm in diameter. The disks were then cleaned with acetone in an ultrasonic bath, carefully dried and coated with Graphit-33 carbon spray (Kontakt Chemie) on both sides. All the disks, except the unpolished reference tungsten, were coated. The graphite coating serves three purposes - increase the absorption of the flash energy to equate the input energy to the deposited energy in the sample, improve the emission of infrared radiation to the detector, and homogenize the surfaces of the test samples and the references used for specific heat determination.

To validate the measurements, two types of standard specimens, alumina and molybdenum, were tested together with the tungsten