Tungsten as Spallation Material at the European Spallation Source.

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Characterization of thermal and mechanical properties of high-energy proton and spallation neutron irradiated tungsten was performed. The work includes studies of unirradiated tungsten from various suppliers and processing routes, to determine which grade of tungsten is the most suitable for use as spallation material at the European Spallation Source (ESS). Present fatigue studies on unirradiated tungsten suggest limiting the maximum stress in tungsten to 100 MPa and choosing rolled and annealed material which has proven to be the most durable. Studies of tungsten in mildly oxidising atmospheres, show that even 5 ppm of oxygen impurity in the helium cooling gas is enough to oxidise it. Based on these results, the upper temperature limit in the target during normal operation was set to 500 °C. However, both temperature and thermo-mechanical stress in tungsten will alter as the properties of the material change due to irradiation. As available data on irradiated tungsten is very limited, large efforts were made to study tungsten irradiated under similar conditions as the future ESS target material. These studies include irradiation induced changes in thermal diffusivity, hardness, ductility, and ultimate tensile strength. Data from these tests show severe embrittlement of tungsten post-irradiation, with virtually zero plasticity in specimens tested up to 500 °C. Hardness tests showed an increase in hardness by more than 50\%, and thermal diffusivity decreased by up to 51\%.

**KEYWORDS:** Tungsten, post-irradiation examination, mechanical testing, European Spallation Source, STIP-V, fatigue, oxidation, thermal diffusivity.

1. Introduction

ESS will use pure tungsten as the spallation material. The target will be irradiated by a pulsed 2 GeV proton beam, depositing 357 kJ/pulse in the tungsten at 14 Hz. During the 2.86 ms pulse, temperature in the tungsten is expected to rise by approximately 100 °C. To distribute the load from the proton beam, the target is designed as a wheel, with a diameter of 2.6 m, rotating at 0.39 Hz. The spallation material is made up of nearly 7000 tungsten bricks with dimensions 80×30×10 mm$^3$. The bricks are separated by 2 mm wide cooling channels where helium gas flows at a rate of 2.85 kg/s.

Pure tungsten was chosen as it has many attractive properties, unfortunately it is also known for its low ductility, low ductile-to-brittle transition temperature (DBTT), poor high-temperature oxidation resistance, and severe irradiation induced embrittlement. In addition, experimental data on properties of tungsten irradiated by high-energy protons is very limited. With the present work, the aim is to estimate the magnitude and nature...
of the irradiation induced changes by characterizing thermal and mechanical properties of high-energy proton and spallation neutron irradiated tungsten. The work also includes studies on unirradiated tungsten for use as a spallation target material where fatigue and oxidation behaviour of tungsten is examined in conditions of relevance to the ESS target.

During the 5-year lifetime of the target, the bricks will be subjected to cyclic thermo-mechanical loading from the beam pulses and occasional beam trips. Fatigue failure of a brick might lead to partial blockage of the cooling channel, causing a hot spot in the target. Temperature increase leads to an increased rate of diffusion driven release of radioisotopes, as well as an increased rate of oxidations. The fatigue life is determined not only by the magnitude of the beam induced stress, but also the material properties. With irradiation, the risk of target failure is greatly increased. Thermal diffusivity has been observed to decrease significantly in irradiated tungsten, which results in larger thermal stresses. In addition, hardness will increase and ductility will decrease, both affecting the fatigue limit.

In the initial stages of the R&D project, the focus was on unirradiated tungsten. The results were published in two papers on fatigue behaviour [1-2], and one on oxidation [3]. The later stages of the project focused on irradiated tungsten. Thermal diffusivity [4]; hardness, ultimate tensile strength, and DBTT [5]; and effects of dynamic loads [6] were studied. Full description of the project can be found in ref. [7]. The present report intends only to give an overview of the main results.

2. Materials

2.1 Unirradiated tungsten

Several types of unirradiated tungsten were studied. The specimens were obtained from different producers and varied in size, surface roughness, and manufacturing techniques. For the oxidation studies, mainly disk-shaped rolled tungsten with a minimum purity of 99.97% was used. The specimens were 20 mm in diameter and 3 mm thick, manufactured by Tian-Long Tungsten and Molybdenum Co., China. Some experiments were carried out using tungsten foil from Plansee, Austria. The fatigue specimens were either forged, rolled and annealed, or sintered and hot isostatically pressed (HIPed).

2.2 Irradiated tungsten

The irradiated tungsten material came from PSI’s STIP-V irradiation campaign [8]. All specimens used in the present work came from the same piece of tungsten, a commercially pure (99.9%), cross-rolled plate with the average grain size 25, 20, and 17 μm (x, y, z). The edge of the plate located closest to the beam centre received a total dose of 28 dpa with 1300 appm He and 7200 appm H, at an average irradiation temperature of 550 °C. The other end of the plate received the corresponding values 1.3 dpa, 37 appm He, 310 appm H and 75 °C. Due to the high radioactivity, only a few of the lowest dose specimens could be studied so far. These include two disks with 6 mm diameter, and six bend bars with dimensions 1×2×8 mm³. The specimens were used for 3-point bend tests, measurements of thermal diffusivity, hardness, and microscopy.
3. Methods

3.1 Fatigue

The objective of the fatigue study was to answer questions such as: how large is the difference in quality between the manufacturers; which processing route produces the best tungsten; which processing route produces the best tungsten; what is the impact of the surface finish technique; how is fatigue life affected by elevated temperatures; and most importantly, what is the maximum allowed stress in the target material. The work was done in two parts. Firstly, rolled vs. forged specimens from Tian-Long were tested at temperatures ranging from room temperature to 480 °C. In the second part, rolled vs. HIPed specimens from Plansee were tested at room temperature. The experimental setup and the runout limit were the same in both studies.

Stress-controlled fatigue testing was performed in the most conservative way possible, i.e. in full tension. The mean stress was therefore approximately the same as the stress amplitude. The load reversal frequency was 25-30 Hz, and the runout limit was set to 2×10^6 cycles.

3.2 Oxidation

The objectives of the oxidation study were to characterise the oxidation behaviour of tungsten at low oxygen partial pressures and temperatures of relevance to ESS, identifying the temperature at which the oxide scale loses its protective nature, and comparing the oxidation rates at different oxygen partial pressures. Oxidation data in the temperature range 400-900 °C, in inert gas mixtures with additions of small amounts of oxygen or water vapour was acquired. Oxygen and water vapour levels ranged from pO2 of 0.5 Pa to 5.1 kPa, and pH2O of 100 Pa to 790 Pa. Most of the experiments were conducted using a thermogravimetric analysis (TGA) unit, in which the specimens were oxidised under isothermal conditions for a duration of 2h per experiment. The oxides were characterised using X-ray diffraction (XRD), Energy-dispersive X-ray spectroscopy (EDS), and Auger Electron Spectroscopy (AES).

3.3 Thermal diffusivity

Some of the fast neutrons produced in the spallation process will be thermalized by the surrounding hydrogen-rich moderators and water coolants and contribute to the formation of rhenium in tungsten as a transmutation product. There will also be a displacement damage of maximum 2 dpa per year in the tungsten. Both are expected to cause a decrease in thermal diffusivity. The measurements in the present study were made on two tungsten specimens irradiated by protons and spallation neutrons to different doses. Using a commercial Netzsch LFA 467 Light Flash apparatus, with a xenon lamp and an InSb IR-detector, their thermal diffusivities could be determined in the temperature range 25-500 °C.

3.4 Mechanical properties

Pure tungsten, from the same irradiation campaign as the specimens in the thermal diffusivity study, was tested in 3-point bending at temperatures 25-500 °C. The specimen sizes were 1×2×8 mm³ and 1×2×4 mm³. The latter ones were prepared from the fractured pieces of the larger specimens, to collect more data over a wider
temperature range. The irradiation doses and test temperatures of the larger test pieces were: 1.8 dpa (350 °C), 2.1 dpa (400 °C), and 1.4 dpa (450 °C). The corresponding values for the smaller pieces were: 1.3, 2.1, and 3.5 dpa (25 °C), 1.8 dpa (350 °C), 2.1 dpa (450 °C), and 2.6 dpa (500 °C). The 3-point bend tests were done on a Zwick universal testing machine with a load capacity of 10 kN. The crosshead travel speed during the tests was 0.1 mm/min. In addition, hardness was measured, and fracture surfaces observed with scanning electron microscopy.

3.5 Effects of cyclic loading by pulsed heavy ion irradiation

Two types of tungsten specimens were irradiated with heavy ions at the at GSI, Darmstadt. The main purpose was to study the behaviour of tungsten under dynamic beam loads comparable to those at ESS. The material was disk-shaped tungsten, 20 mm in diameter. One type was 3 mm thick and the other 26 μm, both were rolled tungsten. The irradiation was done using pulsed, 4.8 MeV/nucleon, gold and uranium ions. The repetition rate of the uranium beam was 1 Hz, the pulse length 150 ms, and the ion intensity 7.5×10⁹ ions/cm²/pulse. The uranium beam was used to study the dynamic response of tungsten under cyclic loading, while the gold beam, with the time averaged ion flux 5×10⁹ ions/cm²/s, was used to study the effect on thermal diffusivity as a function of increasing fluence. The displacement damage in the specimens, calculated by FLUKA, was 0.06 dpa for a thick disk irradiated with uranium, and 0.04 dpa for a foil irradiated with gold. After the irradiation the specimens were hardness tested and studied using various microscopy techniques, such as SEM, AES, and EBSD.

4. Results and discussion

4.1 Fatigue

The fatigue and tensile property studies included forged, rolled, and HIPed specimens, polished and unpolished surfaces, stress- and strain-controlled testing, and testing in ambient and elevated temperatures. At room temperature, all types of specimens failed in a brittle manner. Majority of the specimens did not show any sign of plastic deformation on the fracture surfaces. At 280 °C, fatigue properties were slightly improved and most of the specimens were observed to be in the ductile regime. In general, the rolled specimens showed higher endurance limits, higher density, and higher average hardness compared to the forged specimens, but they were also highly anisotropic. Specimens oriented across the rolling direction were of significantly lower quality. Endurance limits of the rolled tungsten were 237.5 MPa at room temperature and 252.5 MPa at 280 °C. The lowest endurance limit, 125.6 MPa, belonged to the forged specimen type. In the second fatigue study, fatigue and tensile properties of rolled and annealed tungsten were compared to those of sintered and HIPed. Again, rolled tungsten proved to have superior fatigue properties while simultaneously displaying a highly brittle behaviour and strong anisotropy. The UTS of the rolled tungsten from Plansee was nearly 1000 MPa, and the mean fatigue strength 371 MPa. The sintered and HIPed Plansee material had an average UTS of 567 MPa, and mean fatigue strength of 185 MPa.
4.2 Oxidation

The commercially pure inert gas contained oxygen impurities of maximum 5 ppm, which turned out to be enough to oxidise tungsten. A very thin and adherent oxide layer was observed already at 500 °C in pure argon. XRD analysis of the surface confirmed presence of W15O49. Analysis of specimens oxidised in air suggest that a transition from the adherent W16O49 oxide to the porous, nonprotective WO3 occurred between 500 and 600 °C. The implication is that tungsten is to be kept below 500 °C. Below this temperature, there is a possibility that the oxide formed will consist of mainly the adherent type. Temperatures and environments where the tungsten oxide transforms into the porous WO3 type must be avoided. The risk of erosion of the oxide scale, caused by the helium gas flowing at 2.85 kg/s at 1 MPa, will increase significantly if the oxide type changes to WO3.

4.3 Thermal diffusivity

Thermal diffusivity of irradiated tungsten was measured in the temperature range 25-500 °C. The specimens were irradiated at PSI to displacement damage doses of 3.9 and 5.8 dpa. Thermal diffusivity of both specimens was ~35 mm²/s, independent of dose and test temperature. This is 51% lower than thermal diffusivity of the unirradiated specimen tested at 25 °C, and 28% lower at 500 °C. The irradiation induced decrease in thermal diffusivity is attributed to a combination of displacement damage and formation of rhenium as transmutation elements. Both specimens showed similar thermal diffusivity values despite being irradiated to different doses. This implies that the rhenium content, which is expected to be similar in both specimens, is responsible for the main contribution of the degradation. To investigate this further, the irradiated tungsten was annealed at 1000 °C for 1h and remeasured. The heat treatment should have caused partial recovery of the irradiation induced displacement damage. The test results showed that the thermal diffusivity did increase after the annealing. At 25 °C it was 47 mm²/s, and at 500 °C it was 41 mm²/s.

4.4 Mechanical properties

Specimens with doses 1.3-3.5 dpa, irradiated at PSI, were examined using 3-point bending at temperatures 25-500 °C. All irradiated specimens, regardless of dose and test temperature, failed in the elastic regime. The irradiated tungsten has virtually zero ductility and fails at stress levels much below those of the unirradiated specimens. Hardness measurements show significant irradiation induced hardening in all specimens. Unirradiated and low dose specimens (1.3-1.4 dpa) fail in a mixed trans- and intergranular mode. The intergranular fracture mode becomes more dominant at higher temperatures. At 450 °C, both the unirradiated specimen and the 1.4 dpa specimen display a fracture surface with slip bands visible on the grain surfaces, indicating some amount of plastic deformation. Specimens irradiated to higher doses exhibit a rather flat fracture surface, which indicates a very brittle cleavage fracture. Fractographs of higher magnification revealed a substructure consisting of 0.5-2 μm sized grains with grain boundaries angels >10°. This implies that the fracture of the higher dose specimens was caused by crack propagation along the subgrain boundaries.
4.5 Effects of cyclic loading by pulsed heavy ion irradiation

Two types of specimens with thicknesses 3 mm and 26 μm, were irradiated with a pulsed ion beam. The beam pulse induced stress reached 122 MPa in the thick one and 270 MPa in the thin one. The thin specimen was subjected to approximately 1x10⁶ cycles with a stress amplitude of about 50 MPa and did not show any signs of fatigue. Irradiation induced increase in hardness and decrease in thermal diffusivity was observed. These data were used in coupled flow, thermal and mechanical simulations of the maximum temperature and stress in the ESS tungsten brick after irradiation. The calculated results for an irradiated tungsten brick show an increase of more than 20% of the maximum thermal stress. The work included tests of oxide adhesion under the pulsed beam. SEM and AES characterisation of the oxide confirmed loss of >1 μm thickness in the irradiated region, implying that the oxide formed during operation will eventually be released into the cooling loop.

5. Summary

Rolled tungsten shows superior fatigue properties, surviving much higher stress amplitudes than both the forged and the HIPed tungsten. It has nearly zero ductility at room temperature and is highly anisotropic, which means that the orientation of the rolled tungsten bricks with respect to the proton beam needs to be carefully considered. The post-pulse maximum stress should be below 100 MPa, and the post-pulse peak stress amplitude lower than 50 MPa. The temperature in the tungsten bricks should be kept below 500 °C so that significant oxidation can be avoided. Irradiation will cause complete loss of ductility, even at 500 °C, and thermal diffusivity will be reduced by nearly 50%.

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