Int. Journal of Refractory Metals and Hard Materials 60 (2016) 28–36

Contents lists available at ScienceDirect

Int. Journal of Refractory Metals and Hard Materials

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

High-flux plasma exposure of ultra-fine grain tungsten

R.D. Kolasinski [a,*], D.A. Buchenauer [a], R.P. Doerner [b], Z.Z. Fang [c], C. Ren [c], Y. Oya [d], K. Michibayashi [e], R.W. Friddle [f], B.E. Mills [g]

[a] Sandia National Laboratories, Energy Innovation Department, Livermore, CA 94551, USA
[b] University of California – San Diego, Center for Energy Research and Department of Mechanical Engineering, La Jolla, CA 92093, USA
[c] University of Utah, Department of Metallurgical Engineering, Salt Lake City, UT 84112, USA
[d] Shizuoka University, Department of Chemistry, Graduate School of Science, Shizuoka 422-8529, Japan
[e] Shizuoka University, Institute of Geosciences, Shizuoka 422-8529, Japan
[f] Sandia National Laboratories, Energy Nanomaterials Department, Livermore, CA 94550, USA
[g] Sandia National Laboratories, Radiation/Nuclear Detection Materials & Analysis Dept., Livermore, CA 94550, USA

ARTICLE INFO

Article history:
Received 18 December 2015
Received in revised form 26 March 2016
Accepted 10 May 2016
Available online 12 May 2016

ABSTRACT

In this work, we examine the response of an ultra-fine grained (UFG) tungsten material to high-flux deuterium plasma exposure. UFG tungsten has received considerable interest as a possible plasma-facing material in magnetic confinement fusion devices, in large part because of its improved resistance to neutron damage. However, optimization of the material in this manner may lead to trade-offs in other properties. We address two aspects of the problem in this work: (a) how high-flux plasmas modify the structure of the exposed surface, and (b) how hydrogen isotopes become trapped within the material. The specific UFG tungsten considered here contains 100 nm-width Ti dispersoids (1 wt%) that limit the growth of the W grains to a median size of 960 nm. Metal impurities (Fe, Cr) as well as O were identified within the dispersoids; these species were absent from the W matrix. To simulate relevant particle bombardment conditions, we exposed specimens of the W-Ti material to low energy (100 eV), high-flux ($>10^{22}$ m^{-2} s^{-1}) deuterium plasmas in the PISCES-A facility at the University of California, San Diego. To explore different temperature-dependent trapping mechanisms, we considered a range of exposure temperatures between 200 °C and 500 °C. For comparison, we also exposed reference specimens of conventional powder metallurgy warm-rolled and ITER-grade tungsten at 300 °C. Post-mortem focused ion beam profiling and atomic force microscopy of the UFG tungsten revealed no evidence of near-surface bubbles containing high pressure D_2 gas, a common surface degradation mechanism associated with plasma exposure. Thermal desorption spectrometry indicated moderately higher trapping of D in the material compared with the reference specimens, though still within the spread of values for different tungsten grades found in the literature database. For the criteria considered here, these results do not indicate any significant obstacles to the potential use of UFG tungsten as a plasma-facing material, although further experimental work is needed to assess material response to transient events and high plasma fluence.

1. Introduction

Understanding the interaction between the plasmas created in magnetic-confinement fusion devices and the materials that comprise the interior surfaces presents arguably one of the most challenging problems in materials science. A unique issue is the complexity and extraordinary harshness of the fusion environment: the materials must be able to accommodate particle bombardment conditions that in many cases far exceed any existing application. This includes exposure to high-flux deuterium-tritium (D-T) plasmas ($>10^{24}$ D + T m^{-2} s^{-1}), high energy fusion products (e.g. 14.1 MeV n and 3.5 MeV α), impurities, as well as deliberately introduced gas species to promote radiative cooling of the scrape-off-layer. It is not clear that any existing material can satisfy all of the desired performance metrics required for long-term deployment in magnetic-confinement devices, thereby motivating the need for advanced materials and plasma facing component designs [1]. Polycrystalline tungsten has been viewed as a viable starting-point for the ITER tokamak (at present under construction at the Cadarache facility, France), given its high melting temperature, thermal conductivity, and low coefficient of thermal expansion. In terms of its response to plasma exposure, the low sputtering yield and low solubility of hydrogen isotopes in tungsten are also considered advantageous. Nevertheless, conventional polycrystalline tungsten grades suffer from low ductility, and offer only marginal resilience against neutron embrittlement, which may affect their use in higher power

* Corresponding author.
E-mail address: rkolasi@sandia.gov (R.D. Kolasinski).

http://dx.doi.org/10.1016/j.ijrmhm.2016.05.006
0263-4368/

reactors. In addition, recrystallized tungsten is also susceptible to intergranular cracking as a result of transient heat loading and cooling cycles. This could promote crack extension, eventually propagating to the supporting structure [2].

Several strategies have been proposed to improve the properties of tungsten [3–5]. Of particular recent interest is the development of ultra-fine grained (UFG) tungsten materials. A microstructure characterized by tungsten grains <2 μm wide may be realized through the incorporation of small transition metal dispersoids to limit grain growth. The dispersoids can suppress recrystallization embrittlement, and the high density of grain boundaries may serve as sinks for displacement damage arising from neutron and high energy ion irradiation [6]. Several efforts are underway to develop and characterize such materials, including recent work led by Kurishita [6–8] and Miyamoto [9]. Their work focuses on tungsten containing 200 nm diameter TiC (1.1 wt%) or W-TaC (3.3 wt%) dispersoids. Initial testing of the UFG material has demonstrated a lower ductile-brittle transition temperature (240 K). Furthermore, post-mortem analysis of UFG specimens subjected to thermal fatigue loading [10] and thermal shock [11] in electron beam facilities have revealed negligible cracking and surface morphology modification.

While the aforementioned progress is quite promising, optimization of tungsten properties in this manner may affect its performance in other respects. In the present study, we focus on an assessment of (a) plasma-induced surface morphology modification and (b) hydrogen isotope trapping within the material. Because of the low solubility of hydrogen isotopes in tungsten, precipitation of $H_2(g)$ in small, high pressure bubbles and blisters is highly favorable [12]. These structures may grow through a variety of mechanisms, including dislocation loop punching and near-surface crack propagation. This may produce a delaminated layer of material near the surface that is poorly connected to the bulk, and therefore be more susceptible to melting, recrystallization, or flaking during operation. How the modified grain structure of UFG tungsten affects diffusion and trapping of hydrogen isotopes is especially a concern for tritium which, due to its radioactivity, presents a considerable safety hazard if allowed to accumulate within the reactor. Prior investigation of hydrogen-isotope retention in UFG tungsten is limited, and the existing published experimental database for W-TiC contains several contradictory findings. For example, initial high flux plasma-exposures in the PISCES-A linear plasma device revealed very low retention of D within the material compared with pure stress relieved tungsten [9] at an exposure temperature of 300 °C and ion fluence of 5×10^{25} D m^{-2} s^{-1}. Later testing by Zibrov et al. [13] conversely revealed consistently higher trapping in W-TiC and W-TaC alloys for fluences ranging between 10^{22} and 10^{25} D m^{-2} s^{-1}. Since both studies involved the use of similar materials, the reason for the disparity between these two studies is unclear.

In this work, we seek to identify the physics mechanisms underlying hydrogen isotope trapping and surface morphology changes in UFG W materials. For this purpose, we consider a recently-developed material containing Ti dispersoids to serve as inhibitors to grain growth. This article begins with a description of the material preparation and characterization, along with a discussion of high-flux D plasma exposures using PISCES-A linear plasma device at the University of California, San Diego. We then present an analysis of the trapped D assessed by thermal desorption spectrometry, followed by a characterization of the plasma-modified surface morphology using several microscopy techniques. Finally, we conclude with an analysis of the material composition using Auger electron spectroscopy and X-ray photoelectron spectroscopy.

2. Material preparation and plasma exposure

The ultra-fine grained (UFG) tungsten used in this study was fabricated using a unique power metallurgy process. The raw materials consisting of W and Ti powders were mixed with WC balls and milled using a custom-designed high-energy planetary mill (HEPM) [14] for 6 h. The HEPM process generates a 60*g*-force acceleration field that can effectively reduce the crystalline size of tungsten powders to <20 nm. After milling, tungsten powders were compacted into 38 mm diameter disc green parts using a uniaxial die press with pressure up to 200 MPa. The green parts were sintered in an atmosphere-controlled tube furnace. The sintering profile include a reduction step at 700–800 °C for 3 h in flowing H_2 and a sintering step at 1300 °C for 1 h in Ar. The sintered samples have relative density of about 98% of the theoretical value. The high density achieved at such a low sintering temperature was attributed to the extremely high driving force for sintering in the highly active nano-W powder prepared using the HEPM process. The Ti particles at the tungsten grain boundaries were added as grain growth inhibitors resulting in the ultrafine grain size in the samples.

All of the W-Ti UFG samples prepared for this study were cut to a uniform size of 25 mm diameter, 1 mm thick discs by electrical discharge machining (EDM) and grinding. They were then mechanically polished to an rms surface finish of 10 nm (measured by atomic force microscopy.) For comparison, we also considered two pure tungsten reference materials that are widely used for plasma-exposure studies. The first of these is ITER-grade material (Allied Materials Corp.), where the exposed surface is aligned perpendicular to the rolling direction. This produces a microstructure with grains elongated along the thickness of the material to promote better heat conduction away from the surface. The raw material was in the form of a 25 mm diameter rod, from which 1 mm thick specimens were cut via EDM. Details of the ITER-grade specification may be found in Ref. [15]. Specimens of the second reference material were fabricated from a 1 mm-thick warm-rolled tungsten sheet (PLANSEE), where the microstructure is characterized by grain elongation parallel to the surface. An extensive database is available for each reference material, both of which have far larger grain size than the UFG material described above.

To reveal the basic structure of the UFG material and to enable a more quantitative analysis of grain size, we acquired electron backscattering diffraction (EBSD) maps at five randomly selected locations near the center of the sample surface. A representative selection of the EBSD data is shown in the surface normal projected inverse pole figure-based map depicted in Fig. 1(a). The orientation of the individual grains is conveyed by assigning a red/green/blue colors to each Euler angle, which can be translated to orientations through the adjacent color key. In each case a 15.4 μm × 11.5 μm region of the surface was surveyed in a rectangular grid, using 0.1 μm steps. The white regions indicate points where a distinct Kikuchi diffraction pattern could not be resolved. These locations correspond to grain boundaries, where the Ti dispersoids are also present. (Areas of high surface roughness can also lead to indexing errors.) Our attempts to determine the exact phase of the dispersoids using EBSD were ultimately unsuccessful. Since the size of the dispersoids was comparable to the 0.1 μm mapping resolution, this is perhaps not surprising. As will be discussed later in Section 5, we later found that the Ti particles contained O as well as other impurities, and it is likely that this complex combination of constituents would prevent identification of the exact phase. It is important to note that all orientation data presented in the EBSD maps therefore correspond to tungsten grains. An analysis of the orientations did not reveal any obvious evidence of preferred texture. This is consistent with the powder metallurgy fabrication procedure used, where a random distribution of grain orientations would be expected.

Material microstructure is closely linked not only to the mechanical properties, but also to hydrogen diffusion, trapping, and precipitation within the material. To calculate and interpret the grain size distribution, we used data analysis procedures previously developed by Mingard et al. [16] for guidance. We first filtered the EBSD raw data and reconstructed the grain boundaries, using a 10° misalignment threshold. Based on the reconstructed data set, we then calculated the internal area of the individual grains and then binned them according