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Achieving high strength/ductility in bulk W-Zr-Y_2O_3 alloy plate with hybrid microstructure

Z.M. Xie [a,b], R. Liu [a], T. Zhang [a,*], Q.F. Fang [a,b,**], C.S. Liu [a,*], X. Liu [c], G.N. Luo [d]

[a] Key Laboratory of Materials Physics, Institute of Solid State Physics, Chinese Academy of Sciences, Hefei 230031, China
[b] University of Science and Technology of China, Hefei 230026, China
[c] Southwestern Institute of Physics, Chengdu, China
[d] Institute of Plasma Physics, Chinese Academy of Sciences, Hefei 230031, China

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ABSTRACT

A hybrid microstructure design strategy which achieves 6.5 mm thick W-0.2 wt% Zr-1.0 wt% Y_2O_3 (WZY) alloy plates with a DBTT lower than 150 °C is reported. The WZY alloy is brittle at 100 °C with an ultimate tensile strength (UTS) of 798 MPa and shows detectable plastic deformation at 150 °C with a total elongation (TE) ~3.2% and a UTS of 911 MPa. The UTS of this WZY alloy is larger than 580 MPa from RT to 500 °C and the TE increases to ~23.2% at 400 °C. The processing route combining micro-alloying, hot rolling and nano-sized oxide dispersion strengthening leads to a hybrid microstructure with refined equiaxed sub-grains, elongated mother grains and dispersed nanoparticles, which significantly strengthens/toughens the W alloys.

1. Introduction

Tungsten (W) is a kind of refractory metal that keeps its body-centered cubic crystal structure from room temperature (RT) to its melting temperature of 3410 °C. It shows high thermal conductivity (174 W/(m k)), high stability and high hardness/strength with low sputtering yield and low coefficient of thermal expansion. These superior properties are appealing for high temperature applications such as plasma-facing materials (PFMs) in future fusion reactors, solid targets in spallation neutron sources as well as critical components in rockets and missiles [1,2]. However, pure W exhibits a serious embrittlement in several regimes, i.e., low-temperature embrittlement (relatively high ductile-brittle transition temperature, DBTT > 400 °C), recrystallization embrittlement (recrystallization temperature ~1200 °C) and radiation embrittlement [3–5]. These embrittlement drawbacks severely limit the utilization of W based materials especially in high temperature and radiation environments and confine its application in the intermediate temperature region. In order to take advantage of the superior properties of W, it is necessary to significantly mitigate embrittlement and improve the mechanical properties of W based materials.

In recent decades, many efforts have been devoted to improving the toughness and ductility of W alloys. Favorable properties, such as low-temperature ductility and promising machining capabilities, have been achieved in tungsten–rhenium (Re) alloys [6]. But this approach results in softening of W alloy and is costly owing to the expense of Re. Some results have demonstrated that dispersing fine oxide particles such as ThO_2 and La_2O_3 in the W matrix can improve the toughness through strengthening the grain boundaries (GBs) and increasing the recrystallization temperature (RCT). However, they still exhibit poor RT fracture and low ductility with a DBTT above 400 °C [7]. Recently, some new W alloys with excellent performance were developed. For example, the 0.1 mm thick W laminate made of several layers of W foils exhibits ductility at RT [8]. The W-1.1% TiC alloy with fine grains fabricated by grain boundary sliding-based microstructural modification (GSMM W–TiC components [9]) exhibits very high fracture strength up to about 4.4 GPa and appreciable bending ductility at RT. Although some of the above processes have significantly improved the performance of tungsten materials, the small sample size and inferior fabrication efficiency/economy are very limited for the engineering applications.

It is well known that brittle fracture in W mainly occurs along GBs due to the weakness of GBs with random orientations and the segregation and precipitation of gaseous interstitial elements like oxygen (O) and nitrogen (N) at the GBs [10,11]. In order to strengthen the GBs, it is necessary to convert the weak cohesion at random grain boundaries into a strong interatomic bond and reduce the contents of detrimental impurities of O and N as much as possible. Micro-alloying becomes a promising method to strengthen GBs by adding small amount of active elements such as zirconium (Zr) and yttrium (Y) which have high affinity for impurities of O, C, and N and can bind with those impurity

* Corresponding authors.
** Correspondence to: Q. F. Fang, Key Laboratory of Materials Physics, Institute of Solid State Physics, Chinese Academy of Sciences, Hefei 230031, China.
E-mail addresses: zhangtao@issp.ac.cn (T. Zhang), qffang@issp.ac.cn (Q.F. Fang), csliu@issp.ac.cn (C.S. Liu).

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elements to form stable compounds at high sintering temperature. It was confirmed by the recent results of W-Zr and W-Y alloys with an increase of tensile strength and a reduction of DBTT [12,13]. On the other hand, grain refinement could increase GBs area and decrease the average concentration of impurities at GBs, thus improving the strength and toughness of W [9]. It was indicated that dispersing small amounts of thermally stable ZrC/TiC/Y_2O_3 particles into tungsten to form the so-called carbide/oxide-dispersion-strengthened tungsten (ODS-W) can effectively refine the grains and thus fabricate fine-grained W based components exhibiting improved high-temperature strength, recrystallization temperature and creep resistance [14].

Combining the advantages of plastic deformation, micro-alloying and nano-sized oxide dispersion strengthening in powder metallurgy is a good choice for fabricating engineering-applied bulk W alloys. In this work, bulk fine-grained W-0.2 wt% Zr-1.0 wt% Y_2O_3 alloys are fabricated based on an effective powder metallurgy method through mechanical alloying, sintering and hot rolling processes. As compared with some peculiar ways like spark plasma sintering (SPS), hot isostatic pressing (HIP) or microwave sintering, this metallurgy method is more suitable for the production of large size bulk W alloys. The effects of working processes and Zr/Y_2O_3 additions on microstructure and mechanical properties, especially the tensile behaviors, of the tungsten samples are investigated.

2. Experimental details

2.1. Starting materials, ball milling method and fabrication

W-1.0 wt% Y_2O_3 and W-0.2 wt% Zr-1.0 wt% Y_2O_3 (hereafter abbreviated as WY and WZY, respectively) alloys were fabricated from pure W (purity > 99.9% trace metals basis, purchased from Xiamen Tungsten reagent Co., LTD.), ZrH_2 (particle size 10–20 μm, purity > 99.9%), and nano-sized Y_2O_3 (average particle size 50 nm, purity > 99%) powders were ball milled in a planetary ball mill for 4 h in argon atmosphere with ball-to-powder weight ratio of 8:1 and a rotation speed of 240 rpm. The mixed powders were subsequently sintered at 2200 °C at a pressure of 70 MPa for 10 h in vacuum to get sintered billets with grain sizes ranging from 100 to 300 μm. Then the sintered billets were hot-rolled into a plate with thickness of 6.5 mm (from original thickness of 18 mm, through four-step thermomechanical treatment with each step deformation of 15%, 20%, 25% and 30%, respectively, at 1650 °C), width of 140 mm and length of 220 mm.

2.2. Microstructure and relative density

Microstructural observations were examined by high-resolution transmission electron microscopy (HRTEM, JEM–2010) equipped with an energy dispersive X-ray spectrometer (EDS). TEM specimens were prepared by twin-jet in a Struers Tenupol-5. The elaborated metallographic images of the specimens were obtained using an electron backscattered diffractometer equipped on a Zeiss SIGMA field emission scanning electron microscope (FE-SEM) after electrolytic polishing in 5% sodium hydroxide aqueous solution at RT with a constant voltage of 11 V and a current density of 3 mA/mm^2. Electron Backscatter Diffraction Pattern (EBSD) mappings were measured with an acceleration voltage of 20 kV and collected using a CRYSTAL detector (Oxford Instruments, Oxfordshire, UK) to characterize the microstructure of specimens. EBSD was mapped with a testing step size of 100 nm on the chosen area (300 × 300 μm^2). The EBSD images are slightly filtered through a software (HKL Tango) to eliminate the influences of second-phase particles on tungsten grain observations. The corresponding EBSD shows the distribution of tungsten grain size and misorientation angle. Specimens for TEM and EBSD examinations were all cut from the RD–ND surface. RD, TD and ND are short for rolling direction, transverse direction and normal direction, respectively. The second phase particle size has been statistically analyzed using quantitative metallography based on 500 particles from BSD-SEM images of the rolled WY and 800 particles from TEM images of the rolled WZY, respectively. The fracture surfaces of sample were characterized by a field-emission scanning electron microscope (FE-SEM SU8020, Hitachi).

The densities of the as-prepared samples were measured by the Archimedes methods. The relative density is calculated basing on the theoretical densities of 19.3 g/cm^3 (W), 6.49 g/cm^3 (Zr) and 5.01 g/cm^3 (Y_2O_3), respectively, and the results were listed in Table 1. The calculated relative densities of WY and WZY samples are higher than 99%.

2.3. Mechanical properties tests

For tensile testing, the dog-bone-shaped specimens with a cross–section of 1.5 × 0.75 mm^2 and a working length of 5 mm along rolling direction (RD) were prepared. All tensile specimens were mechanically polished and tested at various temperatures from RT to 500 °C using an Instron–5967 machine at a constant speed of 0.06 mm/min in air. Before Vickers micro-hardness tests, the testing specimens cut from RD–ND and RD–TD surfaces were mechanically polished with a 2000 mesh SiC paper and then electrolytically polished in 2% sodium hydroxide aqueous solution at RT with a polishing voltage of 18 V, to obtain the mirror-polished and stress-free surfaces. These polished specimens were then subjected to Vickers micro-hardness testing at RT with a load of 200 g and a dwell time of 15 s.

3. Results and discussion

The Vickers micro-hardness of the rolled WY is 488 HV for RD-ND plane and 486 HV for RD-TD plane, respectively, as listed in Table 1. These almost identical hardness values, to some extent, reflect an isotropic bulk WY plate resulted from the harmonious microstructure of recrystallized state and equiaxed tungsten grain geometry, which will be discussed in the following sections in detail. In the rolled WZY, the hardness is 516 HV for RD-ND and 520 HV for RD-TD plane, respectively, which also show neglectable difference along different testing orientations on a specimen. The hardness of the rolled WZY exhibits an increase of 6% compared to the rolled WY alloys, which intuitively comes from the addition of a small amount of Zr.

The engineering stress-strain curves of the rolled WY and rolled WZY at various temperatures are presented in Fig. 1. As shown in Fig. 1, the rolled WZY shows typical brittle fracture at 100 °C with an ultimate tensile strength (UTS) of 798 MPa as well as a detectable plastic deformation at 150 °C with a UTS of 911 MPa and a simultaneous ductility (total elongation (TE) ~3.2%), indicating a ductile to brittle transition in the range from 100 °C to 150 °C. Defining the DBTT as the lowest temperature at which a sample undergoes a minimum elongation > 0 without failure, the DBTT of the rolled WZY is about 150 °C which is 50 °C lower than that of rolled WY (200 °C). This implies the better toughness of the rolled WZY at relatively low temperature. Despite the tensile strength of the rolled WZY decreasing with the increasing temperature, the UTS values are always larger than 580 MPa from RT to 500 °C.

The UTS and TE are shown as a function of the test temperature in Fig. 2, where the results of SPSed WZY [15], swaged WY [16] and rolled pure W [17] were also presented for comparison. Each data point shown in this figure is the average of results obtained from at least three specimens that were tested under the identical conditions. It is clear that the rolling process significantly raises the strength of materials as compared with swaging or SPSing techniques through comparing the performances of the rolled WY to the swaged WY and especially the rolled WZY to the SPSed WZY. On the other hand, trace Zr addition effectively increases both the tensile ductility and strength of the rolled WZY alloy: (i) both the uniform elongation (~14.2% at 300 °C and ~12.6% at 400 °C), that is, the strain achieved before the stress reaches the maximum point where the necking instability begins, and the total elongation (~18.6% at 300 °C and ~23.2% at 400 °C) to failure have improved over the rolled