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Strong ductile bulk tungsten

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ABSTRACT

Tungsten has long faced the problem of limited to no ductility in bulk polycrystalline form, due in part to the low energy needed for intergranular fracture. This limited ductility can be overcome through processing by dramatically reducing the cross sectional area following a conventional temperature step-down method that improves ductility through elongation of grain boundaries and development of a {110} fiber texture along the length of the material. While this technology is over 100 years old, little progress has been made to improve the ductility of bulk material. In the current investigation plastically deformed bulk tungsten was observed to have a room temperature tensile ductility between 17% and 23% total elongation at failure in bend specimens, and a flexural yield strength near ~ 3 GPa. This improvement in ductility following multipass equal channel angular extrusion processing appears to be caused by a similarity in microstructure and texture to that of ductile tungsten wire. The onset temperature for transition to noticeable ductility is seen to decrease dramatically with the level of plastic strain. The results indicate that severe plastic deformation processing at low homologous temperatures may be an effective way to improve the ductility and toughness of bulk tungsten and other brittle crystalline metals.

1. Introduction

The refractory transition metal tungsten (W) is known primarily for its high density (19.25 g/cc) and high melting point (3422 °C). Tungsten also has a high compressive strength, a high Young's modulus (411 MPa), and a high tensile yield stress (3920 MPa) when drawn to wire [1]. The largest present-day application for tungsten is in the fabrication of tungsten-carbide, a hard material used in cermet cutting tools, for hard-surface components, and for strengthening in iron and nickel based alloys. The special properties of tungsten make it attractive for many applications including cobalt cermet for cutting tools, ballistic projectiles, radiation shielding, ballast weights, and most recently for diffusion barriers in integrated circuits [2]. Despite attractive physical properties, tungsten has found limited use for structural applications because of poor ductility.

Tungsten has the body centered cubic crystal structure, and a ductile-to-brittle transition temperature (DBTT) between 200 and 300 °C [1]. This transition temperature is influenced by working [3–6], orientation [7], texture [8–10], strain rate [11], heat treatment [12], impurities [13,14], test mode [15], and stress concentration [16,17].

In bulk tungsten, poor ductility stems from two characteristics: a preference for intergranular separation caused by weak or embrittled grain boundaries, and a limited number of active slip systems below the ductile-to-brittle transition temperature (DBTT) [18]. In annealed polycrystalline tungsten, failure is dominated by this low energy intergranular separation, because this mode of failure requires less energy than the cleavage of the tungsten crystal lattice (referred to as transgranular cleavage). Elongation of the original grain boundaries through warm working can mitigate this low energy failure mode, where grains have sufficient length to interrupt intergranular crack propagation [19]. Processing to impart such a microstructure is typically done though swaging [3], rolling [14,20–23], and area reduction extrusion at temperatures near but not exceeding the recrystallization temperature [24]. However, while possessing higher fracture toughness at elevated temperatures, tungsten often remains brittle at ambient temperatures and can have a higher DBTT than the unworked material [8]. This behavior is in part attributed to the generation of numerous dislocations during working, which act as barriers for mobile dislocations.

The limited number of active slip systems is the other primary cause of poor ductility in polycrystalline tungsten. In single crystal experiments it was shown that tungsten possess two primary crack propagation systems: along the {100} and {110} planes. It was shown that the {100} planes possess roughly half the fracture toughness of the {110} planes [18]. The higher fracture toughness of the {110} crack system is attributed to the greater nucleation and propagation of dislocations away from the high stress field near a crack tip on the {110} planes,

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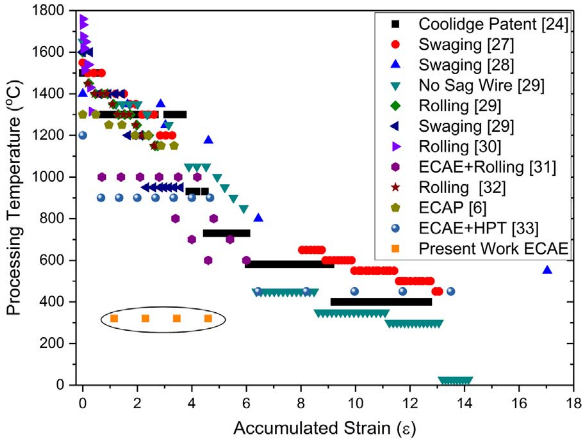

Fig. 1. Tungsten processing data from the literature and this work, converted to accumulated strain at each processing temperature. References [6,24,28–34].

producing blunting of the crack tip. It is only when tungsten has been sufficiently worked that this favorable [110] texture is generated. In order to produce this effect with traditional processing techniques involving oriented deformation, the cross section of bulk material must be reduced substantially, a circumstance that may be undesirable.

William D. Coolidge developed a technique for improving the ductility of tungsten wire for incandescent light bulb filaments and described the process in his 1913 patent on ductile tungsten [24]. This tungsten wire with increased toughness and ductility over previous filament materials was produced through a temperature step down approach illustrated in Fig. 1. For this figure, area reduction is converted into accumulated strain at each processing temperature to allow for comparisons between processing methods for different tungsten products. Various results are shown in Fig. 1 along with the Coolidge patent temperature reduction sequence. The original Coolidge approach involved compacting reduced tungsten powder, followed by sintering through self-resistance heating in a hydrogen atmosphere. The sintered rod was then swaged, area-reduction extruded, and finally drawn at progressively lower temperatures until a wire capable of being spooled and bent was produced. It would only be later in the work by Zay Jeffries [25], that the improved ductility was understood and attributed to the development of an elongated fibrous microstructure. Further work showed that the [110] fiber texture was the main reason for improved ductility [22,26,27].

More recent efforts to improve the strength and ductility of bulk tungsten have focused on grain refinement, and grain boundary elongation through severe plastic deformation (SPD) techniques including high-pressure torsion (HPT) [35], equal channel angular extrusion (ECAE) [36] or pressing (ECAP) [37], rolling [4,21,22,38], swaging [39], and ball milling [40]. These works have illustrated the grain refining ability of severe plastic deformation (SPD) techniques applied to tungsten and the effects on mechanical behavior including increased hardness, increased strength, and reduction in the recrystallization and ductile-to-brittle-transition temperatures.

The limitations of common deformation processing methods provide an opportunity for relatively new SPD processing techniques that plastically strain (work) the material without a reduction in work-piece cross section area. Of these various methods, ECAE is the most promising due to the possibility of large sample dimensions, the uniformity of deformation, high strains capable with multiple processing steps, and the ability to control work-piece temperature, strain rate, and texture [22]. The authors use the acronym ECAE throughout this paper to be consistent with the nomenclature chosen by the originator of the method [41].

Previous work on processing tungsten for improved strength and ductility by ECAE has focused on high temperature deformation following a traditional approach. These studies were based on the idea of grain size refinement for strength and ductility improvements, and not the effects of texture and microstructure/grain elongation have involved deformation steps well above the nominal DBTT of tungsten. The results of this previous work have not been encouraging, in that no bulk tungsten has been produced with notable ductility or with a microstructure similar to that of wire or sheet. The objective of the current work was to identify a method for the fabrication of bulk tungsten with a tensile elongation to failure of at least 10%. By using ECAE processing at low temperature to produce an elongated microstructure, the current work demonstrates that fabrication of bulk polycrystalline tungsten with substantial tensile ductility is possible.

2. Materials and methods

Tungsten rods of commercial purity ~ 99.97% tungsten (W) manufactured by Plansee (Reutte, Austria) measuring 12 mm diameter by 50 mm in length were encapsulated in 25.4 mm square 304 stainless steel cans. The encapsulated as-received W rods were ECAE processed, using a 90° die angle, sliding wall tool, with a 0 ° outer die angle. Processing was done at 320 °C ± 10 °C at extrusion speeds less than 1.0 mm/s, with no rotation between extrusions (referred to as Route A).

Following ECAE, bend test specimens were sectioned from the processed bars (or billets) by wire electrical discharge machining (EDM) with the long axis of the test specimen along the ECAE extrusion direction into samples measuring ~ 2 × 1 × 14 mm^3. The EDM surface was removed by mechanical grinding followed by electrolytic polishing in a 1% NaOH solution.

Electron microscopy was conducted with an FEI Quanta 600 FE-SEM scanning electron microscope (SEM), with secondary and back-scatter electron detectors. Characterization of the microstructure was done on the sample flow plane (side plane of extrusion billet). Quantizations of grain width and average subgrain diameter were accomplished using Fiji software formally known as ImageJ. Grain widths were measured at several locations on optical micrographs of etched samples. Average subgrain diameters were determined by averaging the length and width of as many as 500 individual grains for each specimen.

Because all samples were electro-polished prior to testing, it was possible to examine the deformed and fractured surfaces of specimens after testing without further surface preparation. Failure crack deflection angles were determined by measuring the angle between the loading direction and the average crack propagation direction.

Vickers hardness measurements were taken on the polished flow plane surface of small samples with a Leco Microhardness Tester LM300AT using a 300 g load and a loading time of 13 s. A total of 13 measurements were made on each sample. The highest and lowest values were disregarded when determining the average and standard deviation values.

Three-point bend tests were conducted with a custom-built test apparatus fabricated from H13 tool steel. The support pins were made from precision ground 0.20 mm diameter tungsten carbide (WC) pins. The distance between these roller supports was 7 mm as shown in Fig. 2. The sockets holding the WC rollers were coated with graphite to reduce friction. Rollers were held in grooved channels in the test

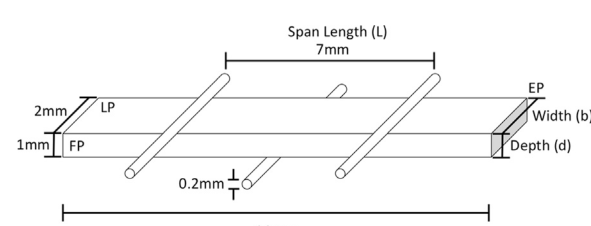

Fig. 2. Diagram of 3-point bend specimen and test apparatus configuration.