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A critical examination of pure tantalum processed by high-pressure torsion

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ABSTRACT

Tantalum, a common refractory metal with body-centred cubic (BCC) crystalline structure, was processed by high-pressure torsion (HPT) at room temperature through different numbers of rotations. Significant grain refinement and high strength were achieved with a reduction in grain size from ~60 μm to ~160 nm and an increase in strength from ~200 to > 1300 MPa. Hardness measurements revealed a high level of homogeneity after 10 turns of HPT but the hardness after 10 turns was slightly lower than after 5 turns indicating the occurrence of some recovery. Tensile testing at a strain rate of 1.0×10^{-3} s^{-1} gave high strengths of ~1200 MPa but little or no ductility after processing through 1, 5 and 10 turns. The introduction of a short-term (15 min) anneal immediately after HPT processing led to significant ductility in all samples and a reasonable level of strength at ~800 MPa.

1. Introduction

Tantalum is a refractory body-centred cubic (BCC) metal having very good room temperature ductility (> 20% tensile elongation) [1]. It is generally considered as an excellent material for use in shaped charges and explosively forged projectiles due to its high density (16.7 g cm^{-3}), superior strength and good ductility over a very wide range of strain rates and temperatures [2]. Because it is chemically inert, tantalum is used widely in the chemical industry particularly as valves, heat exchangers and bayonet heaters [1,3]. Since it is also highly bioinert, it is used for orthopaedic implants [4] and the elasticity of tantalum makes it especially appropriate for use in hip replacements to avoid stress shielding [5]. Generally, there is significant interest in making use of tantalum instead of titanium for medical prostheses but, in order to make Ta comparable to Ti as a potential implant material, it is necessary to improve the material strength.

The grain size of a material is the major microstructural parameter dictating the properties of a polycrystalline solid at room temperature. However, the application of traditional plastic working techniques to tantalum, such as rolling and extrusion, generates relatively coarse-grained microstructures [6,7]. With reference to the Hall–Petch relationship [8,9], it is reasonable to anticipate that the strength and performance of tantalum will be significantly improved if it is possible to produce an ultrafine-grained (UFG) or a nanostructured material.

It is now well-established that significant grain refinement may be introduced in bulk polycrystalline materials through the application of severe plastic deformation (SPD) [10–12] since the grain sizes produced by SPD processing are typically within the submicrometer or even the nanometre range. Various SPD techniques are now available but most attention has been centred on the two procedures of equal-channel angular pressing (ECAP) [13] and high-pressure torsion (HPT) [14]. Extensive experiments have shown that processing by HPT is especially attractive because, by comparison with ECAP, it produces smaller grains [15,16] and a higher fraction of grain boundaries having high angles of misorientation [17]. In practise, processing by ECAP and HPT has been successfully applied to a wide range of metals having face-centred cubic (FCC) and hexagonal close-packed (HCP) crystal structures

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but there are relatively few investigations of the SPD processing of BCC metals. Specifically, for tantalum there are several reports of processing and microstructural refinement using either ECAP [18–21] or HPT [22–26] but the experimental results from the HPT processing are very limited because they concentrate primarily on dynamic testing at strain rates in the range of $\sim 10^3\ s^{-1}$ using a split Hopkinson bar [22], the compression of micro-pillars in a scanning electron microscope [25] and the application of shock compression generated by high-energy laser pulsing [26]. There are two reports documenting hardness measurements in samples of Ta processed by HPT but these investigations were designed specifically to compare the hardness values obtained from a very wide range of FCC, HCP and BCC metals [23,24].

It follows from this summary that there has been no systematic study of the evolution of microstructure and the mechanical properties of tantalum when using HPT processing. Accordingly, the present research was initiated to address this deficiency by providing a detailed evaluation of microstructural evolution in tantalum during HPT processing, measuring the mechanical properties in tensile testing at room temperature using a strain rate of $1.0 \times 10^{-3}\ s^{-1}$ and examining the effect on the tensile properties of short-term (15 min) annealing in the post-HPT condition. Based on these results, the overall objective was to establish a procedure that may be used to enhance the properties of this material for use in bio-applications. An earlier report described some of the preliminary results obtained in this research [27].

2. Experimental material and procedures

The experiments were conducted using tantalum of 99.9% purity which was received in an annealed condition as a rod with a diameter of 10 mm. The rod was sliced into discs with thicknesses of ~1.2 mm and these discs were then ground with abrasive papers to final thicknesses of ~0.8 mm for HPT processing. The processing was conducted at room temperature under quasi-constrained conditions [28,29] through total numbers of turns, N, of 0.5, 1, 2, 5 and 10 using an imposed pressure of 6.0 GPa and a rotational speed for the lower anvil of 1 rpm.

Following HPT processing, discs in both the as-received condition and those processed by HPT were hot mounted in bakelite, ground with abrasive papers, and then a final polish was performed using a colloidal silica solution in order to produce a mirror-like surface. For the as-received sample, the polished surface was etched using the ASTM 163 etchant which consists of 30 mL of water, 30 mL of sulphuric acid (H_2SO_4), 30 mL of hydrofluoric acid (HF) and 3–5 drops of 30% hydroxide peroxide (H_2O_2) with an etching time of 20–40 s. The sample surface was then observed in dark field using an Olympus BX51 optical microscope in order to determine the initial grain structure. The grain size was measured by the linear intercept method using Image J software. The grain structure after HPT processing was examined by electron backscattered diffraction (EBSD) using a JSM6500F thermal field emission scanning electron microscope (SEM). The EBSD patterns were collected using a step size of 50 nm and a cleaning procedure was applied such that the total number of modified points was less than 10% of the total points measured. High-angle grain boundaries (HAGBs) were defined by the software as boundaries having misorientation differences between adjacent measuring points of more than 15° and low-angle grain boundaries (LAGBs) were defined as having misorientation differences of 2–15°. More than 300 grains were measured for estimating the average grain size.

The values of the Vickers microhardness, Hv, were measured on the polished surfaces and the distributions of the hardness values are presented by mapping the values of Hv over the total surface of each disc. In practise, the individual values of Hv were recorded following a rectilinear grid pattern with a separation of 0.3 mm between each consecutive point as described in an earlier report [30]. The hardness measurements were taken using an FM300 hardness tester equipped with a Vickers indenter with a load of 300 gf and a dwell time of 15 s. All of the individual values of Hv were used to construct colour-coded contour maps that provide a clear visual presentation of the distributions in hardness across the surface of each disc.

The X-ray diffraction (XRD) analyses were conducted over the HPT disc surfaces and the samples processed by HPT were evaluated using a Bruker D2 Phaser X-ray diffractometer equipped with a Cu target using Cu Kα ($\lambda = 0.15406$ nm) radiation. The X-ray samples were prepared from the HPT-processed discs by conventional metallographic grinding and polishing and then the discs were slightly etched for 5–10 s using the ASTM 163 etchant to remove the thin deformed surface layer. Scans were performed of θ–2θ from $2\theta = 30$–100° to record the XRD patterns and the crystallite sizes and microstrains were calculated based on the Rietveld method [31] using the Maud software to accomplish the profile fitting. The texture option was applied when using the Maud software and the crystal size and microstrain were used to estimate the values of the dislocation density.

To evaluate the effect of short-term annealing in the post-HPT condition, some samples were annealed for a short time of 15 min at either 873 or 973 K immediately after the HPT processing. These annealing temperatures were selected based on earlier reports presenting the Vickers hardness measurements as a function of annealing temperature for extruded tantalum and showing that the hardness begins to drop from 973 K and reaches a softening plateau at ~1173–1223 K [18,19]. To avoid recrystallisation which may cause strength loss and grain growth in HPT-processed samples, a very short annealing time was used at temperatures of 873 or 973 K.

Tensile specimens were cut from the as-received sample, the HPT-processed samples and the samples subjected to post-HPT annealing. Following earlier practise [32], and in order to avoid any microstructural inhomogeneities in the centres of the discs, two tensile specimens were prepared from each disc using electro-discharge machining with these specimens arranged symmetrically on either side of the disc centre. The miniature tensile specimens had gauge lengths and widths of 1 mm. These specimens were then tested in tension at room temperature using a Zwick 30 KN Proline testing machine operating at a constant rate of cross-head displacement with an initial strain rate of $1.0 \times 10^{-3}\ s^{-1}$.

3. Experimental results

3.1. Microstructure before and after HPT processing

In the initial as-received condition, the material had a uniform equiaxed microstructure with an average grain size of ~60 μm and an average hardness of ~87 Hv.

The microstructures developed during HPT processing are displayed in Fig. 1 where the centre and the edge areas are shown after 0.5 and 10 turns in the top and bottom rows and the central row shows the edge areas after 1 and 5 turns, respectively; the colours denote different grain misorientations as depicted in the unit triangle on the right, low-angle boundaries (2–15°) are shown in yellow and high-angle boundaries (> 15°) are in black. It is apparent from the upper row that the edge experiences very significant grain refinement after 0.5 turn with a measured grain size of ~0.2 μm in Fig. 1 (b) whereas in the centre area in Fig. 1(a) the structure remains coarse and similar to the as-received condition. As processing increases to 1 and 5 turns in Fig. 1(c) and (d), there is some minor additional grain refinement at the edges of the discs with average grain sizes of ~160 and ~140 nm, respectively. After 10 turns in Fig. 1(f) and (e), the