SEVERE PLASTIC DEFORMATION PROCESSING OF REFRACTORY METALS BY EQUAL CHANNEL ANGULAR EXTRUSION

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ABSTRACT

Research efforts to develop high density refractory metals, such as Ta and W, have to a certain extent been hindered by the inability to thermomechanically process the materials to achieve key properties necessary for adequate performance, namely consistent mechanical behavior. The work presented here demonstrates the potential for W, Ta, and other polycrystalline refractory metals to be processed via equal channel angular extrusion (ECAE) to have nanocrystalline microstructures and improved mechanical behavior. Microscopy of the ECAE processed W and Ta show ~300 nm grains in the worked state and enhanced mechanical behavior. Other processing-microstructure-property relationships correlating processing, microstructure, and microhardness are reported here.

1. INTRODUCTION

Ta and W have long been targeted as potential materials for ballistic penetration applications due to their high densities (16.7 g/m^3 and 19.3 g/m^3, respectively) (Magness et al., 1995; Pappu and Murr, 2002). Specifically, W for replacement of depleted uranium kinetic energy penetrators (KEP), and Ta for explosively formed penetrator (EFP) applications, but the research and development of these materials has been limited by the inability to achieve the needed consistent mechanical properties, mainly high strength and high ductility. The problem lies with the inability of conventional thermomechanical processing to achieve uniform and predictable microstructural characteristics such as hardness, grain size, grain morphology and texture in the as-processed refractory metal. In the typical as-processed state, pure refractory metals typically have large grains with non-uniform grain size distributions and extensive texture gradients. Conventional metal processing, such as rolling, swaging, and forging, introduce non-uniform deformation, and thus, non-uniform microstructural properties and mechanical behavior.

Equal channel angular extrusion (ECAE) is a method which could alleviate some of these issues by introducing large, uniform plastic strain into a metal workpiece (R.Z. Valiev et al., 2000). The material undergoes simple shear when passed through two intersecting channels of equal cross section (Fig. 1). Multiple passes can be performed due to the fact that the billet is nearly the same cross-section subsequent to extrusion, and the microstructure can be tailored by accounting for the orientation of the billet (i.e., rotation of the billet along the long axis), between consecutive extrusions.

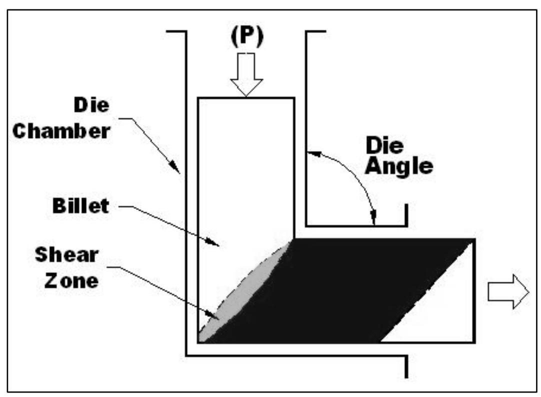

Fig. 1. Schematic of ECAE showing the passing of a billet through equal cross section intersecting channels.

Recent work has showcased one of the main driving forces to ECAE process pure metals and alloys: the ability to obtain a bulk nanocrystalline material. The large strain introduced by multiple extrusions allows grain sizes to be refined to the ultrafine grained (UFG) or nanoscale (NC) regimes. When the grains are so highly refined, high strength and ductility can be concurrently achieved.

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Recent work on NC W processed by ECAE and hot rolling to an average grain size of ~250 nm show behaviors quite different from coarse grained (CG) W. The NC W demonstrates a higher flow stress, enhanced ductility, reduced strain hardening capacity, and reduced strain rate sensitivity as compared to CG W (Wei et al., 2006).

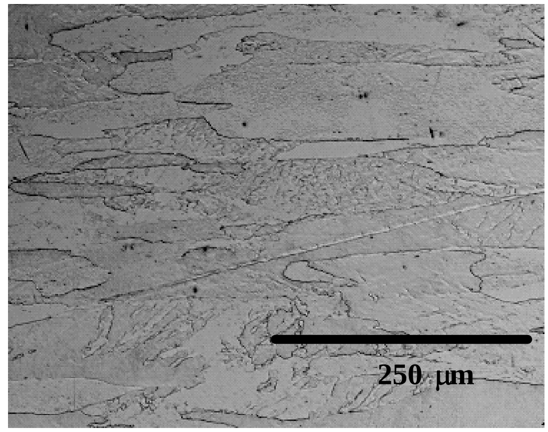
The objectives of this work are to investigate the potential of ECAE processing for grain refinement of CG Ta and W to the UFG/NC scales for improved mechanical behavior. Particularly, the ability of ECAE to break-up the initially large microstructures to the UFG/NC scale, and the effects of these resultant microstructures on the mechanical behavior are investigated.

2. MATERIALS AND METHODS

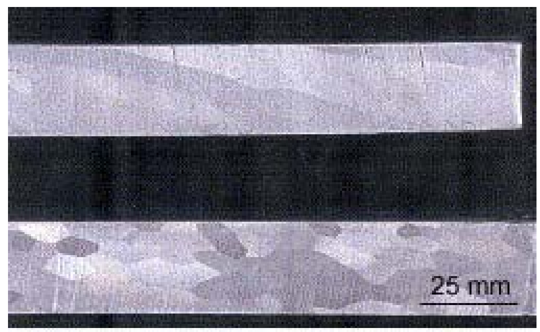
The starting materials used in this study were 99.95% purity W, supplied by Alfa Aesar, Inc. (Ward Hill, NY), and 99.98% purity Ta, supplied by H.C. Starck, Inc. (Newton, MA). The W consisted of elongated grains 50-300 μm in length (Fig 2a) and the Ta consisted of large 5-10 mm diameter elongated grains (Fig. 2b). In both cases, billets were nominally 25 mm x 25 mm x 150 mm in size. ECAE processing was done in lubricated 90° tooling with a channel intersection having sharp inlet and outlet corners under pseudo-isothermal conditions (room temperate for the Ta, and at 800°C for the W). The Ta extrusions were performed at a punch speed of 5 mm/s, and the W was extruded at 25 mm/s. Multipass extrusions were done up to four passes using route A (no rotation about the billet axis between extrusions), route C (180° rotation between extrusions), route B (90° rotation between extrusions), and route E (180° rotation between passes 1 and 2, 3 and 4, and a 90° rotation between passes 2 and 3). Complete extrusion conditions are summarized in Table I.

Table I
Processing conditions for Ta and W

Material	Route	Rate	Temperature
W	1A	25 mm/s	800°C
W	2B	25 mm/s	800°C
W	4E	25 mm/s	800°C
Ta	1A	5 mm/s	23°C
Ta	2C	5 mm/s	23°C
Ta	4E	5 mm/s	23°C



(a)



(b)

Fig. 2. Light micrographs of the as-received starting microstructures for (a) CG W and (b) CG Ta.

Samples for metallography were sectioned from the extruded bars using wire-electro discharge machining, and were prepared using standard polishing and etching procedures. Vickers microhardness (HV) measurements were taken according to ASTM Standard E384 on the flow plane (billet side plane) using a Buehler Micromet (300gmf / 13s) in the case of the Ta, and a Wilson Tukon (500gmf / 20s) in the case of the W. Scanning electron microscopy (SEM) was used to obtain backscattered electron images of the as-worked UFG/NC microstructures (LEO 1530 SEM for the Ta, and Hitachi S4700 FE for the W), and the linear intercept method (ASTM Standard E112) was used to measure the average grain size.

3. RESULTS AND DISCUSSION

Fig. 3 shows the hardening curves for both Ta and W as a function of level of strain (number of extrusions). Firstly, it is observed that the Ta and W hardening curves are very similar with the Ta being ~400 HV softer at every point during the extrusions. The ductile-to-brittle-transition temperature (DBTT) of Ta is well below room temperature (approx. -350°C), whereas, W has a DBTT well above room temperature (100°C - 400°C). Both materials show maximum hardness increases occurs after two passes, with minimal increases on subsequent passes. These results agree with prior work on W processed at 1000°C and 1200°C (Mathaudhu et al., 2008), and prior work with CG Ta (Mathaudhu and Hartwig, 2006).

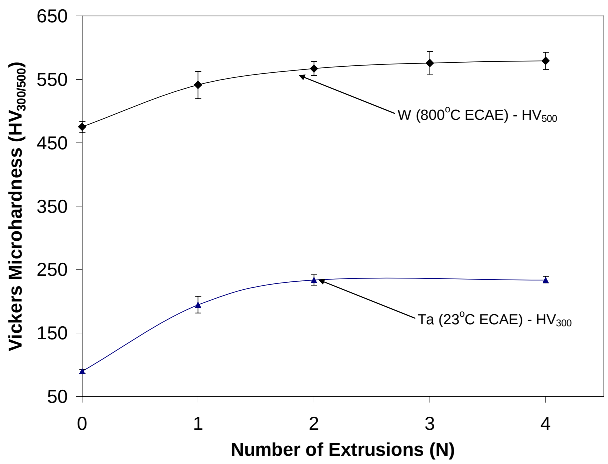
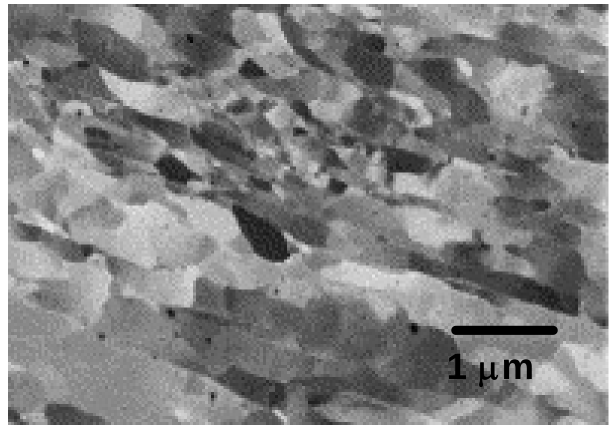
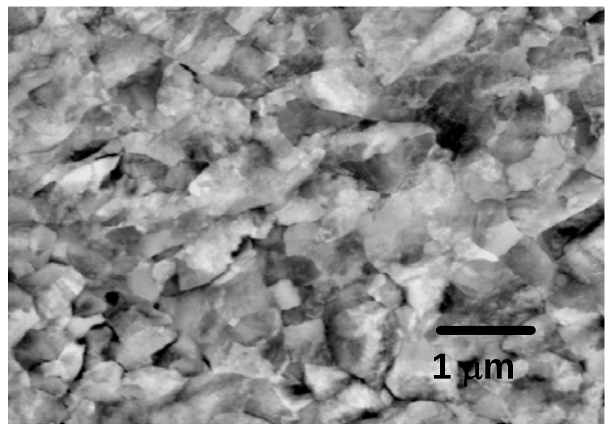


Fig. 3. Vickers microhardness as a function of the number of extrusion passes for Ta and W. The error bars indicate the standard deviation of the measurements.

Previous ECAE work has shown that route E results in equiaxed, fine-grained microstructure in Ta and Nb (Mathaudhu et al., 2005). Backscattered FESEM images of W and Ta processed four passes by route E are given in Fig. 4. For the Ta processed at 23°C, the average grain size was measured to be 373 nm. The average grain size for the W processed at 800°C was measured to be 300 nm. The Ta has a larger variation in grain size distribution, and some large elongated subgrains, whereas the W appears to be more homogeneous and equiaxed. This result is likely due to a combination of a very large grain size in the starting Ta and the predominant casting textures within each large grain. Even with the large strain deformation that ECAE provides after four passes, it is difficult to completely eradicate the effects of the starting large grains. More extrusions, and/or intermediate heat treatments are necessary for better refinement (Mathaudhu et al., 2007). That being said, the level to which the grains are refined in the Ta is far greater than that of the W. Consider that the refinement of the Ta is from –mm to –nm (six orders of magnitude) compared to -μm to –nm (three to four orders of magnitude) for the W for an equivalent number of extrusions.



(a)



(b)

Fig. 4. FESEM backscattered electron images of as-worked UFG/NC microstructures in (a) Ta processed by route 4E at 23°C and (b) W processed with route 4E at 800°C.

The microhardness of a material can be correlated with the yield strength. Therefore a Hall-Petch like relationship has been developed which relates the material microhardness to its grain size by the equation $HV = HV_0 + kd^{-1/2}$, where HV is the measured microhardness, and d is the grain size. HV_0 is related to the friction stress for plastic flow within polycrystalline grains and accounts for lattice friction; constant k is related to the microstructural stress intensity and accounts for dislocation locking effects. Fig. 5 shows the measured Vickers microhardness plotted as a function of $d^{-1/2}$ for both Ta and W. For purposes of extending the results over a useful range of grain sizes, data from as-worked and recrystallized Ta and W given in prior work by the authors (Mathaudhu et al., 2006, 2008) is also plotted.

Observe that k for Ta is half that of the W (69 vs. 138). This means that dislocation pinning in Ta is not as strong as in the W. This may be accounted for by the larger unit cell in Ta ($V_{Ta} = 0.036\ nm^3$) compared to other bcc metals such as W ($V_W = 0.032\ nm^3$). The greater volume of space in the lattice would more easily accommodate interstitial solutes with minimal lattice distortion, and thus result in a much weaker dislocation locking potential.

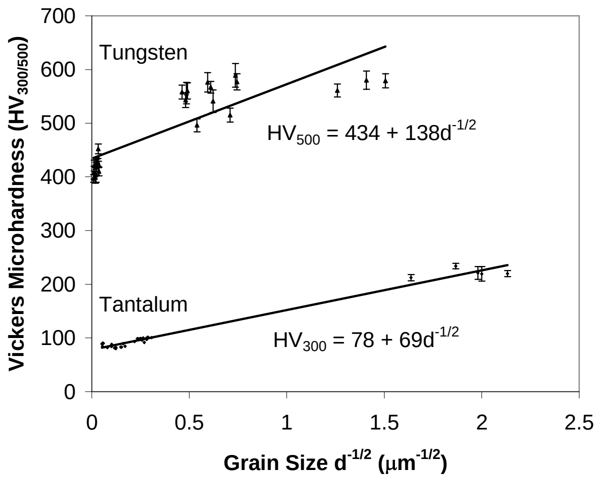


Fig. 5. Hall-Petch plot showing the variation in Vickers microhardness vs. $d^{-1/2}$ for as-worked and recrystallized Ta and W thermomechanically processed by ECAE. The vertical error bars give the standard deviation of the measurements.

Broadly, these equations allow prediction of the mechanical properties achievable in polycrystalline materials when grains are refined to the nanoscale by ECAE processing. Using conversion factors (9.807 x HV = HV in MPa units, and σ_y = HV(MPa)/3), equations (1) and (2) can be formulated which approximate the yield stress as a function of the grain size:

$$\sigma_y = 255 + 226d^{-1/2} \quad : \text{for Ta} \tag{1}$$

$$\sigma_y = 1419 + 451d^{-1/2} \quad : \text{for W} \tag{2}$$

4. CONCLUSIONS

Unique properties demonstrated by NC metals, such as concurrent high strength and ductility, offer unprecedented opportunities for aiding the warfighter. As such, processing methods to obtain bulk nanostructured materials are of interest. The grain refinement potential of ECAE processing for bcc refractory metals, and the subsequent mechanical behavior was examined. Coarse grained Ta and W were processed up to four passes to refine the grain structure. Ultrafine grain structures less than 400 nm were achieved in both materials. Hall-Petch relationships were determined to predict microhardness and yield strength as a function of grain size in both the Ta and W. This high level of grain refinement forecasts the potential of ECAE to effectively break up and refine microstructures, and thus achieve high strengths, in other CG polycrystalline materials of interest for defense applications as well.

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