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Microstructures and recrystallization behavior of severely hot-deformed tungsten

S.N. Mathaudhu[a,b,*], A.J. deRosset[a,b], K.T. Hartwig[c], L.J. Kecskes[a]

[a] U.S. Army Research Laboratory, Aberdeen Proving Ground, MD 21005-5069, United States
[b] Oak Ridge Institute for Science and Education/Oak Ridge Associated Universities (ORISE/ORAU - Maryland),
P.O. Box 53, Aberdeen Proving Ground, MD 21010-0053, United States
[c] Department of Mechanical Engineering, Texas A&M University,
TX 77843-3123, United States

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ABSTRACT

When coarse-grained (CG) tungsten (W) is heavily worked by equal-channel angular extrusion (ECAE), the grain size is reduced to the ultrafine-grained/nanocrystalline regimes (UFG/NC) and the strength and ductility increase. Because of the brittle nature of CG W, the material must be hot-extruded, and, if the temperatures are near the recrystallization temperature (T_{rc}), gains in properties may not be maximized. In this study, the recrystallization behavior of ECAE-processed CG W is examined as a function of the imparted strain (i.e., number of extrusions) and the hot-working extrusion temperature. Up to four ECAE passes were performed in tooling with a 90° channel intersection, and at temperatures of 1000 °C or 1200 °C. Subsequent 60 min annealing of the worked material to 1600 °C allowed for the determination of T_{rc}. Vickers microhardness measurements and scanning electron microscopy, were used to characterize the microstructures in the as-worked and recrystallized states. The ECAE-processed W shows increased microstructural break-up and refinement with increasing strain and decreasing hot-working temperature in the fully worked state. T_{rc} was determined to be ∼1400 °C, which is nearly independent of the number of extrusions and the working temperature. These results show that if ECAE is accomplished below 1400 °C (i.e., at 1000 °C or lower) the attractive properties of the UFG/NC-worked W may be retained. Specifically, below 1000 °C, with increasing strain imparted to the material, high hardness values with a concomitant grain size refinement (∼350 nm) could be expected.

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1. Introduction

Recent studies on W have shown that ultrafine-grained (UFG) or nanocrystalline (NC) microstructures produced by severe plastic deformation (SPD) processing have properties quite different from their coarse-grained (CG) counterparts [1–4]. UFG/NC W, when compared with conventional CG W, shows a higher flow stress, enhanced ductility, reduced strain hardening capacity, and reduced strain-rate sensitivity. To refine the grains to the UFG/NC regimes, SPD processing, specifically, equal-channel angular extrusion (ECAE), was reported to be done at temperatures "around" 1000 °C in a 120° ECAE die to avoid cracking the CG W, and subsequently rolled at temperatures between 600 °C and 800 °C. These elevated processing temperatures are necessary because CG W is well known to have poor workability, and frequently experiences cracking and breakage [5,6].

However, such high working temperatures approach the temperature range within which W is reported to recrystallize (900–1400 °C) [7]. The recrystallization temperature (T_{rc}) is influenced by factors such as starting grain size, impurity content, level of induced plastic strain, and crystallographic texture. As such, care must be taken to process the CG W at the proper temperature: if extrusions are performed at temperatures too near or above the T_{rc}, it may have excellent workability, but conversely, the as-formed UFG/NC microstructure will quickly coarsen and revert to CG microstructure, resulting in a loss of anticipated properties (i.e., high strength and high-strain-rate ductility). The objectives of this work are to explore the recrystallization behavior and grain refinement potential of CG W processed by hot ECAE. In particular, the effects of total plastic strain (i.e., the number of extrusion passes) on the recrystallization temperature and the as-worked grain size are investigated. The results obtained will help establish a thermomechanical processing window to realize the grain refinement

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* Corresponding author at: U.S. Army Research Laboratory, Aberdeen Proving Ground, MD 21005-5069, United States. Tel.: +1 410 306 0813; fax: +1 410 306 0759.
E-mail address: Suveen.mathaudhu@arl.army.mil (S.N. Mathaudhu).

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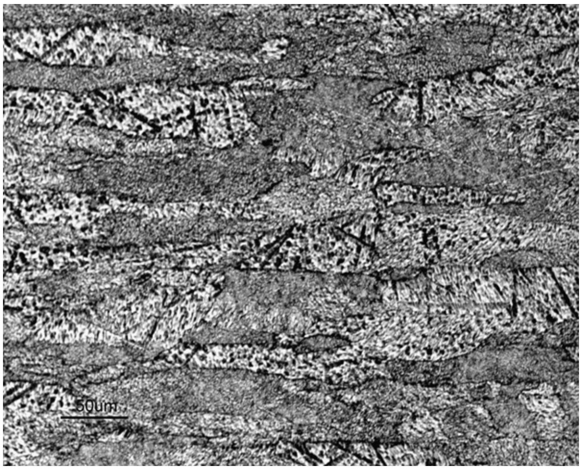

Fig. 1. Micrograph of the as-received 99.95%-pure CG W, showing elongated grains (50–300 μm in length); 50-μm scale bar is located in the lower left-hand corner.

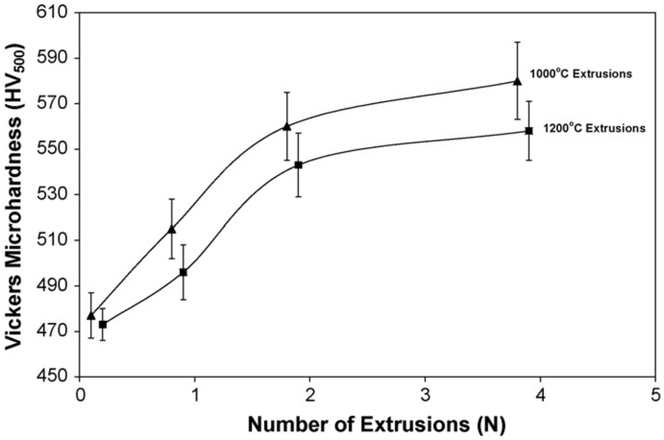

Fig. 2. Vickers microhardness as a function of the number of ECAE extrusions with hot-working at 1000 °C and 1200 °C. Data points are slightly offset so the error bars do not overlap. The error bars indicate the standard deviation of the measurements.

potential of ECAE and thus maximize the desired properties of UFG/NC W.

2. Experimental methods

Pure (99.95%) W rods, produced via powder sintering followed by hot extrusion, with nominal 13 mm diameter × 100 mm length were purchased from Alfa Aesar, Inc., Ward Hill, NY. As shown in Fig. 1, the as-received CG microstructure is composed of elongated grains with widths of 50–300 μm and aspect ratios of about three to four. These rods were processed up to four extrusions in ECAE tooling with a 90° channel intersection angle. The billets were heated for 1 h at temperatures of either 1000 °C or 1200 °C, and quickly inserted into the isothermal tooling, heated to 300 °C. The punch speed during the extrusions was 25 mm/s. Multipass extrusions involved route 2B (90° rotations about the billet axis) and route 4E (180° rotation between passes 1 and 2, 3 and 4, with a 90° rotation between passes 2 and 3).

Wire electro-discharge machine cut samples were annealed for 1 h at temperatures between 400 °C and 1600 °C in a Lindberg-Blue tube furnace under argon cover gas to prevent oxidation. Standard polishing techniques and etching with Murakami's reagent were used to prepare the flow plane of the extruded W samples for metallographic evaluation. Polished samples were then tested for hardness on the flow plane (billet side plane) using a Wilson Tukon Vickers Microhardness tester. Each sample was indented 10 times under a load of 500 gmf for 20 s. A field-emission gun scanning electron microscope (Hitachi S4700 FE) operated at 20 kV was used to obtain grain structures via backscattered electron imaging for the as-worked material.

3. Results and discussion

Microhardness can be quantitatively correlated with yield strength for a polycrystalline metal. Microhardness can also be correlated to a grain size, e.g., a higher microhardness value indicates a smaller grain size (higher yield strength). In turn, a high

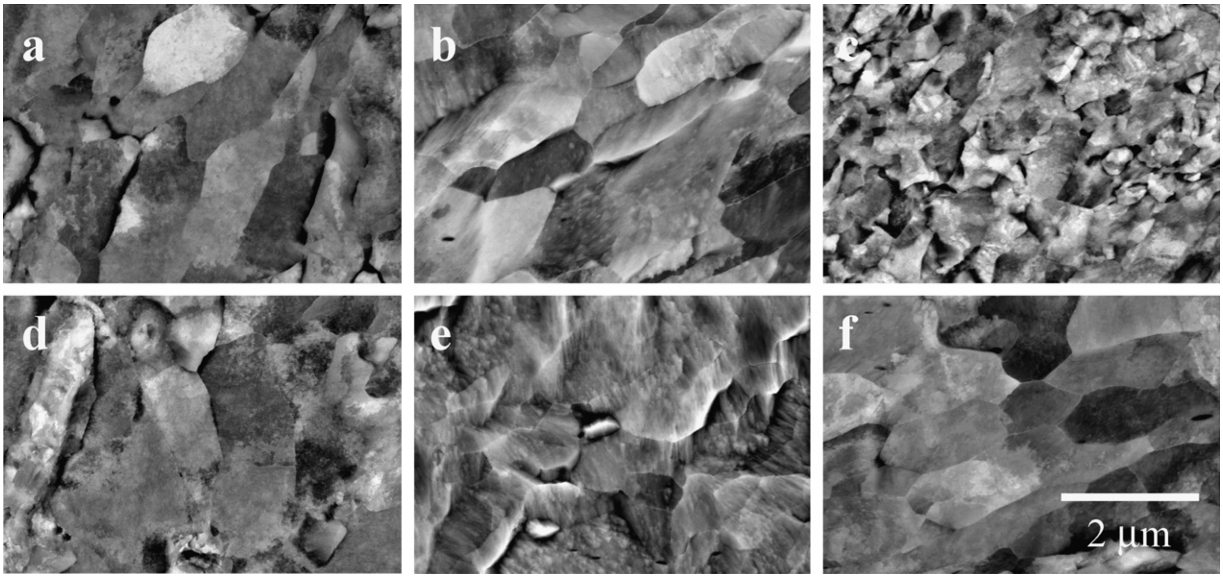

Fig. 3. Backscattered electron micrographs of the as-worked, ECAE-processed W UFG/NC microstructures. The average grain size is given in the parenthesis: (a) route 1A-1000 °C (0.71 μm), (b) route 2B-1000 °C (1.02 μm), (c) route 4E-1000 °C (0.36 μm), (d) route 1A-1200 °C (0.93 μm), (e) route 2B-1200 °C (1.04 μm) and (f) route 4E-1200 °C (1.08 μm).

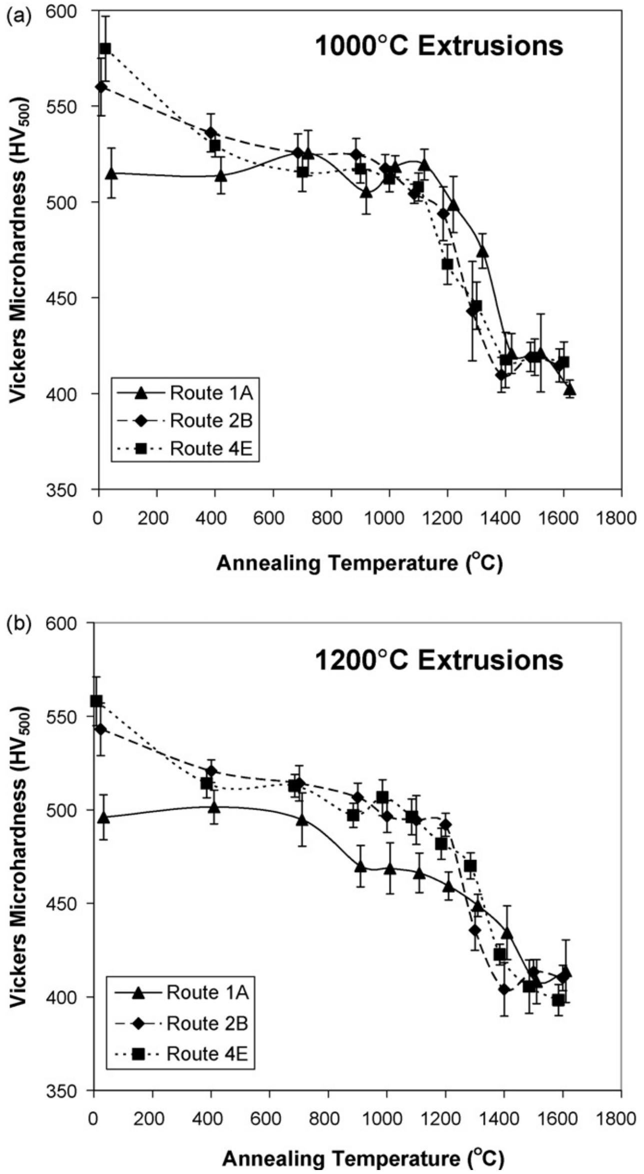

Fig. 4. Recrystallization curves for the ECAE-processed W extruded at: (a) 1000 °C and (b) 1200 °C. The curves demonstrate that the worked W recrystallizes near 1400 °C, independent of the processing temperature. Note, the one-pass material shows an erratic recrystallization behavior, due to inhomogeneous deformation. At two to four passes, recovery and recrystallization events occur similarly. The scale bars indicate the standard deviation of the microhardness measurements.

yield strength corresponds to a fine grain size, as determined by the conventional Hall–Petch relationship [8,9]. Fig. 2 displays the Vickers microhardness values of the starting material as a function of both the number of extrusions and processing temperature. Firstly, note that hardness values for the extrusions performed at 1000 °C are greater than those at 1200 °C. This is to be expected, as the higher temperature likely approaches T_{rc}. In both cases, the hardness reaches a near maximum after two passes, with little further increase from two to four passes. This result indicates that the increased level of strain induced in the W by the higher number of extrusions results in an increased grain refinement. The relative increases in microhardness from two to four passes for the 1000 °C working temperature is higher (~20 HV_{500}) than that of the 1200 °C working temperature (~15 HV_{500}), indicating that the lower working temperature provides a higher level of grain refinement.

These trends predicted by the microhardness are empirically verified by observation of the as-worked UFG/NC microstructures (Fig. 3). The average grains sizes are reported in Fig. 3 as well. It should be noted that the grain sizes reported for the single pass material, in both the 1000 °C and 1200 °C extrusions, have a larger distribution of grain sizes, have fewer internal dislocations, and boundaries that are not as sharp as those produced after subsequent passes. The contrast and demarcations between the grains give a qualitative indication of the misorientation angle at the grain boundary. With this in mind, it can be seen that the 1000 °C extrusions appear to have much sharper grain boundaries, in addition to finer grains than the 1200 °C extrusions (0.36 μm vs. 1.08 μm, respectively), for the same number of passes. The higher number of high angle grain boundaries in combination with a finer grain structure explains the higher hardness of the 1000 °C extrusions. It is also seen that the average grain size decreases with an increasing number of extrusions with very fine, equiaxed, submicrometer structured grains forming after four extrusions at 1000 °C (0.36 μm average grain size). The microstructure developed after four extrusions at 1200 °C, in contrast, is just beginning to show sharp, high angle boundaries similar to what was obtained after two passes at 1000 °C, and correspondingly, the hardness values are similar as well (560 ± 15 HV_{500} for route 2B at 1000 °C vs. 558 ± 13 HV_{500} for route 4E at 1200 °C).

The inability of the extrusions performed at 1200 °C to adequately fragment the starting microstructure to the same degree as the 1000 °C extrusions can be related to the increased dynamic dislocation motion that occurs at the higher processing temperature, and structural relaxation during the 1 h heat treatment performed prior to extrusions. To quantify the effects of the pre-extrusion annealing and high temperatures extrusion process, necessitated the determination of recrystallization curves as a function of both extrusion temperature, and number of extrusion. These recrystallization curves are presented in Fig. 4. The T_{rc} is identified by a sharp elbow at the bottom (soft) end of the curve. Observe that in both cases, the material extruded only a single pass shows a noticeably different behavior than the two or four pass material. The microhardness values are significantly lower to start, and in the 1200 °C case, the material appears to recrystallize at a higher temperature (~1500 °C). Conversely, the curves for the two and four pass extrusions nearly overlap for both hot-working temperatures and show clear recrystallization elbows at ~1400 °C.

From this result, it can be inferred that recrystallization occurs similarly for materials above some particular level of strain (in this case, after two extrusions) or that the dependence of the T_{rc} on the level of deformation strain is weak, but this is contrary to prior results which suggest that the T_{rc} drops with an increasing number of extrusions [10]. The discrepancy could be explained by the fact that the reported materials in Ref. [8] were processed at room temperature, where dislocations and other microstructural features are stable. In contrast, the ECAE-processed W material is hot-worked. The kinetics of grain formation and growth during hot-working prevents significant introduction of dislocations and/or microstructural refinement from occurring with increased number of extrusions. The result of the hot ECAE is decreasing, but very similar grain sizes (~1 μm down to 350 nm). Similar grains sizes would results from a homogeneous distribution of grain nucleation points in the as-worked material, and if all these nucleation points are activated equally, recrystallization would occur similarly, as observed. In any case, T_{rc} for the ECAE-processed W is shown to be at the high end (~1400 °C) of the predicted range for W. More importantly, this is well above the processing temperatures of 1000 °C or 1200 °C used to obtain UFG/NC microstructures.

4. Conclusions

The recrystallization behavior of ECAE-processed W, as a function of the number of passes and hot-working temperature, was determined by an examination of the as-worked microstructure and microhardness in as-worked and annealed materials. The results point to the following conclusions:

(i) The as-worked microstructures show increasing grain refinement and sharper grain boundaries with increasing extrusion passes (from one to four), and with decreasing hot-working temperatures (1200 °C and 1000 °C). The application of four passes at 1000 °C results in an equiaxed, ∼350 nm grain structure.

(ii) The Vickers microhardness increases with increasing strain (number of extrusions) and decreasing hot-working temperature. This observation verifies that finer UFG/NC structures are obtained at higher passes and lower working temperatures. Properties reach near maximum levels after two passes.

(iii) The recrystallization temperature (∼1400 °C), is independent of the hot-working temperature except for the single pass case extruded at 1200 °C, which shows recrystallization at ∼1500 °C. This high $T_{\rm rc}$ indicates that ECAE processing can be performed at 1000 °C where effects (UFG/NC microstructure) are realized.

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