Microstructural Refinement of Bulk Nb and Ta by Severe Plastic Deformation for Composite Superconductor Applications

S.N. Mathaudhu, R.E. Barber and K.T. Hartwig*

Department of Mechanical Engineering, Texas A&M University, College Station, TX 77843-3123

Abstract

Fine grained and homogeneous recrystallized microstructures in precursor Nb and Ta are necessary to avoid filament sausaging and layer fracturing during the production of Nb_3Sn superconductor wires. In this study, the effectiveness of multi-pass equal channel angular extrusion (ECAE) to break up large as-cast microstructures (>5mm diameter columnar grains) in vacuum arc remelted Ta and electron beam remelted Nb is discussed. Extrusions were performed at room temperature in 90° tooling via ECAE with routes C, B_c, and E. Results show significant refinement to average grain sizes of less than 30μm after four extrusions. Four passes via routes E and B_c showed the best potential for refinement due to deformation of the as-cast microstructure on orthogonal planes.

Introduction

Composite multifilamentary Nb_3Sn superconductors are often fabricated with pure Nb and pure Ta precursor components. The Nb begins as a small diameter rod (3-6 mm) surrounded by pure Cu, and ends up after wire drawing and a reaction heat treatment as a small diameter (4 μm diameter) filament of Nb_3Sn. The Ta starts out as a thin sheet (0.38 mm thick) and is reduced to a 3-5μm diffusion barrier layer during wire drawing. Advanced superconducting magnets for high energy physics accelerators and plasma containment for fusion will require low temperature superconductors with finer filaments (submicron diameter) and thinner Ta diffusion barriers (1-3μm thickness) [1,2]. It is extremely difficult to fabricate such conductors without encountering Nb filament sausaging (1) and Ta layer fracture. Filament sausaging leads to a lower critical current density in the composite superconductor, and Ta layer fracture leads to wire breakage and shorter piece lengths during wire manufacture.

The problems of Nb filament sausaging and Ta diffusion barrier fracture are most likely due to 1) a large starting grain size, 2) a non-uniform starting grain size, and/or 3) significant texture variations in the starting microstructure. These causes arise because of the extremely large grained and highly textured microstructure of the original cast metal, and the subsequent ineffectiveness of conventional metal working methods (forging and rolling with intermediate heat treatments) to refine and homogenize the microstructure [3]. Strong texture gradients and banded recrystallized microstructures are common in Nb and Ta wire and sheet products.
Preliminary work on the microstructural breakdown of large grained cast Nb and Ta using severe plastic deformation (SPD) methods has been quite successful [4,5]. In

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* Corresponding Author. thartwig@mengr.tamu.edu

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the case of Nb, bulk material strained to 4.6 by ECAE at room temperature gave a recrystallized grain size of 25 microns but the microstructure contained bands of larger grain size [5]. For vacuum arc remelted (VAR) Ta, an average recrystallized grain size of 15 microns was achieved after an ECAE strain of 4.6 [4]. The as-worked Ta (strained to 4.6) had a microstructure composed of 100-350 nm wide by 200-800 nm long dislocation cells and subgrains containing a high density of dislocations. Comparison of the ECAE processed Nb microstructure with similar fully annealed commercial material shows that the average grain size of the ECAE processed and recrystallized Nb appears to be as fine as, and perhaps finer than, the best commercial material.

The motivation behind the work reported here is the belief that SPD processing of bulk Nb and Ta via ECAE will give a finer and more uniform microstructure than can be produced easily by conventional metal working processes. This improvement could enable the fabrication of better performing and more cost effective low temperature composite superconductors. Supporting evidence for the benefits of processing bulk metal via ECAE to achieve better microstructural refinement already exists in the sputtering target field [6] and encourages further work on refinement of cast microstructures.

Experimental Procedures

The materials used for this research were commercially pure (99.7%) electron beam remelted (EB) Nb and 99.98% pure vacuum arc remelted (VAR) Ta ingots. Both materials consisted of ingots with initial columnar grain structures and large as-cast grains (>5mm diameter for the Ta and >20mm for the Nb). Billets with nominal dimensions of 25mm x 25mm x 150 mm were cut from cast ingots with their long axes parallel to the ingot long axis.

Extrusions were performed at room temperature in sliding wall 90° tooling with a punch speed of 5.1 mm/sec. Multipass processing following routes C, B_c and E was used. Routes C and B_c are well researched, while route E is a hybrid route that involves extrusions via route 2C followed by a 90° rotation, then extrusion via route 2C again so as to deform the microstructure on three orthogonal planes. Post-extrusion anneals (60 min for Nb, 90 min for Ta) were performed under a vacuum of at least 10^{-6} torr. Vickers microhardness measurements were taken with a 300g load for 13 s on a Buehler Micromet. Microstructural characterization was done using a Leica MEF4M stereo wide-field metallograph. Grain sizes were determined using the Image Processing Toolkit (IPTK) plug-in for Adobe Photoshop.

Results and Discussion

Fig. 1 shows the as-worked Vickers microhardness for both Ta and Nb to four passes. It is clear in both cases that significant hardening occurs after one extrusion with continued increases being less for each subsequent extrusion. In the processed Ta, it appears that hardness saturation occurs at four extrusions, which corresponds to an equivalent strain of about five. Different regions of the starting billets will experience different amounts of plastic strain due to their different crystallographic orientations to the ECAE shear plane. Whether or not grains have favorable slip systems will influence how much a localized region will deform [3]. This results in some regions of the billet being severely deformed, accumulating a higher density of dislocations, and work-hardening more than others. The decreasing standard deviation of hardness with increasing number of extrusions indicates that, with an increasing number of extrusions, additional slip systems are activated, thereby homogenizing the strain within the grains composing the billet.

The variations between the initial texture/orientation of the few large grains composing the original billets probably accounts for the differences seen in the Vickers microhardness of the four-pass materials (Ta: 4C and 4E, Nb: 4C, 4E and 4B$_c$).

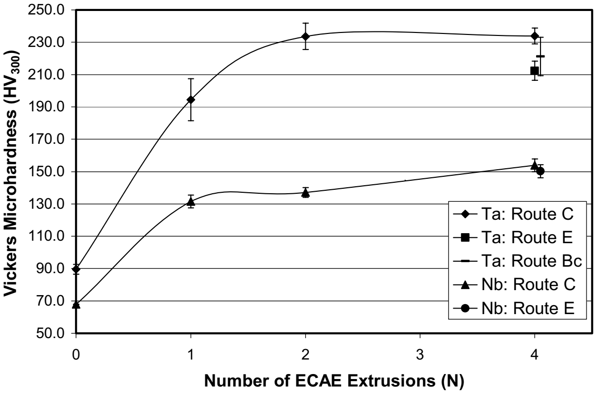
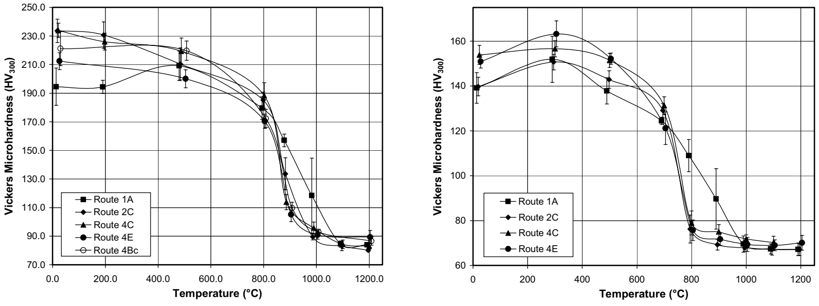


Figure 1. Vickers microhardness measurements as a function of number of extrusions for EB Nb and Ta subjected to multi-pass ECAE at room temperature. Error bars are measurement standard deviation.



(a) (b)

Figure 2. Recrystallization curves (HV$_{300}$ vs. Temperature) for a) VAR Ta and b) EB Nb ECAE processed at room temperature. Error bars are measurement standard deviation.

The recrystallization temperatures (See Fig. 2) are near 1100ºC for the Ta processed by routes 1A and 2C, and the same for Nb processed by route 1C. The rest of the routes recrystallized completely near 1000ºC. Observe that the materials processed by route 1A have large variations in strain even near the recrystallization temperatures, as demonstrated by the large standard deviation in the localized microhardness values. These materials have experienced non-uniform strain and therefore do not recrystallize uniformly, which results in significant grain growth in some regions and higher temperatures for full recrystallization. The maximums occurring on the recrystallization curves near 300ºC for Nb are characteristic of either polygonization or Cottrell atmospheres [7].

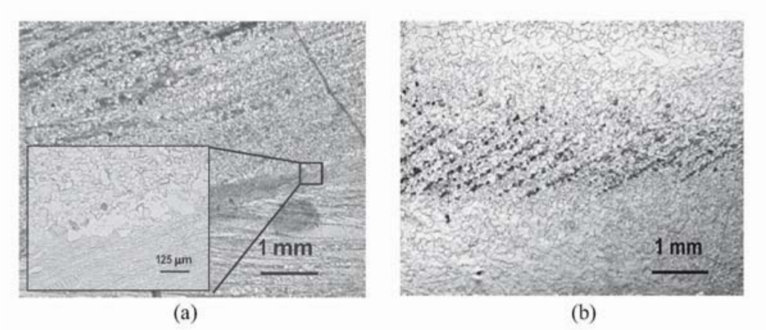
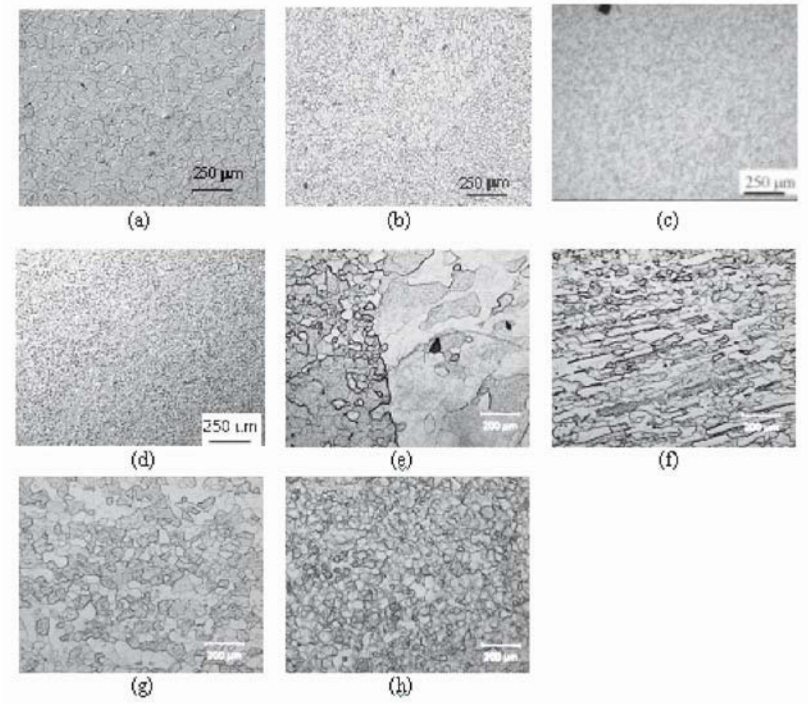


Figure 3. Flow plane optical micrographs of heat treated Ta processed via route 1A at a) 990ºC (55% recrystallized) and b) 1100ºC showing banded and non-uniform microstructure after only one extrusion.

Fig. 3 gives verification of the observations concerning the material processed via route 1A. Fig. 3a shows partial recrystallization occurs due to the non-uniform strain. Full recrystallization has occurred at 1100ºC (Fig. 3b), but banding and grain growth can result in a non-homogeneous distribution of grains in the sample. With an increasing number of extrusions, the average grain size decreases and microstructural homogeneity increases (Fig. 4). It also appears that routes B_c and E result in a finer grain size than route C for an equivalent amount of strain.

As expected, the average grain size decreases with increasing strain (Fig. 5), with microstructural uniformity increasing as well. For both the Ta and Nb, after two extrusions via route C, there are regions that contain the minimum average recrystallized grain size attainable with additional extrusions. These must occur in regions where the initial grain structure is oriented with the shear plane for easy break-up and deformation. The effectiveness of using routes E and B_c over route C involve rotating prior grain regions by 90° that might not have been oriented well to the shear plane, thereby offering other orientations that favor microstructural deformation. It appears that using routes which incorporate billet deformation on three orthogonal planes results in better breakup of large cast microstructures into more fine-grained, equiaxed and homogeneous recrystallized microstructures.



(a) (b) (c)

(d) (e) (f)

(g) (h)

Figure 4. Flow plane optical micrographs of fully recrystallized ECAE processed material: a) Ta: 2C and annealed at 1100ºC, b) Ta: 4C and annealed at 1000ºC, c) Ta: $4B_c$ and annealed at 1000ºC, d) Ta: 4E and annealed at 1000ºC, e) Nb: 1A and annealed at 1100ºC, f) Nb: 2C and annealed at 1000ºC, g) Nb: 4C and annealed at 1000ºC and h) Nb: 4E and annealed at 1000ºC.

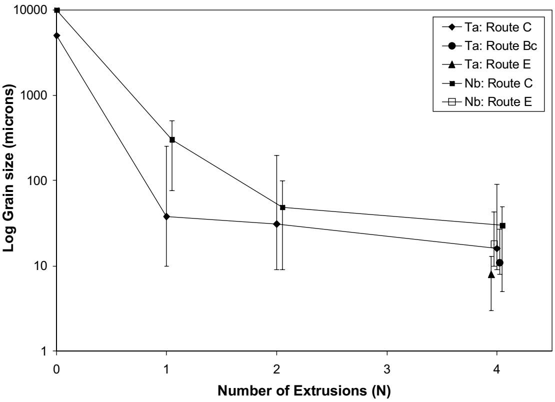


Figure 5. Variation in the fully recrystallized grain size as a function of extrusion route and number of extrusion passes for Ta and Nb processed by ECAE at room temperature.

Conclusions

The effectiveness of ECAE processing for the purpose of breaking up, grain refining and homogenizing VAR Ta and EB Nb has been demonstrated. The results of the study indicate that:

1. ECAE is effective in breaking up as-cast metals allowing the production of micron scale recrystallized microstructures.
2. The small grains observed in some regions of two pass ECAE processed and recrystallized material are an indication of the smallest grain size attainable with further increases in number of extrusions.
3. The intersecting shear planes utilized by routes E and B_c cause multiple slip planes to be activated, thereby increasing the uniformity of deformation and the homogeneity of the recrystallized microstructures.

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