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GRAIN REFINEMENT OF ELECTRON-BEAM-MELTED CRYSTAL BAR ZIRCONIUM BY EQUAL CHANNEL ANGULAR EXTRUSION

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Abstract

Electron-beam (EB) melted crystal bar zirconium has a very low oxygen content (approximately 100 ppm by weight), but a coarse grain structure that needs to be refined to allow successful part fabrication by conventional methods. Cylindrical rods were sectioned from an EB-melted billet by electrodischarge machining, and enclosed in seamless nickel tubing by electron beam welding plugs in each end to make a final billet 9.5 mm in diameter by 50 mm in length. The billet was deformed by equal channel angular extrusion (ECAE) at room temperature in 120° tooling. After extrusion for 4 passes via route B_c the material was annealed in vacuum for 1 h at 550°C. Because of the low oxygen content, the Zr could be successfully deformed by ECAE to high strains at room temperature. The as-cast grains were extensively subdivided by the processing. A fine uniform grain size of approximately 10 microns was achieved after the anneal, with a weak texture of only 2-3 times random.

Introduction

Zirconium is a convenient material to challenge models for plasticity in anisotropic materials. Because of its hexagonal close-packed structure, slip in the basal and non-basal planes will operate at different critical resolved shear stresses. In addition, twinning can play a significant role in deformation, depending on factors such as alloy impurity content, deformation temperature, and strain rate. Thus, the deformation of Zr has been extensively studied (1-4). One of the critical factors is the oxygen content, as O is a potent strengthener of Zr (5,6). Special processing is needed to reduce the O_2 content. Electron-beam (EB) melting is one method of controlling the O_2 content, resulting in typical levels of a few hundred parts per million, as compared to commercial material with over 1000 ppm.

Electron-beam melting of crystal bar zirconium results in high-purity material, but with a very coarse grain size. Extensive thermomechanical processing is needed to refine the grain size to give improved mechanical properties. This study investigated the possibility that an alternate processing method, equal channel angular extrusion (ECAE), could be used to refine the grain size, rather than conventional upsetting and cold rolling.

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Experimental Procedure

Zr billets were sectioned from a remnant of an EB-melted crystal bar Zr billet that was produced in the mid 1980's. The billets were then machined to a diameter of 7.0 mm (0.276 in.) and a length of 35 mm (1.38 in.). Because of concerns about the possible galling of the Zr on the tooling material, the Zr billets were inserted in sleeves made of annealed commercial-purity Ni 200 seamless tubing, 9.5 mm OD by 7.0 mm ID by 50 mm long (0.375 and 0.277 by 1.969 in.). Ni 200 plugs, 7 mm diameter by 7.5 mm long (0.276 by 0.295 in.) were inserted at each end of the sleeve, and these plugs were EB-welded to the sleeve.

The 120°-angle ECAE die was fabricated from H13 tool steel. The inlet channel was 9.53 mm (0.375 in.) in diameter, the outer corner radius at the intersection of the inlet and outlet channels was 4.78 mm (0.188 in.), and the outlet channel was slightly smaller in diameter (9.35 mm [0.368 in.]). The outlet channel had a land that extended 5.1 mm (0.20 in.) from the inner corner; the outer channel then expanded to 9.80 mm (0.386 in.) in diameter.

The billet and tooling were lubricated with a MoS_2-containing grease (Jet-Lube 550 Anti-Seize Lubricant, Jet-Lube, Houston, TX). A short plug approximately 19 mm (0.75 in.) long of C11000 copper was inserted in the die inlet channel at the tail of the billet. The Cu plug at the billet tail allowed the billet to be pushed through the die corner and beyond the outlet channel land without interruption. The Cu plug remained in the die corner, and was expelled by the next extrusion. The billets were extruded at room temperature at a ram speed of 2.5 mm/s (0.1 in./s). After each extrusion, a small bulge of material was noted on the top surface of the billet immediately behind the billet nose; this bulge was removed by abrasive grinding to allow the billet to be reinserted into the tooling. Route B_C (90° rotation of the billet in the same direction) was used, with the same end of the billet being inserted into the die for all passes.

After 4 passes, the billet was divided approximately in half. Sections were taken from one piece for metallography of the as-deformed microstructure in 3 orthogonal axes selected to match the planes of the ECAE tooling. The other piece was annealed in vacuum for 1 h at 550°C. After annealing, this piece was similarly sectioned to show 3 orthogonal views of the annealed microstructure. These microstructures were compared to similar EB-melted crystal-bar Zr that had previously been processed by a conventional upsetting and cold-rolling process.

The annealed materials were examined with electron back-scattered diffraction (EBSD) to determine the textures and grain sizes after annealing. The ECAE-processed material sample surface was perpendicular to the billet axis. The cold-rolled and recrystallized sample was examined on a through-thickness surface. The samples were prepared by standard metallographic procedures with a final polish using colloidal silica; each sample was then lightly swab-etched in a solution of 45 ml water, 45 ml nitric and 10 ml hydrofluoric acid (48%). The microtextures and microstructures were measured using an FEI Phillips XL30 environmental scanning electron microscope outfitted with TSL Orientation Imaging Microscopy (OIM) hardware and software. Scans were typically 400 μm square with a step size of 1 μm. The grain dilation feature of the TSL software was applied to 'dissolve' scan artifacts leaving only the recrystallized structure. The reported grain sizes were calculated assuming circular grains of area equivalent to the actual grains. The textures were calculated on the basis of 5 degree increments using harmonics to L=16.

Results and Discussion

The Zr-in-Ni billets extruded readily, and the Ni can remained intact throughout the 4 passes, with no indication of possible failures of either the welds or the can walls. The load-displacement curves were smooth and unremarkable.

The initial as-cast microstructure consisted of very large grains (Fig. 1), in some cases several mm in size. These grains often contained long, isolated twins. Numerous second-phase particles were present throughout the structure, frequently located on the grain boundaries (Fig. 1). Examination in a scanning electron microprobe suggested these were zirconium carbides.

After ECAE processing, the deformed microstructure was subdivided on a very fine scale and in an apparently homogeneous manner (Fig. 2). In some regions remnants of the prior as-cast grain boundaries could still be discerned, but the interiors of the grains were finely subdivided. The structure had fine linear features which suggests that twinning played a major role in the deformation.

The microstructures of the conventionally processed material are shown in Fig. 3. In the as-rolled condition, the structure is distorted, but deformation appears to be predominantly slip-related, with fewer twin-like features than were observed for the ECAE-processed material. After 4 passes with a 120° die, the ECAE-processed material has received a total strain of approximately 2.6, equivalent to a reduction in thickness by rolling of 90%. Thus, the ECAE-processed material was in a much more heavily deformed condition when it was heat treated, as compared to the cold-rolled material. After heat treatment, the cold-rolled material also had a fine equiaxed microstructure, but the grain size was larger than for the ECAE-processed material.

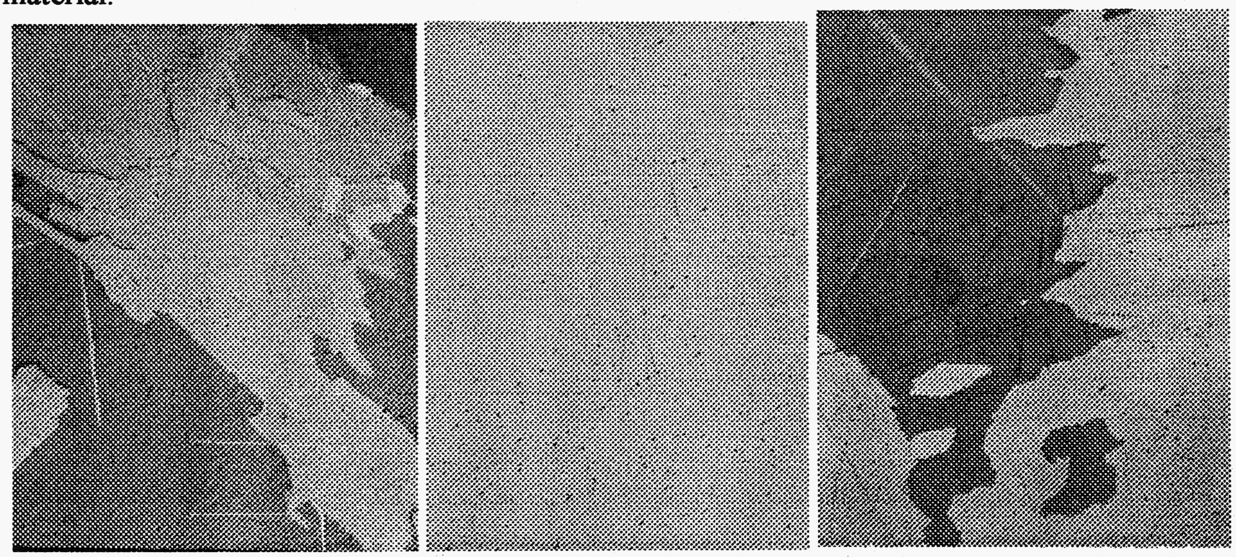

Fig. 1. As-cast Zr structure: Left. polarized light; Center. as-polished, normal illumination, showing second-phase particles; Right. same area as center, under polarized light. (100X)

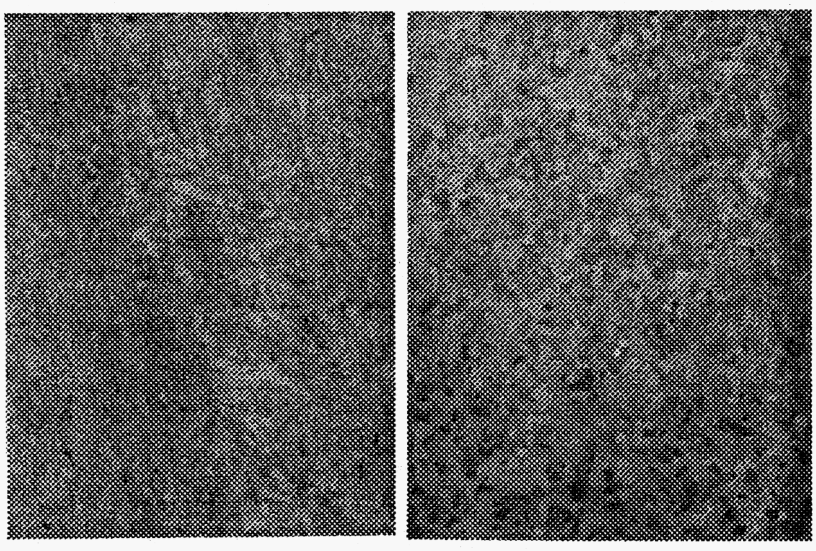

Fig. 2. Structure of ECAE-processed material: Left. after 4 passes; Right. annealed 1 h at 550°C. (200X)

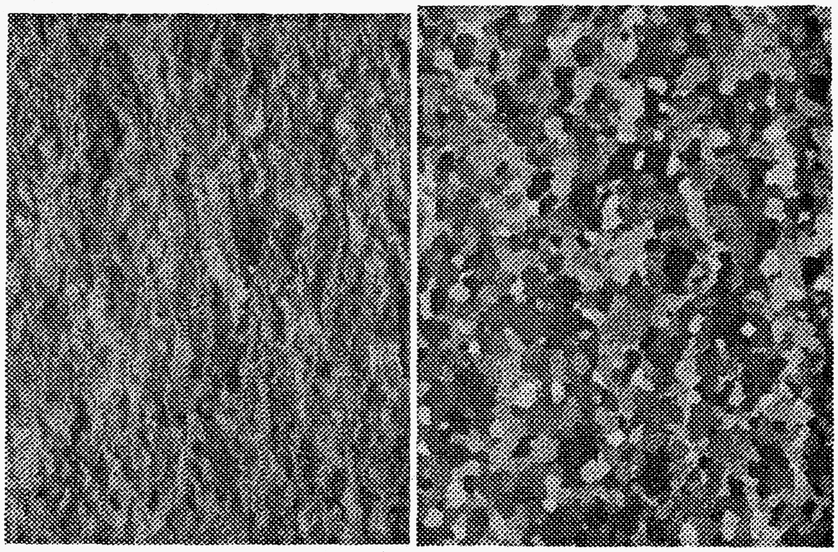

Fig. 3. Microstructure of the cold-rolled material, polarized light: Left. deformed; Right. annealed 1 h at 550°C. (200X)

The calculated grain size from the EBSD scans was determined by establishing a *minimum misorientation* to define grains. Both 5- and 15-degree minimum misorientation criteria were tested, with little change in the calculated grain sizes. Five scans were performed on the ECAE sample, four around the periphery of the cross section, and one in the center. The grain size for the ECAE-processed material was 10 microns +/- 0.5 microns. Three scans on the cold-rolled material, near the two rolling plane surfaces and near the centerline, provided a grain size of 13 microns +/- 0.5 microns.

Strong texture components (as measured by x-ray diffraction) were found in the pole figures of the annealed materials, in the same positions as for the prior deformed materials. The texture of the annealed ECAE material (Fig. 4) was dominated by a single component tilted approximately 60 degrees from the sample normal, with an intensity 2-3 times random; this orientation is consistent with the shear plane during the final pass through the 120° tooling. An interesting feature of these textures was that the c-axis component was not parallel to any of the 4 orthogonal sample orientations used during the 4-pass processing scheme. The texture of the rolled-and-annealed material (Fig. 5), measured in all three positions, was typical of rolled zirconium with the C-axis normal to the rolling plane surface, with the typical lateral spread in intensity. Rotation of the texture data to look down on the rolling plane (rolling plane normal in the center of the pole figures) showed four lobes spread laterally in the rolling and transverse directions. As with the microstructure, the textures were very similar for each of the scans with a maximum around 4 times random.

Commercial-purity Zr has been processed by ECAE, but either at elevated temperature (350°C) (7, 8), or with tooling with an angle of 135° between the inlet and outlet channels (9, 10). The use of the more severe 120° tooling at room temperature is possible because of the very low O_2 content in this EB-melted material. Deformation at room temperature prevents recovery processes that would occur at elevated temperature; this promotes the formation of a finer grain structure during recrystallization than would form after higher temperature processing. The relative ease with which the present EB-melted material was processed suggests that the use of more severe deformation, with tooling with 105 or perhaps 90° angles, may be practical.

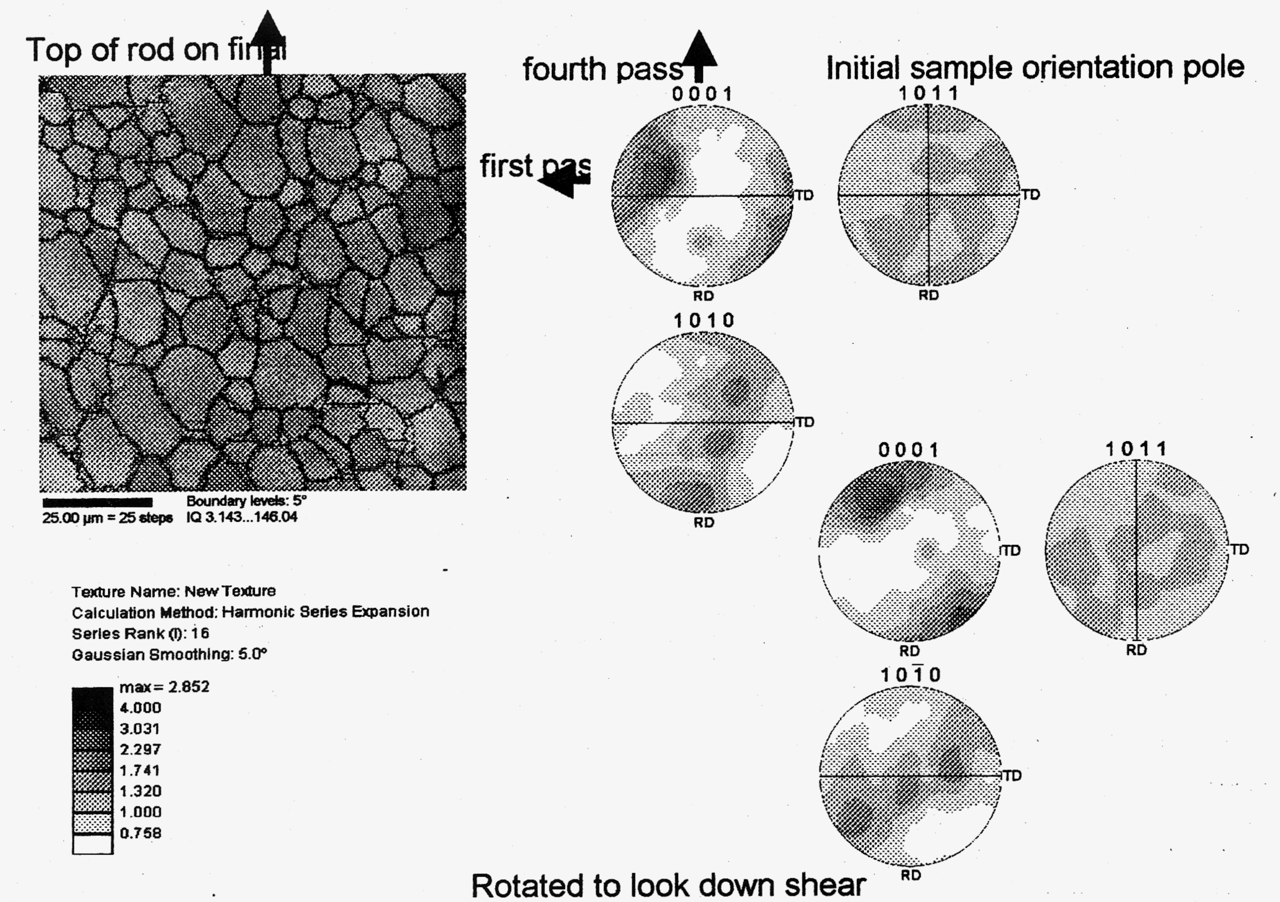

Fig. 4. Texture measurements for ECAE-processed and annealed material.

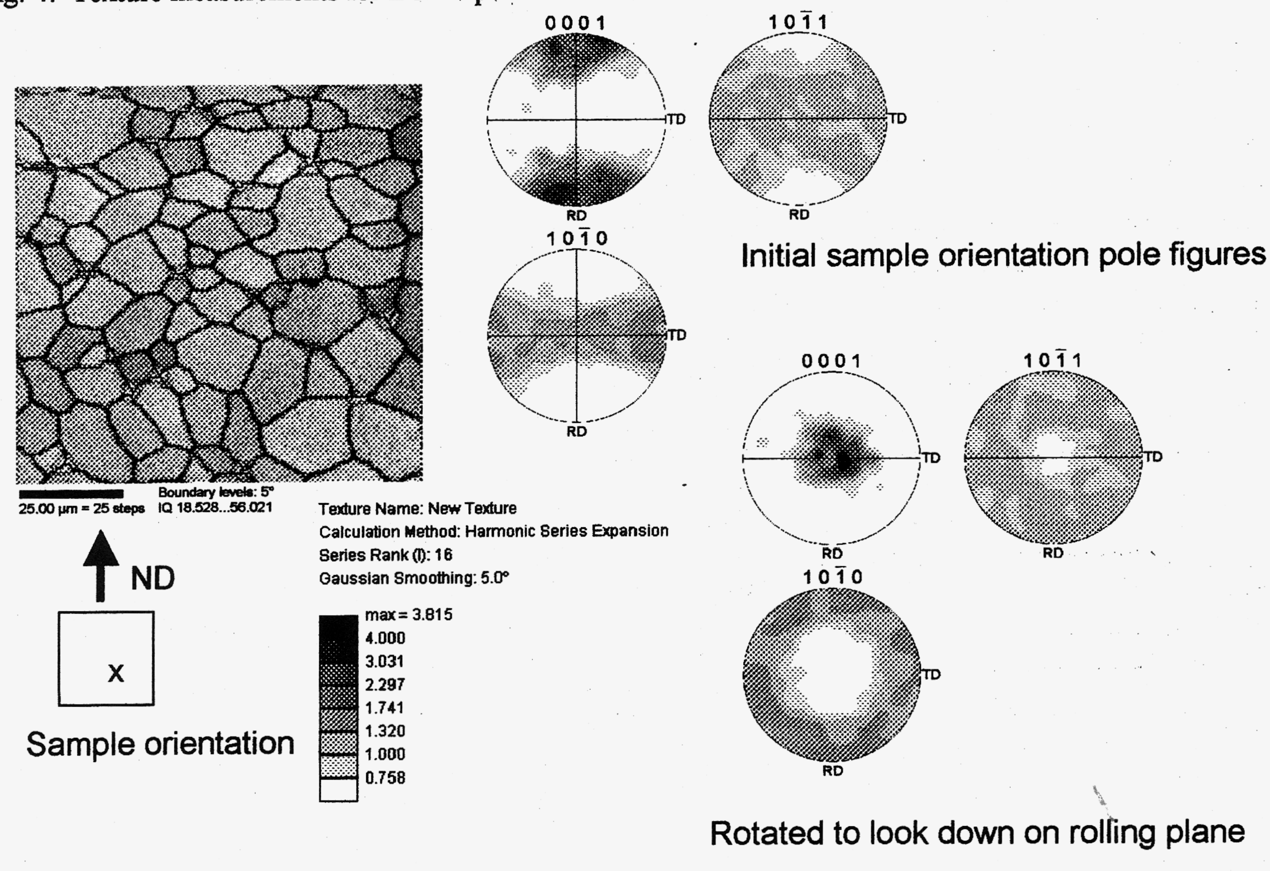

Fig. 5. Texture measurements for cold-rolled and annealed material.

Choi et al. (10) have processed commercial-purity Zr at room temperature with 135° tooling for 4 passes with route B_C, followed by an anneal for 0.5 h at 550°C. This resulted in an increase in the grain size from 0.2 μm in the deformed condition to 6 μm after annealing. The larger grain size obtained in the present work (10 μm) may be the result of the longer annealing time, or more likely the higher purity of the present material. Lower annealing temperatures could be used to achieve a finer grain size for the present EB-melted material. The use of more severe tooling, with a 105 or 90° angle, and/or increasing the number of passes to increase the total strain, should also result in smaller grain sizes after annealing.

Conclusions

High-purity electron-beam-melted crystal-bar Zr has been successfully processed at room temperature for 4 passes by route B_C in 120° tooling. This deformation resulted in a finely and homogeneously divided structure. Annealing in vacuum for 1 h at 550°C produced fine, equiaxed grains approximately 10 μm in diameter, with a weak texture of only 2-3 times random. Conventionally processed material had a slightly larger grain size of 13 μm, and showed a stronger texture, up to 4 times random. More extensive deformation by ECAE, and lower annealing temperatures, should result in an even finer grain size for ECAE-processed crystal-bar Zr.

Acknowledgments

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