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Microstructure evolution and strengthening mechanisms in commercial-purity titanium subjected to equal-channel angular pressing

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ABSTRACT

High-resolution electron backscatter diffraction (EBSD) was applied to examine grain refinement in commercial-purity titanium Grade 4 subjected to equal-channel angular pressing (ECAP) via the Conform technique. This approach enables the production of long-length billets and thus has the potential for commercial application. Microstructure evolution was found to be a relatively-complex process which included several stages. At relatively-low accumulated strains, microstructure changes were markedly influenced by mechanical twinning. However, the concomitant grain refinement suppressed this mechanism, and subsequent microstructure development was dictated by the evolution of deformation-induced boundaries which developed preferentially near the original grain boundaries. The final material produced after an effective strain of ~ 8.4 was characterized by a mean grain size of 0.3 μm, high-angle boundary fraction of 55 pct., a texture of moderate strength, and a yield strength of ~ 1050 MPa. Based on the detailed microstructural analysis, the contributions of various strengthening mechanisms were quantified. The rapid material strengthening during the early stages of ECAP was explained in the terms of a major increase in dislocation density and the extensive formation of the deformation-induced boundaries. With further increments in accumulated strain, however, the dislocation as well as grain-boundary density reached a saturation, thus reducing the hardening efficiency of ECAP at high strains.

1. Introduction

The evolution of microstructure during severe plastic deformation (SPD) and the properties of materials thus processed have received considerable attention in the literature [1–3]. To date, a great number of results has been obtained, and the fundamental principles related to the formation of an ultrafine-grain (UFG) structure and the enhancement of mechanical properties in metals and alloys via SPD have been formulated [1–3]. In particular, it has been shown recently that the formation of a UFG structure in titanium leads to substantial strengthening, thereby making this material attractive for medical applications [4,5].

Most of the research dedicated to SPD of titanium and resulting mechanical properties has been focused on microstructure evolution at high accumulated strains; microstructure development at low and intermediate strains has received less attention.

From a broad perspective, it has been demonstrated that microstructure evolution during equal channel angular pressing (ECAP) of titanium is relatively complex and comprises several stages. At relatively low strains (~ 1 ECAP pass), microstructural changes are markedly influenced by mechanical twinning which provides rapid grain refinement [6–12]. $\{10\bar{1}1\}$ twins are often reported to predominate [6–12], but the twinning mode is expected to vary depending on material purity, deformation temperature, and initial texture. Concomitant grain refinement suppresses twin activity [7,10,12,13], and microstructure evolution at higher strains is presumably governed by other mechanism(s). In particular, the dislocation density increases markedly, reaching ~ 10^{15} m^{-2} [14]. However, a distinct cell structure does not develop [7], and microstructure is characterized by large orientation gradients [13]. The mean grain size estimated from transmission electron microscopy (TEM) observations approaches 0.2 μm [14]. Microstructure evolution at this stage has been hypothesized to be governed by continuous recrystallization [13] but the details of this process are unclear. The results clearly demonstrate, however, that the severely-deformed microstructure is relatively complex and contains dislocation *substructure*. If so, the strengthening effect presumably cannot be explained simply in the terms of the Hall-Petch relation. Indeed, a number of studies have shown that *substructure* hardening may also play a significant role [15–18]. Despite recent progress in this field, however, the microstructure-strength relationship in heavily-deformed titanium

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remains poorly understood.

To obtain a comprehensive idea of microstructure evolution and strengthening mechanisms, a large amount of information on boundary density, types, misorientations as well as crystallographic texture must be obtained. Transmission electron microscopy (TEM), which is typically applied to study substructure, cannot provide sufficient quantitative data. By contrast, high-resolution electron backscatter diffraction (EBSD) appears to be a better choice. Despite its limited angular accuracy (~ 2°), this technique enables quantification of microstructural features over a much larger scale than TEM and thus provide insights that may be more significant from a statistical point of view. In recent years, EBSD has developed markedly, making it a very popular technique. This paper demonstrates the feasibility of high-accuracy EBSD for severely-deformed UFG titanium. The overall objective of the present work was to establish the interrelation between microstructure and strength in SPD-processed titanium. A thorough analysis of grain boundary evolution with increasing strain was made. The results of these measurements were used to elucidate the contributions of various hardening mechanisms to overall material strength.

A recent modification of ECAP technique (the so-called ECAP-Conform process [19]) was used to impart SPD in the present work. This approach enables production of long-length billets and has higher productivity than "classical" (batch-mode) ECAP. Moreover, the application of the Conform scheme allows ECAP of titanium at relatively-low temperatures (~ 200 °C), thus enhancing the strengthening effect.

2. Material and experimental procedures

The material used in the present investigation was commercial-purity titanium Grade 4 whose chemical composition is given in Table 1. The relatively-high interstitial content in this grade imparts significant solid-solution strengthening, thus making it particularly attractive for biomedical applications. In the as-received condition, the material exhibited an equiaxed grain structure with a mean size of ~ 20 μm, a fraction of high-angle boundaries of 95 pct., and a weak $\{hkil\} < 10\bar{1}0>$ crystallographic texture (Supplementary Fig. S1).

The program material was subjected to ECAP via route B_C, i.e. the deformed billet was rotated by 90° in the same direction between each pass, and thus the strain path was changed during deformation. The Conform technique [19] was used, a schematic illustration of which is shown in Supplementary Fig. S2. ECAP was performed on samples measuring 500 mm in length at 200 °C (~ $0.24T_m$, in which T_m denotes the melting point) at a speed of 33 mm/s using a die with a 120° square channel. The effective strain was estimated to be ~ 0.7 per pass. To track microstructure changes with strain, the material was subjected to 1, 2, 4, 6, 8, 10, and 12 ECAP passes. The principal directions of the ECAP geometry are denoted as the longitudinal direction (LD), the transverse direction (TD), and the normal direction (ND) (Supplementary Fig. S2b).

Microstructure characterization was performed primarily by EBSD. To provide a three-dimensional view of the grain structure, samples were cut from both longitudinal and transverse cross sections of deformed billets. EBSD specimens were prepared using conventional metallographic techniques followed by long-term (24 h) vibratory polishing with a colloidal-silica suspension. In all cases, microstructure observations were focused in the central part of the billets. EBSD analysis was conducted with a JSM-7800F field-emission gun, scanning electron microscope (FEG–SEM) operated at 25 kV that was also equipped with a TSL OIM™ EBSD system. To examine the microstructure at different length scales, several EBSD maps with different scan step sizes were acquired for each material condition.

Table 1
Chemical composition (wt%) of the program material.

Ti	C	Fe	O	N	H
Balance	0.05	0.15	0.36	0.007	0.0021

The total statistics of the EBSD measurements are summarized in Supplementary Tables S1 & S2. For each diffraction pattern, nine Kikuchi bands were used for indexing, thus minimizing mis-indexing errors. To improve the reliability of the EBSD data, grains comprising three or fewer pixels were automatically removed from the maps using the grain-dilation option in the TSL software. Furthermore, to eliminate spurious boundaries caused by orientation noise, a lower-limit boundary misorientation cut-off of 2° was used. A 15° criterion was applied to differentiate low-angle boundaries (LABs) and high-angle boundaries (HABs).

Because real microstructures often exhibit a complex mixture of LABs and HABs, confusion can arise with regard to the definition of grains. To avoid this ambiguity, the term "grain" in the present work was applied to denote a crystallite bordered by a *continuous* HAB perimeter. Accordingly, a crystallite delimited by *any* EBSD-detected boundaries (i.e., those having misorientations of at least 2°) was termed a "fragment". Two approaches were used to quantify the grain/fragment size. The first was the classical, linear-intercept method. However, because this technique does not account for the volume fraction of grains with different sizes, the grain-reconstruction method was also applied [20]. In this technique, the grain size is quantified by the measured grain area (in a 2-dimensional section), and the equivalent grain diameter was calculated assuming each grain was a circle.

The EBSD measurements were complemented by TEM observations. For this purpose, thin foils were cut from the deformed material using electric discharge machining, ground to a thickness of ~ 0.1 mm and then jet-polished in a solution containing 5% of perchloric acid, 35% butanol, and 60% methanol. TEM was performed using a JEOL JEM 2100 SEM operated at an accelerating voltage of 200 kV. Further details of the TEM procedures are described elsewhere [14].

To establish the effect of ECAP on material strength, uniaxial tension tests were performed at room temperature. To this end, tensile specimens having a diameter of 3 mm and a gauge length of 15 mm were machined parallel to the LD; other details of the geometry of the specimens are given in Supplementary Fig. S3. To eliminate surface defects, the gauge surface of each specimen was mechanically polished using abrasive papers. Tension tests were conducted in triplicate to failure at a constant crosshead speed of 1 mm/min in a universal testing machine Instron 5982.

3. Results

3.1. Microstructure morphology

Selected portions of typical low-magnification EBSD maps from the deformed billets are shown in Fig. 1 and Supplementary Figs. S4 & S5. In the maps, individual grains are colored according to their crystallographic orientations relative to the LD.[1] For simplicity, only HABs are shown (black lines). In longitudinal sections, the grains were elongated along a particular direction. In contrast, the grains tended to retain a nearly-equiaxed shape in transverse sections, although the microstructural pattern had become somewhat complex after 12 ECAP passes. To a first approximation, therefore, the three-dimensional grain shape was similar to a fiber. The inclination angle to the LD in the longitudinal section was close to 41°, in good agreement with theoretical predictions for a 120-degree ECAP die [21]; thus, the grain shape mirrored the imposed shear strain during processing. These observations are consistent with other reports in the literature [6,8,13,14,18,22], and indicate that microstructure evolution was highly influenced by the geometric effect of strain.

Fig. 1 also illustrates significant grain refinement during ECAP. To examine this process in greater detail, high-resolution EBSD maps were acquired from the deformed material; typical examples are shown in Fig. 2 and Supplementary Fig. S6. In these maps, LABs, HABs, and

[1] Here and hereafter, a reader is referred to on-line version of this paper to see figures in color.