Microstructural characteristics of pure gold processed by equal-channel angular pressing

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Experiments were conducted to evaluate the microstructural characteristics of pure (4N) gold processed by equal-channel angular pressing using routes A or B. Using atomic force microscopy and X-ray diffraction, it is shown that, although these two routes lead to similar dislocation densities of \( \sim 1.5 \times 10^{15} \text{ m}^{-2} \) and similar average grain sizes of \( \sim 460-490 \text{ nm} \), there are significant differences in the shearing patterns and in the densities of planar faults.

Keywords: Atomic force microscopy (AFM); X-ray diffraction (XRD); Deformation structure; Equal-channel angular pressing (ECAP); Gold (Au)

Severe plastic deformation (SPD) is an effective tool for producing ultrafine-grained (UFG) metals and alloys [1,2]. At the present time, the SPD procedure of equal-channel angular pressing (ECAP) is used most frequently because it is relatively easy to establish in any materials laboratory, it has a potential for scaling up to large samples and it produces reasonably homogeneous microstructures without any reduction in the cross-sectional dimensions of the samples [3]. To date, the effect on the microstructure of the various ECAP strain paths has been investigated for several face-centered cubic (fcc) metals, including Al [4–9], Ni [10] and Cu [11]. However, only limited information is available for Au [12,13], and for this metal there are no direct measures of either the dislocation densities after ECAP or the densities of any planar faults. Accordingly, the present research was undertaken to evaluate the microstructural characteristics of 24 carat (99.99% purity) Au processed by ECAP for four passes using either route A, in which the sample is pressed repetitively without rotation, or route B, in which the sample is rotated by 90° in the same sense between each separate pass [14].

The processing by ECAP was conducted at room temperature using samples having lengths of \( \sim 70 \text{ mm} \) and cross-sectional areas of \( 10 \times 10 \text{ mm}^2 \). Pressing was conducted at a velocity of 0.1 mm s\(^{-1}\) using an ECAP die containing an internal channel bent through an abrupt angle of 90° and with an outer sharp corner so that the angle denoting the arc of curvature was 0°. It can be shown that these conditions give an equivalent strain of \( \sim 4.6 \) after a total of four passes [15].

Following ECAP, specimens were prepared for examination using atomic force microscopy (AFM) and X-ray diffraction. All observations were taken on the plane parallel to the side face at the point of exit from the die, generally designated the Y plane [3]. This plane was first mechanically polished to a mirror-like finish in three steps using 1 µm and 60 nm diamond paste and a 20 nm colloidal silica suspension. The surface was etched for 3 min at room temperature (20°C) in a solution of 17 g potassium cyanide, 3.75 g potassium ferrocyanide, 3.75 g potassium sodium tartrate, 3.5 ml phosphoric acid and 1 ml ammonia in 250 ml water. The surface topographies were examined using an atomic force microscope (Solver P47H) operating in the contact mode with conductive silicon cantilevers having a resonant eigenfrequency of 14–27 kHz. Additional microstructural observations were also undertaken.
using X-ray diffraction line profile analysis. The X-ray line profiles were measured using a high-resolution diffractometer (Nonius, FR 591) with Cu Kα1 radiation and the line profiles were evaluated through use of the extended convolutional multiple whole profile fitting procedure [16]. In this method, the experimental profiles are fitted using theoretical profile functions calculated on the basis of a model microstructure where it is assumed that the lattice strains are caused by dislocations and planar faults. Through the use of this method, it is possible to obtain information on both the density of dislocations, ρ, and the density of planar faults, β.

The larger micrographs in Figures 1 and 2 show typical AFM images of the microstructures of the Au samples processed by ECAP using routes A and Bc, respectively, where the pressing direction is horizontal, the billet is moving from left to right and the bottom edges of the images correspond to the pressing, or X, direction. It is apparent from these micrographs that processing by ECAP produces a relatively homogeneous grain structure and, from inspection of the magnified smaller images, the average grain sizes for both processing routes are of the order of ~500 nm. It is apparent also that the grains have a slightly elongated configuration after processing through route A but that they are reasonably equiaxed after processing through route Bc; a similar trend was reported earlier in the processing of pure aluminum [5,6]. The AFM images reveal the formation of distinctive shear bands during ECAP. These mesoscopic traces, labeled Band 1 in Figures 1 and 2, are reasonably aligned with the ideal shearing plane when the sample passes through the die. Similar bands have been reported in experiments on other polycrystalline fcc metals processed by ECAP, including Al [17], Cu [18,19] and Ni [19], and the results are also consistent with data reported for the pressing of Al single crystals [20]. During subsequent deformation in tensile testing, earlier experiments on pure Au processed by ECAP showed that this shear localization leads to the development of macroscopic deformation bands [12].

![Figure 1](image1.png)

**Figure 1.** Typical AFM images and a representative surface vertical profile taken on the Au sample processed by ECAP using route A.

![Figure 2](image2.png)

**Figure 2.** Typical AFM images and a representative surface vertical profile taken on the Au sample processed by ECAP using route Bc.
In practice, very careful inspection revealed a significant difference in the distributions of shear bands formed in these two separate processing routes. Thus, in route A the band distribution is relatively inhomogeneous and there are wide separations between the most well-delineated bands. This is demonstrated in Figure 1, where labels are placed on the individual wide bands at A1, A2, etc. These coarse bands are a natural consequence of the relatively small number of shearing directions available after processing through four passes using route A [21]. By contrast, the rotation of the sample through 90° between successive passes in route Bc leads to a finer and more uniform distribution of the shear bands, as shown in Figure 2. These differences are also clearly revealed in measurements of the surface vertical profiles taken along the paths labeled Route A and Route Bc in Figures 1 and 2, where these paths lie perpendicular to the ideal shear plane. These profiles are shown in the small insets recording the measured profiles over total distances of 10 μm. In addition, close inspection of Figure 2 for route Bc shows the presence of a secondary set of faint shear bands lying perpendicular to Band 1; this secondary system is labeled Band 2. The difference in shear band formation between routes A and Bc for four passes of ECAP is due primarily to the difference in the total angular ranges containing the active slip traces for these two processing routes since these angular ranges are 37° and 63° when recorded on the Y plane for routes A and Bc, respectively [21]. Thus, the concentration of shearing over a narrower angular range in route A leads to the formation of a broader set of parallel shear bands whereas the larger angular range in route Bc permits the operation of a secondary shearing system.

The densities of dislocations and the planar faults may be determined using X-ray line profile analysis. Figure 3 shows a typical X-ray diffraction pattern for the sample processed by route A using a logarithmic intensity scale for the 2θ range between 34° and 40°. There is clear evidence for a very small peak in the left-hand tail of the 111 fundamental reflection of Au and this occurs at a position given by 2θ ≈ 36.2°. Earlier experimental results have shown that similar small peaks appear when planar faults having an hexagonal closed-packed sequence are present in the microstructures of fcc or diamond crystal structures [22]. The position of this peak may be determined as 2θ = 2(\text{arcsin}(0.82/\lambda))$, where \( \lambda \) is the X-ray wavelength and \( \lambda \) is the lattice parameter of the cubic crystal [22]. Taking the lattice parameter of Au as \( \lambda = 0.4079 \text{ nm} \) and the wavelength of Cu Kα1 radiation as \( \lambda = 0.15406 \text{ nm} \), the value calculated for 2θ is 36.1°. This calculated value is in excellent agreement with the experimental result shown in Figure 3 and thereby confirms that the small peak at 2θ = 36.2° is associated with planar faulting.

The microstructural parameters obtained from the line profile analysis are summarized in Table 1, including the dislocation densities and the planar fault densities for each processing route. Also included in Table 1 are the measured average grain sizes and, using earlier reported data [12], the values of the yield strengths for each condition. These results show that there are significant densities of dislocations and planar faults for both processing conditions, and this suggests that the shear occurring during ECAP is accommodated through the introduction both of dislocations in plastic deformation and planar faulting due to the exceptionally low stacking-fault energy of 45 mJ m⁻² for pure Au. It is also apparent from Table 1 that the processing route or strain path has only a minor influence on the individual values of the dislocation densities although the slightly higher density after processing by route Bc may be due to the larger angular range of shearing directions for this route. Thus, in route A the shearing occurs over a narrow angular range, and this leads to greater strain hardening in each shear band and a consequent higher activation of planar faulting during plastic deformation. Despite these differences, both the grain sizes and the total dislocation densities, in addition to the tensile stress–strain curves [12], are only slightly dependent upon the processing route.

Considering the influence of the microstructural characteristics on the mechanical properties, it is noted that the yield strength of severely deformed fcc metals is often given as the sum of the contributions of the so-called geometrically necessary dislocation boundaries and the incidental dislocation boundaries, representing high and low angle misorientations, respectively, where the hardening effects of these two boundary types are described by the conventional Hall–Petch and Taylor terms, respectively [23]. However, it was shown recently that, for fcc metals having submicrometer grain sizes

![Figure 3. A typical X-ray diffraction pattern for the sample processed by route A.](image)

| Table 1. Microstructural parameters for pure Au processed by ECAP for four passes using routes A or Bc |
|----------------------------------|-----------------|-----------------|-----------------|-----------------|
| Sample                          | Average grain size, \( d \) (nm) | Dislocation density, \( \rho \) \( \times 10^{14} \text{ m}^{-2} \) | Planar fault density, \( \theta \) (%) | Yield strength, \( \sigma_y \) (MPa) |
| Route A                         | 490 ± 30        | 15 ± 2          | 0.42 ± 0.04     | 230 [12]        |
| Route Bc                        | 460 ± 30        | 17 ± 2          | 0.28 ± 0.04     | 245 [12]        |
between 100 and 1000 nm after processing by ECAP, these two contributions to the yield strength may be incorporated into a single modified Hall–Petch relation [24]. In this modification, the yield stress, \( \sigma_y \), and the grain size, \( d \), for high-angle boundaries are related through an expression of the form:

\[
\frac{\sigma_y}{\mu} = C_1 + C_2 \left( \frac{d}{b} \right)^{-0.77},
\]

where \( \mu \) is the shear modulus, \( b \) is the magnitude of the Burgers vector, and \( C_1 \) and \( C_2 \) are constants. The results of this analysis are shown in Figure 4, where the solid points are taken from an earlier report [24], the line is fitted using Eq. (1) and the two open points show the results for the two samples of Au tested in the present experiments. Figure 4 confirms the general validity of the modified Hall–Petch relationship for use also with pure Au.

In summary, pure Au was processed by four passes in ECAP using routes A and B, and the deformed billets were examined to determine the shearing patterns, the grain sizes and the densities of dislocations and planar faults. The results show that the grain sizes and dislocation densities are almost identical for both processing routes but the distributions of shear bands depend critically upon the strain path. It is demonstrated that the values of the yield stresses and the measured grain sizes after ECAP are consistent with a normalized Hall–Petch relationship proposed earlier for UFG fcc metals.

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