J Mater Sci (2009) 44:1656–1660
DOI 10.1007/s10853-009-3278-1

LETTER

Twinning and dislocation activity in silver processed by severe plastic deformation

Jeno Gubicza · N. Q. Chinh · J. L. Lábár · Z. Hegedűs · T. G. Langdon

Received: 17 December 2008 / Accepted: 14 January 2009 / Published online: 4 February 2009

Introduction

Severe plastic deformation (SPD) provides attractive procedures for producing ultrafine-grained (UFG) metals in bulk form [1, 2]. One of the most frequently used methods is equal-channel angular pressing (ECAP) where it is possible to produce relatively large bulk UFG metals having dimensions of several centimeters in all directions [3]. It is now well established that the high strength of metals processed by ECAP is due to their high dislocation densities and small grain sizes [4]. There have been extensive reports documenting the evolution of the microstructure during the processing by ECAP of pure face-centered cubic (fcc) metals having medium or high stacking fault energies (SFE). Thus, it was reported that in metals such as Al and Cu, the grain size reaches a minimum value and the dislocation density saturates after about 4 passes of ECAP [4]. Nevertheless, very little information is available at present describing microstructural evolution during ECAP in pure fcc metals where the SFE is very low.

It was shown recently that in Ag, where the SFE is only ~ 16 mJ m^{-2} [5], the dislocation density was $\sim 46 \pm 5 \times 10^{14}$ m^{-2} after 8 passes of ECAP [6, 7] where this density is exceptionally high by comparison with other fcc metals, such as Au or Cu [6, 8–10]. The high dislocation density in Ag is a consequence of the very low SFE because the annihilation of dislocations is hindered by their high degree of dissociation into partials. The present investigation was therefore initiated to provide a comprehensive report on the evolution of grain structure and dislocation and twin densities when Ag is processed to very high strains up to a maximum of 16 passes through the ECAP die. As will be demonstrated, the experimental results are consistent with, and provide the first direct support for, a theoretical model of deformation in fcc metals with low SFE developed earlier by Müllner and Solenthaler [11].

J. Gubicza (✉) · N. Q. Chinh · J. L. Lábár · Z. Hegedűs
Department of Materials Physics, Eötvös Loránd University, Pázmány Péter s. 1/A, 1117 Budapest, Hungary
e-mail: gubicza@metal.elte.hu

J. L. Lábár
Research Institute for Technical Physics and Materials Science, P.O. Box 49, 1525 Budapest, Hungary

T. G. Langdon
Departments of Aerospace & Mechanical Engineering and Materials Science, University of Southern California, Los Angeles, CA 90089-1453, USA

T. G. Langdon
Materials Research Group, School of Engineering Sciences, University of Southampton, Southampton SO17 1BJ, UK

Experimental material and procedures

High-purity 99.99% Ag billets, having lengths of ~ 70 mm and diameters of ~ 10 mm, were homogenized for 60 min at a temperature of 741 K where this corresponds to $0.6 \cdot T_m$, where T_m is the absolute melting point of Ag. The billets were then processed by ECAP at room temperature through totals of 1, 4, 8, and 16 passes using a pressing velocity of 8 mm s^{-1} and a solid die with an internal channel angle of 90° and an outer arc of curvature of $\sim 20°$. For these angles, it can be shown that one pass corresponds to an equivalent strain of ~ 1 [12]. The pressings were conducted using route B_c in which the billet is rotated about its longitudinal axis by 90° in the same direction between each pass [13, 14].

The microstructures were examined by X-ray line profile analysis on the transverse sections perpendicular to the axes of the billets. The measurements of X-ray diffraction lines were performed using a special high-resolution diffractometer (Nonius FR591) with CuKα_1 radiation ($\lambda = 0.15406$ nm) and the line profiles were evaluated using the extended Convolutional Multiple Whole Profile (eCMWP) fitting procedure [15]. This method gives the dislocation density and the twin-fault probability with good statistics. The twin-fault probability is defined as the fraction of the twin boundaries among {111} lattice planes along their normal vector. The microstructures produced by ECAP were examined using a Philips CM-20 transmission electron microscope (TEM) operating at 200 kV. The TEM samples were mechanically thinned to ~50 μm, cooled to liquid nitrogen temperature and then thinned with 6 keV Ar^+ ions from both sides until perforation. Finally, the thin damaged surface layer was removed using 2 keV Ar^+ ions. The relatively large grain structures of the initial sample and the specimen processed through 1 pass were investigated using a JEOL JSM-25SII scanning electron microscope (SEM). Samples were prepared for SEM by exposing the polished surfaces of the polycrystalline samples to a beam of 10 keV Ar^+ ions at an angle of 30° to the surface. This high-angle ion-etching was used to roughen the surface and reveal the grain structure because it is known that the etching rate depends on the crystal orientation such that the surface morphology is enhanced by the different etching rates of local surfaces oriented at different angles to the ion beam [16].

Experimental results and discussion

Figure 1a shows an SEM image of an Ag sample before ECAP where the mean grain size is ~10 μm. After 1 pass the grain size was reduced to ~5 μm as shown by the SEM image in Fig. 1b. The microstructures of various samples are shown in Fig. 2 using TEM. The interior of a grain is given in Fig. 2a for the sample processed through 1 pass where both dislocations and twins are observed. The white arrows denote the locations of twin boundaries and the mean spacing between these boundaries was measured as ~200 nm. After 1 pass of ECAP, X-ray line profile analysis was used to measure the dislocation density and the twin probability as $16 \pm 2 \times 10^{14}$ m^{-2} and 0.1 ± 0.1%, respectively. It should be noted that this twin probability is in good agreement with the value of ~0.12% calculated from the mean twin spacing of ~200 nm and from the spacing of 0.236 nm between neighboring (111) planes. The mean grain size was reduced to ~160 ± 50 nm after 4 passes as shown in Fig. 2b and this size remained essentially unchanged within experimental error after 8 passes (~200 ± 50 nm) and 16 passes (~190 ± 50 nm) as illustrated in Fig. 2c and d, respectively.

Figure 3 shows the measured grain size plotted as a function of the number of passes through the die together with the dislocation density and the twin probability determined by X-ray line profile analysis, where the equivalent strain is directly proportional to the number of ECAP passes. The experimental results show that after 4 passes the dislocation density and the twin probability increase to $37 \pm 4 \times 10^{14}$ m^{-2} and 0.7 ± 0.1%, respectively. However, whereas the dislocation density saturates after 8 passes at $46 \pm 5 \times 10^{14}$ m^{-2} and then decreases to $25 \pm 3 \times 10^{14}$ m^{-2} after 16 passes, the measurements show the twin probability increases to 0.9 ± 0.1% after 8 passes and then further increases to 1.5 ± 0.1% after 16 passes. Thus, there is a very significant difference between the observed trends for the dislocation density and the twin probability in polycrystalline Ag processed by ECAP.

Careful examination shows these differing evolutions of the dislocation and twin densities with increasing strain are consistent with the theoretical predictions of a model

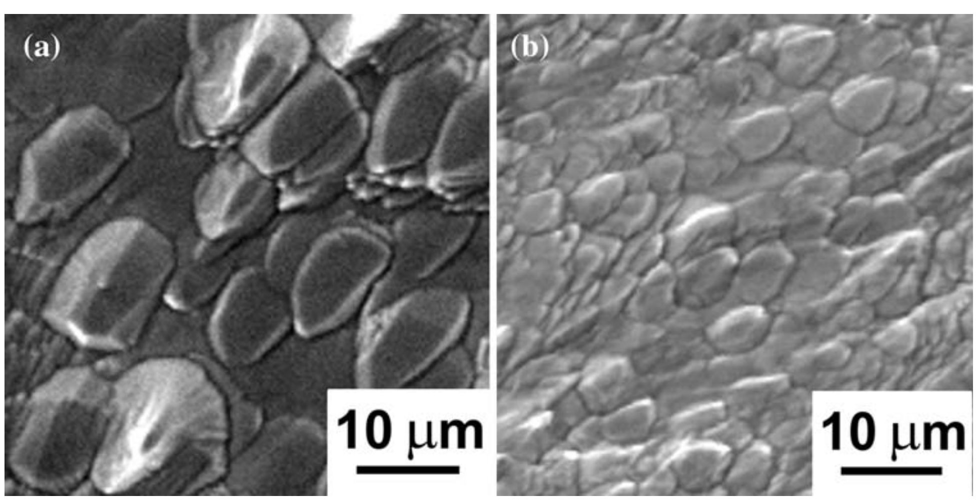

Fig. 1 SEM images of the microstructure **a** in the initial state before ECAP and **b** in the sample processed through 1 pass

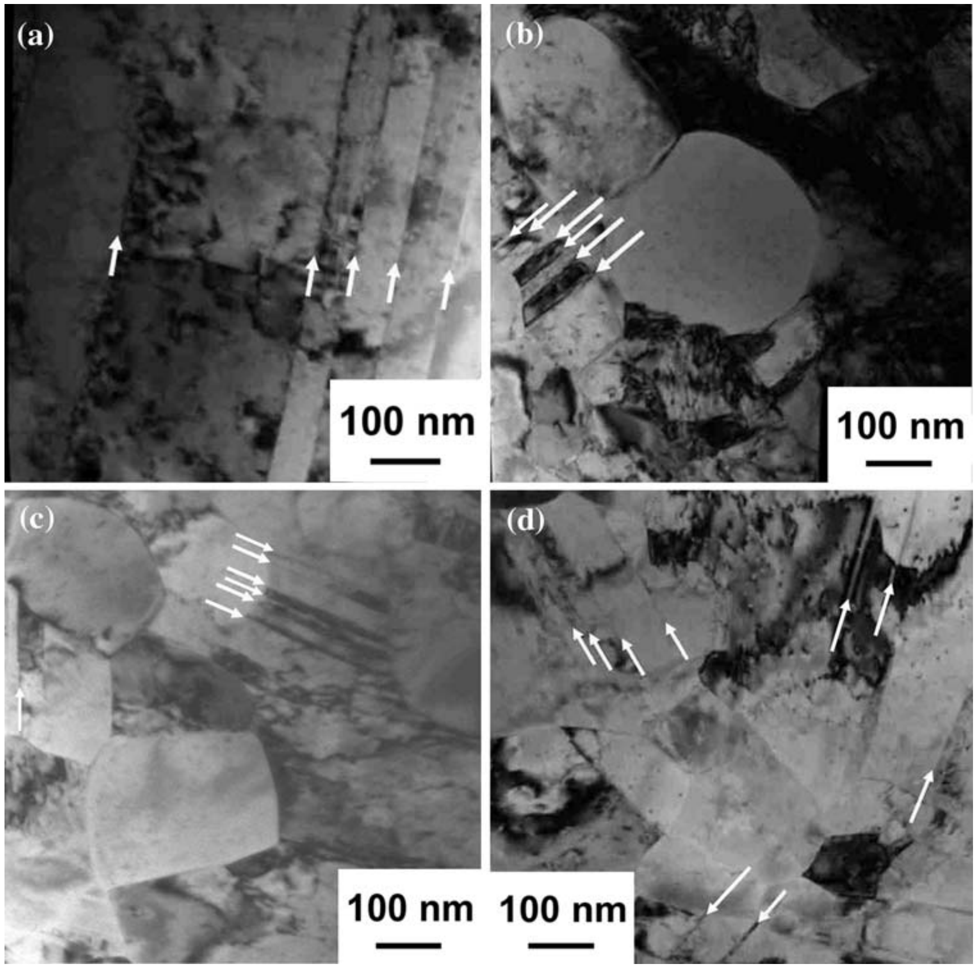

Fig. 2 TEM images taken after **a** 1, **b** 4, **c** 8, and **d** 16 passes of ECAP: the white arrows indicate the presence of twin boundaries

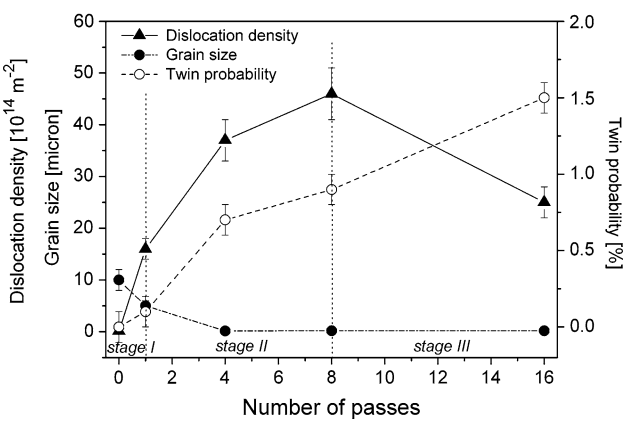

Fig. 3 The variation of the grain size, the dislocation density, and the twin probability with the number of ECAP passes and therefore with the equivalent strain

developed earlier by Müllner and Solenthaler [11] for the plastic straining of fcc metals and alloys having low SFE. At relatively low strains, the deformation behavior in these materials is controlled mainly by the planar glide of dissociated dislocations and the dislocation density increases rapidly while twinning is marginal. As the deformation proceeds, dislocation pile-ups form at glide obstacles such as Lomer-Cottrell locks and grain boundaries. In fcc metals having very low SFE, these glide obstacles strongly hinder the further activity of lattice dislocations because the nonconservative motion of dislocations is difficult due to the high degree of dissociation. At these obstacles, if the local stress exceeds the critical stress required for twin nucleation, plasticity continues by twinning. With increasing strain, as in ECAP, the dislocation density further increases, together with the locations of these high stress concentrations, leading to an increased density of twins. However, these twins also obstruct the glide of lattice dislocations thereby hindering the operation of dislocation sources and leading to a reduction in the rate of dislocation production.

The present results demonstrate there is a well-defined saturation in the dislocation density and thereafter, at higher strains, the dislocation density decreases while the twin probability continues to increase. This reduction in the dislocation density at high strains may be explained in terms of the nature of the twinning mechanism. Several possible mechanisms have been proposed for twin formation including (i) by a pole mechanism [17], (ii) by the dissociation of lattice dislocations into Schockley and Frank partials at Lomer-Cottrell barriers [18], (iii) by the nucleation of three-layer twins by dissociation of co-planar lattice dislocations into Schockley partials [19], and/or (iv) by the emission of twinning partials from grain boundaries [20, 21]. In the fourth process, it may be assumed that the partials form by a dissociation of non-geometrically necessary lattice dislocations in the grain boundaries [20]. All of these mechanisms of twinning are based inherently on the dissociation of lattice dislocations into twinning partials such that, if these partials then move to the grain boundaries, the formation of twins will contribute to the annihilation of lattice dislocations. As a consequence, the decrease in the dislocation density between 8 and 16 passes, as shown in Fig. 3, is associated with an increase in the twin concentration. It is important to note that the dislocation sources are incapable of compensating for the dislocations that have disappeared by twinning because the operation of the sources is also obstructed by the newly nucleated twins. It is possible that untwinning may occur during the deformation process, due to the interaction between the dislocations and twins [11], but the present increase in the twin probability with increasing strain suggests that twin production is the dominant process. It should be noted that dynamic recovery mechanisms may also contribute to the decrease of the dislocation density in Ag after 8 passes in a manner similar to that shown for Cu processed by more than 8 passes in ECAP [8, 22]. The high stresses at twin tips may assist the cross-slip of dissociated dislocations in Ag which plays an important role in recovery, whereas for Cu, twinning remains marginal even when the ECAP processing is continued to extremely high strains [8, 22]. However, for the case of Ag where the SFE is very low, the influence of twinning on the dislocation density must also be considered as noted in the earlier discussion.

In an analysis of the evolution of the dislocation and twin densities as a function of strain, it is interesting to note that the earlier theory divided the deformation of fcc metals with low SFE into four clearly defined stages [11], where these stages are not related to the well-established deformation stages that describe the plasticity of fcc single crystals with high SFE. For fcc metals with low SFE, the dislocation density increases rapidly at low strains in stage I while the twin probability is very low: this corresponds to the situation prior to the first pass in Fig. 3. Thereafter, the twin density increases at a high rate as a function of strain in stage II and the dislocation density further increases but the rate of increase is gradually reduced with increasing strain: this stage II continues up to about 8 passes for Ag processed by ECAP. In stage III, the twin density further increases while the dislocation density decreases due to the effect of twinning and/or dynamic recovery, where this stage occurs between 8 and 16 passes of ECAP in Fig. 3. Müllner and Solenthaler [11] predicted a stage IV at even higher strains where the twin probability also decreases but this region was beyond the upper limits of the present investigation.

Conclusions

In summary, the evolution of the microstructure was studied in Ag processed by ECAP for up to a maximum of 16 passes where Ag was selected because of the exceptionally low value for the SFE. The results show the change in the microstructural characteristics are consistent with a theoretical model proposed earlier for the deformation of fcc metals and alloys with very low SFE [11]. Up to 1 pass of ECAP there is a rapid increase in the dislocation density while twinning is marginal (stage I), between 1 and 8 passes both the dislocation density and the twin probability increase significantly (stage II) and between 8 and 16 passes the twin probability further increases but there is a corresponding reduction in the dislocation density (stage III). It is concluded that the contribution of twinning to plasticity increases significantly with increasing strain in fcc metals, such as Ag, where the SFE is very low.

Acknowledgements This work was supported in part by the Hungarian Scientific Research Fund, OTKA, Grant Nos. K67692 and K71594 (JG and NQC) and in part by the National Science Foundation of the United States under Grant No. DMR-0243331 (TGL). In addition, JG is grateful for the support of a Bolyai János Research Scholarship of the Hungarian Academy of Sciences and NQC thanks the Hungarian-American Enterprise Scholarship Fund for support. The authors thank Andrea Jakab for the preparation of the TEM samples and JG is grateful to Prof. Geza Tichy for helpful discussions.

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