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Principles of self-annealing in silver processed by equal-channel angular pressing: The significance of a very low stacking fault energy

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ABSTRACT

Experiments were conducted to evaluate the long-term microstructural stability of silver after processing using equal-channel angular pressing (ECAP). The results show that an ultrafine-grained microstructure is produced by ECAP at room temperature but there is self-annealing in the form of recovery and recrystallization during long-term storage at room temperature. In practice, the very low stacking fault energy of silver results in a high degree of dislocation dissociation and thereby hinders recovery by cross-slip and climb. The experiments examine the evolution of microstructure and the mechanical behavior as a function of the storage time after different numbers of ECAP passes. The results demonstrate that the degree and kinetics of self-annealing depend upon the number of passes imposed in ECAP.

1. Introduction

Bulk nanocrystalline (NC) and ultrafine-grained (UFG) materials are currently a major focus in materials science due to their unique properties in comparison to their coarse-grained counterparts [1–3]. Specifically, an important feature of NC and UFG materials is their high strength at ambient temperatures. In practice, the long-term stability of these UFG microstructures is an important prerequisite if these materials are used in any industrial applications. Thus, if the fine grains become coarsened during their service lifetime, their unique properties including their high strength will be lost.

It is generally considered that NC and UFG materials are reasonably stable at their production temperature. Nevertheless, it was shown for nanocrystalline Cu and Ag layers, having a thickness of ~1–20 μm and processed by electroplating, that recrystallization occurs at room temperature (RT) within ~1–2 days of their production [4–7]. This phenomenon is known as "self-annealing". It was suggested that during layer processing the organic additives reduce the interfacial energy anisotropically and additionally pin the grain boundaries thereby stabilizing the small grain size [4,5]. However, the additives disappear from the layers in several hours after plating and this increases the grain boundary energy and leads to an increased driving force for recrystallization. Nanocrystalline Cu and Ag samples processed by the consolidation of nanopowders also show self-annealing at RT during periods of several days after their production [8,9].

One of the most frequently used method for producing bulk UFG metals by severe plastic deformation (SPD) is equal-channel angular pressing (ECAP) where it is possible to produce materials having dimensions of several centimeters in all directions [10]. These materials also exhibit self-annealing and it was shown recently that the UFG microstructure in Cu samples processed by ECAP at RT becomes partially recrystallized during storage at the temperature of processing [11–13]. For example, large recrystallized grains were observed in 99.96% purity Cu 8 years after processing by ECAP [13]. Experiments on 5N purity Cu showed that recrystallization occurred only 2 months after ECAP and the lower recrystallization time for the pure material was explained by the non-availability of the pinning effect of alloying elements on grain boundaries and dislocations [11]. There is also a similar report of partial recrystallization in the severely deformed surface region of a thin wire-drawn copper, having a diameter of 0.1 mm, where this was strongly affected by the impurity concentration [14].

An earlier study reported experiments on pure Ag where the stacking fault energy (SFE) is very low (~16 mJ m^{-2} [15]) and the

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dislocation density after 8 ECAP passes was $\sim 46 \pm 5 \times 10^{14}\,m^{-2}$ which is very high by comparison with other fcc metals such as Al, Au or Cu [16]. This exceptionally high dislocation density is a direct consequence of the very low SFE of Ag because the annihilation of dislocations is hindered by their high degree of dissociation into partials. The result also matches reports on the alloying of Cu with Zn where there is a large increase in the dislocation density due to the reduction of SFE with increasing Zn concentration [17,18]. Some earlier experiments on Ag showed self-annealing of the UFG microstructure at RT which occurred several months after processing through 8 passes of ECAP [19].

The present investigation was initiated to provide a detailed evaluation of the significance of self-annealing in Ag of 99.99% purity when processing by ECAP. The evolution of the microstructure and the consequent mechanical properties were systematically investigated as a function of the time of storage following processing by ECAP through totals from 1 to 16 passes.

2. 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 (corresponding to $0.6T_m$, where T_m is the absolute melting point of Ag). Following homogenization, these billets were pressed through totals of 1, 4, 8 and 16 passes of ECAP using route B_C at room temperature with a pressing velocity of 8 mm s^{-1} and a solid die having an internal channel angle of 90° and an outer arc of curvature of ~20° at the intersection of the two parts of the channel. In this configuration, one pass corresponds to an equivalent strain of ~1 [20]. Following ECAP, the billets were stored at RT and the microstructures and mechanical behavior were examined as a function of the time of storage for periods up to a total of 4 months.

Microstructures were examined periodically by X-ray line profile analysis on transverse sections cut perpendicular to the axes of the billets. The measurements of the X-ray diffraction lines were performed using a special high-resolution diffractometer (Nonius FR591) with CuKα_1 radiation ($\lambda = 0.15406$ nm). The line profiles were evaluated using the extended Convolutional Multiple Whole Profile (eCMWP) fitting procedure [21]. In this method, the diffraction pattern is fitted by the sum of a background spline and theoretical peak profiles. The profile functions are calculated as the convolution of the theoretical size and strain peak profiles. In addition, the theoretical size and strain peak profile functions are calculated on the basis of a model of the microstructure. Based on this model, the crystallites have spherical shape and log-normal size distribution, and the lattice strains are assumed to be caused by dislocations and twins. This method gives both the dislocation density and the twin fault probability with good statistics, where the twin fault probability is defined as the fraction of the faulted {1 1 1} planes along their normal vector. The microstructures of selected samples were examined using a Philips CM-20 transmission electron microscope (TEM) operating at 200 kV. The TEM samples were mechanically thinned to ~80 μm, cooled to liquid nitrogen temperature and then thinned with 6 keV Ar^+ ions from both sides until perforation. Finally, a thin damaged layer was removed using 2 keV Ar^+ ions.

The hardness of the samples was measured using a Vickers microhardness indenter in a Shimadzu-DUH 202 machine with an applied load of 2000 mN. The deformation behavior was studied using uniaxial compression testing with a computer-controlled hydraulic mechanical testing MTS 810 machine. For all of these tests, the direction of compression was parallel to the longitudinal axis of each billet.

3. Experimental results

3.1. Characterization of the microstructure in silver immediately after ECAP

The initial mean grain size of the Ag samples was ~10 μm prior to ECAP but this was reduced to ~5 μm after 1 pass of ECAP. The TEM image in Fig. 1(a) demonstrates that the grains contain both dislocations and twins after processing through 1 pass, where some of the twin boundaries are denoted by white arrows. The dislocation density and the probability of twins were estimated after 1 pass as $\sim 16 \pm 2 \times 10^{14}\,m^{-2}$ and $\sim 0.1 \pm 0.1\%$, respectively, as determined from X-ray line profile analysis. After 4 passes, the grain size decreased to $\sim 160 \pm 50$ nm as shown in Fig. 1(b) and the grain size remained essentially unchanged within experimental error after 8 passes (200 ± 50 nm) and 16 passes (190 ± 50 nm) as illustrated in Fig. 1(c) and (d), respectively. This invariance with increasing numbers of ECAP passes matches recent detailed measurements taken on a high-purity (99.99%) aluminum processed by ECAP through 4–12 passes [22].

Measurements using X-ray line profile analysis after 4 passes showed increases in the dislocation density and the twin probability to $\sim 37 \pm 4 \times 10^{14}\,m^{-2}$ and $\sim 0.7 \pm 0.1\%$, respectively. The dislocation density saturated after 8 passes at $\sim 46 \pm 5 \times 10^{14}\,m^{-2}$ and the twin probability increased to $\sim 0.9 \pm 0.1\%$, whereas after 16 passes the dislocation density was reduced to $\sim 25 \pm 10 \times 10^{14} m^{-2}$ while the twin probability further increased to $\sim 1.5 \pm 0.1\%$. The hardness of the initial sample prior to ECAP was $\sim 0.40 \pm 0.03$ GPa but the hardness increased to $\sim 0.89 \pm 0.05$ GPa after 1 pass and a maximum hardness of $\sim 1.07 \pm 0.05$ GPa was attained after 4 passes. Thereafter, at higher numbers of passes, the measured hardness remained unchanged within experimental error.

3.2. Self-annealing of the severely deformed microstructure during storage at room temperature

Fig. 2 shows the microhardness of samples processed by different numbers of ECAP passes as a function of the time of storage at room temperature, where the lower horizontal line denotes the hardness of the initial sample. It is apparent that the hardness after 1 pass remains unchanged within experimental error even after storage for 4 months. By contrast, the hardness gradually decreases with increasing storage time for the samples processed by 4, 8 and 16 passes thereby demonstrating that the severely deformed microstructure is inherently unstable and subject to self-annealing, most probably by recovery and recrystallization, during long-term storage at room temperature.

True stress–logarithmic strain compression curves are shown in Fig. 3 for samples processed through 1, 4, 8 and 16 passes both immediately after ECAP and after 4 months in storage: for comparison, a stress–strain curve for the initial unpressed condition is also shown. Immediately after 1 pass of ECAP the stress–strain curve shows only a negligible hardening with increasing strain and the compression behavior is similar also for the samples pressed through 4, 8 and 16 passes. After storage for 4 months at RT, there is a similar stress–strain curve with negligible hardening after 1 pass of ECAP whereas the samples pressed through 4–16 passes show a reduction in the yield strength and a very significant strain hardening. This softening of the samples confirms that the change in microhardness evident in Fig. 2 is not related only to a surface effect but rather it reflects a genuine microstructural change occurring during storage at RT. The results in Fig. 3 show that the magnitude of the decrease in flow stress increases with increasing numbers of ECAP passes and this is consistent with the variation of the microhardness values recorded in Fig. 2.