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Thermal behavior of copper processed by ECAP with and without back pressure

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ABSTRACT

Samples of electrolytic tough pitch (ETP) pure copper were subjected to 12 passes of Equal-Channel Angular Pressing (ECAP) at room temperature with and without back pressure. Subsequent annealing was performed to evaluate the influence of back pressure during ECAP on the thermal behavior of ultrafine-grained copper. The microstructural and hardness changes caused by annealing were characterized by orientation imaging microscopy (OIM) and microhardness measurements. The application of back pressure resulted in an earlier drop in hardness upon annealing, which is believed to be associated with a lower critical temperature for the initiation of recrystallization and a rapid coarsening of microstructure. Regardless of whether back pressure was applied or not, structure coarsening during short-time annealing of ECAP-processed copper was governed by discontinuous static recrystallization. This is seen as a result of microstructure heterogeneity. Analysis of recrystallization kinetics was carried out based on observations of the microstructure after annealing in terms of the Avrami equation. The magnitude of the apparent activation energies for recrystallization in the absence of back pressure and in the case of back pressure of 100 MPa was estimated to be ~99 kJ/mol and ~91 kJ/mol, respectively. The reasons for reduced activation energy in the case of processing with back pressure are discussed.

1. Introduction

Equal Channel Angular Pressing (ECAP) as one of severe plastic deformation (SPD) methods has enjoyed great popularity over the last two decades. This technique has the capability to produce bulk ultafine-grained (UFG) materials with dimensions sufficient for potential practical application. For example, high-purity copper processed by ECAP was employed to manufacture premium quality sputtering targets [1,2]. However, UFG materials manufactured by SPD methods are metastable in nature, and some of them readily recrystallize even at ambient temperature [3,4]. As a result of deformation to giant plastic strains involved in SPD processing, a large energy is stored in the material, owing to a high concentration of crystal lattice defects, such as vacancies [5], dislocations [6], and non-equilibrium grain boundaries [7]. From the viewpoint of industrial application, the investigation of the thermal stability of the severely deformed materials, i.e. their resistance to the microstructural restoration processes (recovery and recrystallization), is a necessary step in evaluating their suitability.

A considerable amount of work has concentrated on the thermal stability of ECAP processed materials, in which the influential factors can mainly be put in two categories: (i) extrinsic, processing-dependent parameters (the magnitude of plastic strain [8–10] as well as the strain path, including the processing route [11–13] and the post-ECAP deformation [14,15] and subsequent treatment during annealing [16]), and (ii) intrinsic, i.e. material-dependent parameters, such as the purity [17] and the stacking fault energy (SFE) [18]. These studies show that both the extrinsic and the intrinsic variables play an important role in thermal stability of UFG materials [19].

The application of back pressure (BP) during ECAP processing has attracted wide attention due to the profound advantages it provides. Thus, it has been reported that back pressure assists in achieving greater grain refinement, improvement in mechanical properties and production of high density bulk materials, as well as preventing fracture initiation [20–23]. An important factor is an increase in the fraction of high-angle grain boundaries and a more uniform grain structure the material attains if back pressure is imposed [20,23]. Despite the importance of the ability of these structures to resist restoration, studies of thermal stability of ECAP-induced structures produced under back-pressure are rather scarce. In our preliminary investigation [24], the effect of back pressure on thermal stability was studied for the case of ECAP-processed copper. Thermal stability was found to decrease

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marginally when back pressure was imposed during ECAP. The objective of the present work is to provide a detailed evaluation of post-deformation thermal annealing of copper processed by ECAP with and without back pressure. The evolution of the microstructure with annealing time and the ensuing mechanical properties were investigated in a systematic way as a function of the annealing temperature. Analysis of the recrystallization kinetics in terms of the recrystallization mechanisms was also conducted. The outcomes of the experiments and their interpretation are presented below.

2. Experimental

Electrolytic Tough Pitch (ETP) copper billets of 99.9 wt% purity were studied. Prior to deformation by ECAP, rectangular samples of dimensions $20 \times 20 \times 110$ mm^3 were annealed at 600 °C for 2 h to obtain a homogenous coarse-grained structure. The resulting microstructure is presented in Fig. 1. The average grain size was about 28 μm and the corresponding microhardness was 45.5 kgf/mm^2. ECAP with and without a back pressure of 100 MPa were conducted at room temperature up to 12 passes using an angular die having a channel intersection angle of 90° (see Fig. 2). In the process the sample was rotated about the long axis of the billet by 90° in the same sense between the passes (route Bc ECAP [25]). After deformation, the billets and disc-shaped samples prepared from them were kept in a conventional freezer at −18 °C to suppress potential coarsening of the microstructure. Samples were then annealed at different temperatures using an oil bath, followed by immediate water quenching.

All examinations were conducted on the transverse sections of the middle parts of the billets. A scanning electron microscope (JEOL JSM-7001F FEG SEM) equipped with the Oxford Instruments HKL Channel 5 software package was used to perform microstructural characterization and average grain size calculation. Samples for characterization by electron back scattered diffraction (EBSD) were first ground on 2400-grit SiC paper, then mechanically polished using 3 μm and 1 μm diamond suspension before being electro-polished. Electro-polishing was conducted at 20 °C in an electrolyte of 25% orthophosphoric acid, 25% ethanol and 50% distilled water at a voltage of 7 V with a current of ~140 mA. Misorientations below 2° were excluded from the analysis due to the limitations of the angular resolution of the EBSD technique. At least two EBSD maps for different locations were obtained for each condition to truly reflect the microstructure. Each map contained at least 10,000 grains/subgrains. The average grain size was analyzed using a misorientation threshold angle set at 5°, which means two

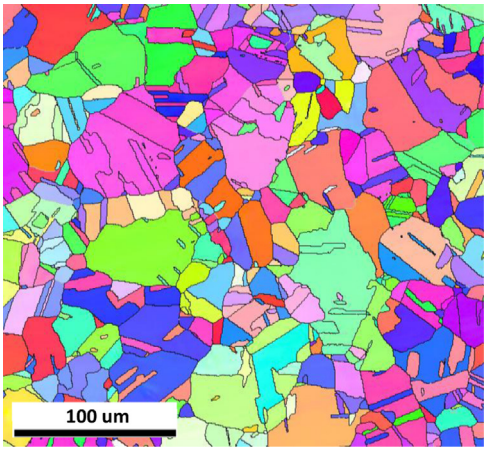

Fig. 1. Microstructure of the initial material.

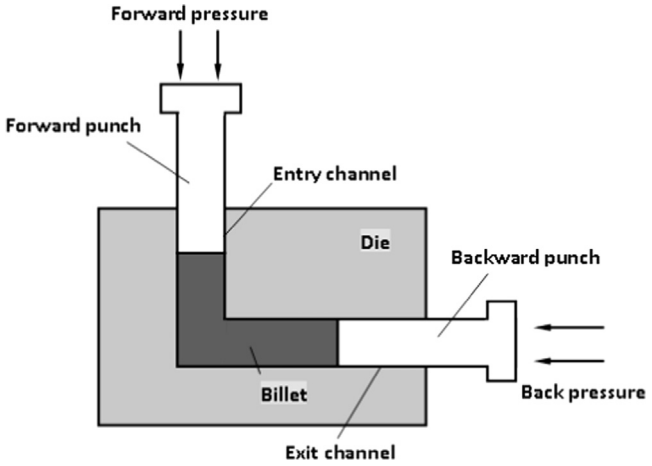

Fig. 2. A schematic diagram of the ECAP process with back-pressure.

neighboring scan points were considered to belong to two different grains if the misorientation between them exceeded 5°. Recrystallized (RXed) and unrecrystallized (unRXed) grains were identified based on the grain orientation spread (GOS) misorientation-based EBSD method [24], whereby GOS values were calculated by averaging the deviations of the orientation of individual points within a grain from its average orientation [26]. In this method, the GOS criterion was first applied to each EBSD map by using a cut-off value, θ_c, determined by the change in slope of the normalized cumulative distribution versus internal misorientation [27]. The microstructure was thus divided into two subsets: the one with internal misorientation exceeding θ_c (i.e. with high stored energy) was classified as the deformed grains; the other with the misorientation angles below θ_c (low stored energy) was considered as a combination of recrystallized and recovered grains. The latter was further partitioned into recrystallized and recovered grains via a grain size cut-off criterion; the threshold was obtained by tracking the slope change of the normalized cumulative grain size distribution, similar to the GOS criterion.

Transmission electron microscopy (TEM) analysis was performed on thin foil samples prepared by a twin-jet polisher in the aforementioned solution at around 0 °C using a Tecnai G2 T20 Twin TEM operating at an acceleration voltage of 200 kV. The average grain size was calculated based on at least 400 (sub)grains per sample utilizing the equivalent circle diameter (ECD) method, where the (sub)grain size was defined as the diameter of a circle with the same area as that of the grain it represents. An aperture diameter of 10 μm was used for selected area diffraction (SAD) pattern acquisition.

The Vickers microhardness was measured by Duramin A-300 hardness tester with a load force of 300 gf and 10 s holding time. The measurements for each condition were performed over a rectilinear grid comprising 16 points with the spacing of 0.5 mm between the points. For tensile tests, cylindrical samples were cut from the longitudinal sections of the middle parts of the billets. The specimens had a gauge length of 10 mm, all other dimensions being scaled down proportionally from those specified in ASTM standard E 8M. At least two room temperature tensile tests were performed for each condition with a constant nominal strain rate of 3×10^{-3} s^{-1} using an Instron 4505 machine.

3. Results

3.1. Deformation microstructure

The characterization of as-deformed microstructures was carried out by means of TEM. As shown in Fig. 3, irrespective of the