A Transmission Electron Microscopy Study on Subgrain and Grain Formation during Equal-Channel Angular Pressing of an 1200 Aluminium Alloy

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Abstract

Equal channel angular pressing (ECAP) was used to produce sub-micron size grain structures in a 1200 commercially pure aluminium alloy. ECAP was conducted at room temperature following the so-called route B_c . A nano-structure was achieved after 6 ECAP passes and the High Angle Grain Boundary (HAGB) misorientation constantly increased reaching a ~70% fraction, among the total amount of boundaries after 8 passes. Transmission electron microscope inspections revealed the mechanism of transformation from a Low-Angle Boundary subgrain microstructure (achieved after the early ECAP passes) to a HAGB structure (refers to the material subjected to the 8th pass). The results attained through ECA-pressing were compared to the ones published in cold-rolling of the same 1000 aluminium alloy series.

1. Introduction

The advantages of fabricating materials for structural components having sub-micron size grained microstructure lie in the increased strength, low-temperature superplasticity and net-shaped bulk production. These potentialities have widely promoted considerable scientific and industrial interest in the past two decades [2-11]. Among them, equal channel angular pressing (ECAP) is, at present, one of the most promising techniques. ECAP have basically the same potentialities and microstructure refining efficiency as of cold-rolling (CR), but as a primary advantage, the materials subjected to ECAP maintain their shape irrespective of the number of passes used to acquire a sub-micrometric grained structure. Due to the geometric constraints of the die, the billet deforms in shear within a small area at the channels intersection and the material is deformed by pure shear [2]. Shear strain per pass through the die is determined by the channels angle and intersecting curvature [5,6,9-11]. Moreover, since the billet shape and section area remain constant, there is no geometric restriction to the maximum strain, i.e. the maximum number of passes to which the material can be subjected. Yet, the limitation lies in a grain refining saturation that occurs after a number of passes which basically depends on the material and on the type of "route", i.e. different ways of rotating the billet after the first pass and before each subsequent pass, used to ECA pressed the material [8]. Actually, extensive TEM studies [5-11] showed the influence of the different ECAP routes, which can either produce almost equiaxed homogeneous microstructures, or induce a more

effective grain refining effect. At the same time, only in recent years, a systematic study on the role of shear, introduced by ECAP, on the subgrain and grain evolution was dedicated [3,12,13,14]. Under severe plastic deformation (SPD), dislocation boundaries evolve within a regular pattern of grain subdivision belonging to two scales [15-22]. A larger scale includes long, continuous dislocation boundaries, commonly called geometrically necessary boundaries (GNBs), and a smaller scale, called incidental dislocation boundaries (IDBs) [19]. Geometrically necessary boundaries play an important role in the deformation process. In order to accommodate the imposed strain during deformation, additional dislocations arrange to form geometrically necessary high-dislocation regions which are associated to dislocation interactions and slip patterns that develop within a deforming grain [18].

The average spacing between dislocation boundaries is a common microstructure parameter in deformed and severely deformed materials, due to its inverse relationship with the flow stress, as extensively documented in literature [16-18, 21]. Yet, a systematic study of misorientation angles and the early formation process of subgrain has been less extensively characterized (e.g. [15,18]). Indeed, the kinetics of subgrain growth depends primarily on the distribution of subgrain size and on the magnitude and distribution of the energies and mobility of the low-angle boundaries (LABs).

The present paper reports a TEM study on the early steps of subgrain formation within high-angle grain boundary (HAGB) networks in a severely deformed AA1200 commercially pure Al-alloy. Moreover the microstructure deformation mechanism, induced by ECAP, has been put in comparison to the structure evolution generated by CR.

2. Experimental

The chemical composition of the AA1200 commercially pure Al-alloy is as follows: 0.7% Si, 0.3% Fe, 0.1% Zn, 0.05% (Cu+Mn). The material was cast and machined by Hydro Aluminium (Norway) in form of rod-shaped bars 10 mm in diameter and 10 cm in length.

ECA-pressing was carried out at room-temperature using a solid die fabricated from a block of SK3 tool steel (Fe-1.1%C) and endowed with two cylindrical channels intersecting at an angle Φ = 90° and a curvature Ψ = 20°, thus inducing an equivalent strain of $\varepsilon \approx 1$ (ε = 1.055) at each pass [1,2].

TEM samples were sectioned along the longitudinal rod axis. Thin TEM foils were prepared by grinding 1-mm thick slices down to a thickness of 70-90 μ m, and then they were thinned by means of a double-jet electropolisher, using a solution of 20% HCINO₄ and 80% of methyl at -15°C and 24V. The TEM inspections were carried out using a Philips CM20 operating at 200 kV and equipped with a double-tilt specimen-holder and energy dispersive spectrum (EDS) micro-analysis. Thin foils were tilted to align the specimen at low-index zone axis almost parallel to the incident beam direction, yielding a better image contrast. For Moiré fringes angular misorientation measurements, a full description is reported in [23,24].

3. Results and Discussion

Figure 1 shows a TEM dark-field (DF) image, along with the corresponding SAED patterns, of the material subjected to 1 ECAP pass. The microstructure was characterised

by the formation of several new High-Angle Grain Boundaries (HAGB) and a diffuse network of Low-Angle Boundaries (LAB) subgrains, decorating the grains interior (se also Fig. 2 (a)). The substructure basically originated from the induced shear bands and was mainly oriented along the deformation shear direction, i.e. at an angle of ~45° to the pressing direction. Figure 2 (a) to (d) shows the microstructure of some of the severely deformed samples. The mean width and value of the observed Moiré fringes remained practically unchanged over the different ECAP passes.



Figure 1: Low-magnification TEM dark-field (DF), and the corresponding SAED patterns of the material subjected to 1 ECAP pass.



Figure 2: BF-TEM images showing the microstructure of some of the severely deformed samples; 1 pass (a), 2 passes (b), 8 passes (c), 7 passes (d). A very fine subgrain embedded by diffuse Moiré fringes is reported (d), and the configuration of intersecting two subgrains, producing a rather complex Moiré fringes intersection. A and B subgrains produce two different Moiré fringes system with the belonging grain due to their different reciprocal orientation. Moreover, they overlap to some extent (A/B in the figure) producing a third different Moiré fringes system in between.

Figure 2 (a) shows the grain boundary formation in a material subjected to 1 ECAP pass. Figure 2 (b) reports (indicated by arrow) a typical example of subgrain boundary formation by the dislocation rearrangement along wall configurations (after 2 passes). A complex low-angle boundary system was documented, in which a very fine subgrain is embedded by diffuse Moiré fringes (material subjected to 7 passes: Fig. 2 (d)). The very low-angle boundary formation eventually transformed at first into medium, and then to high-angle boundaries, with increasing strain (i.e. ECAP passes). Figure 2 (c) shows the elongated grained structure characterizing the 8 passes material. New fine grains were also found to form near the original grain boundaries, where strain incompatibility occurs between adjacent grains leading to a rather complex local deformation pattern [8].

Figure 3 shows the plot of GNBs and IDBs misorientation as a function of the strain in which IDBs have a trend practically flat with increasing strain, whilst GNBs revealed a constant positive trend whose slop was calculated to be 0.44. CR of the same 1000 aluminium alloys series [19] reported values for GNB and IDB of 0.7 and 0.4, respectively. This discrepancy is due to a non-uniaxial deformation induced by ECAP, respect to the uniaxial occurring in CR. As a matter of facts, the deformation in ECAP follows the shear bands induced by the geometry of the two channel intersection of the die. More specifically, the route B_c , involving a rotation of +90° at each pass, induces a deformation essentially in two mutually almost perpendicular directions at every pass, and reversing the direction sense at every two passes.



Figure 3: GNBs and IDBs misorientation as a function of the true strain.



Figure 4: Open and solid circle data points refer respectively to commercially pure Al drawing data by Hansen (1969) and to commercially pure Al cold-rolled data by Schuh et al. (1974) [25], while solid up-triangle data points refer to 1200 ECApressed material. Solid connecting square data points refer to HAGBs (solid) and LABs (open) fraction.

This peculiarity results in the formation of quasi-equiaxed grains at each even pass which break-up, if not destroy, the long bamboo-like grain, formed essentially by GNB structure, allowing the high-angle GNBs to propagate in between the formerly produced long bamboo-like structures. Thus, most of the IDBs are replaced by GNBs causing the former to remain practically constant in misorientation with increasing strain, because most of the medium-angular-range IDBs transform into GNBs.

This microstructure evolution is also responsible for the lower increasing of GNBs misorientation. The process of transformation of IDBs into GNBs is rather slower in CR due to the uniaxial straining which continues to shrink, with the strain, the lateral GNBs

spacing without let them cross to connect to a neighbouring parallel GNB, which would increase GNBs misorientation at the expense of IDBs.

The described microstructure had basically the same aspect as the one subjected to highstraining by cold rolling. Severely deformed cold-rolled commercially pure aluminium alloys exhibit the same bamboo-like very elongated grains decorated by diffuse transverse and almost parallel LABs which resemble to section out the long bamboo grains [15].

Moreover, cold-rolled commercially pure aluminium exhibits different microstructure evolution with increasing strain. At low strains the majority of the grains are subdivided into typical cell blocks (CBs) with dense dislocation microbands (MBs) having an angle of ~40° respect to the rolling direction. With large strain (typically for 2.5 and above) the microstructure evolves into lamellar dislocation boundaries (LBs) almost parallel to the rolling direction. At the medium range, the structure is mixed with some grains having a cell block structure and others subdivided by lamellar boundaries typical of large strain microstructure [22]. CBs are typically 10-times wider than MBs and contain approximately equiaxed cells several times larger than those in MBs. The misorientation between neighbouring CBs is much larger than between cells within a CB or MB. [15,19,20]. In the majority of the cases, GNBs are separated by only one dislocation cell and the microstructure resembles a bamboo-like structure in which IDBs are located across the GNBs having boundaries almost perpendicular to those of GNBs [15,19,20]. Some IDBs, at large strains, may develop such high misorientation angles to become GNBs. Thus, the boundaries may be integrated into lamellar structure or interconnect lamellar boundaries [20].

In brief, the flow stress may be expressed as the sum of strength contributions from the GNBs (mainly constituted by MBs and LBs), and from IDBs forming low-angle boundaries between the GNBs. GNBs behave as strong barriers to the slip due to their high dislocation concentration. On the contrary, the lower angle IDBs are assumed to be penetrable by mobile dislocations. This way, the flow may take place within the channels formed by the lamellar boundaries with a slip length related to the distance between the lamellar boundaries [21,22].

The evolution of the microstructure during ECAP deformation can be described as the formation and evolution of cell block structures. At low and medium strains, the CBs, formed by grain subdivision, are delineated by extended dislocation boundaries having misorientation angles significantly larger than that between individual cells [26]. By increasing the strain, these blocks become thinner; correspondingly, the spacing between the CB boundaries decreases and misorientation across them become larger. Eventually, large misorientation can develop across closely spaced CB boundaries [26]. At very large strains, a layered CB structure is still a characteristic feature, although equiaxed structures are also observed. The building blocks of these equiaxed structures are subgrains with typically both low and medium angle boundaries which eventually, under further straining, transform into high angle boundaries acquiring a grain microstructure character.



Figure 5: Non-equilibrium and equilibrium grain boundaries. Material subjected to 3 ECAP passes: BF (a), DF (b).

(a)

Figure 4 reports the substructure evolution and the HAGB fraction as a function of the true strain. In spite of increasing of the HAGB fraction with the strain, a considerable fraction of new fine grains revealed bent boundaries also at the maximum strain (i.e. 8), not straight boundaries (Fig. 5). This is due to an instability state of such grains, revealing that the fine grained microstructure was not still stable at a strain of 8. In other words, the number of ECAP passes, that is the maximum strain, had to be prolonged to eventually reach a saturation limit for the formation of new HAGBs, as the fraction of HAGBs obtained after the 8th pass was ~70% of the total amount of boundaries decorating the microstructure. The authors believe that the microstructure of the material subjected to the minimum strain belonging to the saturation limit does not bear a considerable fraction of bending grain boundary. That is, the grain instability is lost when a saturation limit is reached. A picture model describing the microstructure evolution with the ECAP passes was proposed in a related manuscript [27,28].

4. Summary

The early subgrain-to-grain evolution, induced by equal channel angular pressing in a commercially pure 1200 aluminium alloy, was investigated. Transmission electron microscopy techniques were extensively used in order to look in detail the process of formation of very low-angle boundaries (i.e. less than $1.5 - 2^{\circ}$) which are hard, if not impossible, to be detected by field-emission gun scanning electron microscopy technique equipped with electron back-scattered diffraction.

- The early very low-angle boundaries formation were mostly characterized by producing Moiré fringes due to their angular mismatch; these fringes were measured yielding the exact value of the lattice angular misorientation across each boundary. They ranged 0.4 to 5.2° unrespectable of the number of ECAP passes.
- ii) The maximum fraction of HAGB after 8 ECAP passes was of ~70% and thus a saturation limit had still to be reached. This is the major reason for having observed, still after 8 ECAP passes, a certain amount of very low-angle subgrains (producing Moiré fringes) and a considerable fraction of bended grain boundaries evidencing an instability state of the microstructure.
- iii) The microstructure evolution, induced by ECAP, was compared to the one involved in CR aluminium alloys. To some aspects, the two deformed microstructure have similar properties. In particular the sub-structure trend calculated for the ECA-pressed material followed exactly the trend reported in literature by Hansen (1969) and Schuh (1974) for

(b)

commercially pure aluminium alloys. As a counterpart, the IDBs and GNBs misorientation trend with the strain was found to substantially differ from the trend found by Liu, Huang, Lloyd and Hansen of the same alloy subjected to cold rolling, the discrepancy being basically due to the non-uniaxial deformation induced by ECAP, respect to the uniaxial occurring in CR, and to the deformation peculiarity induced by the route B_c .

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