Microstructural Evolution in a Commercial Al-Mg-Sc Alloy during ECAP at 300°C

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Abstract. Microstructural evolution taking place during equal channel angular pressing (ECAP) was studied in a commercial coarse-grained Al-6%Mg-0.4%Mn-0.3%Sc alloy at a temperature of 300°C ($\sim 0.6 T_m$). Samples were pressed using route A to a total strain of 12 and quenched in water after each ECAP pass. ECAP at moderate-to-high strains leads to the formation of a bimodal grain structure with grain sizes of around 1 and 8 µm and volume fractions of 0.3 and 0.6, respectively. The development of new-grained regions has been shown to result from a concurrent operation of continuous dynamic recrystallization that occurs during deformation and static recrystallization that occurs during each ECAP cycle by the exposure of the as-deformed material in the die kept at 300°C for around 1.5 minutes. The microstructural development during warm-to-hot ECAP is discussed in terms of the enhanced driving force for recrystallization, resulting from the evolution of high-density dislocation substructures due to the localization of plastic flow and inhibition of recovery in the present alloy.

Introduction

The processing of Al alloys through equal-channel angular pressing (ECAP) has been known to result in the production of a submicrocrystalline grain structure [1]. This technique is now gaining recognition with the incorporation of the concept of intense plastic deformation (IPS) and shows promises of producing high performance bulk materials, which may be successfully utilized in industrial applications. In addition, ECAP is now considered to be one of the most valuable scientific tools for studying the microstructure formation during large strain deformation. For cold-to-warm ECAP of Al alloys, it is well acknowledged that the new grains evolve in accordance with continuous dynamic recrystallization (cDRX) [2]. Namely, some low-to-medium angle subboundaries, which are sometimes called *deformation bands* or *geometrically necessary boundaries*, may be generated in the original grain interiors at relatively low strains due to strain heterogeneity and result in grain subdivision [3,4]. They are progressively transformed into high-angle boundaries with further deformation leading to a full development of new grain structures at large strains [4].

However, some heavily alloyed Al alloys are categorized as hard plastic materials; i.e. they show high resistance to deformation during ECAP because of their limited ductility and formability at low- to-intermediate temperatures [e.g. 5]. In contrast, by increasing the pressing temperature the plastic workability may be improved and some additional benefits may be obtained due to the decreasing strength of the material. With these advantages, ECAP performed at elevated temperatures could become a very promising technical procedure with great commercial potential. Unfortunately, the effect of ECAP temperature on microstructural development is still unclear due to the lack of related experimental data, especially for heavily alloyed Al alloys. Factors affecting grain refinement, as well as *the mechanisms* of deformation-induced grain formation in such materials, are currently being debated and are not clear. Therefore, the aim of this present research is to study the microstructural evolution in a commercial coarse-grained Al–6%Mg–0.3%Sc alloy subjected to ECAP at 300°C (about $0.6 T_m$). A pre-extruded rod,

which may be the most suitable object for evaluating the potentiality of grain refinement under *industrial conditions*, was used as the starting material for this work [6]. Specific attention was given to clarify the main features of the new grain structure developed in this material and to discuss the mechanisms of new grain formation in detail.

Experimental Procedure

A commercial Al-Mg-Sc alloy with a chemical composition of Al–6%Mg–0.4%Mn-0.3%Sc (in mass %) was used in this study. It was fabricated by casting in a steel mold and then homogenized at 520°C for 48 h. The as-received alloy was first extruded at 390°C to a strain of 0.7, followed by annealing at 400°C for 1h. After extrusion and annealing, the alloy was composed of partially recrystallized fine grains developed along coarse elongated grains lying parallel to the extrusion axis (see Fig. 1(a)). The coarse grains had a size of around 170 and 70 μm in longitudinal and transverse directions, respectively. The fine grains had an average size of 4.4 μm and a volume fraction of 0.35. Two types of dispersion particles were identified by TEM, coherent Al_3Sc dispersoids with an average size of about 10-20 nm, and incoherent Al_6Mn precipitates of about 200 nm [6]. Samples for ECAP were machined parallel to the extrusion axis into rods with a diameter of 20 mm and a length of 100 mm. ECAP was carried out repeatedly at 300°C up to a strain of 12 with a strain of about 1 in each pass using route A, i.e. orientation of the rods was not changed at each pass. The pressed samples were cooled in water after each passage through the die. The mean time interval between the entrance of the heated sample into the ECAP die and its immersion after pressing into water was about 1.5 min. Deformed microstructures were examined in the central regions from a section parallel to the pressing direction using optical microscopy, TEM and EBSP techniques, which are described in detail elsewhere [6].

Results

Deformation microstructure. A series of typical microstructures (a) before ECAP ($\varepsilon = 0$) and (b) - (d) evolved at various strains is represented in Fig. 1. It is seen that the original coarse grains that are present
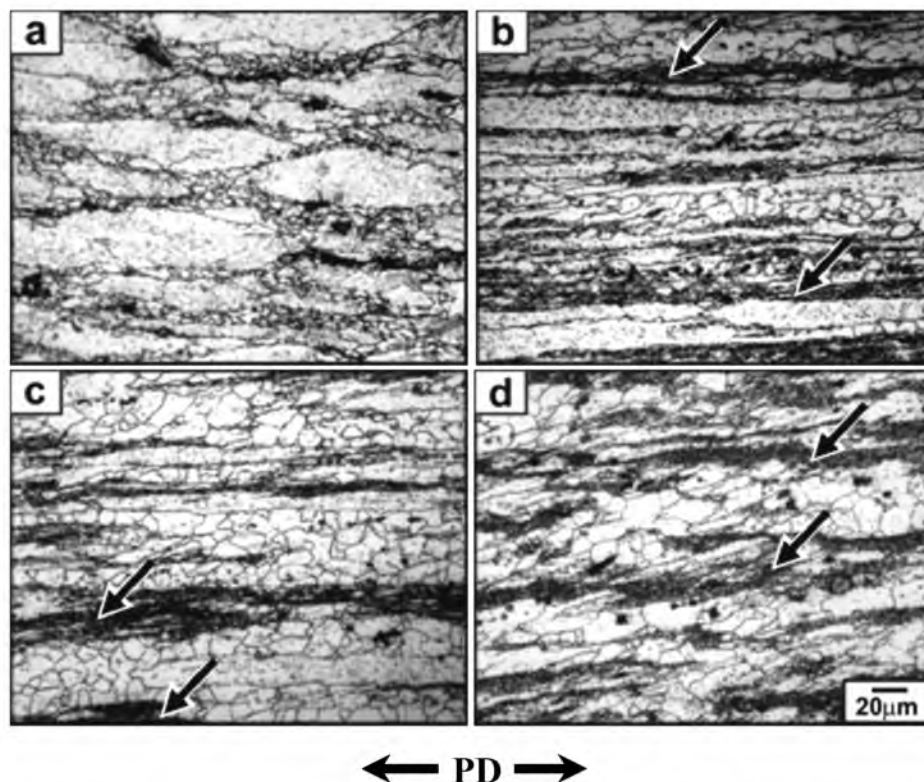

← PD →

Fig. 1 Typical optical microstructures developed in Al-6%Mg-0.3%Sc-0.4%Mn alloy after ECAP at 300°C: (a) ε=0; (b) ε=1; (c) ε=4; (d) ε=12. Dark regions arrowed in (c) and (d) are composed of fine equiaxed grains with the grain size of around 1 μm. PD indicates the pressing direction.

in the initial structure (Fig. 1(a)) are pancaked along the pressing direction (PD) and, concurrently, new finer grains are created by ECAP, first in the former mantle regions and then on all the original coarse grains (Figs. 1(b)-(d)). Note that the newly evolved ECAP microstructure is highly non-uniform and consists of two structural components with a bimodal distribution of the grain size at all strains investigated. One of them is comprised of fine-grained bands, which appear in dark-color regions aligned along the PD, as shown by arrows in Figs. 1(b)-(d). The crystallites in these bands are around 1 μm. The other is composed of relatively coarse grained regions with grains of around 8 μm, which maintain an essentially equiaxed shape after each ECAP pass. Furthermore, the deformed original grains were also present at all strains investigated. At moderate-to-high strains above 8, the volume fractions of fine- and coarse-grained components were as high as 0.3 and 0.6, respectively. It is clear that no material with a submicron grain size could be obtained by ECAP at 300°C. The two types of grained structures with significantly different grain sizes are assumed to be originated from different structural mechanisms operating under the present ECAP conditions. Let us consider the development of both grain structures in more significant details.

OIM microstructures. Fig. 2 shows typical OIM pictures taken at a larger magnification for the samples processed to (a) ε=1 and (b) ε=12. Here the different grayscale levels indicate the different crystallographic orientations and the orientation differences (Θ) between neighboring grid points, $2°<\Theta<4°$, $4°<\Theta<15°$ and $\Theta>15°$ are marked by thin white, narrow black and bold black lines, respectively. Several important results that were derived from the EBSP analysis can be described as follows.

First, highly inhomogeneous deformation can be introduced into the present alloy during ECAP, leading to high strain and misorientation gradients in the grain interiors (see Fig. 2(a)). The EBSP measurements showed that the internal lattice distortion angle that developed in the mantle regions after the first ECAP may be more than $2°/\mu m$; which corresponds to the minimal stored dislocation density of around 4×10^{14} m^{-2} [3]. Such highly distorted areas may also develop frequently in some local regions within the strain-hardened original grains after large strains. This suggests that dynamic and static recovery hardly occur in the present alloy at 300°C and sequentially, structural changes during repeated ECAP are mainly affected by a strong strain accumulation that is applied in each ECAP pass.

Secondly, Fig. 2(a) shows that new boundaries with low-to-moderate (narrow lines) and even high angle (bold lines) boundary misorientations evolve in areas with high local lattice rotations. The crystal orientation is changed and/or altered in the regions fragmented by these boundaries thus, they may correspond to the boundaries of deformation or microshear bands, as discussed in detail elsewhere [7]. Several sets of these subboundaries that developed in various directions can be observed more clearly in areas adjoining the original mantle regions, where frequently elongated new grains are evolved, as indicated by arrows in Fig. 2(a). With further ECAP to ultrahigh strains, such fine grains surrounded by high-angle boundaries are frequently but inhomogeneously developed in colonies accompanied by the
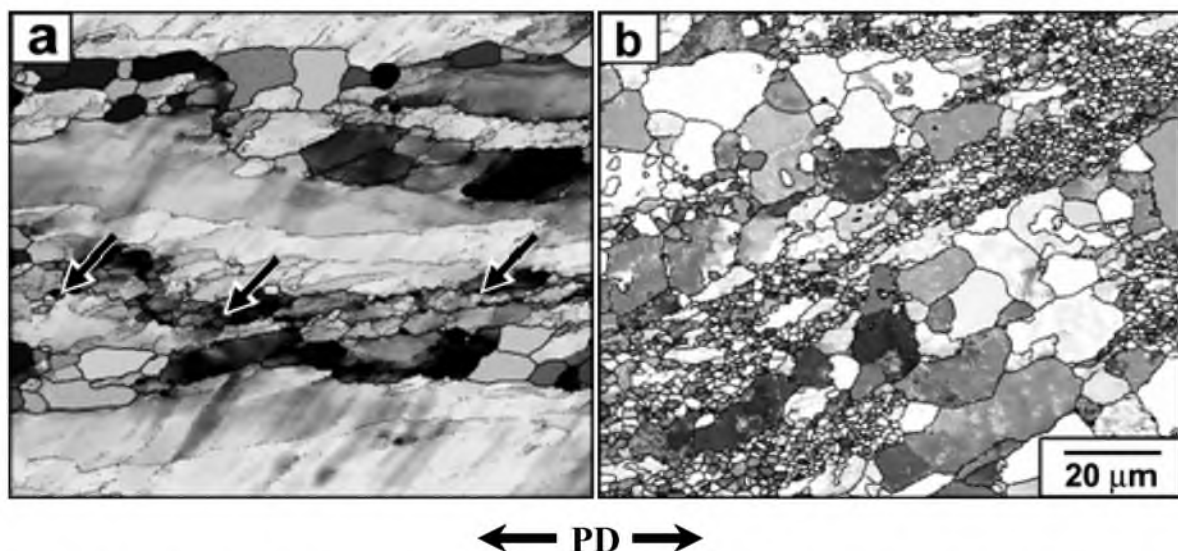

← PD →

Fig. 2 Typical OIM microstructures developed in Al-6%Mg-0.3%Sc-0.4%Mn alloy during ECAP at T=300°C: (a) ε=1; (b) ε=12. PD indicates the pressing direction.

evolution of elongated crystallites and deformation bands (see Fig. 2(b)). It is known [4-7] that evolution of deformation bands can play a key role in the occurrence of grain refinement during IPS in some metallic materials; in other words, a gradual increase in the number and misorientation of these bands and their conversion into high-angle boundaries can result in the formation of a new grain structure through cDRX [4,6,7]. The present data suggest that a similar scheme of new grain formation can be used to describe the microstructural evolution in the fine-grained structural component during ECAP of the current Al-Mg-Sc alloy at 300°C.

Thirdly, Fig. 2 shows that coarse-grained structural components mentioned in Fig. 1, appear frequently after the first ECAP pass at the sites of the original fine-grained structure in the mantle regions (see Figs. 1(a) and 2(a)). The fraction of these grains increases rapidly with repeated ECAP to consume some parts of the deformed microstructure that especially exhibit a large level of the lattice distortion and/or misorientation gradients. Note that such grains can mainly be classified as statically "recrystallized" ones [see 8], because their average internal lattice distortion angle does not exceed 0.2°/μm and their coincidental index (CI) was measured to be normally more than 0.5-0.6. Some of them show interior low-angled boundaries (white lines), as shown in Fig. 2(b). However, by referring to the almost identical equiaxed shape of most of these grains, which appears in all stages of deformation (see Fig. 1), we can suggest that they can be reproduced during each ECAP pass by static, rather than dynamic processes.

TEM microstructures. Typical TEM microstructures evolved in the samples pressed to ε=4 (see Fig. 1(c)) are illustrated in Figs. 3(a)-(c). These show (a) banded dislocation arrays with a high density of lattice dislocations; (b) well-developed (sub)grain structures composed of crystallites separated by moderate-to-high angle dislocation (sub)boundaries (see the SAED pattern inserted); and (c) coarse recrystallized grains, which are formed in various parts of the ECAPed material at moderate-to-high strains. Note that (i) the dislocation density in some areas with large amounts of lattice dislocations introduced by ECAP was evaluated as 10^{14}-3×10^{14} m^{-2}, although it may often be so high, that separate lattice dislocations can hardly be resolved by the TEM technique, as shown in Fig. 3(a); (ii) there are relatively high dislocation density values within small (sub)grains represented in Fig. 3(b), which contrast with the virtually dislocation free interiors of coarse recrystallized grains denoted in Fig. 3(c). The latter
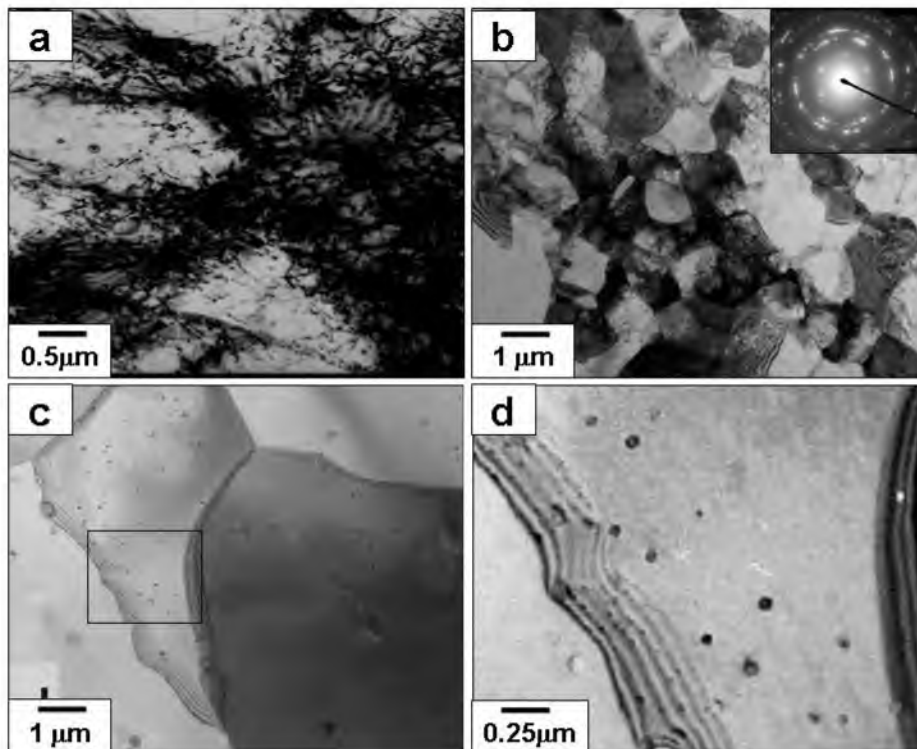

Fig. 3 Typical TEM microstructures developed at ε=4 in Al-6%Mg-0.3%Sc-0.4%Mn alloy during ECAP at 300°C; SAED pattern in (b) was taken from the area with a diameter of 5 μm; (d) shows an enlarged portion outlined in (c).

suggests that by comparing them to the fine-grained (sub)structures in Fig. 3(b), those coarse grains have not suffered any remarkable strain after their formation. Thus, the structures in Fig. 3 indicate (a) the inability of the material to recover completely; and (b) and (c) some independent evidence that during warm-to-hot ECAP of a heavily alloyed Al alloy, new grains may form both dynamically and statically by mechanisms of evolution of dislocation subboundaries and nucleation and limited grain growth, respectively.

Fig. 3(d) shows the enlarged portion outlined in Fig. 3(c). There is some strong interaction between migrating boundaries of recrystallized grains and nanoscale dispersoid particles, which are located in the grain boundary regions and pin the boundaries. It is worth to suggest that in the present Al-Mg-Sc alloy, these dispersoids may be the coherent $L1_2$-Al_3Sc particles that were described in previous *Sections*. However, the precipitates in Fig. 3(d) hardly exhibit any "coffee bean-like" contrast corresponding to the coherent particles in the TEM images [6] and also have an unexpectedly large size of around 60-80 nm.

Discussion

We conclude from the present observations that the development of a new grain structure in the Al-Mg-Sc alloy during ECAP at 300°C can result from the simultaneous action of cDRX that occurs during deformation followed by static recrystallization that occurs through annealing of the as-deformed material by its exposure in the ECAP channel[1] and/or reheating between passes [5]. However, the formation of "coarse" statically recrystallized grains even in a material containing a respectable amount of fine coherent particles, which, as known, can serve as very effective pinning agents and, therefore, prevent nucleation and grain growth [2,9] is rather surprising. A criterion for the restraining effect of small intermetallic particles to influence recrystallization was proposed in [2] in terms of the driving force for recrystallization, F_d ($= 0.5Gb^2\Delta\rho$), and the restrained force of the particles, F_r ($= 1.5 \, f\gamma / R$), where G is the shear modulus; b is the Burgers vector; $\Delta\rho$ is the dislocation density; f is the volume fraction of particles; γ is the grain boundary energy; R is the particle radius.

Driving force. For the present deformation conditions, the values of the parameters for the calculation of F_d are given as follows: *Burgers vector*, b=2.8×10^{-10} m [10]; *shear modulus at T=300°C*, G=2.16×10^{10} Nm^{-2} [10]. A substitution of these values leads to F_d ~10^5 Nm^{-2} for $\Delta\rho=10^{14}m^{-2}$ and F_d ~10^6 Nm^{-2} for $\Delta\rho=10^{15}m^{-2}$.

Restrained force. To evaluate the restrained force of the Al_3Sc particles, F_r, the following values were used: *the grain boundary energy*, γ=0.3 Jm^{-2} [11]; *the volume fraction of the Al_3Sc phase in Al at 300°C and the 0.3 wt % Sc, in accordance with the lever rule*, f=7.06×10^{-3} [9]; *the particle radius*, R~5×10^{-9}-10^{-8} m (see *Experimental* section). The calculation resulted in the F_r values lying in the interval ~3×10^5– 6×10^5 Nm^{-2}. For the occurrence of recrystallization, F_d was determined to be $\geq F_r$ [2,12], i.e. $\Delta\rho$ must be at least $\geq 3\times10^{14}m^{-2}$. Thus, despite the very approximate nature of the calculations, a highly stored dislocation density is apparently required to "trigger" recrystallization in the present Al-Mg-Sc alloy. Also note that Al-based alloys having high stacking fault energy are normally considered to be the "recovery materials", in which dynamic/static recovery operating extensively under warm-to-hot deformation conditions can effectively decrease the dislocation density stored during deformation and suppress recrystallization [2].

However, certain areas with an enhanced dislocation density can develop in the present alloy (see Fig. 3(a)); therefore, the static recrystallization takes place under warm-to-hot ECAP conditions. This may be caused by a very inhomogeneous shear deformation with intense deformation/shear bands that are introduced by the ECAP, as shown, for instance, in Figs. 2(a) and 3(a), and reduced mobility of the lattice dislocations due to their strong interaction with the Al_3Sc particles and also the Mg atoms present in the solid solution [13]. Because of the low rate of recovery, the dislocation rearrangement becomes difficult and ECAP carries a high density of dislocations. Also note that the nucleation of the statically

recrystallized grains occurs more likely in the mantle regions, where the strain gradients are greater [e.g. 5-7]. On the other hand, the occurrence of recrystallization accompanied by the grain boundary migration may weaken the restraining effect of the Al_3Sc phase. It has been pointed out [2] that the passage of high-angled boundary through the coherent particle results in the particle losing coherency. The latter may in turn lead to a rapid particle coarsening due to coagulation, as it was noticed in Fig. 3(d), and, hence, decrease F_r. At larger strains, this creates a large possibility for "coarse" recrystallized grains to regenerate easily in the same local places after each ECAP pass.

In contrast, in the other places where the microstructural conditions are not satisfied with the inequality $F_d \geq F_r$, i.e. the stored dislocation density is lower and/or the restraining effect of secondary particles is larger, no static recrystallization takes place. Instead, the progressive evolution of the microstructure may occur through a gradual strain accumulation within deformation/shear bands in each ECAP pass and result in a transformation of their boundaries into high-angle ones in accordance with cDRX mechanisms, which are discussed in details elsewhere [4,6,7].

Summary

Microstructural evolution in a commercial Al-6%Mg-0.3%Sc alloy subjected to ECAP to a total strain of 12 at 300°C was examined in this work. The main results can be summarized as follows:
(1) A bimodal grain structure with grain sizes of around 1 and 8 μm and volume fractions of 0.3 and 0.6, respectively, develops during ECAP at moderate-to-high strains. The development of two structural components with significantly different grain sizes results from the simultaneous operation of continuous dynamic recrystallization (cDRX) that occurs during deformation, and static recrystallization that occurs during each ECAP pass by the exposure of the as-deformed material in the ECAP channel held at 300°C.
(2) Intense shear deformation applied during each ECAP pass results in high strain / misorientation gradients and introduces deformation and/or microshear bands in the original grain interiors. Structural changes in some parts of the deformation microstructure are characterized by the evolution of these deformation bands, followed by the formation of new fine grains at high strains in accordance with cDRX.
(3) Coarser statically recrystallized grains first form in the original mantle regions that developed during a previous thermomechanical treatment, and then in some parts of the grain interiors to consume areas with high local dislocation density. In the current heavily alloyed Al alloy, an enhanced driving force for recrystallization is provided even under warm-to-hot deformation conditions due to the strong localization of plastic deformation introduced by the ECAP and the inhibition of recovery by the presence of coherent dispersion particles and a high concentration of Mg atoms in the solid solution.

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