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Precipitation and mechanical properties of supersaturated Al-Zn-Mg alloys processed by severe plastic deformation

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Abstract. Supersaturated Al-4.8Zn-1.2Mg-0.14Zr and Al-5.7Zn-1.9Mg-0.35Cu (wt.%) alloys were processed by Equal-Channel Angular Pressing (ECAP) at 200°C. The crystallite size distribution and the characteristic parameters of the dislocation structure of both Al matrix and precipitates were determined by X-ray diffraction line profile analysis, which has been complemented by transmission electron microscopy (TEM) observations. The results show that severe plastic deformation promotes the precipitation process and consequently has a strong influence on the strength of these alloys.

Introduction

Severe plastic deformation (SPD) techniques are generally applied for obtaining ultrafine-grained (UFG) microstructure in bulk metals and alloys [1]. Equal-Channel Angular Pressing (ECAP) is the most often used method among SPD procedures since it results in homogeneous UFG microstructure without contamination and changing the dimensions of the bulk specimen [1]. For understanding the mechanical behavior of materials produced by ECAP it is necessary to characterize their microstructure.

It is well known that X-ray diffraction line profile analysis is an effective tool for studying the microstructure of UFG materials, in which both the small crystallite size and the long-range strain fields of the lattice defects cause broadening of line profiles. In the SPD processed materials, where the lattice distortions are primarily caused by dislocations, the strain broadening of the line profiles can be expressed in terms of the characteristic parameters of the dislocation structure [2,3].

In this study the microstructure of Al-4.8Zn-1.2Mg-0.14Zr and Al-5.7Zn-1.9Mg-0.35Cu (wt.%) alloys processed by ECAP at 200°C is investigated. The crystallite size and the dislocation density for both the matrix and the precipitates are determined by X-ray diffraction line profile analysis. Morphology, size and dispersion of precipitates are studied by transmission electron microscopy (TEM). Furthermore, the yield strength of the materials is correlated to the characteristic parameters of their microstructure.

Experimentals

Billets of Al-4.8Zn-1.2Mg-0.14Zr and Al-5.7Zn-1.9Mg-0.35Cu (wt.%) alloys were processed by ECAP. Before ECAP processing, the material was subjected to solute heat treatment at 470 ºC for 30 minutes and water-quenched (Q), resulting in supersaturated solid solution. Cylindrical rods of 10 mm in diameter and 70 mm in length were pressed through the ECAP die with 90° intersecting channels. Eight passes were performed following route B_C (the billet was rotated around its longitudinal axis by 90° between intermediate passes). The ECAP procedure was carried out at 200°C because at lower temperatures the high rigidity of the specimens results in crack formation and breaking of the billets. Owing to the relatively high temperature of ECAP, precipitates are formed from the supersaturated solid solution. Several important mechanical and precipitation properties of these alloys after quenching have been reported before [4,5].

The phase composition of the specimens was determined by X-ray diffraction using a Philips X'pert powder diffractometer with a Cu anode. For studying the microstructure, the X-ray line profiles of both the Al matrix and the precipitates were measured on the cross section of the billets after the last ECAP pass by a high-resolution double-crystal diffractometer (Nonius, FR 591) using CuKα_1 radiation. The peak profiles were evaluated by the Multiple Whole Profile (MWP) fitting procedure described in details in Ref. [6]. In the MWP method, the Fourier coefficients of the experimental profiles are fitted by the theoretical functions which are calculated on the basis of a model of the microstructure. In this model the crystallites have spherical shape and their size distribution is given by a log-normal function, furthermore dislocations are regarded as the main lattice defects. As the results, the method gives the area-weighted mean crystallite size ($\langle x \rangle_{area}$) and the density of dislocations (ρ).

The microstructure was also investigated by a JEOL-200CX transmission electron microscope (TEM) operating at 200 kV. The TEM foils were taken from the centre of the cross section perpendicular to the axis of the output channel of the last ECAP pass. The yield strength of the specimens was determined as one-third of the hardness measured on the cross section of the billets by Vickers indentation.

Results and discussion

Figure 1a shows the X-ray diffraction pattern taken on the cross section of the Al-5.7Zn-1.9Mg-0.35Cu specimen processed by 8 ECAP passes. Beside the strong peaks (denoted by open circles) corresponding to the reflections of the Al matrix, several weaker peaks (denoted by solid squares) were also observed indicating the existence of hexagonal $MgZn_2$ precipitates (η phase). The $MgZn_2$ particles were formed from the supersaturated solid solution during high temperature ECAP in both Al-4.8Zn-1.2Mg-0.14Zr and Al-5.7Zn-1.9Mg-0.35Cu alloys investigated here. The η precipitates develop usually during ageing from the Guinier-Preston (GP) zones through the intermediate η' phase [7,8]. The particles of η phase are regarded as incoherent precipitates in the Al matrix, contrary to the coherent GP zones and η' phase [8,9].

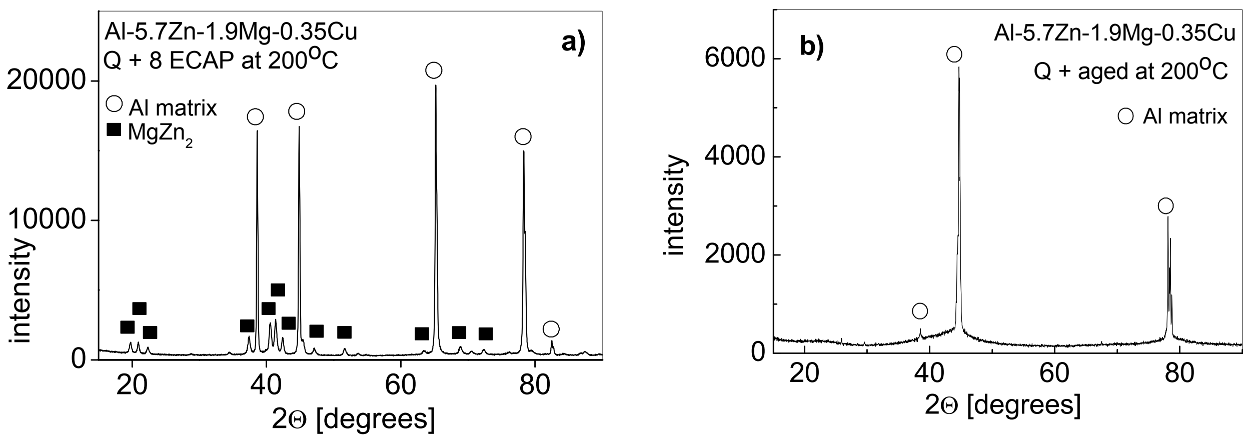

Figure 1: *The X-ray diffractograms taken on the Al-5.7Zn-1.9Mg-0.35Cu specimens (a) quenched and processed by ECAP at 200°C and (b) quenched and aged at 200°C for 20min.*

In order to reveal the effect of severe plastic deformation on the precipitation in Al-Zn-Mg alloys, the initially annealed solid solutions were also aged at 200°C for 20 min which is the approximated duration of the ECAP procedure. The X-ray diffractogram taken on the aged Al-5.7Zn-1.9Mg-0.35Cu sample (see Fig. 1b) shows that $MgZn_2$ precipitates have not formed yet after 20 min ageing at 200°C without any deformation. Comparing the diffractograms in Figs. 1a and 1b, we can conclude that the formation of η phase was promoted by SPD. For the Al-4.8Zn-1.2Mg-0.14Zr specimen the same phenomena was observed, although very weak reflections of $MgZn_2$ appear.

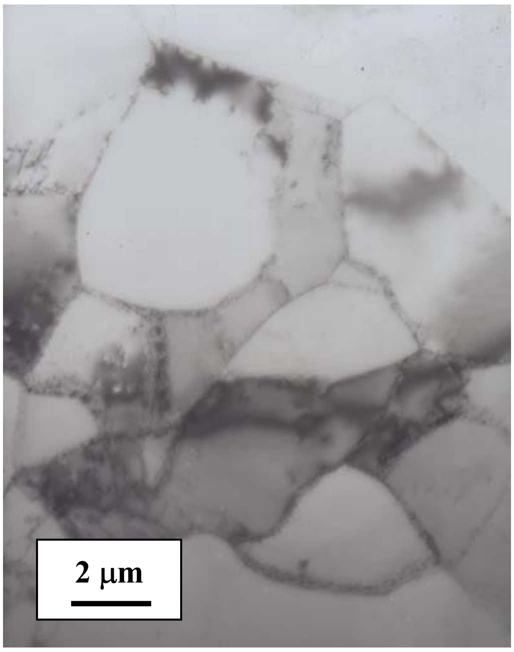

Figure 2: *TEM micrograph for the quenched and aged Al-4.8Zn-1.2Mg-0.14Zr specimen.*

In Fig. 2 the TEM image of the quenched and aged Al-4.8Zn-1.2Mg-0.14Zr specimen shows that its grain size is about 4 μm which is 10 times higher than the value obtained after ECAP (see Fig. 4a). The higher density of dislocations and grain boundaries in the ECAP processed specimens may act as precipitate-forming sites which facilitate the formation of η phase. The X-ray diffractogram in Fig. 1b reveals a strong texture in the matrix of the quenched and aged specimen which was developed formerly during the casting and extrusion of the initial sample. The texture diminished during ECAP as it can be seen from the comparison of Figs. 1a and 1b.

Figure 3 shows the *111/222* pair of X-ray line profiles of the Al matrix and the *00.2/00.4* reflections of $MgZn_2$ precipitates of the Al-5.7Zn-1.9Mg-0.35Cu specimen processed by 8 ECAP passes. The broader *222* reflection compared to *111* peak indicates the existence of strain broadening of the profiles for Al matrix. At the same time the *00.2* and *00.4* reflections of $MgZn_2$ phase are practically the same, indicating that the strain broadening of the line profiles is negligible for the η precipitates. This can be explained by the incoherency of η particles with the matrix so that the dislocations are moving around the η particles (bypass mechanism), but do not cut through precipitates.

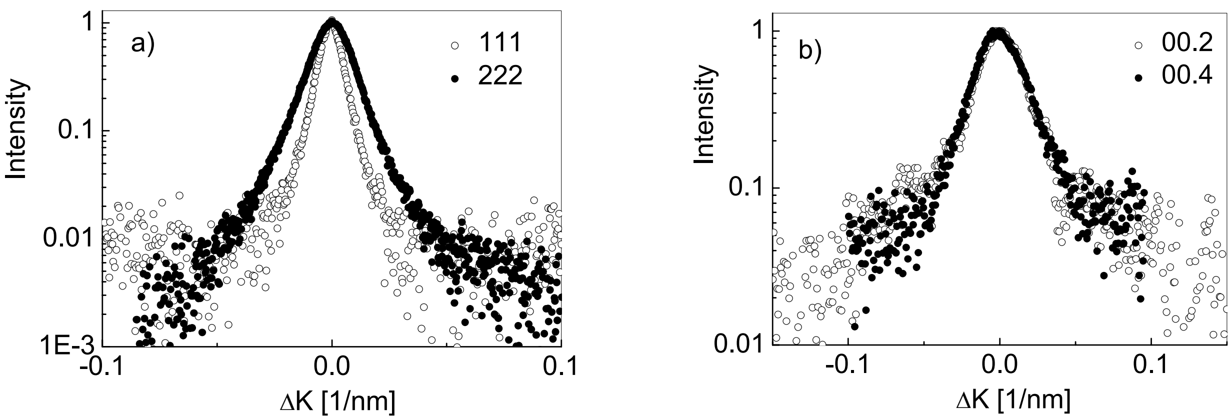

Figure 3: *The 111/222 pair of X-ray line profiles of the Al matrix (a) and 00.2/00.4 reflections of $MgZn_2$ precipitates (b) of the Al-5.7Zn-1.9Mg-0.35Cu specimen processed by 8 ECAP passes.*

The line profiles of both the Al matrix and η precipitates of the ECAP processed specimens were evaluated by MWP fitting procedure and the results are listed in Table 1. The area-weighted mean crystallite size of the matrix of the Al-4.8Zn-1.2Mg-0.14Zr alloy is 165±15 nm which is higher than the value of 119±14 nm obtained for the Al-5.7Zn-1.9Mg-0.35Cu specimen. The values of the dislocation density for the two alloys agree within the experimental error. The area-weighted mean crystallite size of the $MgZn_2$ precipitates in the Al-4.8Zn-1.2Mg-0.14Zr sample is higher than that obtained for Al-5.7Zn-1.9Mg-0.35Cu alloy. The dislocation structure of the precipitates was not evaluated from the diffraction peaks because of the negligible strain broadening of their line profiles.

Table 1: *The parameters of the microstructure and the yield strength (σ_Y).*

Alloy	Phase	$\langle x \rangle_{area}$ [nm]	ρ [10^{14} m $^{-2}$]	σ_Y [MPa]
Al-4.8Zn-1.2Mg-0.14Zr	matrix	165±15	3.2±0.4	290
	$MgZn_2$	30±3	-	
Al-5.7Zn-1.9Mg-0.35Cu	matrix	119±14	3.4±0.4	350
	$MgZn_2$	22±2	-	

Figure 4 shows TEM micrographs taken on the microstructure of both alloys after ECAP. The grain size of the matrix for the Al-4.8Zn-1.2Mg-0.14Zr and Al-5.7Zn-1.9Mg-0.35Cu alloys determined from the TEM images are 500 and 300 nm, respectively, which are 2.5-3 times higher than the values obtained from X-ray analysis. This is a general observation because of the subgrain-structure of the grains for metals processed by SPD techniques. The crystallite size determined from X-ray line profiles corresponds rather to the size of subgrains or dislocation cells which is smaller than the grain size observable in TEM micrographs [9]. The average size of the precipitates in the Al-4.8Zn-1.2Mg-0.14Zr and Al-5.7Zn-1.9Mg-0.35Cu alloys obtained from the images are 30 and 20 nm, respectively. These values are in good agreement with those determined by line profile analysis, confirming that the $MgZn_2$ particles are single crystals, i.e.- contrary to the grains of the matrix - they have no subgrain-structure. The average distance, *D* between the precipitates is approximately 120 nm in the Al-4.8Zn-1.2Mg-0.14Zr alloy and 80 nm in the Al-5.7Zn-1.9Mg-0.35Cu alloy. Experimental results show that the precipitates in the Cu-containing alloy have smaller size and finer dispersion, than in the other alloy. This difference is a consequence of the different alloying composition of the two specimens. The higher concentration of the alloying elements and especially the Cu addition hinder the coarsening of precipitates [10]. Furthermore, it has also been shown before that the addition of Cu into Al-Zn-Mg alloys, in the one hand, increases the upper temperature of GP zone-η' phase transition, and on the other hand, increases the density of GP zones [4,5], which have a strong influence on the formation of η' particles, leading to finer dispersion of η precipitates.

Considering the strengthening effect of non-cutable particles, such as η phase precipitates, it is well known that both the decreasing particle-distance (*D*) and increasing precipitate-size (x) increase the hindering effect of the particles on the movement of dislocations [11]. The effect of particle-distance is, however, stronger than that of precipitate-size. In the present investigation, as both the average size of precipitates and the distance between the η particles in the Al-5.7Zn-1.9Mg-0.35Cu alloy is lower than that obtained for the Al-4.8Zn-1.2Mg-0.14Zr alloy by the same factor (2/3), the precipitate structure of the Cu-containing alloy has higher hindering effect on dislocation motion, resulting in lower recovery rate during high temperature ECAP process.

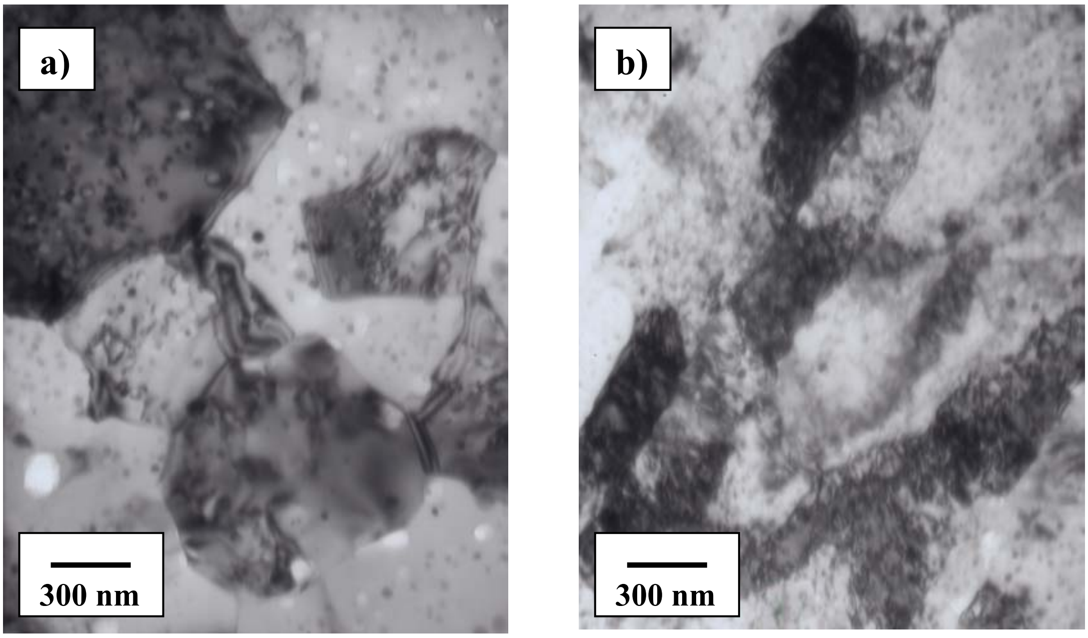

Figure 4: *TEM micrographs for the specimens (a) Al-4.8Zn-1.2Mg-0.14Zr and (b) Al-5.7Zn-1.9Mg-0.35Cu processed by 8 ECAP passes.*

The shape of η phase particles seems to be influenced by the ECAP processing. Zhao et al. [12] have observed needle-like precipitates after subsequent ECAP at RT and ageing at 200°C, while in our experiments only spherical particles were formed during ECAP at 200°C. According to the recently reported results [5] the addition of Cu into Al-Zn-Mg alloys results in both spherical and ellipsoidal GP zones and η' precipitates so that the spherical particles are practically Cu-free, while the ellipsoidal ones contain a significant amount of Cu atoms. This is the reason of the observation of both spherical and needle-like precipitates after the mentioned static ageing of Zhao et al. [12], if taking into account the GP zone → η' → η subsequences. In our experiments the lack of needle-like particles is certainly the consequence of the dynamic ageing process. It can be supposed that the η' phase nucleus, which can be formed in the early stage, during the first pass of ECAP process at 200°C, are sheared and cut by dislocations introduced by severe plastic deformation. The η' phase precipitates formed mainly along {110} planes and then cut by dislocations along the most active {111} planes may be divided into smaller particles, leading to the formation of smaller and rather spherical η phase precipitates during subsequent ECAP at 200°C.

In the alloys investigated here, beside the strengthening effect of dislocations, the precipitate-hardening should be also considered. Assuming the additivity of the different strengthening contributions, the yield strength can be estimated by the following formula [13]:

$$\sigma_y = \sigma_0 + \alpha M^T G b \rho^{1/2} + 0.85 M^T \frac{Gb \ln(x/b)}{2\pi(D-x)}, \tag{1}$$

where σ_0 is the friction stress (20 MPa is taken), α is a constant (α=0.33 is taken), G is the shear modulus (G=26 GPa), b is the length of the Burgers vector of dislocations (b=0.2865 nm), M^T is the Taylor factor (M^T=3 for untextured polycrystalline materials), and x is the average size of precipitates. The different contributions to yield strength can be calculated by using the values of D and x obtained from TEM micrographs and the dislocation density determined by line profile analysis. The strength contributions originated from the dislocation density (second term in Eq. 1) are almost the same, 133 and 137 MPa for Al-4.8Zn-1.2Mg-0.14Zr and Al-5.7Zn-1.9Mg-0.35Cu alloys, respectively, as there is practically no difference between the dislocation densities in the two matrix. The third term in Eq. 1 caused by the dislocation-precipitation interaction gives 156 and 214 MPa for Al-4.8Zn-1.2Mg-0.14Zr and Al-5.7Zn-1.9Mg-0.35Cu alloys, respectively. The smaller particles with more dense distribution causes higher strength for Al-5.7Zn-1.9Mg-0.35Cu alloy. The sum of the three components gives the total strength of 309±20 and 371±25 MPa which are in good agreement with the values determined by mechanical test (290±10 and 350±10 MPa) for Al-4.8Zn-1.2Mg-0.14Zr and Al-5.7Zn-1.9Mg-0.35Cu alloys, respectively.

Summary

High temperature (200°C) ECAP technique was applied to produce ultrafine-grained microstructure in Al-4.8Zn-1.2Mg-0.14Zr and Al-5.7Zn-1.9Mg-0.35Cu alloys. Experimental results obtained by X-ray diffraction and TEM have shown that the high temperature severe plastic deformation promoted the formation of $MgZn_2$ (η) precipitates from the supersaturated solid solution. It was found that the strain broadening of the line profiles for η precipitates is negligible, indicating that the dislocations do not cut through precipitates which can be explained by the incoherency of η particles with the matrix. The Al-5.7Zn-1.9Mg-0.35Cu alloy contains smaller $MgZn_2$ particles with lower distance between them compared to Al-4.8Zn-1.2Mg-0.14Zr which results in higher strength in the former alloy.

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