Aging behavior of ECAP processed AZ80 Mg alloy

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Abstract. Equal-channel angular pressing (ECAP) can produce ultrafine grain structures in metals. The processing can also dissolve second phases through mechanical alloying effects over the equilibrium solubility of alloying elements. Therefore, one can enhance mechanical properties by combining ECAP and subsequent precipitation treatment by proper aging. In this preliminary study, an AZ80 Mg alloy was investigated. The original extruded bars were subjected to ECAP at 473 K for 4 passes to achieve a significant grain refinement down to the submicrometric regime. The possibility of exploiting the aging effect to improve mechanical strength of the alloy was studied by the following two different methods. The first method consisted of ECAP processing the samples followed by aging. The second method consisted of performing a solution treatment prior to ECAP processing and then the final aging of the samples. Micro-hardness measurements and microstructure analyses showed that re-precipitation of the Mg\textsubscript{17}Al\textsubscript{12} phase can occur during warm temperature ECAP and aging in the AZ80 alloy at grain cores in a more finely dispersed form. This precipitation behavior can potentially generate a significant contribution to the strength of the UFG alloy.

1. Introduction
Magnesium and Magnesium alloys are attractive materials for use in electronics, transportation, and biomedical fields because of their excellent properties such as low density, high specific strength and stiffness [1,2]. Recently, it has been reported that ultrafine structure and improved mechanical properties are achievable in magnesium alloys by equal channel angular pressing (ECAP), which is a typical severe plastic deformation (SPD) technique [3–5]. Grain refinement is very important in magnesium alloys because they have poor formability and limited ductility at room temperature due to their hexagonal close-packed crystal structure [3] . It is well known that the AZ80 alloy could be age hardened and its precipitation behavior has already been studied [6]. In the present investigation an extruded AZ80 alloy was ECAP treated under two different conditions, namely with and without prior solution heat treatment. An artificial aging treatment to promote strengthening-particle precipitation was then applied. The synergistic effects of the solution heat treatment, of ECAP and of aging on microstructure and hardness of the AZ80 alloy were investigated in this paper to provide some preliminary data about dispersion strengthening effects potentially available in type-AZ ECAP-processed Mg alloys.

2. Materials and experimental procedures
2.1 ECAP processing
A commercial AZ80 extruded alloy supplied in the form of bars having a diameter of 15 mm was used in this experiment. Cylindrical samples used for ECAP process were machined from the extruded bars to a diameter of 10 mm and a length of 100 mm. The die used for ECAP processing featured two channels intersecting at an angle of 110° with an angle of 20° as outer arc of curvature. The die was heated by 4 electrical resistance heaters homogeneously distributed around the vertical channel and at intersection point of the two channels. ECAP was conducted for up to 4 passes at 473 K. After each pass, the samples were rotated by 90° about their longitudinal axis according to route Bc [7].
2.2 Heat treatment
The aging response was investigated in the as-extruded billet, in solution heat treated samples and in ECAP-processed samples after 4 passes with and without prior solution heat treatment. The possibility of exploiting the aging effect to improve mechanical strength of the alloy was studied by following two different methods. Method A consisted of first ECAP processing the samples at 473 K and then aging them at 403 K at different times up to 120 hours. Method B consisted of first solution heat treating the samples at 693 K for 2h, followed by quenching into water, ECAP processing at 473 K and final aging at 403 K.

2.3 Microstructure observation
The samples used for microstructural analyses were cut perpendicular to the extrusion direction, polished and etched by a solution of 4.2 g picric acid, 80 ml ethyl alcohol, 10 ml acetic acid and 10 ml distilled water. Optical microscopy and scanning electron microscopy (SEM) both by conventional W filament SEM and by Field Emission Gun (FEG-SEM) were used to observe the microstructures and grain boundary features.

2.4 Vickers hardness
Micro-hardness testing was performed using a Vickers indenter with a load 200 gf for 15 s on samples ground with 2500 grit paper. Each data point is the average of at least 15 measurements.

3. Results and discussions
3.1 Solution heat treatment
Fig. 1a and b shows the microstructure of as-extruded and solution treated AZ80 alloy samples, respectively. The stringers of precipitates observed in Fig. 1a are recognized on the basis of EDS analyses as Mg17Al12 phase (one of the two phases forming the eutectic together with Mg-rich solid solution). They are clustered in the as-extruded samples and aligned along the extrusion direction (vertical in the micrographs) and occasionally in isolated form as coarse single particles at grain boundaries. After solution treatment (see Fig. 1b), these precipitates almost completely disappeared and the average grain size grew from 15 µm to about 25 µm, indicating that grains slightly coarsen during the solution treatment. Fig.1c, d and e show the microstructure of solution treated (at 693 K for 2h) and aged samples (at 403 K for 2h, 24h and 120h, respectively). It is confirmed that in the solution treated samples the relatively large Mg17Al12-phase precipitates substantially re-dissolved into the matrix and also the grain boundaries appear substantially free (see Fig 1b). In contrast, after the aging treatment at 403 K for 2h (see Fig. 1c) small precipitates became visible at grain boundaries. Further aging at the same temperature led to the onset of discontinuous precipitation in the form of a lamellar constituent nucleating from grain boundaries (see Fig. 1d for aging times of 24 h). With increasing the ageing time the number of nucleation sites for discontinuous precipitation increases and, furthermore, the already formed discontinuous phases grow toward grain cores (Fig. 1d and e). Such preferential formation and the growth of Mg17Al12 discontinuous precipitation was also observed in other research works [8–10]
Fig. 1 Microstructure of AZ80 alloy: (a) as-extruded, (b) solution heat treated (SHT); SHT and aged at 403 K for: (c) 2h, (d) 24h, (e) 120h.
3.2 Vickers micro hardness

Fig. 2 summarizes the hardness evolution of as extruded and of solution treated samples as a function of number of ECAP passes. In both conditions, a significant improvement in hardness is recorded in the two first passes, followed by a stage of slower increase.

![Fig.2 Microhardness behavior of ECAP processed samples](image)

Fig. 3 shows the aging curves of the AZ80 alloy in the as extruded condition and after solution treatment. As expected, the extruded alloy showed a reduced response to aging while in the solution treated alloy, aging induced a relatively larger increase in hardness even though the starting hardness values were lower.

![Fig.3. Aging response of as-extruded and solution treated samples](image)

Fig. 4 shows the Vickers micro-hardness of the AZ80 alloy processed by solution treatment and ECAP for 4 passes as a function of aging time at 403 K. Aging after 4 ECAP passes generally induced a drop in hardness showing that, in general terms, the aging treatment is
ineffective on overall alloy strengthening when it was already severely deformed at temperatures higher than the usual aging levels.

![Fig.4 Aging response of ECAP processed samples](image)

Finally, Fig. 5 summarizes and compares the micro-hardness values obtained in the different samples as a function of the combination of treatments above described. It is clearly observed that ECAP and the related grain refining generate the largest strengthening effect. Peak aging also induces a remarkable improvement in strength of coarse-grained alloys, while no substantial overall effect is observed in ECAP processed and aged samples.

![Fig.5 Peak micro-hardness values of AZ80 alloy samples in the different conditions investigated](image)

3.3 Microstructure of ECAP processed samples

Fig. 6 shows the microstructure of solution heat treated and ECAP processed samples and of non-solution treated ECAP processed samples. For the non-solution heat treated sample, the grain size could be qualitatively evaluated to be in the sub-micrometer scale based on
evidence of grain contours visible on the micrographs, confirming that ECAP is a powerful technique to obtain a fast reduction in grain size of the AZ80 alloy. Moreover, the Mg17Al12 precipitates, which were elongated along the extrusion direction in the starting condition (see Fig. 1a), became fragmented into smaller and rounded particles during ECAP processing at 473 K (see Fig. 6a). It is also believed that re-precipitation of alloying elements might occur in a much finer form during following ECAP passes.

In contrast, in solution heat treated and ECAP strained samples (Fig. 6b), the microstructure is quite different, containing relatively coarse grains, which are the residual part of the original structure, surrounded by much finer grains. It is proven that the microstructural evolution of Mg alloys during processing by ECAP depends strictly on the nature of the initial structure. In other words, grain refinement in Mg alloys is characterized by the nucleation of fine grains along pre-existing boundaries by dynamic recrystallization (DRX) [11]. It is also worth mentioning that in the solution heat treated and ECAP condition, a large number of tiny (less than 100 nm in size) Mg17Al12 precipitates formed homogeneously in the coarse regions along with the coarser precipitates located in the zones occupied by the ultra-fine structure (see Fig. 6b and 6c). It is therefore supposed that in recrystallized areas grain boundary density, which is considered as high speed diffusion path for elements, the diffusion of solute atoms is faster and thereby growth of precipitates occurs to a higher degree. It must also be clarified that this re-precipitation already occurs during ECAP processing (at 473 K) without any deliberate aging treatment. Furthermore, the results give evidence that ECAP at high temperature preferably leads to spherical shaped precipitates owing to surface energy considerations [12]. Fig. 6d and e show the precipitates morphology after 72 h of aging of Fig. 6a and b, respectively. As seen the precipitate size and morphology generated by ECAP did not undergo any changes, implying that during aging at 407K precipitates induced by ECAP remained relatively stable.

Fig. 6 SEM microstructure of ECAP processed AZ80 at 473K: (a) non-solution heat treated, (b) solution heat treated, (c) specified area in solution heat treated at higher magnification
showing the fine $\text{Mg}_17\text{Al}_{12}$ precipitates inside the coarse grains and bigger ones at DRX zone, (d) non-SHT and aged at 403K for 72 h, (e) SHT and aged at 403K for 72 h

4. Conclusions

From present investigation, the following conclusions can be drawn.

1. An AZ80 Mg alloy was successfully ECAP processed at 473 K showing a significant increase of hardness after 4 passes. Concurrently, the grain size was reduced from 15 $\mu$m to the sub-micrometer size range.

2. In the as extruded condition, the eutectic constituent composed of $\text{Mg}_17\text{Al}_{12}$ particles and $\alpha$-Mg solid solution was mainly clustered in stringers and elongated along the extrusion direction whereas after solution treatment the $\text{Mg}_17\text{Al}_{12}$ particles re-dissolved into the matrix.

3. Aging at 403 K initially led to decoration of grain boundaries, followed by discontinuous growth of a lamellar structure toward grain cores in distinct grains.

4. Considering hardness evolution as a function of the different material conditions, it was observed that ECAP and the related grain refining generate the largest strengthening effect. Peak aging also induced a remarkable improvement in strength of coarse-grained alloy, while no substantial overall effect was observed in ECAP processed and aged samples.

5. In the non-solution treated ECAP processed AZ80 the $\text{Mg}_17\text{Al}_{12}$ precipitates, elongated along the extrusion direction in the starting condition, were replaced by a fine and homogeneously distributed population of round shape precipitates. On the other hand, in the solution heat treated and ECAP processed alloy, re-precipitation from the supersaturated solid solution occurred in a finely dispersed form at the coarse grain interiors with limited presence of grain boundary phases. Moreover, the size of $\text{Mg}_17\text{Al}_{12}$ phases at DRX-ed area was considerably bigger due to the fast nucleation and growing of precipitates.

6. Even though the combination of ECAP and aging was not effective considering hardness, compared to aged only samples a significant modification was observed in precipitate shape and distribution. ECAP processing was shown to be effective in suppressing discontinuous re-precipitation during following aging. The lamellar structure was replaced by a fine and homogeneously distributed population of precipitates which is supposed to be more favorable for enhanced ductility, and corrosion properties of the alloy.

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