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Effect of Pre-Aging on the Microstructure and Strength of Supersaturated AlZnMg Alloys Processed by ECAP

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Abstract. This work is focused on the effect of the combination of natural aging and severe plastic deformation (SPD) produced by Equal-Channel Angular Pressing (ECAP) on the microstructure and strength of supersaturated AlZnMg alloys. Following a solution heat-treatment and quenching into water at room temperature, samples were naturally aged for different time periods and then processed by ECAP. The microstructure and mechanical properties of these samples are described and discussed. This investigation leads to proposing an interesting application of ECAP for supersaturated alloys. Using the shear bands created by ECAP in only one pass and applying appropriate subsequent aging treatments, composite-like microstructures can be achieved in conventional age-hardenanble Al alloys.

Introduction

The chemical composititon and the aging conditions influence effectively the decomposition of the supersaturated solid solution (S.S.S.) in the Al-Zn-Mg ternary system [1-4]. The precipitation in these alloys has been the subject of many experiments concentrating mostly on the later stages of precipitation, and consequently the role and formation of the main precipitates have been determined. For the early stages of the decomposition in these alloys relatively few experimental results are available [5].

It is well-known that the precipitation and strength of supersaturated AlZnMg alloys can be effectively changed by the application of pre-strain to the samples. Usually, a small amount of pre-strain is introduced by conventional tension or compression [6]. Recently, severe plastic deformation (SPD) by Equal-Channel Angular Pressing (ECAP) has also been applied to change the microstructures and improve the mechanical properties of supersaturated alloys [7-10]. For precipitate-hardened AlZnMg materials, however, the application of SPD is often complicated. On the one hand, because of the hardening effect of precipitates (e.g. Guinier-Preston (GP) zones and/or other metastable particles), samples often break during ECAP at room temperature (RT). Therefore, in several cases supersaturated AlZnMg samples were processed by ECAP mostly at elevated temperatures. On the other hand, at high temperatures the precipitation is often uncontrolled and a recovery of the ultrafine-grained microstructures may occur.

In this paper the effect of the combination of natural pre-aging and severe plastic deformation (SPD) produced by Equal-Channel Angular Pressing on the microstructures and strength of supersaturated of Al-4.8Zn-1.2Mg-0.14Zr, Al-5.7Zn-1.9Mg and Al-5.7Zn-1.9Mg -1.5Cu (wt.%) is investigated.

Experimental Materials and Procedures

Billets of polycrystalline Al-4.8Zn-1.2Mg-0.14Zr, Al-5.7Zn-1.9Mg and Al-5.7Zn-1.9Mg-1.5Cu alloys were first subjected to solution heat-treatment at 470 ºC for 30 minutes and water-quenched, resulting in S.S.S. Following quenching, the samples were naturally aged at RT for different time periods prior to ECAP processing. Cylindrical billets of 10 mm in diameter and 70 mm in length were pressed through an ECAP die with 90° intersecting channels at RT for different numbers of passes in route Bc (the billet was rotated around its longitudinal axis by 90° between intermediate passes) at a constant displacement rate of 8 mm/s. Following the ECAP process, some samples were subjected to aging at 100 ^{o}C or 140 ^{o}C for 24 hours. Several important mechanical and precipitation properties of these alloys after quenching were reported earlier [10-12].

Samples for microhardness measurements were mechanically and electrolytically polished, measurements were carried out using a Vickers indenter in a Shimadzu-DUH 202 depth-sensing ultra-microhardness machine operating with 2N maximum load.

The microstructure was 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

1) Microscopic effect of natural aging – Plastic instabilities

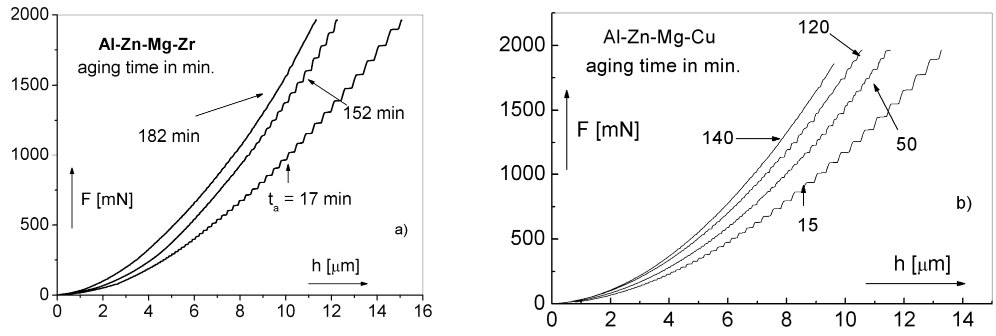


Figure 1: *The effect of the natural aging on the indentation load-depth curves taken by depth-sensing microhardness tests on a) Al-Zn-Mg-Zr and b) Al-Zn-Mg-Cu alloys*

Typical indentation load-depth (*F-h*) curves obtained on Al-Zn-Mg-Zr and Al-Zn-Mg-Cu alloys in the early stage of natural aging are shown in Fig.1. Considering both the position and the shape of the indentation curves, experimental results show, on the one hand, the increasing hardness of the samples with increasing aging time, indicating the early formation of GP zones after quenching. On the other hand, it can be seen that at the beginning of the natural aging characteristic steps appear in the indentation depth-load (*h-F*) curves for both alloys. The step formation is a typical plastic instability effect similar to the Portevin-Le Châtelier effect or to jerky flow [13].

The instability load-depth steps observed in the very early stage of the natural aging (within 20 min after quenching) of both alloys are relatively regular, similar to those found in a stable solid solution Al-3Mg alloy [13,14]. However, with increasing aging time, the steps occur less frequently and become more irregular and finally, after a certain time, t_i, they disappear completely. The values of t_i are between about 100-200 minutes and in the lower concentration (Al-Zn-Mg-Zr) alloy the steps are retained for somewhat longer.

The occurrence of plastic instabilities is caused by the interaction of diffusing solute atoms with moving dislocations, a phenomenon called dynamic strain aging (DSA) [15-18]. This implies that the presence of the load-indentation depth steps is connected to a certain solute concentration in the alloys. The formation of the instability steps at this stage of the process indicates that the GP zones are not strong enough to suppress the DSA effect. The occurrence of the steps, therefore, characterizes the transition (with lifetime of t_i) from the SSS to a SSS + GP zone structure, when the effect of GP zones on moving dislocations becomes dominant compared to that of the solute atoms. The value of t_i, therefore, gives important information about the changes of the microstructure of supersaturated AlZnMg alloys in the range of early formation of GP zones.

2***) Macroscopic effect of natural aging – Shear bands during ECAP process***

Figure 2: *The surface topography of the billets after 1 ECAP pass in the case of a) Al-Zn-Mg-Zr sample quenched and aged at room temperature for only 10 minutes (Q+RT 10 min.) b) Al-Zn-Mg-Zr sample, (Q+RT 7 days) and c) Al-Zn-Mg-Cu sample, (Q+RT 7 days)*

Figure 2 shows three billets of the two alloys after 1 pass in ECAP. It is obvious that natural aging has significant influence on strain localization and crack formation in the ECAP process. While in the case of short (about 10 minutes) RT aging the surface of the samples remains smooth (Fig. 2a), indicating a similar SPD process to that taking place in polycrystalline pure metals, the aging for 7 days resulted in intensive formation of macroscopic shear bands in the lower concentration AlZnMgZr sample (Fig. 2b) and even catastrophic cracking in the more concentrated AlZnMgCu sample (Fig. 2c). This shows that age-hardenable AlZnMg alloys are diificult to ECAP process, similarly to some magnesium alloys [19]. It should be noted that macroscopic shear bands, similar to those of samples in Fig. 2b, were observed in single-crystal Al [20] but for a different reason.

As a consequence of the nature of the ECAP process, shear bands are always formed. In the case of polycrystalline pure metals or in solid solution alloys, however, because of high mobility of dislocations, the shear bands are formed only on microscopic scales and do not lead to the failure of the sample. Pure Al or Cu [21] and solid solution Al-3Mg alloys can be pressed by ECAP up to 8 passes without catastrophic (macroscopic) cracks in the sample. In the case of supersaturated alloys, however, the effect of precipitates must also be taken into account. The macroscopic, even catastrophic, shear bands shown in Fig. 2 reflect unambiguously the combined effect of GP zones and SPD by ECAP. Results shown in both Figs 1 and 2 confirm that in order to avoid catastrophic shear bands caused by GP zones during ECAP the samples have to be processed by ECAP i) immediately after quenching, at least within the mentioned lifetime, t_i, when the effect of GP zones is not so strong or ii) at elevated temperatures where GP zones (and other strengthening precipitates) cannot be formed. When applying the high temperature process (case ii), however, uncontrolled precipitation or undesirable recovery may occure in the samples. In the case of processing at RT, even immediately after quenching (case i), it has also to be taken into account that the high amount of dislocations produced already in the first ECAP pass can strongly accelerate the formation of GP zones. Therefore, there may be a detrimental effect of GP zones in subsequent passes. In the present work, when the samples were processed immediately (within 10 minutes) after quenching the lower concentration Al-Zn-Mg-Zr alloy could be pressed by 7 passes and the more concentrated Al-Zn-Mg-Cu samples could undergo 3 passes in ECAP. This latter result may explain the report of a similar composition sample processed by ECAP for only 2 passes at room temperature [7]. The results of the ECAP processing experiments and the microhardness values of the materials are summarized in Table 1.

Table 1: *Microhardness and estimated yield strength of supersaturated AlZnMg samples investigated under different conditions*

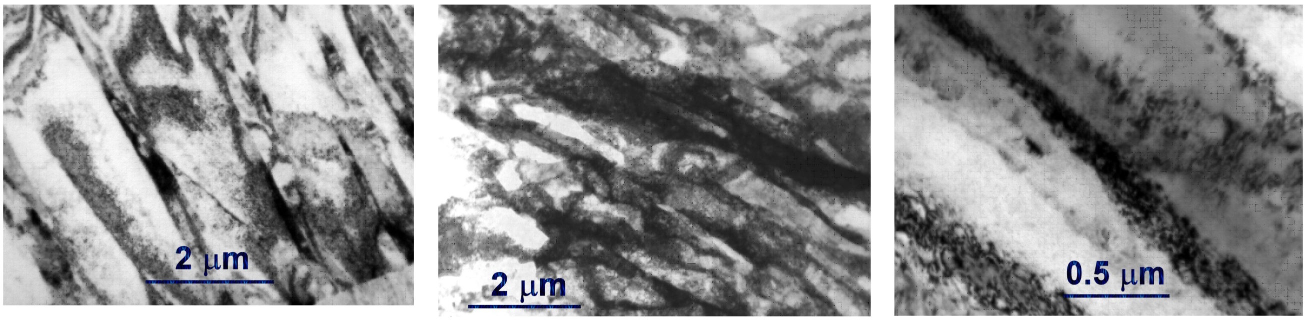
Alloy	Aging time at RT	Actual number of passes by ECAP in route B_c	Number of passes to failure	Subsequent aging	Hardness [MPa] ± 5%	Estimated yield strength [MPa]
1.	1 week	1	1	-	2000	~670
2.	1 week	1	1	100 ^{o}C (24h)	2020	~670
3.	10 min.	3	3	-	2080	~690
Al-5.7Zn-1.9Mg-1.5Cu 4.	10 min.	1	-	-	2070	~690
5.	10 min.	1	-	100 ^{o}C (24h)	2020	~670
6.	10 min.	1	-	140 ^{o}C (24h)	1550	~520
(*Peak strength in conventional mater.)* 7.	2 months	-	-	100 ^{o}C (24h)	1480	~490
8.	10 min.	4	4	-	1540	~520
Al-5.7Zn-1.9Mg 9.	10 min.	4	4	100 ^{o}C (24h)	1690	~560
10.	10 min.	4	4	140 ^{o}C (24h)	1280	~430
11.	1 week	3	3	-	1350	~450
12.	1 week	1	-	-	1360	~450
13.	1 week	1	-	100 ^{o}C (24h)	1390	~460
14.	1 week	1	-	140 ^{o}C (24h)	1150	~380
15.	1 day	4	4	-	1340	~450
16.	1 day	1	-	-	1320	~440
17.	1 day	1	-	100 ^{o}C (24h)	1410	~470
Al-4.8Zn-1.2Mg-0.14Zr18	1 day	1	-	140 ^{o}C (24h)	1140	~380
19.	10 min.	7	7	-	1310	~440
20.	10 min.	4	-	-	1370	~460
21.	10 min.	1	-	-	1320	~440
22.	10 min.	1	-	100 ^{o}C (24h)	1240	~410
23.	10 min.	1	-	140 ^{o}C (24h)	1070	~360
(*Peak strength in conventional mater.)* 24.	2 months	-	-	100 ^{o}C (24h)	1140	~380

As can be seen from Table 1, the maximum number of ECAP passes in route B_c up to failure depends strongly both on the composition of the materials (compare samples 3, 8 and 19) and on the aging time at RT (see samples 11, 15 and 19), confirming the effect of GP zone structures.

From the point of view of strengthening effect, the application of ECAP processing can improve the strength (hardness) by 20-40 percent, compared to the peak hardness of the conventionally age-hardened materials (cf. the hardness of samples 4 and 7, as well as that of samples 12 and 24). However, for a given alloy composition, the strength gained after ECAP seems to depend neither on the time period of pre-aging at RT (see samples 1 and 4, or samples 12, 16 and 21) nor on the number of passes of ECAP (see samples 1 and 3, as well as samples 19, 20 and 21). This latter result suggests that in order to improve the strength by ECAP it is sufficient to process quenched AlZnMg materials in only 1 or 2 passes. This finding may have important practical significance, because the application of less passes in ECAP leads not only to time saving but also it reduces the possibility of formation of micro-cracks often occuring during SPD. It should be noted that the application of more passes (e.g. up to 4 passes) is more important if the increase in strength is brought about by the Hall-Petch effect as a consequence of grain-refinement [21] or in the case of deformation mechanisms like grain-boundary sliding [22-24]. According to these experimental results, the application of several passes of ECAP in supersaturated AlZnMg alloys should be done in the very early stage of natural aging to prevent the formation of Guinier - Preston zones which leads to severe strain localization and subsequent crack initialization.

3) Further effects of ECAP process on microstructure of supersaturated AlZnMg alloys

TEM micrographs (Figure 3) show some typical structures of microscopic shear bands in AlZnMgZr samples processed by 1 pass of ECAP (same samples shown in Figs. 2a and b).



a) b) c)

Figure 3: *Microstructure of Al-Zn-Mg-Zr samples deformed by 1 pass in ECAP in the case of a) Q+RT 7 days, and b), c) Q+RT 10 min. in different magnifications.*

The TEM investigation also revealed the important role of the natural aging. While RT aging for relatively long times, e.g. for 1 week (see Fig. 3a), resulted in typical shear bands with spacing of about $2\mu m$ after 1 pass of ECAP, for shorter aging times (10 min) thinner ($\sim 0.5\mu m$) bands (see Figs 3b and 3c) can be observed. It should be noted that the collective operation of the microscopic shear bands will cause the appearance of macroscopic bands as shown in Fig. 2. More systematic investigations should be conducted but these preliminary results suggest that the thickness of micro-shear bands could be established by controlling the period of natural aging. It would also be worth studying the structure (see Fig. 3c) of the individual micro-bands, which are, in fact, bands having relatively low dislocation density (brighter strips in Fig. 3c), surrounded by regions having relatively high dislocation densities (dark strips). Considering a sandwich-like structure, one can conclude that different kinds of precipitates, the formation of which is strongly affected by pipe-diffusion along dislocations, could be formed in different parts of the sandwiches, producing composite-like microstructures in conventional supersaturated AlZnMg alloys. This question will be discussed in more detail in a later paper.

Summary

The effect of the combination of natural pre-ageing and the severe plastic deformation produced by Equal-Channel Angular Pressing on the microstructures and strength of supersaturated AlZnMg alloys was investigated. It was shown that in order to improve the strength by using ECAP, it is sufficient to process supersaturated AlZnMg alloys up to only 1 or 2 passes, reducing the possibility of formation of micro-cracks often formed during SPD. Furthermore, the application of only 1 pass in ECAP leads to the formation of a sandwich-like structure, the characteristics (e.g. the thickness of the layers) of which can be controlled by the periods of natural aging. It is anticipated that the composite-like microstructures can be produced in conventional supersaturated AlZnMg alloys by applying appropriate artificial aging to samples processed by ECAP.

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