Evaluation of the Hardening and Softening Effects in Zn–21Al–2Cu with as Cast and Homogenized Microstructure Processed by Equal Channel Angular Pressing

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Abstract In this work it is reported the evolution of the microstructure of the as-cast and homogenized Zn–21Al–2Cu samples, after 2 and 6 passes in an equal channel angular pressing (ECAP) at room temperature. A homogenization treatment for 24 h at 350 °C was applied in the as-cast samples and then they were deformed. An annealing heat treatment was made in all samples after ECAP process. One of the main results is that the homogenized and deformed samples showed a uniform fine-grained microstructure after annealing, while as-cast samples without annealing presented only some regions with fine-grained microstructure. The micro segregation level was higher in the as-cast samples in contrast to the homogenized ones even after annealing. Vickers microhardness measurement on

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M.A. Meyers et al. (eds.), *Proceedings of the 3rd Pan American Materials Congress*,
The Minerals, Metals & Materials Series, DOI 10.1007/978-3-319-52132-9_69

samples after deformation is smaller than the original material indicating a softening, however after the annealing treatment the microhardness increased, indicating that there was a slight hardening of the material.

Keywords Severe plastic deformation · Work softening · Annealing hardening

Introduction

The Zn–Al eutectoid alloy has been extensively studied, because it has a low melting point, good castability, wear resistance, low forming temperature, high strain rate sensitivity and high plastic deformability because presents the superplasticity phenomenon [1, 2]. This alloy forms a lamellar microstructure during solidification, so that to form a fine equiaxed-grained microstructure both, stages of plastic deformation and annealing treatments are required [2, 3]. Numerous thermo-mechanical treatments have been studied for eutectoid alloy, these have enabled to obtain fine-grained microstructure which is required for this alloy can be deformed superplastically, most of these treatments first considered homogenizing the as cast alloy, followed of multistage of rolling [3] or extruding [2] and then different annealing treatments. An atypical phenomenon has been observed in Zn–Al eutectoid alloy as well as alloys modified with 0.3 and 2% by weight copper, this phenomenon consists in a work-softening and anneal-hardening of these materials [2–4]. For potential applications, it is of interest find grain refinement processes that allow to obtain a fine grain microstructure from as cast alloy, with fewer processing steps. The aim of this work is to study the effect of extrusion in constant angular channel (ECAP) on the microstructure of an Zn–Al eutectoid alloy as well as Zn–22%Al–2%Cu, comparing the effect in both as-cast and homogenized microstructure. We assessed whether these alloys show the work softening and annealing hardening effect, observed for thermomechanical processes involving rolling and extrusion. The results are compared with those obtained by Yang et al. [3] for the Zn–Al eutectoid alloy and the eutectoid modified with 0.3% by weight copper. The effect of copper content on atypical behavior of these alloys and the advantages of processing by ECAP for obtaining a fine grained microstructure were studied.

Experimental Procedure

A frequency induction furnace was used to produce the Zn–22Al–2.5Cu alloy by melting the corresponding quantities of Zn (99.99%), Al (99.99%) and Cu (99.96%). The melt was poured into a graphite crucible in air and casting it into cylindrical bars of 19 mm in diameter and 35 mm in length. After that, some bars were subjected to a homogenization treatment at 350 °C for 24 h in air atmosphere. Cast and homogenized samples were subjected to an equal channel angular

Fig. 1 Tools used during the ECAP process

extrusion (ECAP) in a die with two cylindrical channels of an equal diameter (15.8 mm). The die consisted of two channels intersecting at an angle (φ) of 90° and an outer angle (ψ) of 36° (Fig. 1). A FEM simulation was carried out using the DEFORM 3D software. A similar geometry of the die and sample were employed. An experimental flow curve obtained from compression test of the alloy was loaded to the software. All samples were extruded by 2 and 6 passes with a ram velocity of 5 mm/min and by using B_c route.

The lubricant used was MoS_2 and it was applied in both channels for each pass. The process was carried out in a universal testing machine Shimadzu AG-I 600 kN at room temperature. Load-displacement curves were registered during the ECAP process. Then, deformed samples were annealed at 270 °C for 30 min. Longitudinal sections of annealed and deformed samples were ground and polished in order to characterize their microstructure in a JEOL 6610 LV scanning electron microscope. Vickers hardness was evaluated in a Shimadzu HMV-G21DT micro-hardness testing machine using 1.96 N for 15 s. At least 8 values were taken for each specimen to obtain an average.

Results and Discussion

Effect of SPD on Microstructure and Microhardness

Figure 2 exhibits the cast microstructure at low and high magnification. For example in Fig. 2a it can be seen the dendritic pattern composed by the zinc rich phase (η) and aluminum rich phase (α). The former can be seen in a bright contrast and the latter in a grey contrast. In the incise b of the same Fig. 2, it can be distinguished two zones (with laminar and granular morphology) product of the eutectoid reaction surrounded by η phase. Figure 2c, d shows the same dendritic pattern with some preferred orientation visible from left to right, which can be

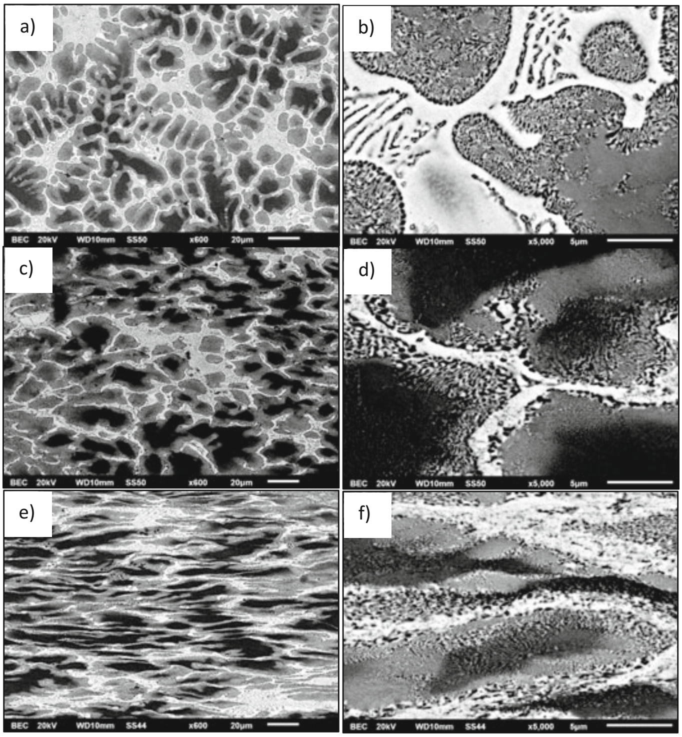


Fig. 2 SEM micrographs at low and high magnification of the samples with initial cast morphology, **a**, **b** without deformation, **c**, **d** with 2 passes, **e**, **f** with 6 passes

attributed to the 2 passes of ECAP that were applied to this sample. In Fig. 2e, f it can be noted that the deformation pattern is more accentuated in the microstructure, however the microsegregation level is still very significant. It is also evident that bright and grey areas exhibit a more fine grained morphology inside. This change can be associated to a combination of both mechanisms: mechanical twist effect of the α phase and dynamic recrystallization of η phase [3].

Figure 3 shows the homogenized microstructure of the samples. It can be noted (incises a and b) that a laminar morphology results but there are also some areas of η phase that remain with the original dendritic morphology. In the incises b and c, the microstructure corresponding to 2 passes of ECAP is shown. The micrograph of the incise d exhibits how the lamellar microstructure is now distorted as a result of the application of the severe deformation. In some areas, the initial morphology is very clear, however in other regions, the distance between the layers of both phases are no more distinguishable, a fact that is proposed as an evidence of the mechanical twist effect caused by ECAP. Micrographs corresponding to 6 passes of ECAP are presented on Fig. 3e, f. As it can be seen, there are still some areas of η phase which is in the initial dendritic morphology, while the majority of the

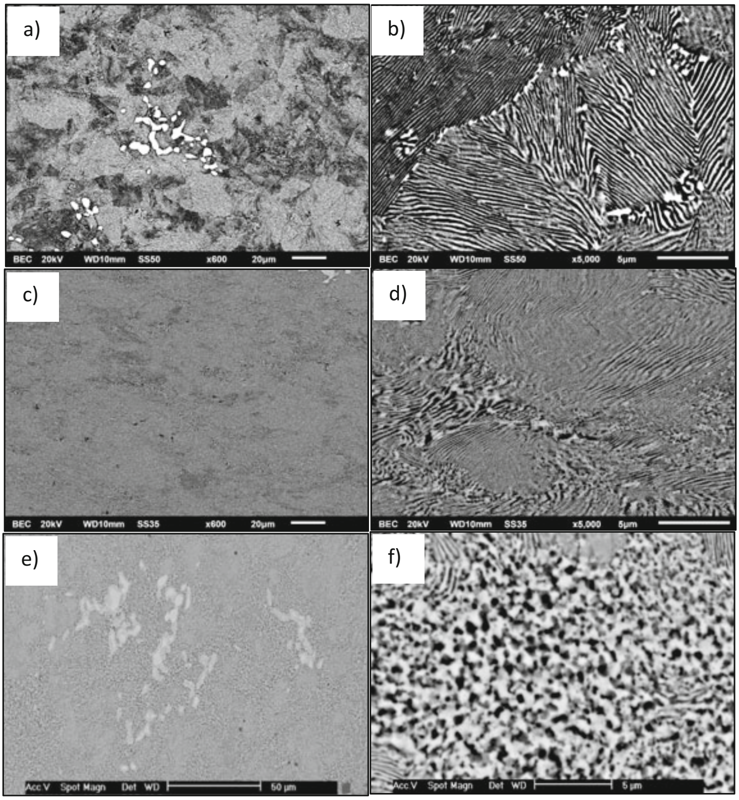


Fig. 3 SEM micrographs at low and high magnification of the samples with homogenized morphology, **a**, **b** without deformation, **c**, **d** with 2 passes, **e**, **f** with 6 passes

microstructure shows a fine granular morphology of α and η. As it has been already mentioned before, it is believed that this microstructure is the result of the mechanical twist effect of the aluminum rich phase and the dynamic recrystallization of zinc rich phase. Yang et al. [3] have demonstrated by means of DSC runs that the recrystallization temperature of the α phase was 337 °C and the corresponding to η phase was −12 °C. Consequently, it is proposed that the former phase is fractured and mixed along the microstructure while the latter experiments dynamic recrystallization during severe plastic deformation at room temperature. This is the explanation about the fine microstructure shown in Fig. 3f. It is important to note that even for 6 passes, there are still some areas that remain with a lamellar morphology due to the deformation heterogeneity as it is shown in Fig. 4. In this case, it may be necessary to subject the sample to longer homogenization treatments and/or more passes of ECAP in order to see if these areas disappeared with higher strain.

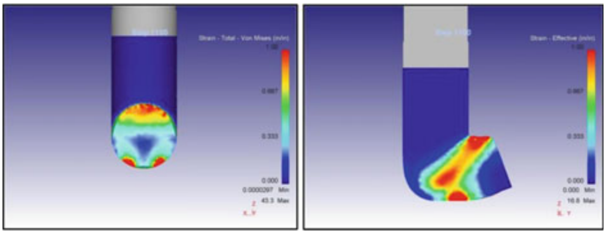


Fig. 4 Strain effective contour maps for 1 pass of ECAP obtained by finite element method simulation

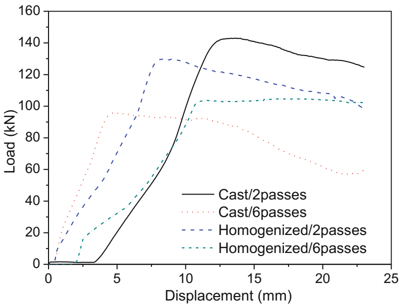


Fig. 5 Load-displacement curves for 2 and 6 passes of ECAP in the cast and homogenized state

El-Danaf et al. [5] have recently reported a similar work softening phenomenon in an eutectic Pb-Sn alloy subjected to ECAP processing. They concluded that a combination or fracture of eutectic crystals and recrystallization of α and β phases lead to a change in morphology from lamellar to fine granular after 4 passes of ECAP. On Fig. 5 it is presented the behavior of load during the ECAP process for 2 and 6 passes for the cast and homogenized samples. In both cases it can be noted that there is a significant reduction in the required load for the process. For example, in the case of the cast samples there is a reduction from 140 to 95 kN while for homogenized samples the reduction is from 132 to 100 kN. The reductions observed can be associated to the recrystallization of η phase as has been already mentioned in the precedent section. It is proposed that there is a major drop in load in the cast samples in comparison to the homogenized ones ought to the higher microsegregation in the former.

Figure 6 exhibits the behavior of the microhardness of the cast and homogenized samples as they are subjected to several passes of ECAP. These are compared with results from the literature [3] on which Zn–22Al and Zn–22Al–0.3Cu alloys microhardness values are reported for lower strain values. As can be seen the general tendency is a decrement of the microhardness as the strain increases.

It is important to point out that in this work it has been confirmed that the softening experimented during deformation is still valid for higher values of deformation in comparison to the values reported in the literature. It is also

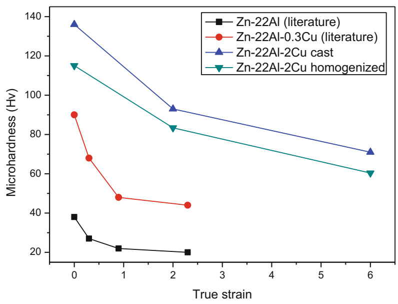


Fig. 6 Microhardness graphs for the alloy studied in this work compared with values from the literature

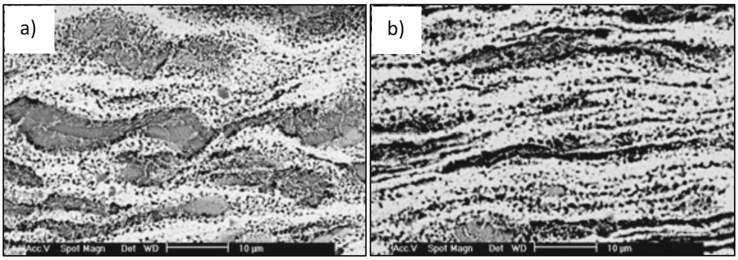


Fig. 7 SEM micrographs of the cast samples subjected to 6 passes of ECAP for **a** 30 min and **b** 120 min of annealing at 275 °C

important to mention that the Cu content has a significant effect in the hardness of the alloy since our values are higher in both cases: cast and homogenized compared to the ones reported in [3]. The general phenomenon observed in the microhardness of the deformed samples supports the hypothesis about the recrystallization of the Zn rich phase during the ECAP process. It is proposed that the higher hardness values exhibited for the cast specimens are due to the presence of a higher microsegregation in comparison with the homogenized samples.

Evolution of Microstructure and Microhardness During Annealing

Figure 7 shows the microstructure of samples with 6 passes of ECAP when they are subjected to 30 and 20 min at 275 °C. As it is observed, the microsegregation level is still present in the entire sample (Fig. 7a, b). Also it is evident that there are some areas on which there exists a microstructure with fine grains. On the other hand when the homogenized sample with 6 passes of ECAP is annealed for the same time at 275 °C the resultant microstructures are shown in Fig. 8. It is evident that for 30 min the microstructure has fine grains with a size close to 1 µm (Fig. 8a).

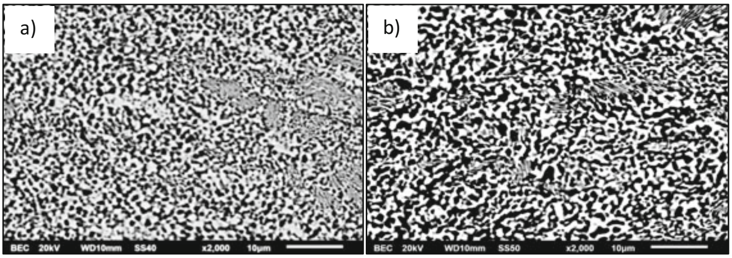
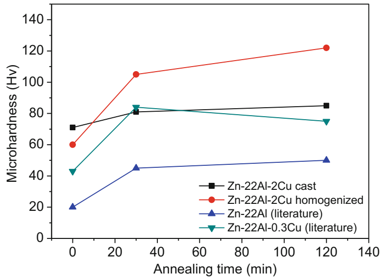


Fig. 8 SEM micrographs of the homogenized samples subjected to 6 passes of ECAP for **a** 30 min and **b** 120 min of annealing at 275 °C

Fig. 9 Comparison of the hardness behavior of the experimental samples and literature values during the annealing time



Also there are still some areas with lamellar morphology that did not transform to fine grains. This result is associated to the heterogeneity in deformation present in the sample even for 6 passes. Figure 8b shows the corresponding microstructure for 120 min of annealing. It is clear that the fine grains have coarsened and also zones with laminar morphology remained in the microstructure.

Figure 9 shows the microhardness behavior of the cast and homogenized samples (with 6 passes of ECAP) annealed for 30 and 120 min at 275 °C. These results are compared to the ones presented in Ref. [3]. It can be noted that the hardness increases in both samples as the time is higher. However the increment is significantly higher in the case of the homogenized sample in comparison to the cast one. This result can be attributed to the initial morphology in the samples. We have already demonstrated that even for 6 passes of ECAP the cast sample possessed a mixed morphology of dendritic and granular arrays and also the microsegregation level is evident while the homogenized sample showed fine grain morphology with some isolated islands of lamellar microstructure (Fig. 3f). According to [3] as annealing time is higher there is an increment in the high angle boundaries quantity in the microstructure which in turn lead to the reduction of the heterogeneous nucleation sites of dynamic recovery which causes the hardening observed.

In our case, we proposed that the increment in the relative amount of high angle boundaries is higher in the homogenized sample in comparison to the cast sample, however a deeper study about the quantification of this parameter is carried out at present to prove this assumption. Zhang et al. [2] have associated this anneal hardening phenomenon to a phase transformation of Zinc-rich phase (α_2) to equilibrium phases Al-rich α_1 and the Zinc-rich η. However it is stated that this mechanism is not operative in the system studied because the phase α_2 is improbable to form during the slow cooling conditions that prevail during cast and homogenization processes.

Conclusions

It was demonstrated that work softening in an Zn–22Al–2Cu is valid even for true strains close to 6 in both samples: cast and homogenized.

It was found that homogenized samples showed a fine grain microstructure (less than 1 μm), while in the case of cast sample the microsegregation pattern and zones with initial dendritic microstructure remained after 6 passes of ECAP.

It was found that during the annealing of cast and homogenized samples with 6 passes of ECAP, there was an annealing hardening phenomenon which was attributed to the increment in the relative amount of high angle boundaries in both samples; however it was observed that this increment was more significant in the case of the homogenized samples.

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