FORMATION OF HOMOGENEOUS ULTRA-FINE GRAIN STRUCTURES IN ALUMINIUM ALLOYS BY EQUAL CHANNEL ANGULAR EXTRUSION

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ABSTRACT Equal channel angular extrusion (ECAE) was used to deform aluminium alloys to very large strains, with the aim of seeing if the technique could be used to form ultra-fine, homogeneous, grain structures in inhomogeneous cast starting materials. The local homogeneity of deformation after one pass was characterised by studying a grid scribed on the internal surface of a split sample. The homogeneity of flow within the die and the progressive microstructural changes were investigated for shear strains of up to 17. The electron back scattered diffraction (EBSD) technique was used to study the grain structure evolution and boundary misorientations during ECA extrusion at 200°C and on annealing at 300°C. An ultra-fine homogeneous grain structure was found to be formed, by the break up of the original grains, at shear strains of about 17. The mechanism involved the formation of high angle boundaries during grain fragmentation, as well as the large increase in high angle boundary area due to distortion of the original grains. However, on annealing discontinuous grain growth occurred resulting in a bimodal grain size distribution.

Keywords: *ECAE, Equal Channel Angular Extrusion, Ultra-Fine Grain, High Strain, Continuous Recrystallisation, Geometric Dynamic Recrystallisation*

1. INTRODUCTION

Ultra-fine grained alloys can have exceptionally beneficial mechanical properties [1-5]. One method of achieving very fine grain sizes in aluminium alloys is through the promotion of continuous recrystallisation by the application of very high plastic strains [4,6,7]. In conventional processes such as rolling, or wire drawing the amount of strain is limited by the dimensional change of the work piece. Using torsion, higher strains have been achieved although the volume of the material deformed is usually very small. Recently a processing route known as Equal Channel Angular Extrusion (ECAE) has attracted a great deal of attention [2-4]. In this process the billet is sheared through an angle which determines the level of strain for each pass. High strains of 10 or more can be obtained by multiple passes through the die without any change in the billets dimensions [2-4]. It has been widely claimed that during ECA extrusion the majority of the sample only undergoes a simple in-plane shear [2-4], although to date there have been few studies of the homogeneity of plastic flow during ECAE extrusion [9]. According to Segal [2], for uniform deformation, the shear strain per pass (γ) can be calculated for a given internal die angle (2ϕ) from the relationship:

$$\gamma = 2\operatorname{cotan}\phi \qquad (1)$$

However, it is difficult to reconcile these claims with the fact that a sheared billet must change its shape, unless the direction of shear is reversed each pass, and there must thus be secondary deformations required to accommodate the dimensional changes due to the shear. Furthermore, although it is widely accepted that ultra-fine grain structures can be produced using this technique [2-4], there has been little work directed towards understanding the mechanisms by which this occurs. In this paper work has been carried out, using simple model alloys, aimed at determining whether ECA extrusion can be used to produce homogeneous ultra-fine grain structures in non-homogeneous starting materials. Using a novel hydraulically clamped split die, the homogeneity within the process itself has been studied, with scribed grid samples, and the effects of friction investigated. Preliminary investigations have been carried out into the physical processes by which an ultra-fine grain structure can evolve, and as to whether an isotropic ultra-fine grain structure can be produced in inhomogeneous starting materials by the use of this technique.

2. EXPERIMENTAL PROCEDURE

Table 1 shows the nominal chemical composition and starting conditions of the aluminium alloys used in this study, which were provided by Alcan Int. The alloys were based on a solid solution of Al-3%Mg with various levels of intermetallic particles, depending on the, Zr, Cr, and Fe content. The Zr and Cr additions were used to inhibit recrystallisation. The three materials were used to give different starting grain sizes in order to ascertain whether the process deformed the samples by a simple homogeneous shear, even after multiple passes. Alloy A was DC-cast with a grain size of 200μm. Alloy B was cast in a book mould and had an initial grain size of 50μm. The high Fe content Alloy C, was DC-cast and cold rolled to 80% and annealed at 400°C to achieve a fine recrystallised initial grain size of 20μm, by particle stimulated nucleation.

Table 1- Chemical composition, as-cast grain size and the condition of the alloys used in the study.

Material	Al	Mg	Zr	Cr	Fe	Grain Size	Condition
A	Balance	3	0.2	-	0.6	200μm	DC-cast
B	Balance	3	0.3	-	-	50μm	Book mould
C	Balance	3	-	0.2	0.2	20μm	Rolled and annealed

The ECAE rig was designed with two rams, one either side of the die, capable of operating in stroke or load control. The die was split along its centre line and held in a hydraulic clamp so that the samples could be removed quickly following deformation. The die took 15 mm diameter samples and had a 120° angle between the channels, which according to equation 1 produces a shear strain of 1.15 after each pass. An Al sample split along its centre line and with a precision grid scribed on one half of the centre plane was used to measure the local deformation in the die, in the plane of the shear, for one pass at room temperature. Both halves were held together using locking rings before extruding halfway through the die. After partial extrusion the deformed grid was used to measure the strains and their local variation. The effect of friction was studied by comparing a grid deformed with a clean die surface and using PTFE tape and spray lubrication, to produce low friction, with one deformed when the die had become coated with aluminium through extensive use and the sample was only wrapped in PTFE tape. Because of the results of these experiments (see section 3.1), samples for microstructural analysis were subsequently deformed with a back pressure in the exit channel, applied by the second ram, to ensure that the die was completely filled and were well lubricated with PTFE tape. Extrusion of the three alloys was carried out at 200°C by using heaters in the die, which controlled the temperature to ± 2°. Samples were deformed by up to 15 passes, with the sample always being replaced in the die with exactly the same orientation each time. The microstructural evolution of alloy A was studied in detail at increasing levels of strain, and after post extrusion annealing at 300°C, by optical microscopy of anodised samples and back scattered electron orientation mapping, using a JMS-6300 JEOL SEM with an EBSD system.

3. RESULTS AND DISCUSSION

3.1 Homogeneity of Deformation. Scribed grids were used to study the homogeneity of deformation in the billet after one pass (after more than one pass the grids could no longer be recognised). The deformation was investigated for both high and low die surface friction conditions. The general results of the extruded grids were in very close agreement with previously reported FE model predictions [4]. Comparing Fig.1c, the 'low friction' to Fig.1d, the 'high friction' case it can be seen that with low friction the shear strain distribution in the die deformation zone is less homogeneous than for the high friction condition. This is because in the low friction condition the specimen slides more easily and there is insufficient constraint to fully fill the die channels bottom corner. As a consequence the sample does not experience a uniform strain across its section. Near to the top corner the majority of the deformation is a shear and this zone extends some way down through the sample. However, at the bottom corner, the shear component is negligible and the sample is mainly stretched through an arc developing a tensile plastic strain on the bottom surface, which is reversed once the material has moved past the corner. Because the sample is deformed over an arc, the net shear strain experienced by the top part of the sample, that has been largely uniformly, sheared is less intense than when the die is fully filled and was measured to be 0.93, compared to 1.15 calculated for a uniform shear (using equation 1). For the high friction case, the sample still does not fully-fill the bottom die corner, but the arc over which deformation takes place is much reduced, leading to a larger proportion of the sample width that has undergone a homogenous shear and a higher overall shear strain per-pass measured as 1.0 from the grid. The ECAE process is thus rather unusual in that to obtain a uniform shear deformation in the sample friction is actually desirable. Alternatively, a back pressure can be imposed on the exit side of the die. When a 5kN back pressure was used the grid results were more homogeneous than for the high friction case, filling of the die was complete, and the shear imposed on the sample was measured as 1.1 which was even closer to that theoretically expected from equation (1).

Fig.1b illustrates another cause of inhomogeneity in ECAE processing. As the sample does not change shape significantly during ECAE processing, but is progressively more sheared by each pass, the shear deformation must be accommodated within the constraints of the external surface of the extruded specimen. As the strain is increased, the elongated grains are thus forced to wrap around at the end of the specimen, a similar effect has been observed by Wu et al. using a plasticine model [9]. This accommodation can lead to a local zone where the lamellar like flow of the grains breaks down, as seen in Fig.1b. However the majority of the deformation of the sample, even after 15 passes, still appears to be due to the development of an accumulative homogenous shear, leading to a highly directionally deformed grain structure similar to that seen in rolling or extrusion. In terms of the generation of the deformation structure, the additional strains required for the accommodation are probably small, in terms of the development of the deformation structure, to the enormous shear strains, although further investigation is needed to confirm this supposition.

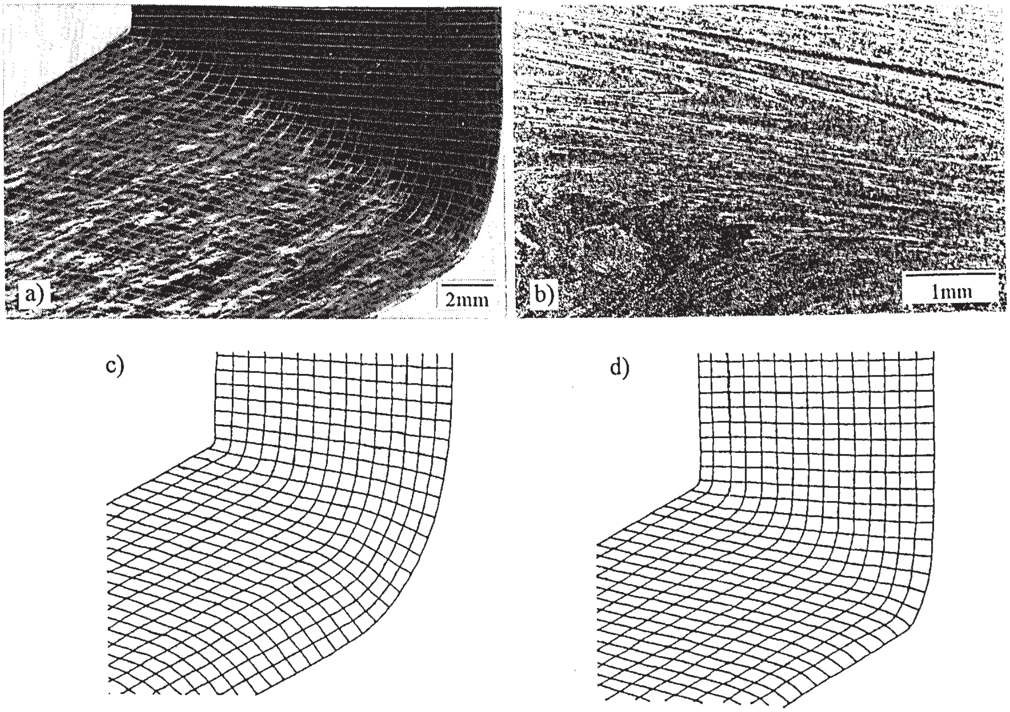


Figure 1 *The homogeneity of deformation in the ECAE process. In a) a photographed partially deformed grid scribed on a split sample is shown for the high friction case, after one pass and in b) the metal flow near the sample end, as seen in an optical micrograph of an anodised sample, after 15 passes. In c) and d) magnified traces of the partially deformed grids are given for the low and high friction case respectively.*

The validity of the assumption that the majority of the deformation is by a uniform shear was further investigated by measuring the grain widths as a function of the number of passes and comparing them to predictions made, based on the calculation of the geometric shape change of an element for a homogeneous shear with a die angle of 120°, using the relations given by Segal in [2]. Fig. 2 shows the graph of the expected grain width calculated for progressive increase in strain after each pass for different initial grain sizes for materials A, B and C, deformed at 200°C. An example of the change in grain shape is shown in figure 3a, after one pass, and after multiple passes. The microstructural changes are discussed in more detail below. For the deformed cast structures the original grain widths could be recognised from the position of the eutectic particles. At high strains (greater than 6 passes) the original grains were very difficult to distinguish, and the measurements became increasingly subjective. It can be seen from the graph that up to 6 passes (corresponding to a shear strain of 6.9) the reduction in the measured grain widths are comparable to the calculated values, which suggests that the assumption that the majority of the deformation is caused by shear in the die is reasonable. An important conclusion that can be made from figure 2 is that, in common with other shear processes (e.g. torsion [10]), the rate of reduction in grain width reduces with accumulative shear strain. As some mechanisms of recrystallisation, that are thought to give rise to the formation of ultra-fine fine grain structures at high strains, are dependent on the impingement of the original deformed grain boundaries [8,11,12] this is particularly significant. For example, with a large initial grain size (e.g. 200μm, material A in figure 2) the deformed grains will only approach a separation of the order of the subgrain size required for geometric dynamic recrystallisation at impossibly large strains, or at very low values of the Zener-Hollomon parameter. However, sample C might be expected to approach this condition after 7 passes.

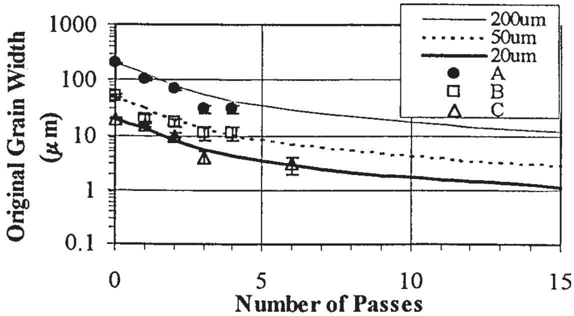
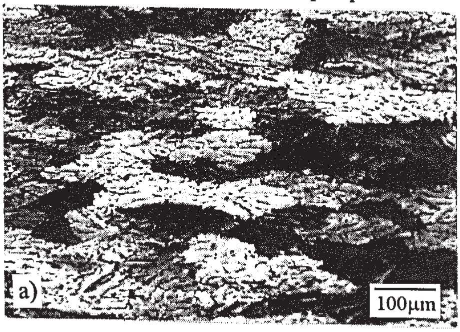


Figure 2 *Measured grain width, as a function of the number of passes, for different original grain sizes. The lines are calculated predictions based on a uniformly applied shear of 1.15 per pass.*



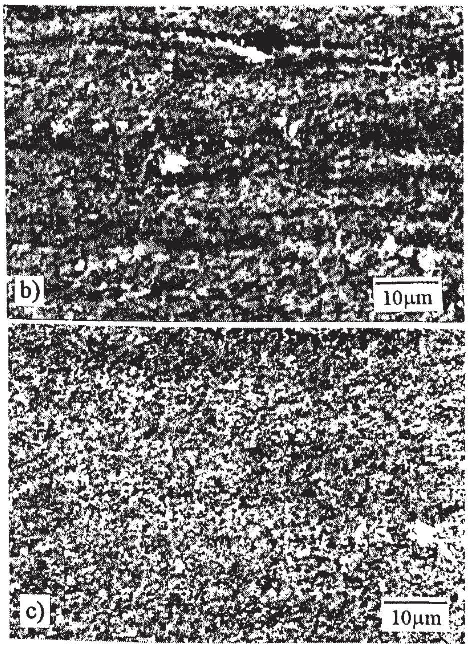


Figure 3 *Material A deformed through the ECAE die at 200°C. In a) an optical image of an anodised sample after one pass, showing the sheared originally equiaxed cast grains, in (b) an SEM back scattered image after 10 passes (γ = 11.5) and in (c) after 15 passes (γ = 17).*

Microstructural Evolution of the Deformed Samples To see if very high strain ECA extrusion could be used to produce ultra-fine grained homogeneous samples from an inhomogeneous casting, alloy A was deformed at 200°C up to a shear strain of 17 (15 passes). SEM images of the deformed structures are shown in figures 3b & c for two different strain levels, of γ = 11.5 (10 passes) and γ = 17 (15 passes). From figure 3 it can be seen that the deformed structure for a strain of 11.5 is highly directional and resembles that seen in rolling. But at the higher strain of 17 the structure appears to be largely isotropic and comprised of an ultra-fine microstructure with grain/sub-grain size of the order of 1μm. Whether the microstructure is comprised of an ultra-fine-grain structure, or is made up of subgrains, can not be readily determined from back-scattered images alone. However, there is no obvious remnant of the banded deformation structure seen at lower strains (Fig. 3b). EBSD could not be reliably carried out on the deformed samples because the "grain" size was too small, but was used after annealing at 300°C (below). TEM imaging showed that when the deformed structure was observed at a higher resolution it is less homogenous and isotropic than the SEM images would suggest. In the TEM the microstructure could be seen to be comprised of very fine asymmetric grains, typically with an aspect ratio of around three, containing a few low angle boundaries perpendicular to the direction of grain alignment and grain width of 0.3μm. A typical example is shown arrowed in Fig. 4a and is very similar to that noted by Harris et. al, for pure aluminium deformed at room temperature [4]. The exact mechanism of formation of such heavily deformed microstructures is still under investigation, but is thought to be related to the large increase in high angle boundary area generated by distortion of the original grains and deformation banding within the grains [4,6,7]. On lightly annealing, such structures are known to reorganise themselves continuously into a high angle grain boundary network by eliminating some of the low angle boundaries and forming more equiaxed grains with less distorted grain boundaries [13]. This is a very different process to that expected at higher temperatures due to Geometric Dynamic Recrystallisation, and has occurred at a strain lower than that required for impingement of the original cast grain boundaries (Fig. 2).

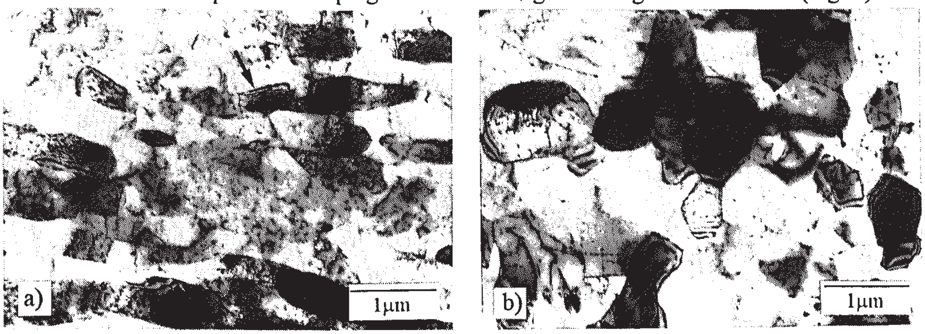


Figure 4 *TEM micrographs of material A deformed to a shear strain of 17 at 200°C (a) and after annealing at 300°C (b).*

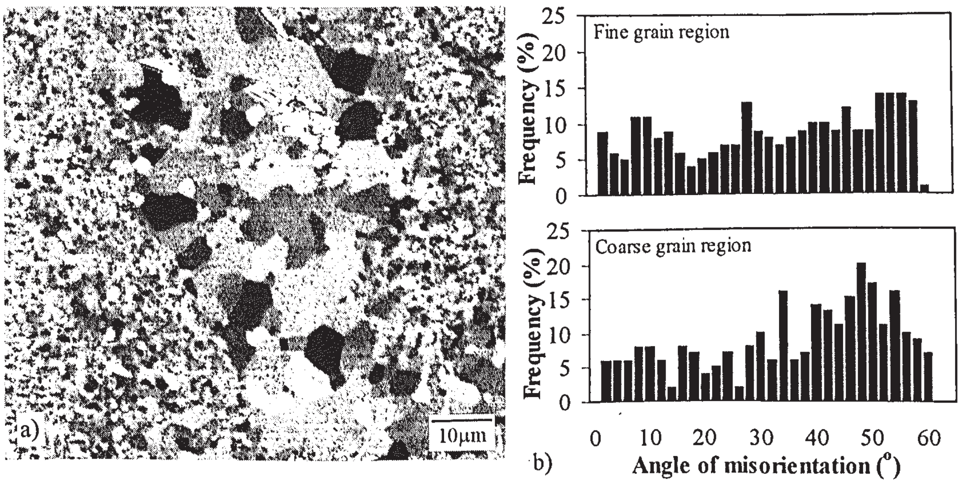


Figure 5 *SEM back scattered image of alloy A deformed to a strain of 17 and annealed for one hour at 300°C. In (b) boundary misorientation distributions are shown for the two regions in (a), comprised of course and fine grains.*

Annealing Behaviour By deformation at 200°C, to the very high strain of γ = 17, it has been shown above that it is possible to produce a material with an almost isotropic heavily deformed structure comprised of ultra-fine grain fragments containing few low angle boundaries. To assess the stability of this structure the heavily deformed sample was annealed at 300°C, for 1 hour. In Fig. 5a a back scattered SEM image of the annealed material is shown, revealing an inhomogeneous grain structure. The linear intercept grain sizes were 5.4 μm for the coarse and 0.4 μm for the fine regions. The fine grained and coarse grained regions in Fig. 5a were analysed in more detail by orientation mapping in the SEM. From Fig. 5b it can be seen that both fine and coarse areas exhibit very similar grain boundary misorientation distributions with a large proportion of high angle boundaries. The average grain boundary misorientation was found to be 34° in the fine grain region and 36° in the coarse grain region. The boundary misorientation distributions, and comparison of the TEM micrographs in Fig. 4, show that both the coarse and fine grained regions have continuously recrystallised upon annealing to give equiaxed grains (Fig.4b) with a high average misorientation. However, it is apparent that in some regions the grains have coarsened more rapidly than others leading to discontinuous grain growth (Fig.5a). Similar observations have been reported by Markushev et al. [5]. EDX analysis of the Mg level, across the sample, showed the Mg level to increase in the region of the larger grains, indicative of solute banding remaining from the cast structure. Furthermore Zr would be expected to partition in the opposite sense to Mg in the casting. The presence of Mg increases the stored energy in the vicinity but also decreases the grain boundary mobility. It is thus more likely that a non-uniform distribution of Zr is responsible for the discontinuous growth behaviour, by preferential Al_3Zr precipitation in the fine grain regions, inhibiting boundary mobility. However, more work will be needed to confirm this hypothesis.

4. CONCLUSIONS

The homogeneity of deformation in the ECAE process was found to be highly dependent on friction, as was the level of shear imposed on the sample per pass through the die. Although the majority of deformation in ECA extrusion appears to be by a homogeneous in plane shear, for the sample to maintain a constant geometry additional deformations are required to accommodate the progressive shape change that this shear generates.

On deformation to shear strains of ~ 17 an ultra-fine grain structure was formed by the increase in high angle boundary area, due to the distortion of the original grains, and the formation of high angle boundaries by grain fragmentation. This produced what appeared to be a relatively homogeneous ultra-fine, isotropic, grain structure, but in the TEM was found to still contain a few low angle boundaries, within slightly asymmetric grain fragments. On annealing the structure was unstable and coarsened discontinuously to give a bimodal grain size distribution, which was related to an inhomogeneous solute distribution in the original casting.

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REFERENCES

[1] D.J.Lloyd and D.Kenny: Acta Metall., 28 (1980), 639.
[2] V.M.Segal: Mat. Sci. and Eng., A197 (1995), 157.
[3] R.Z.Valiev, A.V.Korznikov and R.R.Mulyukov: Mater. Sci. Eng., A168 (1993), 141.
[4] C.Harris, S.M.Roberts, P.B.Prangnell: Proc. 3rd Int. conf. on recrystallisation and related phenomena, REX96, ed. T.R.McNelley, (MIAS), California, (1997), 587.
[5] M.V.Markushev, C.C.Bampton, M.Yu.Murashkin and D.A.Hardwick: Mat. Sci. and Eng., A234-236 (1997), 927.
[6] A.Gholinia, J.Sarkar, P.J.Withers and P.B.Prangnell: Submitted to Acta Metall., (1997).
[7] A.Gholinia, S.I.Hulley and P.B.Prangnell: Proc. 3rd Int. conf. on recrystallisation and related phenomena, REX96, ed. T.R.McNelley, (MIAS), California, (1997), 537.
[8] F.J.Humphreys and M.Hatherly: Recrystallisation and related annealing phenomena, pub. Elsevier Science Ltd., Oxford, (1995).
[9] Y.Wu and I.Baker: Scripta Mater., 37 (1997), 437.
[10] J.G.Sevillano, P.Van Houtte and E.Aernoudt, Prog. Mater. Sci., 25 (1980), 69.
[11] M.D.Drury and F.J.Humphreys: Acta Metall., 34 (1986), 2259.
[12] H.J.McQueen, O.Knutstad, N.Ryum and J.K.Solberg: Scripta Metall., 19 (1985), 73.
[13] F.J.Humphreys and H.M.Chan: Mat. Sci. and Tech., 12 (1996), 143.