The Effect of Initial Grain Size on the Generation of High Angle Boundaries During the Equal Channel Angular Extrusion of Aluminium to High Levels of Plastic Strain

Chris Harris[1,2], Phil B. Prangnell[1], and Xiaoming Duan[2]

[1]*Manchester Materials Science Centre, University of Manchester / UMIST, Grosvenor Street, Manchester, M1 7HS, U.K.*
[2]*EA Technology Ltd., Capenhurst, Chester, CH1 6ES, UK.*

ABSTRACT

Equal channel angular extrusion was used to cold deform commercially pure aluminium samples, with two very different initial grain sizes, in order to quantify the effect of the starting grain size on the deformation structure formed at very high strains. The development of high angle boundary area, average low angle and high angle boundary separations, and boundary misorientation distributions, were measured using EBSD orientation mapping, up to an applied effective strain of 10. It was found that the deformed states became identical, for the two different grain sizes, at strains greater than 6 and that the high angle boundary separation converged on the low angle boundary spacing (subgrain size) at very high strains. At low strains, the rate of formation of high angle boundaries was much greater for a coarse grained sample. The geometric increase in high angle grain boundary area, due to the distortion of the original grains, was found to be only a small proportion of the high angle boundary area generated in the coarse grained samples, the majority of high angle boundaries being produced by grain fragmentation. However, the geometric increase in high angle grain boundary area became more important as the initial grain size was reduced.

Keywords: *ECA extrusion, aluminium, ultra-fine grain size, high angle grain boundaries, EBSD*

INTRODUCTION

Ultra-fine grain (UFG) materials are of great technological and scientific interest due to their potentially superior properties compared to alloys with conventional grain structures [1]. Superplasticity at high strain rates and increased strength are some of the claimed benefits of an UFG structure [2,3]. It has been shown that an effective method for producing ultra-fine grained materials is through promoting continuous recrystallisation by deforming alloys to very high levels of plastic strain (typically greater than ε_{true}=5). This form of recrystallisation occurs once there are no true sub-grains (having a surface of entirely low angle boundary) left in the deformed structure, all the crystal fragments having a significant proportion of their perimeter consisting of high angle boundary [4]. When this condition is reached, the deformed structure recrystallises continuously upon annealing, forming a high angle boundary network with a grain size related to the crystal fragment size [4]. Normally the high strains required to invoke continuous recrystallisation can only be produced by rolling alloys to very thin foils, or in torsion testing, where the resultant samples are very small and their properties very difficult to determine. A technique, initially developed by Segal and colleagues [5], for the deformation of materials to very high plastic strains is that of Equal Channel Angular (ECA) extrusion. The ECA extrusion process deforms the sample by shear in a constant cross-section die, so that the billet dimensions are maintained constant and there is no geometric limit to the strain that can be developed. This method has been investigated by a number of workers [1,2,3,6] and aluminium alloys deformed via ECA extrusion to effective strains of 5-10 have produced grain sizes in the range 0.3-2 μm [6,7].

To date, there has been a relatively little quantitative analysis carried out on the development of the deformation structure and its continuous recrystallisation at high plastic strains. During plastic deformation, it thought that the high angle grain boundary area is increased by two mechanisms. The geometrically required increase in area as the original grains become distorted in proportion to the specimen shape change [8], and the formation of high angle grain boundaries within the grains

from intense deformation bands, due to grain fragmentaion [8]. A very high increase in high angle boundary area is required before continuous recrystallisation can take place. Obtaining quantitative microstructural data of the rate of formation of high angle boundaries during deformation has previously been both difficult and laborious. However, with the introduction of automated electron back scattered diffraction (EBSD) measurement of grain orientations in the SEM statistically significant data can now be readily obtained from suitable materials [7]. Through the extensive use of EBSD orientation mapping, this paper will hopefully answer some of the questions that surround the formation of an UFG microstructure in aluminium alloys.

EXPERIMENTAL METHOD

All the experiments were conducted using commercial purity aluminium (99.75% Al), provided by EA Technology Ltd., and cast at two different cooling rates, to give samples with grain sizes of 1.5mm and 150μm and a random texture. Cylindrical samples, 12 mm in diameter and 50mm in length, were machined from the centres of the castings. The samples were then subjected to ECA extrusion at room temperature. The die used in the experiments had a die angle of 135°, between the two channels. The strain per pass is dependent on the processing conditions as well as the die angle (see for e.g. [7]). However assuming shear is homogenous, the theoretically expected shear strain per pass was 0.8 and the effective strain 0.5 [7]. Specimens with the two initial grain sizes of 1.5mm and 150μm were deformed for 2, 6, 10, 16 and 20 passes corresponding to effective strains of 1, 3, 5, 8, and 10 respectively. A molybdenum disulphide lubricant was used and extrusion was repeated, maintaining a constant strain path, until the desired strain for each specimen was reached.

For microstructural characterisation the specimens were sectioned and examined in the plane of shear, along the axis of the billet. The specimens were studied in a JMS-6300 JOEL SEM with an integrated EBSD system. Automated grain orientation measurements were carried out over areas of 200μm x 150 μm. Because the resolution of the orientation determination in the SEM was 0.2μm in the direction of the tilt axis and about 0.6 μm in the transverse direction, an insufficient percentage of patterns could be indexed with the as deformed samples. The samples were consequently lightly annealed at a modest temperature of 200°C for 10 min., prior to analysis in the SEM, to allow a controlled level of recovery and improve the percentage of successfully indexed patterns to an acceptable level. A minimum of 7,500 and maximum 30,000 data points were collected over any scanned area. Grain boundary misorientation's, between adjoining grains were calculated using the 'ICE' and 'VMAP' analysis packages. From the misorientation data, the percentage of high and low angle grain boundaries was obtained (by convention, low angle boundaries were defined as boundaries of lower than 15° misorientation). A map showing only the high angle grain boundaries was then produced and the boundary length per unit area measured. This experimentally obtained value, assumed to represent the grain boundary area per unit volume, was compared to the theoretical geometrical increase in the grain boundary length per unit area. The geometrical increase in grain boundary length was simply calculated from the change in length of the perimeter of a cubic grain being deformed by pure shear using the relations given in [5].

RESULTS AND DISCUSSION

Figure 1 shows the high angle grain boundary maps and the misorientation distributions that were obtained from the orientation area scans, as a function of strain, for the two materials with the different cast grain sizes of 150 and 1500 μm. Figures 1a-1c shows results for the coarser grained sample and figures 1d-1f for the fine grained sample, at strain intervals of 1, 3 and 10 respectively. In the samples deformed to a strain of one (figures 1a and 1d) it is already evident that even at this stage there has been some generation of high angle grain boundaries within the grains. In figure a, the area covered by the map is within one initial cast grain width and would exhibit no high angleboundaries unless some new boundaries had been generated within the grains. Because of the smaller initial grain size in the second material, the area in figure 1d would accommodate approximately 2 orignal grain widths, however, in this sample again more high angle boundaries have been formed than can be attributed just to the distrotion of the original cast grains. At this strain the majoirty of the boundaries within the sample are low angle, with only a small percentage of high angle boundaries, as can be seen from figure 2 where the percentage of high and low angle boundraies are plotted against strain. Figure 2 reveals that during the early stages of deformation there is a dramatic increase in the fraction of low angle boundaries, as would be expected for a conventional "low" strain deformation process. In figure 2 it can further be seen that the fraction of

high angle boundaries steadily increases with higher levels of strain, leveling off in both materials at about 80% on approaching strains of 10.

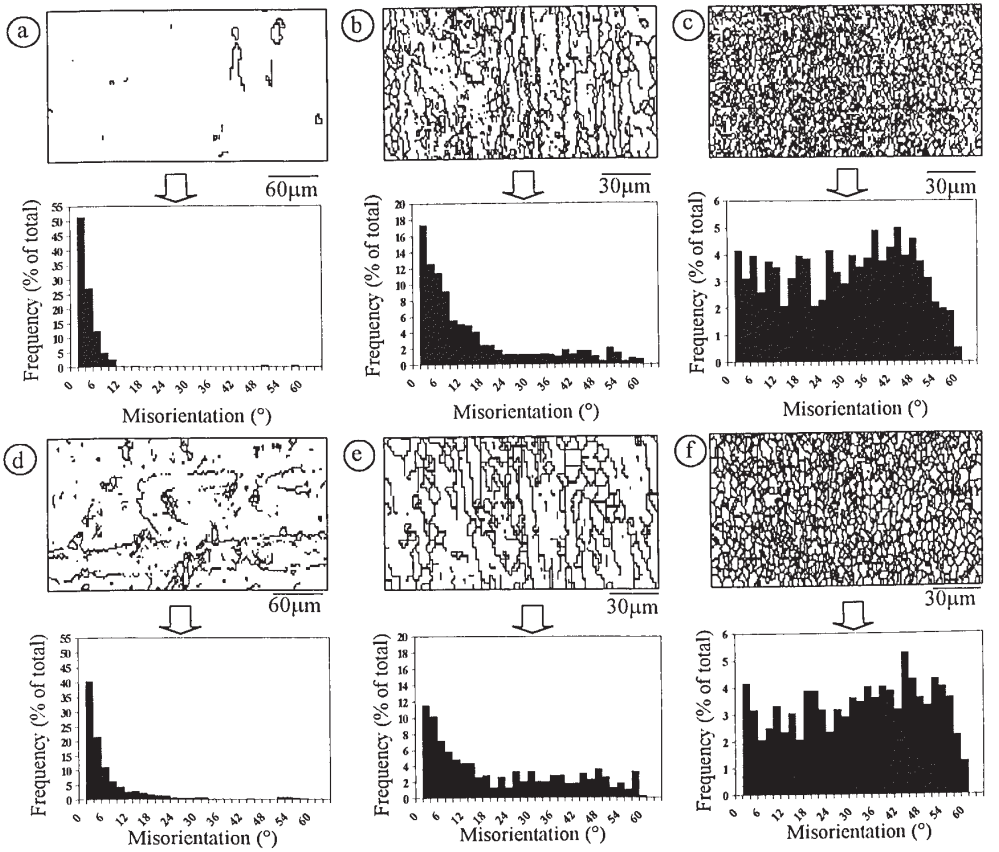

Figure1: *High angle grain boundary maps, and the corresponding boundary misorientation histograms, for the samples with a very coarse 1500 μm initial cast grain size (a-c) and a much finer 150μm cast grain size (d-f) deformed to strains of 1, 3 and 10 respectively.*

At intermediate strains (effective strain = 3, figures 1b and e) there is a noticable increase in the density of high angle boundaries (HAGBs) in both materials, but they form a lamellar grain structure aligned in the direction of deformation, typical of that seen in rolled materials. At this stage there still appears to be a slightly higher fraction of low angle boundaries in the coarse grained sample, compared to the fine grained. From the relevant boundary misorientaion distributions (figure 1b, 1e) it can be seen that the misorientations of the HAGB that have been generated at this strain are in the range 15°-30°. In a previous study by Hughes and Hansen [9], at simliar strains, the origins of this type of HAGB formation was attributed to heterogeneous defromation within the grains, where sepearate parts of the grain roate towards different stable texture components. As the strain is further increased there is a progressive shift towards a higher mean boundary misoreintaion. In figures 1 c and f, it can be seen that at very high strains a homogeneous network of high angle grain boundaries are formed, with no noticable difference between the two materials, and a grain size of around 3-5μm. At this high strain, and after the annealing treatment, the samples thus appear to have contiuously recrystallised. The grain boundary misorientation distribuiton can also be seen to be approaching that of a 'Mackenzien' distribution expected for a random assembly

of grains [10]. Interestingly, the high angle boundary maps, misorientaion distributions, grain sizes, and textures were all found to be identical for both materials, despite the two very different starting grain sizes.

The fact that the final microstructure at very high strains is independent of initial grain size, is illustrated in figure 3 where the mean linear intercept boundary separations, obtained from the misorientation maps, are plotted for both high and low angle boundaries as a function of strain. In the coarse grained, cast sample, the initial high angle boundary separation is obviously far greater than for the fine grained. As the subgrain size is known to change very little for strains greater than 1 [11], in the strain ranges investigated, it is not surprising that the mean low angle boundary separations are virtually constant (within the experimental scatter) and are the same for both specimens. In contrast, the high angle grain boundary separations rapidly decrease with strain and for both materials converges with that of the low angle boundary separation (subgrain size) at very high strains. This convergence appears to take place at a significantly lower strain for the material with the finer initial grain size, occurring at an effective strain of around 3-4 in the 150 μm and 5-6 in the 1500 μm grain size materials respectively. The point of convergence of the subgrain size with the high angle boundary separation might be considered to be where the material approaches the condition required for continuous recrystallisation. However, at this point in the deformation process an individual crystal may be bounded by low angle boundaries predominantly, by a mixture of low and high-angle boundaries, or predominantly by high angle boundaries. Previous work has shown [7] that it is only after the specimens have been deformed to higer strains (greater than 7 in general) that the individual crystal boundaries in both samples are predominantly high angle and that the samples can be genuinely described as being continuously recrystallised.

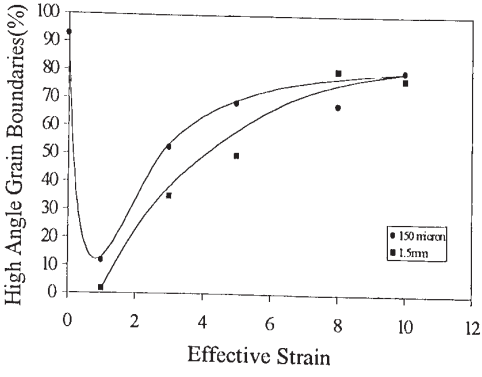

Figure 2 *Percentage of low (<15°) and high (>15°) misorientation angle boundaries, as a function of strain, for the two materials with initial grain sizes of 1500 and 150 μm,.*

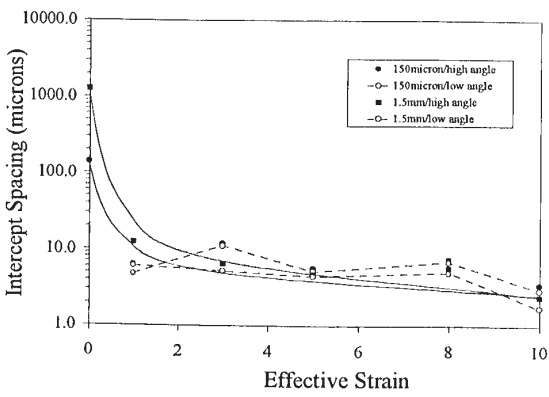

Figure 3: *Low and high angle boundary mean linear intercept spacings, for the samples with initial grain sizes of 1500 μm and 150 μm, as a function of strain.*

From figure 1 it has been inferred that new high angle grain boundary area has been created by the formation of high misorientaion boundaries within the grains by a process of grain fragmentaion. During plastic deformation, the original cast grains must change their shape in proportion to the emposed strain, resulting in a geometrically required increase in high angle boundary area. This process is thought to be extremely important during elevated temperature deformation, where grain fragmentaion does not occur due to the more homogeneous nature of plastic flow at higher tempearatures, and can ultimtely lead to geometric dynamic recrystallisation [12]. The increase in grain boundary area, attributable to this effect, must be strongly related to the initial grain size. To see if the generation of geometrically required grain boundary area makes a significant contribution to the overall increase in area with strain at room tempearture the geometrically required area was calculated (see experimental method) and comapred to that measured from the high angle boundary maps, examples of which are shown in figure 1. The measured and calculated increases are depicted in figure 4 for the two different initial grain sizes.

From figure 4 it can be seen that because the samples have very different cast grain sizes, there is a large initial difference in the grain boundary area per-unit volume. In the lower strain range, typical of conventional metal working processes, the measured high angle boundary area in the large grained material increases much more rapidly than in the fine, the disparity between the two reducing with strain. In line with the previously presented data, the measured grain boundary areas converge at strains greater than 6 and approach a limiting value at very high plastic strains. Because with shear deformation the distortion of the initial grains reduces in rate with accumulative strain the geometrically required increase in grain boundary area also decreases in rate. In both

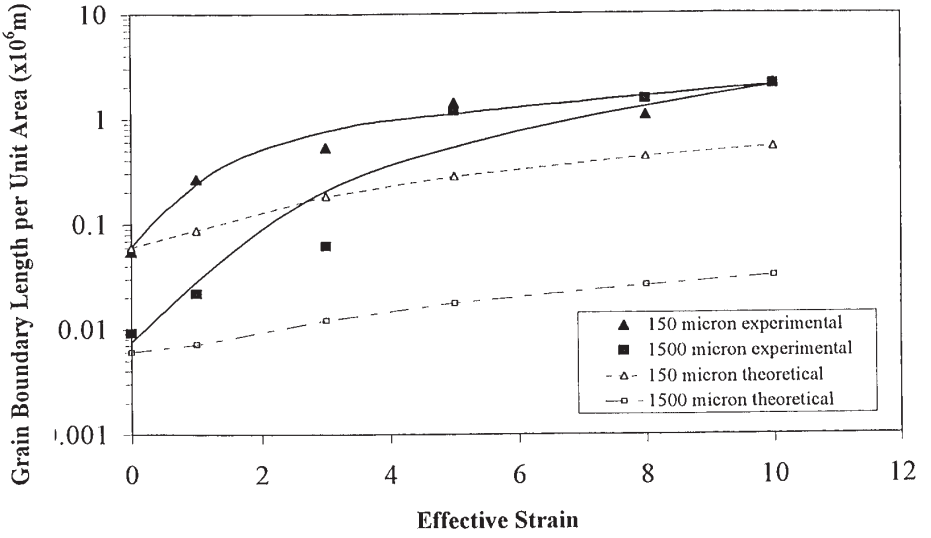

Figure 4: *The measured grain bounday area per unit volume plotted against effective strain for the two samples, with initial grain sizes of 1500 and 150 μm. The broken lines depict the calculated geometrically required increase in grain boundary area for the two different grain sizes.*

materials the theoretically expected, geometrically required increase in grain boundary area is small relative to the amount measured. In the material with the larger initial grain size the effect is minimal and the increase in area is only 1% of the total. Nevertheless, in the material with the smaller initial grain size, the geometric effect is significant and accounts for 20% of the total increase in high angle grain boundary area at an effective strain of 10. The difference between the actual grain boundary area, and that calculated, is direct evidence of the importance of grain fragmentation which has been linked to grain subdividison into different textural components, resulting in the generation of new grain boundaries within grains [9,13]. The exact mechanism of this process is complex and is not currently fully understood. However, in large grained materials a larger volume of the grains are less constrained by their neighbours and are more likely to behave like single crystals, possibly resulting in a greater degree of the grain subdividison into different textural components. By extrapolation from the level where the high angle grain boundary areas converge and tend to a limit at high strain it is possible to predict that a material with a starting grain size of 20 μm could generate sufficient high angle boundary area simply by distortion of its original grains to continuously recrystallise at a strain of 10. It is hypothosised that this would occur without any grain fragmentaion, although this has yet to be experimentally verified.

When considering figures 1-4 as a whole, it is apparent that during deformation to high strains the material converges on a stable grain fragment size, irrespective of the starting grain structure. resulting in a far more homogenous deformation structure than that seen at low strains. Figure 3 suggests that this fragment size is the same order as that of one subgrain and is thus controlled by the smallest identifiable crystal fragment within the deformation structure. It has to be noted that these levels of strain are not normally reached during conventional defromation processes, which

rarely use strains in excess of 4. Also, in commercial themomechanical processing it is already well known microstructural variables, such as the initial grain size, have a pronounced effect on the development of the deformation structure and the subsequent recrystallisation behaviour.

CONCLUSIONS

Equal channel angular extrusion has been used to deform commercially pure aluminium samples, with two very different initial grain sizes to very high plastic strains. Due to the limitations of the resolution of the EBSD system, used the samples were lightly annealed before examination. However, the extensive use of orientation mapping in the SEM has proved extremely revealing as it has allowed the generation of high angle boundary area and boundary misorientations to be quantified as a function of strain.

Both materials were found to produce a very fine homogenous grain structure by continuously recrystallisation at high strains, resulting in remarkably similar microstructures, irrespective of their different initial grain sizes. However, this process appeared to occur at lower strains in the fine grained material. The high angle boundary separation converged with the low angle boundary spacing at high strains (greater than 6) as a result of the generation of additional high angle boundaries during plastic deformation. Mechanistically this takes place by the geometrically required distortion of the original grains and grain subdivision.

The geometrically required increase in high angle grain boundary area, due to the distortion of the original grains has been found to be only a small proportion of the high angle boundary area generated, in the case of a coarse grained sample, the majority of high angle boundaries being produced by grain fragmentation. However the geometrically required increase in grain boundary area becomes far more important as the initial grain size is reduced and may dominate for initial grain sizes less than 20 μm.

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