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Intermediate annealing of pure aluminum during cyclic equal channel angular pressings

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Intermediate heat treatment was employed to anneal pure Al samples subjected to cyclic Equal Channel Angular (ECA) pressing in order to investigate the microstructural change and tensile properties of as-pressed and annealed samples. As-pressed samples were annealed at 200°C for 1 hr after 2 and 4 passages. Some annealed samples were pressed further to compare the changes in the properties of pure Al with the same equivalent strain induced by cyclic ECAP but having different thermal history. Intermediate annealing in the cyclic repetitive ECA pressing appears to be an effective method to form an equiaxed, fine-grained structure with high misorientation angle. It decreases the aspect ratio of subgrains while keeping the grains size unchanged. Annealing followed by further pressing results in an increase in the aspect ratio but small decrease in the grain size. It is speculated that recrystallization of ECA pressed aluminum is responsible for the significant increase in the misorientation angle.

1. Introduction

For deforming alloys to severe plastic strains, constant billet geometry gives a big advantage over conventional metal working processes such as rolling and extrusion, where the work piece is always being reduced in one dimension. In the equal channel angular pressing (ECAP), there is no geometric restriction to the maximum strain that can be reached and alloys have been processed to strains greater than 10 using this method [1, 2]. With this unique feature, the applicability of ECAP has been currently expanded for various kinds of materials.

It has been shown that a material with a coarse grain size can be refined to have a fine grain size through intense plastic deformation. With the large accumulative strain while keeping the shape of a material, the ECAP is very effective in producing ultra-fine grain size even in a bulk form [3–5]. The introduction of an ultra-fine grain size may lead to significant change in the mechanical properties of the material.

Most of the works to date have been trying to understand the development of fine grain size with high misorientation angles by employing different processing routes and cyclic pressings. They have revealed that subgrains form in the early stage of cyclic pressings and these subgrains subsequently evolve with further pressings into an array of grains separated by high angle grain boundaries with further pressings [6, 7]. In spite of the effective shearing deformation, there seems to be a limit in the reduction of grain size with increasing number of ECA pressings [5].

The recent work using a die with channel angle of 135° has made the suggestion that two or three passes followed by the low-temperature annealing can be useful to break down an initial coarse structure, refine the grain size and increase the strength of the material [8, 9]. Although effective shear strain makes it possible to create fine grains with high-angle boundary through repeated pressings, it seems reasonable to think that suitable annealing following ECAP results in the formation of stable subgrains with high misorientation angles that may facilitate the multiple slip to promote further plastic deformation without cracking. Furthermore, it has an additional advantage that the load needed to press the sample becomes lower than that of the previous path during the cyclic pressing: which makes cyclic pressing difficult especially for materials with strong strain hardening behavior.

Unfortunately, up to date, majority of the studies on the ECAP has focused on the deformation structures and the mechanical properties of various alloys deformed to such enormous strains. There is little work on the effect of annealing between passages during repeated ECA pressings.

In this study, intermediate annealing was performed on some ECA pressed samples in order to investigate

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the change in the microstructure and to compare the tensile properties of as-pressed and annealed samples. We tried to understand the effect of intermediate annealing on the grain size refinement and the development of stable subgrain structure before successive ECA pressing.

2. Experimental procedures

The material used in this study was a pure Al with 99.9% purity. An ingot was prepared by melting Al pellets and cast into a rectangular mold ($80 \times 70 \times 120$ mm^3). The ingot was extruded to produce a bar (18 mm in diameter). Extruded samples were then heat treated at 500°C for 15 hrs to create the equiaxed grain structure with grain size of about 200 μm. Prior to ECA pressing, the surface of the samples was polished using SiC paper (1000 grit).

The pressing die designed for this study has two channels having the same square cross-section with dimensions of 12×12 mm^2 and meeting in an L-shape configuration at an angle of 90°. The angle defining the arc of curvature at the outer point intersection of the two channels was 0°. The resultant shear strain with this die can be predicted to be 1.15 from the proposed equation [10]. ECAP was conducted at room temperature with a speed of 10 mm/min. All pressings were done using the route A where samples were not rotated between each pressing. Billets were pressed to 1, 2, and 4 passes, and annealed at 200°C for 1 hr. For the comparison, other samples pressed 1 or 2 passes at first were annealed at the same condition and then continuously pressed to additional 1 or 2 passes respectively. To make a concise indication of the processing to which a specimen has been subjected, hereafter we use "D1 + D1" indicating a sample subjected to 1 pass ECA pressing and annealed at 200°C for one hour and ECA pressed for additional 1 pass.

Following the pressing, small specimens were taken from the side faces at the exit from the die (y-plane described in [11]). Microstructure of the ECA pressed pure Al was examined by TEM (Philips CM30) operated at 200 kV. For TEM observation, final thinning was done by electro jet-polishing in a solution consisting of 30% nitric acid and 70% methanol at −30°C. The samples were prepared for electron back scattering diffraction (EBSD) by removing the top surface with SiC paper followed by an electro-chemical polishing using colloidal silica to obtain a good surface finish. A scanning electron microscope (JEOL 6400) equipped with EBSD was used for misorientation measurements. EBSD image was constructed with three different colors representing the three different crystal orientations ($\langle 200\rangle$, $\langle 220\rangle$, $\langle 111\rangle$) with spatial resolution of 0.25 μm and angular resolution of 1°. Cut off angle was set to 1.5°. Plate type tensile specimens with a gauge length of 10 mm were machined from the samples. The surface of the specimens was finished with 1000 grit SiC paper in order to lessen the surface effect. Tensile tests were performed at room temperature with a tensile test machine (Shimadzu, AG-10TE) at 2 mm/min constant crosshead speed so that the initial strain rate for the test is about 3/sec. An obvious yield point was not observed and 0.2% proof strength was used as yield strength. Tensile properties were measured by averaging three results obtained from each condition.

3. Results

Fig. 1 shows the TEM images of as-pressed and annealed samples after two and four pressings. Each pressing gives a strain of 1.15 so that these two samples were subjected to the resultant strain of 2.3 and 4.6 respectively. The sample after two passings has a deformation banded structure comprising the submicron grains that are elongated in the direction of shear indicated by an arrow. A lot of dislocations are usually localized at the subgrain boundaries. Cyclic pressing up to four times gradually breaks up the band structure to produce fine equiaxed subgrain structure. Compared to the as-pressed samples, both annealed samples (Fig. 1c and d) seem to have more round shape of subgrains separated by clear boundary.

TEM observations of intermediate annealed samples followed by further pressing can be seen in Fig. 2. It is noted that the intermediate annealed sample shows more elongated subgrains than those of as pressed ones (Fig. 1a and b), in spite of the same resultant shear strain exerted by ECAP. Concerning that relatively equiaxed subgrain appears in annealed sample, it is supposed that further pressing after intermediate annealing causes re-elongation of round shape of subgrain formed during annealing.

Size and aspect ratio of subgrains measured from the TEM images are summarized in Table I. Both values decrease with number of pressings owing to the formation of fine and equiaxed subgrains. The shape of grains changes from an elongated to an equiaxed shape, meanwhile the size of the grains decrease with increasing strain. It is obvious that annealing decreases the aspect ratio as predicted in Fig. 2 while maintaining the grain size almost the same. Intermediate annealed samples (D1 + D1, D2 + D2) show smaller grain size and larger aspect ratio than those of as-pressed samples (D2, D4) respectively, in spite of the same resultant strain.

Recently, electron back scattering diffraction (EBSD) method has been widely used to evaluate the misorientations among fine subgrains because this technique not only examines large areas but also gives an accurate orientation distribution of each grain [7, 12, 13]. By employing this technique, the orientation distributions with process conditions are examined and represented in Table II.

TABLE I Grain size and aspect ratio of pure Al with each processing condition

Process condition	Grain size (μm)	Aspect ratio
D1	1.08 ± 0.1	2.75 ± 0.25
D2	0.88 ± 0.03	2.24 ± 0.18
D1 + D1	0.85 ± 0.08	2.3 ± 0.2
D2+	0.88 ± 0.1	2.14 ± 0.15
D4	0.79 ± 0.1	1.45 ± 0.14
D2 + D2	0.77 ± 0.08	1.5 ± 0.18
D4+	0.80 ± 0.12	1.38 ± 0.15

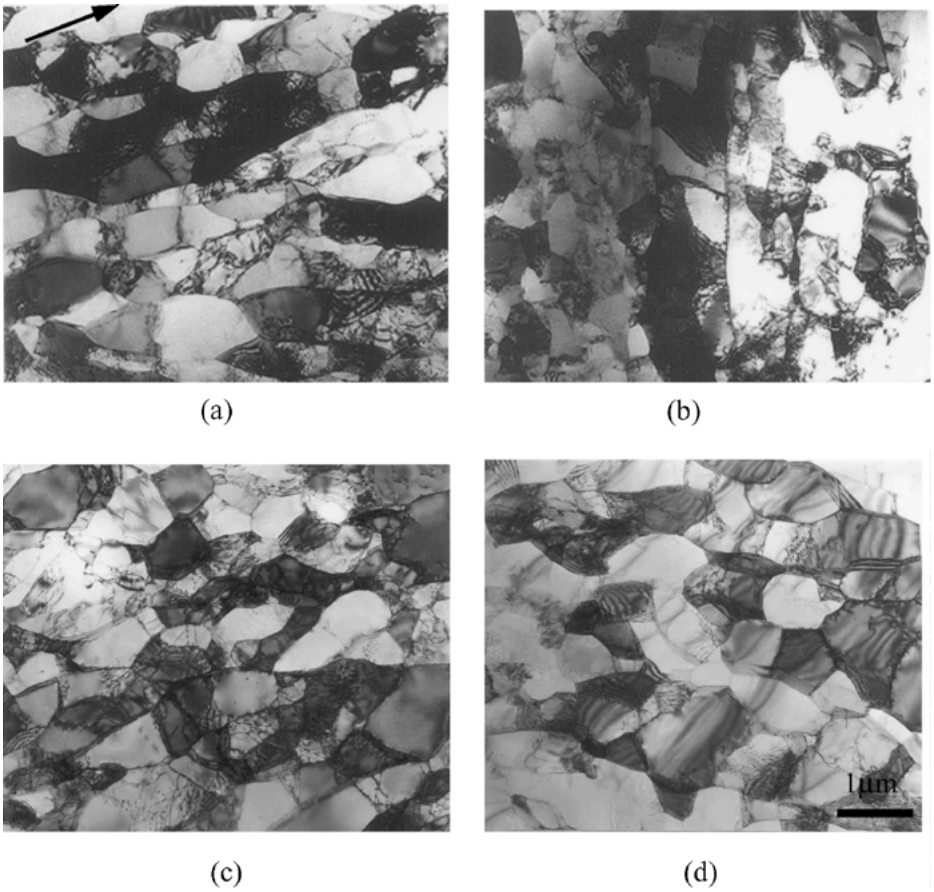

Figure 1 TEM observation of typical microstructures of as-pressed (a) and (b) and annealed (c) and (d) after cyclic ECA pressings up to two (a) and (c) and four (b) and (d) times. (a) D2, (b) D4, (c) D2+, and (d) D4+.

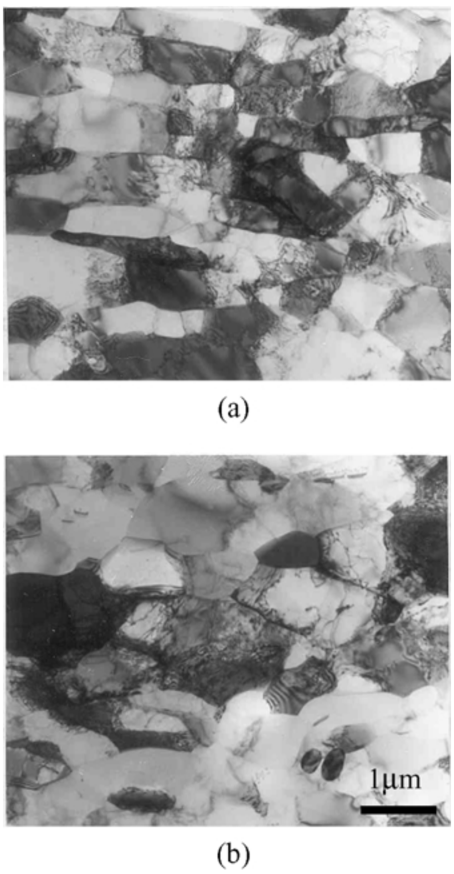

Figure 2 TEM images of samples intermediate annealed followed by further pressing. (a) D1 + D1 and (b) D2 + D2.

Table II shows the fraction of grain misorientation angles higher or lower than 15 degree. About 400 orientation measurements were done to determine the misorientation distribution. Spatial resolution of the technique is small enough to evaluate the misorientation angles between fine grains of 0.8 μm in diameter. As expected, the fraction of high angle boundaries increases with number of cyclic ECA pressings. In addition, annealing drastically increases the fraction of high misorientation angle, which is thought to be the significant role of annealing in the cyclic ECA pressing. It should be noted that almost 70% of grains appear to have high angle boundaries in the D4+ sample. Such a high fraction of misorientation was difficult to obtain without annealing process as it was reported that the same fraction of misoriention angle was not achieved until 16 cyclic pressings [12].

TABLE II Fraction of misorientation angles below and above 15 degrees at each processing condition

Process condition	Fraction of low angle (<15°) misorientation	Fraction of high angle (>15°) misorientation
D2	0.92	0.08
D1 + D1	0.7	0.3
D2+	0.62	0.38
D4	0.57	0.43
D2 + D2	0.52	0.48
D4+	0.32	0.68

TABLE III Tensile properties of cyclic ECA pressed and annealed samples

Process condition	Ultimate tensile strength (MPa)	Yield strength (MPa)	Elongation (%)
D0	67 ± 5.5	28 ± 4.8	39 ± 2.8
D1	112.6 ± 4	108.2 ± 3	16.8 ± 1.5
D2	136 ± 4.6	131 ± 4.5	15.4 ± 3
D1 + D1	133 ± 4.2	126 ± 3.2	18.6 ± 1.8
D2+	110 ± 4	102.5 ± 3.5	20.1 ± 1.6
D4	140 ± 4.8	132 ± 4.0	13.1 ± 5
D2 + D2	137.5 ± 4.5	130 ± 4.2	13.7 ± 3.4
D4+	124.4 ± 3	112 ± 2.8	14.2 ± 4

The tensile properties of the samples at each condition are shown in Table III. They vary with number of pressings in a similar manner expected for large strain deformation. For as-pressed samples, strength increases rapidly with first two passes, but further pressing results in very little additional strengthening, which is identical to the previous work [5, 8]. They show a general effect of annealing that elongation to failure increases at the expense of decrease in tensile strength. Small improvement of ductility is found in intermediate annealed sample (D1 + D1 and D2 + D2) while the tensile strength decreases very little compared to D2 and D4 respectively. Intermediate annealing followed by further pressings gives better ductility. Plausible explanation on the mechanical and microstructural differences attributed to intermediate annealing has to be given by the mechanism of high angle boundary formation under two different processes, i.e., mechanical and thermal process.

4. Discussion

It is well documented that ECA pressing causes drastic increase in the dislocation density as well as rapid change in microstructure even at the beginning of cyclic pressings owing to high strain induced by effective shear deformation [14]. The strain energy caused by ECAP should be much higher than those of conventional deformation processes. From this point of view, it is plausible that intermediate annealing is a potential method to control both microstructure and tensile properties.

As seen in Tables I and II, intermediate annealing appears to be very effective in producing stable subgrain with high angle misorientations. For the annealing effect, it is obvious that annealing after cyclic pressing drastically increases the misorientation angle because 70% of grain boundary is revealed to have high angle misorientation in a four cyclic pressed sample followed by annealing. Generally, annealing process consists of recovery (polygonization) and recrystallization (grain nucleation and growth). These thermally activated processes can give rise to high angle grain boundaries in ECA pressed samples. Since recovery involving the polygonization occurs by rearrangement of boundary dislocations, it just produces low angle boundaries with more stable dislocation configurations. Even though such change in subgrain boundary configurations cannot produce high angle boundaries, it may act as an effective nucleation site for recrystallization. It is well known that recrystallization can produce strain-free grains with high angle boundaries through nucleation and growth process. The driving force for such process is stored elastic energy and can be activated by the heat, e.g., annealing of severely deformed materials [15].

Annealing of ECA pressed sample can satisfy abovementioned conditions for recrystallization. If subgrain boundaries provide nucleation sites, recrystallized grains may have almost the same size with parent subgrains but having high angle grain boundaries. As a result of such recrystallization, one will expect slight decrease in strength but increase in elongation. Furthermore, if additional pressing is applied, grain elongation can be observed from the grains whose slip planes aligned favorably to the direction of applied stress causing increase in aspect ratio. Therefore, it is speculated that recrystallization of ECA pressed aluminum is responsible for significant increase in the misorientation angle as well as modifications of mechanical properties and aspect ratio of grains. It is known that dynamic recovery is so dominant that recrystallization is difficult to occur in pure Al. However, concerning that the purity of Al sample used is not high of 99.9% and high strain is introduced owing to ECAP, it is likely that recrystallization is considered to give high misorientation angles of grains in our ECA pressed samples. More detailed characterization is necessary to confirm the role of intermediate annealing on the microstructural modification.

5. Conclusions

Apart from the fact that applied load to deform the material through a ECAP die is greatly reduced by introducing intermediate annealing, it is found that intermediate annealing in the repetitive ECA pressings is regarded as an effective method to form an equiaxed fine grain structure with high misorientation angles. Annealing decreases the aspect ratio of subgrains while keeping the grain size the same. The distinctive effect of annealing is to increase the fraction of high misorientation angles by 20% compared to that of as-pressed samples. Intermediate annealing followed by further pressing results in an increase in the aspect ratio while small decrease in the grains size. It should be emphasized that intermediate annealing makes improvement of ductility without sacrifice of tensile strength by increasing misorientation angle of subgrains.

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