

Grain Refinement in As-Cast 7475 Aluminum Alloy under Hot Equal-Channel Angular Pressing

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Microstructural evolution taking place during equal channel angular pressing (ECAP) was studied in an as-cast 0.16%Zr modified 7475 aluminum alloy at a temperature of 673 K. The structural changes are characterized by development of deformation bands due to large strain inhomogeneity occurring through the ECAP die. Repeated deformation leads to an increasing of number and misorientation angle of the boundaries of deformation bands, finally followed by formation of new fine grains at high strains. The misorientation angle distribution for newly developed boundaries shows a single peak type at relatively low misorientations in low strain and changes to a bimodal distribution with two peaks at low and high misorientations in moderate strain. Pressing to a strain of 12 leads to a full development of new grains surrounded by medium to high angle boundaries with an average size of about 1.7 μm . It is concluded that grain refinement occurs by a deformation-induced continuous reaction, that is essentially similar to continuous dynamic recrystallization.

Keywords: aluminum alloy, high temperature, equal channel angular pressing (ECAP), deformation band, continuous dynamic recrystallization

1. Introduction

Metallic materials with ultra-fine grained microstructures have many advantages of mechanical properties, *e.g.* an increasing of strength,^{1,2)} low temperature and/or high strain rate superplasticity³⁻⁶⁾ and thermal stability of such structures,^{6,7)} *etc.* Methods for the production of ultra-fine grained metals and alloys have been paid attention with important scientific and commercial interest.¹⁾ For producing submicron-grained materials by imposing very large plastic strains, several techniques, including multi-axial forging,^{8,9)} accumulative roll-bonding¹⁰⁾ and equal-channel angular pressing (ECAP),¹⁻⁷⁾ *etc.* are available at present time. The procedure of ECAP, originally developed by Segal *et al.*,¹¹⁾ consists of materials severely deformed by repeated shearing in a special die with two channels of equal cross-section intersecting at an angle of 90° or higher.^{11,12)} In conventional metal working processes, like rolling and extrusion, the dimensions of billet are always reduced with deformation and, say, a true strain of about 5 can be obtained in foils. In contrast, ECAP has a big advantage for producing structural products because of pressing to strains greater than 10 without any changes in their dimensions.

It has been reported^{7,12-14)} that fine-grained structures are developed in aluminum alloys during ECAP at low to high temperatures. It was discussed in¹²⁻¹⁴⁾ that fine-grain formation can result from occurrence of continuous dynamic recrystallization. Microstructures evolved at an early stage of pressing consist of many parallel bands of subgrains surrounded by low angle boundaries. With further deformation, these subgrains change to equiaxial grains separated by high angle boundaries. Several mechanisms of ultra-fine grain formation during ECAP have been proposed,^{7,12-14)} however, these are currently a matter of some debate and is not clear.

The aim of the present work is to study the evolution characteristics of subcrystalline structures in an as-cast

0.16%Zr modified 7475 aluminum alloy during ECAP at a temperature of 673 K ($\approx 0.7T_m$, where T_m is the melting point). The 7475 aluminum alloy was used as a tested material, because the authors studied grain refinement processes taking place under axisymmetric hot compression of the same aluminum alloy.¹⁵⁾ The evolution processes of fine grains with high angle boundaries are analyzed and the mechanisms are discussed in detail.

2. Experimental Procedure

The alloy used was a 0.16%Zr modified 7475 aluminum alloy with the following chemical composition (in mass%): 6.04Zn, 2.46Mg, 1.77Cu, 0.23Cr, 0.16Zr, 0.03Si, 0.04Fe, 0.03Mn and the balance is Al. It was fabricated by direct chill casting and homogenization at 768 K for 20 h at the Kaiser Center for Technology. The initial microstructure was composed of dendrite lamellas lying parallel to the ingot axis with an average size in the range from 1 to 10 mm in longitudinal direction and from 100 to 200 μm in transverse direction, as shown in Fig. 1. Some boundaries of lamellar grains were rather smooth and straight, and the others corrugated. Samples for ECAP were machined parallel to the ingot axis into rods with a diameter of 20 mm and a length of around 100 mm. ECAP was carried out using a circular die in cross-section with a diameter of 20 mm, which was preheated at 673 K. The die had a channel in an L-shaped configuration with an angle of 90° between the two channels and an angle 90° at the outer arc of curvature at the point of intersection. These angles lead to a strain of about 1 in each passage through the die. The samples heated at 673 K were pressed repeatedly up to a strain of 12 by using Route A. Route A^{11,12)} denotes the process of repetitive pressing without rotation of the billet in each pass. The pressed samples were quenched in water after each deformation.

Samples for optical, electron back scattering diffraction pattern (EBSP) and transmission electron microscopy (TEM)

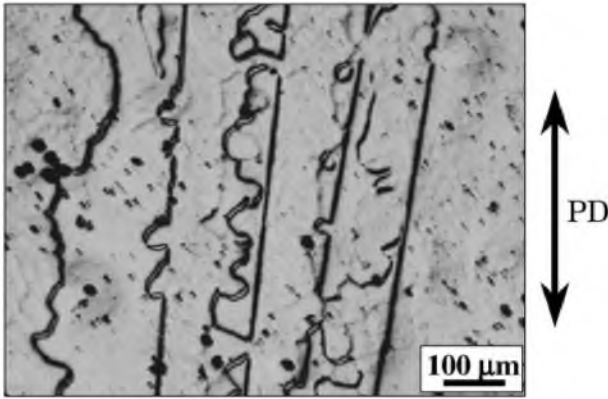


Fig. 1 Initial structure of 7475 Al alloy fabricated by direct chill casting and homogenized for 20 h at 768 K. PD indicates the direction of pressing parallel to the ingot axis.

analysis were cut from central places in longitudinal section of the pressed samples in parallel to pressing direction. The metallographic analysis was carried out using Olympus PME3 in polarized light after etching by standard Dick-Keller etchant. EBSD investigations were performed in Hitachi S-3500H SEM with OIM™ software provided by TexSem Lab., Inc. Specimens for TEM examination were mechanically ground to a thickness of about 200 μm and electropolished in a solution of 30% HNO₃ and 70% CH₃OH at a temperature of -30°C using a Tenupol-3 twin-jet polishing unit. They were then examined using a JEM-2000FX TEM operating at 200 kV. Average crystallite size was measured by a linear intercept method. The misorientation of (sub)grain boundaries were studied using a conventional Kikuchi-line technique.¹⁶⁾ The total number of boundaries analyzed was from 60 to 80 in each sample.

3. Experimental Results

3.1 Deformed Microstructure

Typical microstructures developed after a first pass of ECAP at 673 K are shown in Fig. 2. These are taken in different places, *i.e.* inner side (top), center and outer side (bottom) in longitudinal section of the ECAP sample. It can be clearly seen in Fig. 2 that ECAP results in large inhomogeneity in strain distribution developed in longitudinal section. Similar results have been recently reported by Terhune *et al.* for pure aluminum deformed by ECAP in Route B_c at room temperature.¹⁷⁾ The spacing between original grain boundaries and the angle between initial boundaries and pressing direction decrease from top to bottom in cross-section. Due to such inhomogeneous deformation characteristics of ECAP, the results presented hereafter were those obtained only from a central part in the longitudinal section of ECAP samples.

A series of typical optical microstructures evolved at different strains under ECAP is presented in Fig. 3. A structure at a strain of 2 is characterized by pancaked initial grains with recovered subgrains, which are in line roughly along pressing direction (Fig. 3(a)). Further straining to $\epsilon = 3$, many deformation bands and substructures are developed in elongated grain interiors (Fig. 3(b)). New fine grains are evolved along the original grain boundaries and

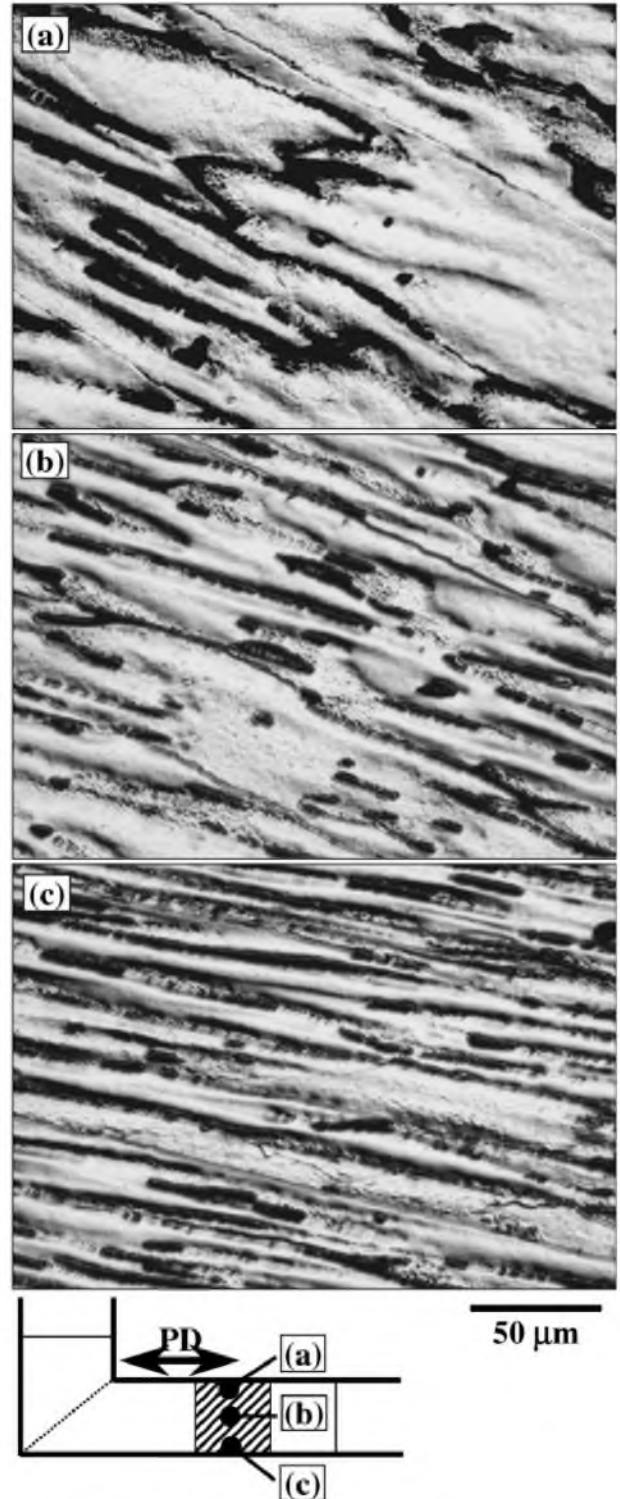


Fig. 2 Typical microstructures in longitudinal section developed in different places of ECAP 7475 Al alloy. The sample was pressed to a strain of 1 at 673 K. PD means the pressing direction. (a) Inner side (top), (b) center and (c) outer side (bottom) in longitudinal section of ECAP sample.

also deformation bands. Further deformation to $\epsilon = 4$ results in increasing of the number of deformation bands and evolution of fine crystalline in the whole area (Fig. 3(c)). The microstructure evolved at $\epsilon = 12$ (Fig. 3(d)) is more complicated and cannot be analyzed in detail under optical microscopy. It can be seen in Fig. 3(d) that the microstructure

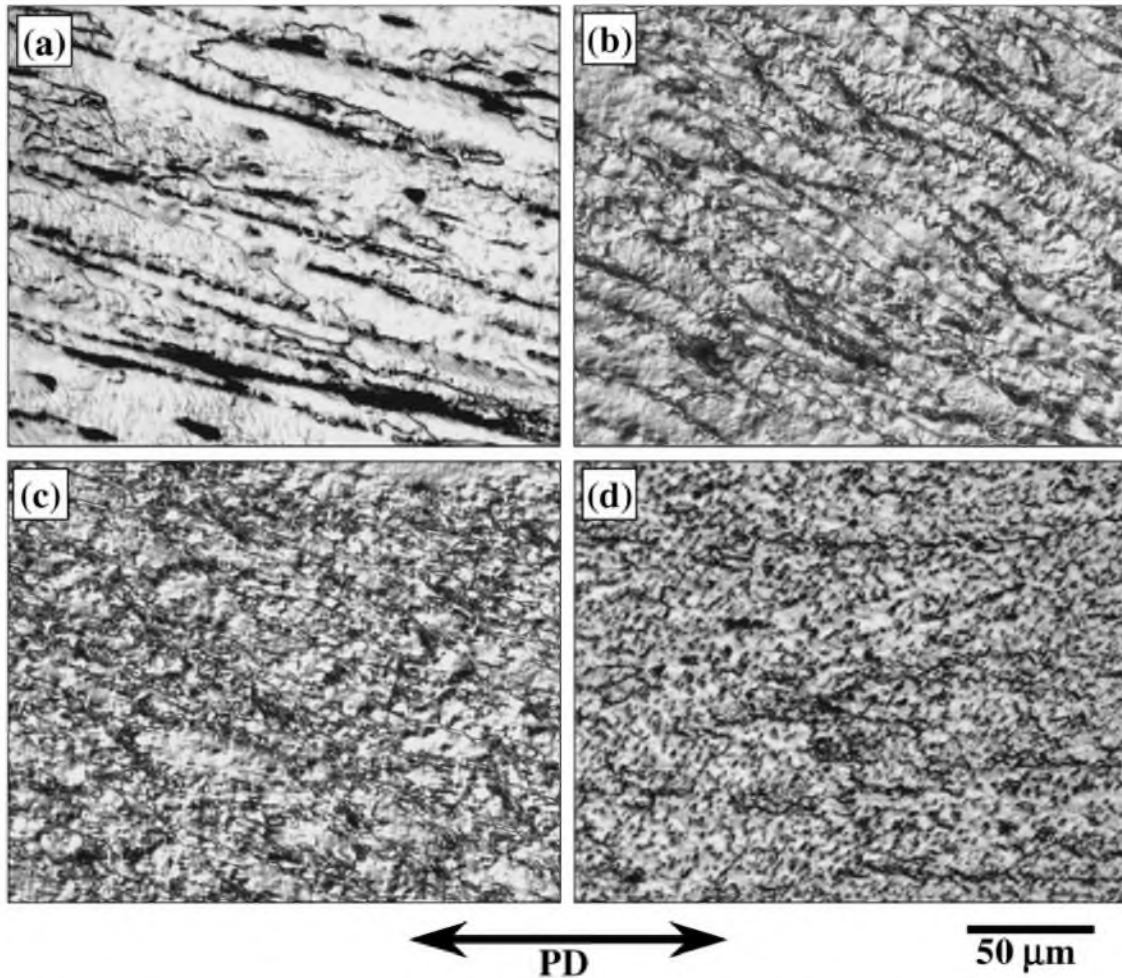


Fig. 3 Microstructural changes of 7475 Al alloy during ECAP at 673 K. PD means the pressing direction. (a) $\varepsilon = 2$, (b) $\varepsilon = 3$, (c) $\varepsilon = 4$ and (d) $\varepsilon = 12$.

in high strain looks like a powdered aggregate with a granule size of submicron scale. This may suggest that a full development of fine-grained structure takes place at such large strains.

3.2 OIM Microstructure

Typical orientation imaging microscopy (OIM) pictures of the 7475 alloy deformed to various strains are represented in Fig. 4. In these OIM maps, orientation differences (θ) between neighboring grid points, $\theta > 3^\circ$, $\theta > 5^\circ$ and $\theta > 15^\circ$ are marked by a thin white, narrow and bold black lines, respectively. It can be seen in Fig. 4(a) that subgrains with low angle boundaries are developed in elongated initial grains accompanied with some medium angle boundaries. The latter are considered as the boundaries of deformation bands observed in Fig. 3. Such deformation bands are reported also to be developed in pure aluminum after first pass of ECAP at room temperature.¹⁷⁾ New fine grains are frequently but inhomogeneously developed along grain boundaries and newly formed deformation bands with medium to high angle boundaries at $\varepsilon = 3$ (Fig. 4(b)). Further straining to above $\varepsilon = 4$, the evolution of new grains proceed in the whole area, but still not completely.

Figure 5 represents the point-to-point misorientation ($\Delta\theta$) developed along the lines T_1 and T_2 in Figs. 4(a) and (b),

respectively. Here the point-to-point misorientation defines a relative difference of crystal orientation between two adjacent scan points. It can be seen in Fig. 5(a) that $\Delta\theta$ exceeds 6° at several local places, while it ranges from 1 to 3° in the other ones. The formers are considered to be similar to the geometrically necessary dislocation boundaries (GNBs) or boundaries of deformation bands evolved in grain interiors.¹⁸⁾ The other low angle boundaries are those of conventional subgrains developed. With increasing of strain to $\varepsilon = 3$ (Fig. 5(b)), $\Delta\theta$ rapidly rises and are ranges from 20° to 50° , and also a distance between deformation bands decreases, although they developed inhomogeneously, as can be seen in Fig. 4(b). It will be summarized from these results that inhomogeneous deformation characteristics of ECAP introduces locally lattice bending and rotation in coarse grain interiors, leading to frequent formation of some GNBs or deformation bands. The misorientation and number of boundaries of deformation bands rapidly rise with repeated ECAP, finally followed by evolution of new grains near or along the boundaries of initial pancaked grains and deformation bands with high misorientations.

3.3 TEM microstructures

Typical TEM microstructures evolved under early deformation are presented in Fig. 6. Main microstructural changes

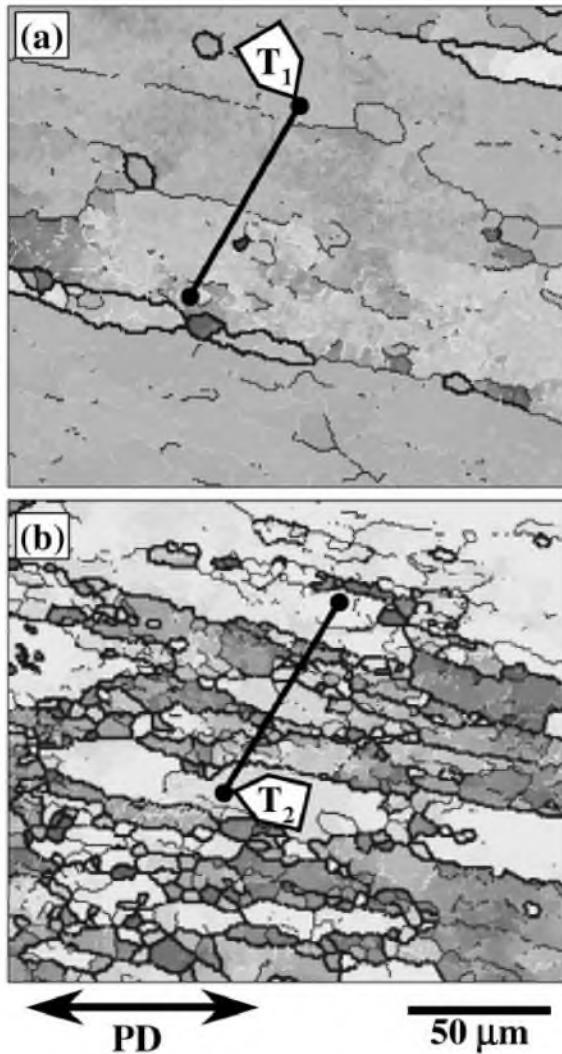


Fig. 4 OIM micrographs of 7475 Al alloy pressed to (a) $\varepsilon = 1$ and (b) $\varepsilon = 3$ at 673 K. Thin white lines correspond to boundaries of misorientation $> 3^\circ$, thin black lines $> 5^\circ$ and bold lines $> 15^\circ$, respectively. PD means the pressing direction.

up to a strain of 2 (Fig. 6(a)) are characterized by homogeneous formation of subgrains with low angle boundaries, which are rather elongated along pressing direction. After 3 passes of ECAP, *i.e.* $\varepsilon = 3$, the misorientation of dislocation subboundaries increases and some boundaries with high angle misorientation above 10° are evolved accompanied with many second phase particles along the boundaries and in grain interiors (Fig. 6(b)).

A typical TEM microstructure evolved at a strain of 8 is represented in Fig. 7. Here (sub)grains developed can be categorized to the following two types. One is the elongated (sub)grains with rather high angle boundaries, which are evolved in regions containing rich secondary particles (Fig. 7(b)). The other one is more equiaxed crystallites with low to medium angle boundaries and developed in regions with relatively free particles (Fig. 7(c)). The former crystallite size is smaller than the latter, *i.e.* they are about $1 \mu\text{m}$ and $1.7 \mu\text{m}$, respectively. The misorientation analysis suggests that high angle boundaries (HABs) are developed faster in rich particle areas and the average misorientation angle was about 33° . In contrast, the network of low to medium angles boundaries is

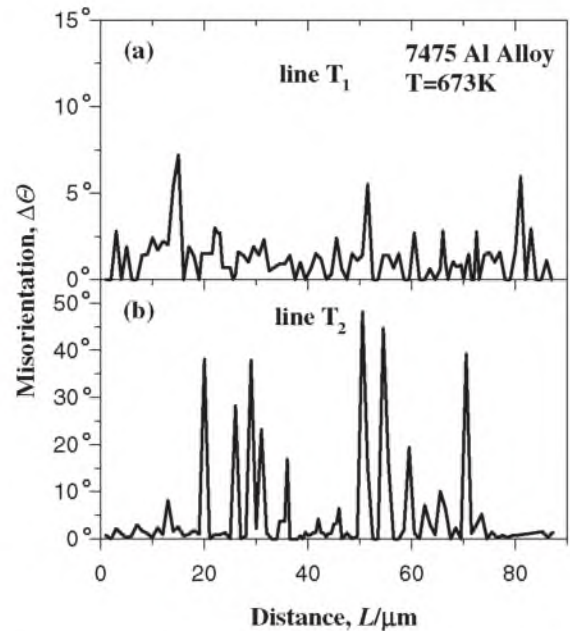


Fig. 5 Point-to-point misorientation ($\Delta\theta$) developed in original grain interiors of 7475 Al alloy pressed to (a) $\varepsilon = 1$ and (b) $\varepsilon = 3$. They were measured along the lines T_1 and T_2 indicated in Figs. 4(a) and (b), respectively.

formed in particles free regions and the average angle was about 14° . Such a mixed grain structure changes completely to uniform equiaxed grains with HABs only after 12 passes of ECAP at 673 K. One of such typical TEM microstructures is presented in Fig. 8. It should be noted in Fig. 8 that equiaxed crystallites with an average size of about $1.7 \mu\text{m}$ as well as second phase particles are developed homogeneously in the whole area.

Figure 9 shows strain dependence of the distribution of misorientation angles for strain-induced dislocation boundaries developed under repeated ECAP. It can be seen in Fig. 9 that the majority of misorientation angles are lower than 10° at strains up to 2 and then the fraction of HABs with more than 15° starts to increase at strains of 3 and 4. The fraction of low angle boundaries rapidly drops and conversely that of HABs rises with further straining to $\varepsilon = 8$ and $\varepsilon = 12$, resulting in development of a bimodal misorientation distribution with two peaks (Figs. 9(e) and (f)). It should be noted in Fig. 9(f) that the misorientation distribution developed at $\varepsilon = 12$ is roughly similar to a random one for annealed cubic metals predicted by Mackenzie,¹⁹⁾ although the fraction of low angle boundaries is relatively large.

Changes in the average misorientation, θ_{av} , of strain-induced boundaries, the crystallite size, d , and the aspect ratio with repeated pressing are summarized in Fig. 10. The average misorientation, θ_{av} , measured by EBSD and TEM techniques is indicated by solid and open symbols, respectively. Both the results are almost the same within experimental scatter. It can be seen in Fig. 10(a) that the average misorientation increases gradually until $\varepsilon = 2$ and then rapidly up to around 15° at $\varepsilon = 3$, followed by gradual rise with further deformation. A rapid increase in θ_{av} after third pass of ECAP can be connected closely with development of new fine grains surrounded by HABs (Figs. 3 and 4). On the

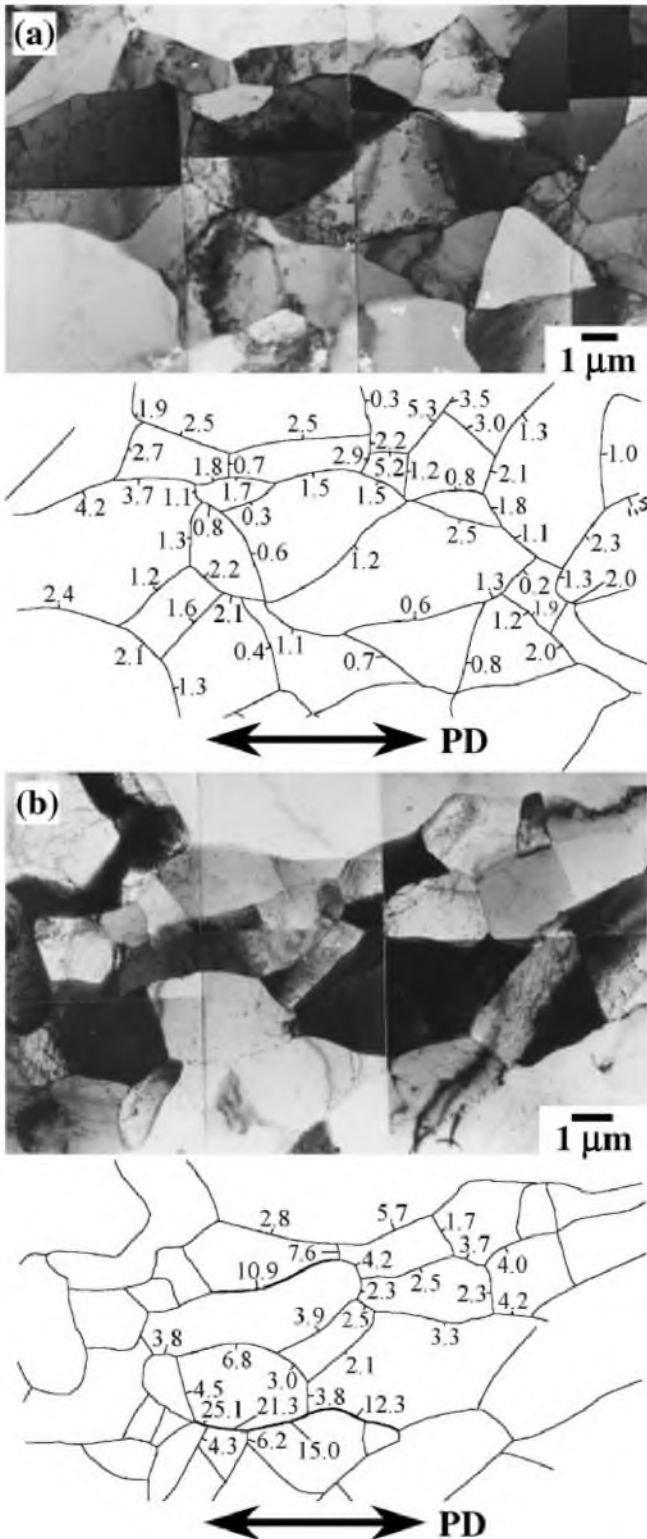


Fig. 6 Typical TEM microstructures of 7475 Al alloy with repeated ECAP at 673 K. The numbers indicate the misorientation angle in degrees. PD means the pressing direction. (a) $\epsilon = 2$ and (b) $\epsilon = 3$.

other hand, (sub)grains elongated toward pressing direction are changed to roughly equiaxed ones accompanied with decrease in their average size by early ECAP. (Sub)grains with an average size of about $1.7\mu\text{m}$ and an aspect ratio of about 1.3 are developed only after $\epsilon = 4$ (Figs. 10(b) and (c)), while the boundary character changes gradually with defor-

mation to further high strains. It is interesting to note in Fig. 10(b) that the “elongated” size of crystallite approaches the “transverse” one, which does not change with deformation.

4. Discussion

The process of grain refinement taking place under equal-channel angular pressing (ECAP) can be summarized from the experimental results described above as follows.

- (i) Inhomogeneous deformation characteristic of ECAP (Fig. 2) can introduce locally lattice bending and deformation bands in coarse grain interiors.
- (ii) The misorientation of the boundaries of deformation bands and (sub)grains rapidly rises after three passages of ECAP.
- (iii) New grains are evolved near or along the boundaries of initial grains and newly developed deformation bands with HABs.

These results (i), (ii) and (iii) suggest that formation of new fine grains during ECAP can be resulted from grain fragmentation process accompanied by frequent evolution of deformation bands. It was reported under severe plastic deformation at low to moderate temperatures ($T < 0.5T_m$)^{8,9,14,17} that original grains are subdivided by deformation bands or geometrically necessary boundaries (GNBs), followed by evolution of fine crystallite components in high strain, and such a process can play an important role in grain refinement. Under intense plastic deformation at low temperatures, several GNBs, such as microband, kink or deformation band, *etc.*,²⁰ are easily developed and the number and the average misorientation of these boundaries increases with straining, finally leading to evolution of new grains at high strains.^{8,9} The main mechanism of grain refinement can be directly associated with grain splitting by formation of internal GNBs followed by their transformation into high angle boundaries (HABs). Let us discuss here whether the same mechanism can operate even under hot ECAP.

It is well known that with increasing of temperature slip takes place more homogeneously, and so results in reduction of the tendency for grain fragmentation. Even if strain gradients are developed during hot deformation, they could rapidly disappear due to frequent operation of dynamic recovery and also grain boundary sliding (GBS), and so such strain induced GNBs could be hardly developed.²¹ By the way, the authors reported recently that inhomogeneous deformation takes place in the present coarse-grained 7475 Al alloy under axisymmetric hot compression and results in development of strain gradients accompanied with local lattice rotation, and finally frequent formation of GNBs at $\epsilon \leq 1.4$.¹⁵ They discussed that such strain gradients can be developed by GBS taking place with different rates along straight and corrugated grain boundaries (Fig. 1). This is the reason why deformation bands following strain gradients are easily developed even after rather low strain during hot deformation.

Deformation under ECAP in itself leads to development of inhomogeneous strain distribution in the whole area of deformed sample, as shown in Fig. 2. Repeated hot ECAP leads to increasing of the number and misorientation of the

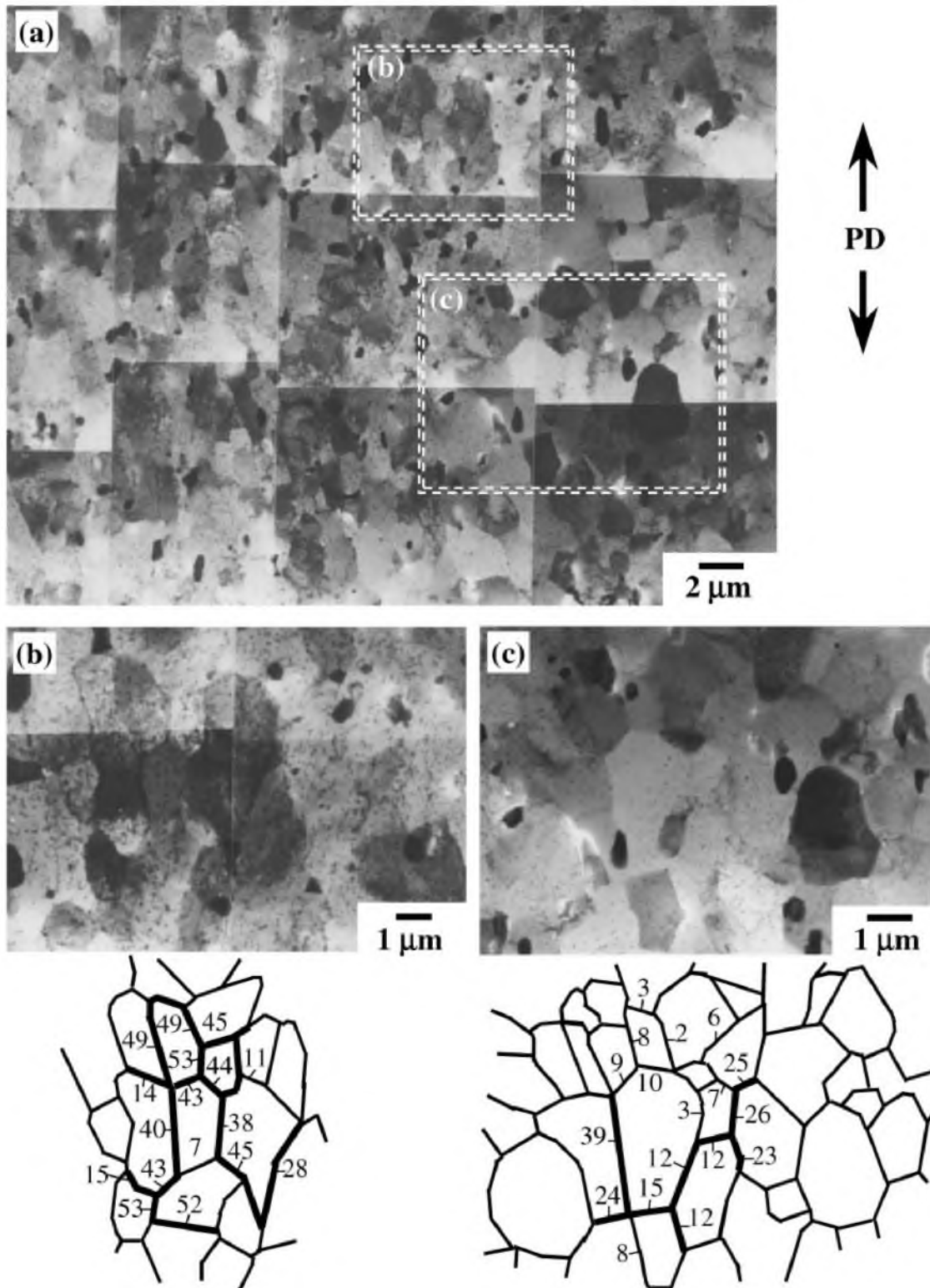


Fig. 7 TEM micrograph of new grains evolved in 7475 Al alloy deformed to a strain of 8 at 673 K. PD means the pressing direction. Note the grains developed in regions containing rich secondary particles in (b) and the (sub)grains evolved in regions with relatively free particles in (c). The numbers in (b) and (c) indicate the misorientation angle in degrees.

boundaries of deformation bands (Figs. 4 and 5). It was reported in Route A of ECAP^{13,22)} that shear introduced at an angle of 45° in longitudinal section takes place in two mutually orthogonal shearing planes. Changes in shearing plane with repeated pressings can result in formation of deformation bands with different orientations.²³⁾ Mutual

crossing between new deformation bands and previous ones as well as original grain boundaries leads to continuous fragmentation of coarse grains into misoriented fine domains. The boundary misorientations of these domains grow rapidly with increasing of strain, finally followed by their transformation into HABs. It is interesting to note in Fig. 10 that a

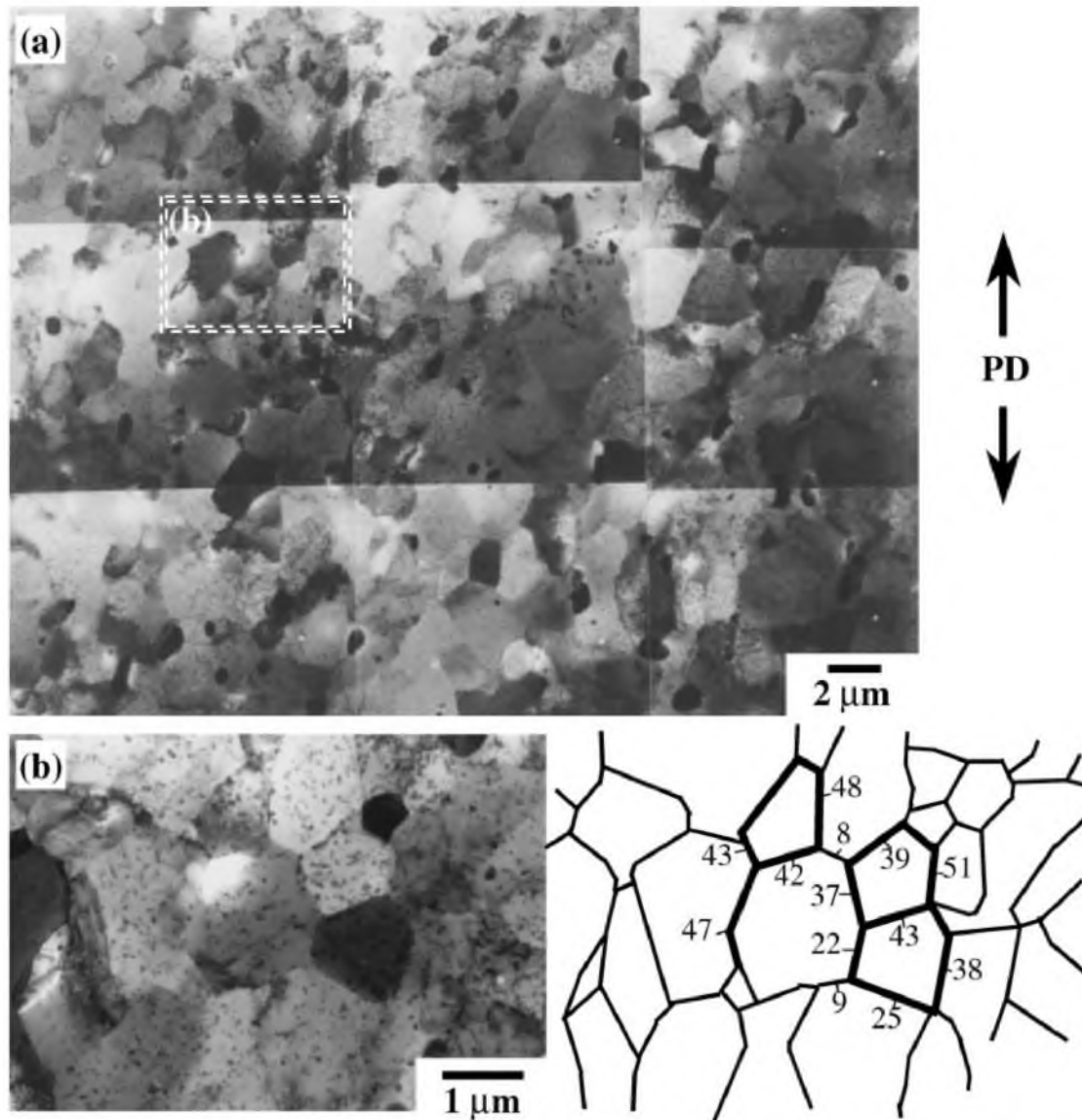


Fig. 8 TEM micrograph of microstructure evolved in 7475 Al alloy deformed to a strain of 12 at 673 K. The numbers indicate the misorientation angle in degrees. PD means the pressing direction.

series of such processes can take place effectively only after 3 passes of ECAP. Concurrently, lattice rotation occurs locally in the places of intersection of deformation bands. GBS also takes place first near initial grain boundaries and subsequently along deformation bands with HABs, leading to progressive (sub)grain rotation. Such series of continuous processes leads finally to development of new fine grains in high strain. It is concluded, therefore, that new fine grained structure developed under hot ECAP can result from a kind of deformation-induced continuous reaction taking place in submicron scale, that is similar to continuous dynamic recrystallization (cDRX).

It should be noted that an important role of secondary phase particles can promote cDRX under high temperature ECAP of the present 7475 Al alloy. They provide a thermal stability of dislocation structures^{14,15)} and prevent any relaxation of strain gradients. It is known that the particles of η -phase ($Mg(Zn_2,AlCu)$), S -phase (Al_2CuMg), T -phase ($Al_{32}(Mg,Zn)_{49}$) and the $Al_3(Zr/Cr)$ dispersoids are presented in 7475 Al alloy at 673 K.²⁴⁾ It is clearly seen in Figs. 6 and 7

that many precipitates are located along strain-induced boundaries with high angle misorientation, which are formed faster in the regions containing rich secondary phases particles (Fig. 7). This suggests that these particles may restrict effectively an ability of lattice dislocation to long-range rearrangement and thus retard or prevent any relaxation of evolved microstructures. Therefore, they can accelerate increasing of the misorientation of strain-induced boundaries and their conversion into HABs. It is concluded, therefore, that a stabilization effect of dislocation substructure due to second phase particles should play an important role in operation of cDRX during hot ECAP of the present 7475 Al alloy.

5. Conclusion

Microstructural evolution an as-cast 0.16%Zr modified 7475 aluminum alloy was studied in equal-channel angular pressing (ECAP) at temperature of 673 K ($0.7T_m$). The main results can be summarized as follows:

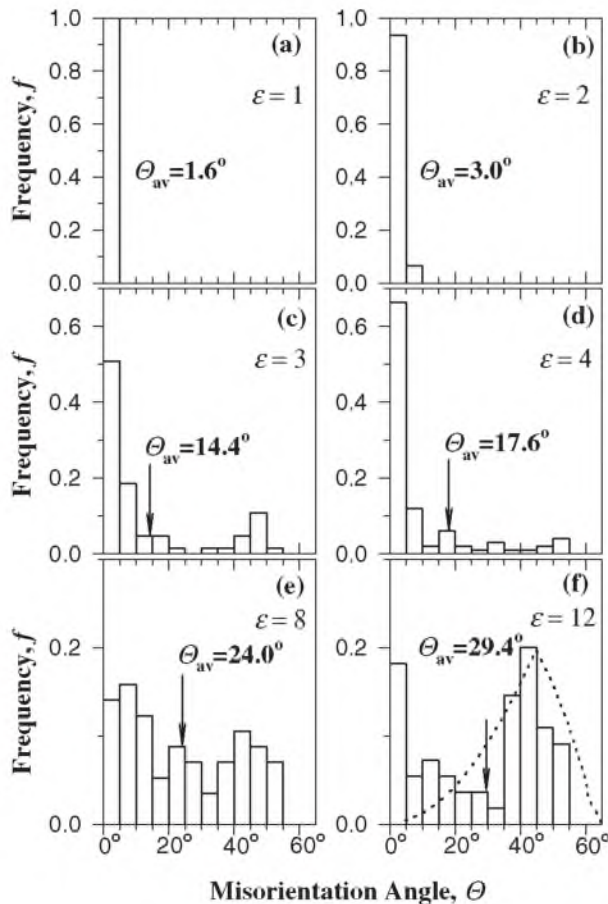


Fig. 9 Change in the misorientation distribution of deformation-induced boundaries with repeated ECAP for 7475 Al alloy. The samples were pressed to various strains at 673 K. The broken line indicates the random misorientation distribution by Mackenzie.¹⁹⁾ (a) $\varepsilon = 1$, (b) $\varepsilon = 2$, (c) $\varepsilon = 3$, (d) $\varepsilon = 4$, (e) $\varepsilon = 8$ and (f) $\varepsilon = 12$.

- (1) ECAP results in considerable grain refinement. A new fine-grained microstructure with an average crystallite size of about 1.7 μm develops at large strains above 6.
- (2) Inhomogeneous deformation characteristics of ECAP lead to formation of deformation bands after first pass through the ECAP die. Repeated ECAP results in mutual crossing, increasing of number and misorientation of deformation bands.
- (3) Repeated ECAP results in gradual shift of misorientation distribution of newly developed boundaries toward higher angles, leading to increasing and approaching of an average misorientation angle of 30° in high strain.
- (4) The processes in (2) and (3) lead to transformation of boundaries of deformation bands into high angle boundaries and evolution of new fine grains in high strain. It is concluded that grain refinement takes place by a deformation-induced continuous reaction, that is essentially similar to continuous dynamic recrystallization.

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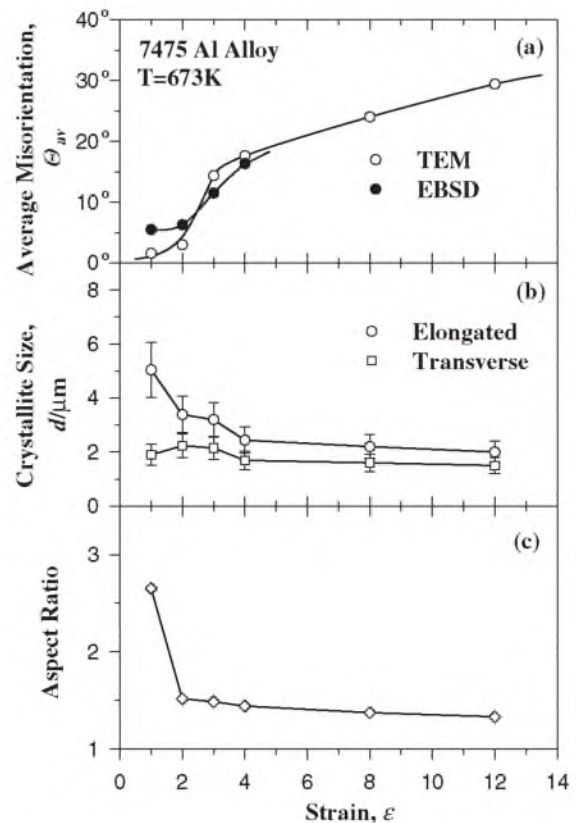


Fig. 10 Changes in (a) average misorientation angle, Θ_{av} , of strain-induced (sub)grain boundaries, (b) crystallite size, d , and (c) aspect ratio with repeated ECAP at 673 K for 7475 aluminum alloy.

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