Effect of Processing Temperature on Microstructure Development during ECAP of Al-Mg-Sc Alloy

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Abstract. Microstructural evolution taking place during equal channel angular pressing (ECAP) was studied in a commercial coarse-grained Al-6%Mg-0.4%Mn-0.3%Sc alloy in a temperature interval 200-450°C (~0.5-0.8 T_m). Samples were pressed using route A to a total strain of 12 and quenched in water after each ECAP pass. Uniform fine-grained microstructures with the average grain sizes of 0.7 and 2.5 μm, are almost fully evolved at high ECAP strains at 250°C and 450°C, respectively, while ECAP at 300°C (~0.6 T_m) leads to the formation of bimodal grain structure with fine grains of around 1 μm and relatively coarse grains of around 8 μm. The latter are developed due to the occurrence of static recrystallization during "keeping" time in the ECAP channel and/or reheating between ECAP passes. The microstructural development under warm-to-hot ECAP conditions is discussed in terms of the large potential for grain boundary migration resulted from an overlapping of accelerated grain boundary mobility at high pressing temperatures and enhanced driving force for recrystallization, which is caused by a strong inhibition of dynamic recovery in a heavily-alloyed Al alloy.

Introduction

There have been several works to date showing that ECAP is an effective method for grain refinement at low-to-high temperatures [1]. Microstructural changes in Al and its alloys have been studied at various temperatures in previous papers [e.g. 1-12]. The formation of new grains during ECAP has been proposed as a result of the transformation of deformation–induced low angle boundaries (LABs) that are formed at earlier stages of deformation into high-angle boundaries (HABs), accompanied by dynamic recovery. This mechanism is similar to *in-situ* or *continuous dynamic recrystallization* (cDRX) [13,14]. It is suggested in [1-3,6-8,11,14] that the transition of LABs into HABs can be controlled by the recovery rate, which is accelerated with increasing the temperature. On the other hand, it was found that final grain size and the fraction of LABs in dilute Al alloys are generally increased with increasing ECAP temperature [e.g. 3,6,11]. Unfortunately, the major factors controlling such microstructural development, as well as the precise mechanisms operating during warm-to-hot ECAP, are still unclear due to the lack of related experimental data. This picture becomes yet much more complicated by looking at some heavily-alloyed Al alloys, in which dynamic recovery is additionally inhibited by the presence of large number of dispersed particles and/or substitutional atoms in the solid solution [e.g. 1-3,4,6,8,9]. It has been shown that the new grain formation can be delayed, and also relatively finer grains with more diffused dislocation (sub)boundaries can be evolved in such alloys at low-to-moderate temperatures [e.g. 3,9]. Besides, a *sudden* formation of a microstructure with *unexpectedly* large grain size was sometimes reported for Mg-alloyed Al alloys ECAPed in the large-temperature range [4,6,8]. All those mentioned above allow us to emphasize that the deformation temperature may be one of the most complicated and least understandable processing parameters of ECAP.

The main aim of the present work is to study the effect of deformation temperature on microstructural changes and new grain development taking place in an Al-6%Mg-0.3%Sc alloy during warm-to-hot ECAP in a wide temperature range. This alloy contains a respectable amount of Mg and Sc, both of which can significantly inhibit dynamic recovery even at elevated temperatures [1-3,9]. It has been previously shown by some of the present authors [5,10,12] that the microstructural behavior of this alloy during ECAP is significantly different at various temperatures; this is summarized in the current work. Also, a special attention has been paid to analyze the relationships between strain-induced grain structures and pressing temperatures in a wide temperature range of ECAP. The main factors promoting strain-induced new grain formation at various temperatures are analyzed and discussed in details.

Experimental Procedure

The alloy used had the following chemical composition; Al-6%Mg-0.3%Sc-0.4%Mn (in mass %). It was fabricated by casting into a steel mold and then homogenized at 520°C for 48 hrs. Extrusion was performed at 390°C to a strain of 0.7, followed by annealing at 400°C for 1h. After extrusion and annealing, the alloy was composed of a non-uniform partially recrystallized microstructure with a bimodal distribution of the grain size, namely, coarse elongated grains lying parallel to the extrusion axis, and fine equiaxed grains in their mantle regions. The size of coarse grains was around 170 and 70 μm in longitudinal and transverse directions, respectively. The fine grains had the average size of 4.4 μm and the volume fraction of 0.35. Coherent Al_3Sc dispersoids with a size of about 10-20 nm and incoherent Al_6Mn precipitates of about 200 nm were identified in this structure [10]. Samples for ECAP were machined parallel to the extrusion axis into rods with Ø20 mm and a length of 100 mm. ECAP was carried out repeatedly at 150-450°C to ε=12 with a strain of about 1 in each pass using route A, i.e. orientation of rods was not changed at each pass. More details for this ECAP procedure are described elsewhere [10,12]. Deformed microstructures of the samples were examined in the central regions of a section parallel to the pressing direction using optical microscopy, TEM, SEM and X-ray analysis.

Results

Feasibility of ECAP at elevated temperatures. It was impossible to apply more than 5-6 ECAP passes to the present material at 150°C without including several macrocracks. After 5, exceptionally 6 passes, severe macrocracks appeared parallel to the shear plane and spaced about 3-5 mm from each other. The possible reason for this failure is the very inhomogeneous deformation with intense shear bands introduced by ECAP [8]. At 200-450°C, in contrast, ECAP was successfully performed to 12 passes.

Microstructures developed at high strains. The typical optical microstructures developed at ε=12 and at temperatures (a) 250, (b) 300 and (c) 450°C at are represented in Fig. 1. The associated X-ray diffraction patterns, which were analyzed at the specimen surface areas with a diameter of 1mm are shown in Figs. 2(a), (b) and (c), respectively. It can be seen that uniform fine-grained microstructures are created by ECAP at 250 and 450°C at high strains; those appear in dark- and dark-to-gray regions in Figs. 1(a) and (c). The grain sizes and the volume fractions of new grains were as high as ~0.7 μm and 0.76 at 250°C and ~2.5 μm and 0.85 at 450°C. Thus, it can be evident that grain refinement takes place during ECAP under these temperature conditions. In contrast, a bimodal grain structure with significantly different grain sizes is evolved after 12 ECAP passes at 300°C (Fig. 1(b)). One component of this structure comprises fine-grained bands aligned along the pressing direction. The size of the crystallites evolved in these bands is about 1 μm; their volume fraction is about 0.3. The other grain component is composed of relatively coarse grains of about 8 μm and the volume fraction of about 0.6. The size of these grains is unexpectedly large and most of them have an essentially equiaxed shape. The later suggests that they have not suffered any remarkable strain after their formation. It is seen in Fig. 2 that the X-ray diffraction patterns that correspond to the as-ECAPed states at 250, 300 and 450°C show the continuous diffraction rings; this can be an additional independent evidence that some misoriented (sub)structures are almost fully evolved in the present alloy during ECAP in a wide temperature range.

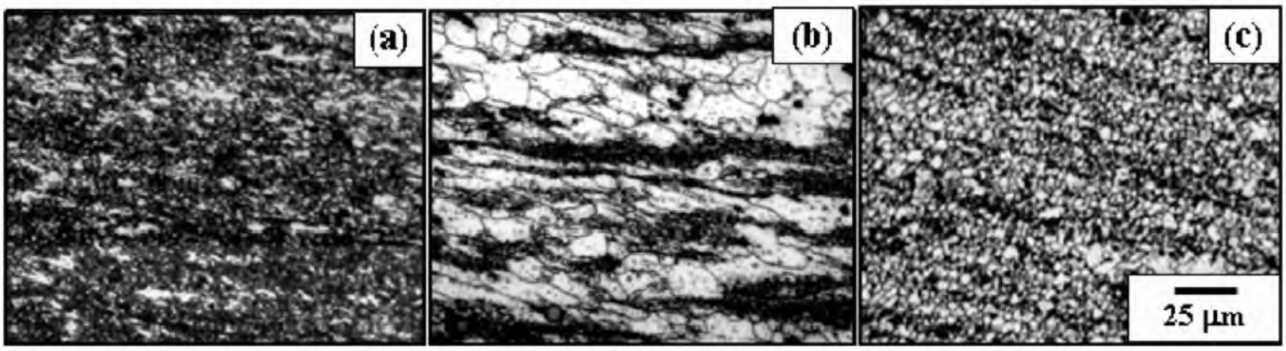

Fig.1 Typical microstructures formed in Al-6%Mg-0.3%Sc alloy after 12 ECAP passes:
(a) 250°C, (b) 300°C, (c) 450°C. The pressing direction is horizontal.

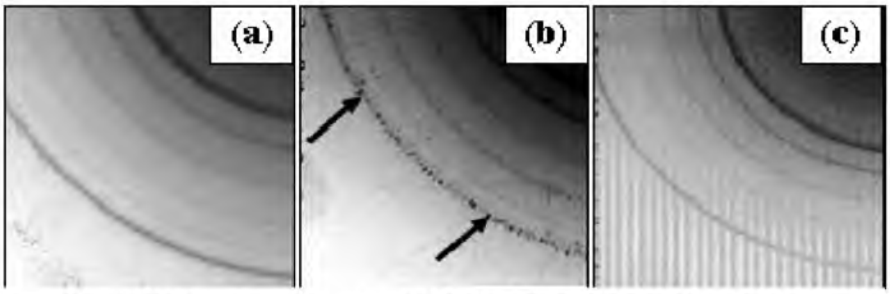

Fig. 2 Diffraction patterns of Al-6%Mg-0.3%Sc alloy after 12 ECAP passes: (a) 250°C, (b) 300°C, (c) 450°C.

However, it can be seen in Fig. 2 that the diffraction patterns at 250 and 450°C exhibit rather diffuse lines on the diffraction rings; that may be related to the large strain gradients and/or high lattice distortions introduced by ECAP, while the picture taken at 300°C displays very sharp diffraction spots around the rings, as arrowed in Fig 2(b). Those indicate that some coarse undeformed grains may be present in the examined material at this temperature.

Temperature dependences of the grain size and the volume fraction of new grains developed after 12 ECAP passes are represented in Figs. 3(a) and (b), respectively. It can be seen in Fig. 3(a) that the mean size of the new fine grains is *gradually* increased with increasing temperature from about 0.4μm at 200°C to about 2.5μm at 450°C. This may be related to the acceleration of dynamic recovery under raising deformation temperature conditions, as it will be discussed latter. It is remarkable in Fig. 3(a) that the size of the new fine grains (~1 μm), that are developed in the present alloy during ECAP at 300°C, is in a good agreement with a dependence of the grain size on *T*. In contrast, the size of the coarse grains (~8 μm) is much larger and, thus, is far away from the general dependence. It can be assumed, therefore, that at all temperatures investigated, the same mechanisms of new grain formation can be responsible for the development of the new fine-grained structures, while, there is a “processing window” at around 300°C, in which the structural behavior during ECAP is complicated by the *sudden* development of unexpectedly large grains. The volume fractions of new grains, developed at large strains, are also *gradually* (except 300°C) increased with increasing processing temperature from about 0.63 at 200°C to about 0.85 at 450°C (Fig. 3(b)). This suggests that the kinetics of grain refinement in the present dispersoids-containing Al alloy is accelerated at larger temperatures. Note that such positive temperature dependence of the grain refinement kinetics during high-temperature ECAP may contrast with that reported earlier for some conventional dilute or commercial Al alloys, in which increasing ECAP temperature resulted in a profound decrease of the volume fraction of new grains and/or fraction of HABs [e.g.3,11]. This question will be also discussed in the *next Section.*

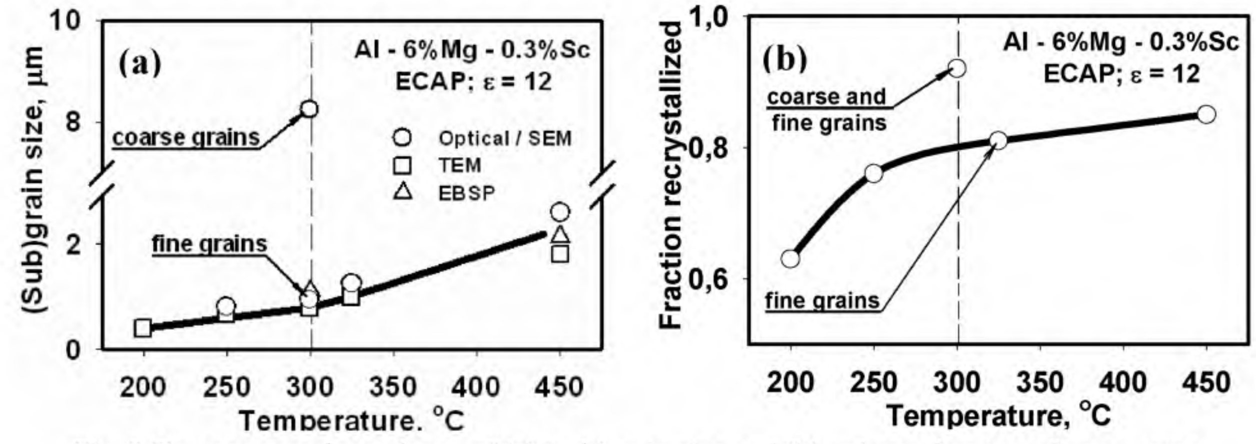

Fig. 3 Temperature dependence of (a) (sub)grain size and (b) volume fraction of new grains developed in Al-Mg-Sc alloy during ECAP at ε=12.

Discussion

We can conclude in the present observations that Al-6%Mg-0.3%Sc alloy exhibits rather complicated structural behavior during ECAP at elevated temperatures. *First*, the material shows low ductility at a relatively high ECAP temperature of 150°C (~0.46T_m) due to a strong localization of deformation and cracking along the shearing direction. *Second*, grain refinement takes place during ECAP at 200-450°C (~0.5-0.78T_m); in this temperature interval, the size and the volume fraction of new grains are continuously increased with increasing temperature. *Third*, there is a "processing window" around 300°C (~0.6T_m), in which the structural development during ECAP is characterized by the appearance of a coarse-grained structure with much larger grain size than that developed at a higher temperature. Note that these coarse grains may be related to a "*sudden discontinuous grain growth*", which was reported earlier for some Al-Mg alloys ECAPed at elevated temperatures [4,6,8]. In the *current Section*, these features of the microstructural development during ECAP will be discussed in details.

The evolution process of new fine grains, occurring in the present alloy during ECAP at T = 250 to 450°C, is represented and discussed in the previous works [5,10,12]. It has been shown that high strain and misorientation gradients can be introduced into the present alloy in the earlier stages of ECAP and, concurrently, new low-to-moderate and moderate-to-high angle boundaries that are related to the boundaries of deformation- and/or microshear bands evolve often in the areas with high local lattice rotations. It has been pointed out [10,12] that evolution of deformation bands can play a key role in occurrence of grain refinement during ECAP of the present alloy. Namely, several sets of these bands that are developed in various directions fragment original grains into small separate misoriented domains. A gradual increase in the number and misorientation of the boundaries of deformation bands and their conversion into HABs can result in the development of the new fine-grained structure through cDRX [13].

It can be noted in this connection that the similar schemes of the new grain formation related to cDRX were also employed in the recent works [e.g.7,9,11] to describe the fine-grained structure development in some Al alloys at low-to-moderate ECAP temperatures. However, the heterogeneous deformation introduced by ECAP should normally decrease with increasing temperature [e.g.3,7,8]. Dislocation motion occurs more homogeneously at elevated temperatures and, even if any strain gradients are formed on a mesoscale due to a specific deformation mode introduced by ECAP they should rapidly disappear by frequent operation of some relaxation processes, including dynamic recovery and grain boundary sliding [14]. As a result, deformation bands following high strain gradients could scarcely develop under hot ECAP conditions [e.g.15]. Hence, it was reported that the kinetics of grain refinement in conventional Al alloys is significantly retarded with increasing temperature [e.g. 3,11]. In contrast, since the present alloy has a relatively high content of Mg and also Sc and Mn additions; deformation bands can be frequently developed even at elevated temperatures. A strong interaction of the lattice dislocations with Mg atoms in Al solid solution and/or with Al_6Mn- and, mostly, Al_3Sc dispersion particles [2,5,9,10,12] can restrict dislocation rearrangement within deformation bands and, thus, provides a thermal stability of the dislocation structures introduced by ECAP.

On the other hand, it is well known that cDRX is controlled by the rate of dynamic recovery. In this way, increasing temperature may be very important for acceleration of dynamic recovery to provide grain refinement. It has been concluded [9] that strong inhibition of recovery in high Mg- and/or dispersoid-containing alloys at low temperatures makes deformation bands more diffuse and so, represses their transformation into HABs. Note also that too strong inhibition of recovery in the present alloy at 150°C results in the strong localization of the plastic deformation within the deformation bands followed by the premature failure of the material [8]. It is apparent that an elevated deformation temperature, in contrast, may promote a higher rate of dislocation rearrangement within the boundaries of deformation bands. This allows a greater possibility for them to become permanent boundaries, and, thus, accelerates the kinetics of grain refinement (Fig. 3(b)). It can be also generally considered that accelerating dynamic recovery at larger temperature may assist the formation of coarser and more equiaxed grains, as it is evident in Fig. 3(a).

Coarse-grain formation at 300°C. Another structural mechanism that results in a "*sudden*" grain growth in the present alloy can be related to static recrystallization. It has been shown in the recent work [12] that the latter may start to occur under warm-to-hot ECAP conditions as an alternative restoration process, when dynamic recovery is strongly inhibited by the structural factors. Static recrystallization may accompany grain refining during ECAP at elevated temperatures, since an important feature of this technique is that the ECAPed samples *always* undergo a spontaneous static annealing both inside the heated die and also between ECAP passes, since the deformation temperature should be maintained during the whole ECAP procedure. An enhanced driving force for recrystallization is produced even under warm-to-hot deformation conditions due to the factors that were discussed in the previous *Section*, i.e. (i) strong localization of plastic deformation introduced by ECAP and (ii) inhibition of recovery by the presence of coherent dispersion particles and Mg atoms in the solid solution; both may provide locally a high dislocation density, which gives rise to a large stored deformation energy [12,14]. In this *Section*, we would like to discuss, why the development of the coarse grains in the present Al-Mg-Sc alloy takes place only in the narrow "processing window" at around 300°C, while no recrystallization was noticed under lower and larger ECAP temperature conditions (Fig.1).

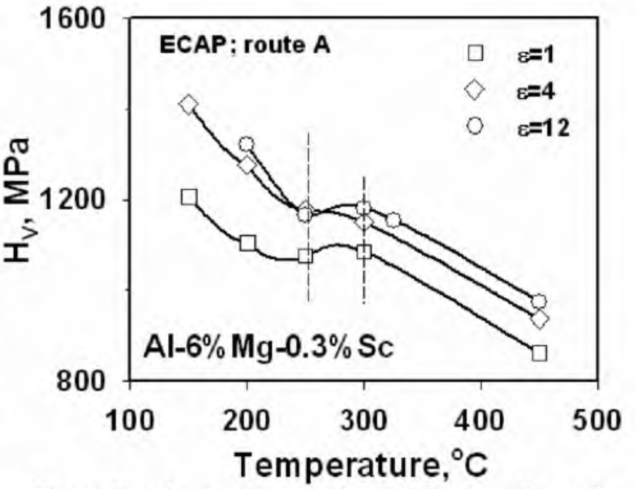

Fig. 4 Hardness changes in Al-Mg-Sc alloy after ECAP. At 300°C, only the hardness of deformed/fine-grained component was evaluated.

The dependence of the room temperature Vickers microhardness of the present alloy *vs* ECAP temperature at ε = 1, 4 and 12 is represented in Fig. 4. Note that the hardness after ECAP at 300°C was examined only in the deformation/fine-grained microstructure to provide the compatible data with those obtained at other deformation temperatures. It is seen in Fig. 4 that at all ECAP strains investigated, the hardness of the present material monotonically drops in the temperature intervals from 150 to 250°C and from 300 to 450°C; this may be normally related to acceleration of the rate of restoration processes in deformation structures, controlled by dynamic recovery [14]. On the other hand, at the temperature ~300°C, at which the static recrystallization takes place in the present material, hardness is changed scarcely. The latter indicates the inability of the material to recover completely within dislocation substructures developed at this ECAP temperature. It is interesting to note that the temperature interval from 250 to 300°C may coincide with the vicinity of the solvus point of the Al-6%Mg alloy, which is about 280°C (Fig. 5). It is also remarkable to see in Fig. 5 that amount of Mg in the solid solution is rapidly increased from about 3 to about 6% with temperature increasing from 200 to 300°C, i.e. up to ~3 mass% of Mg may additionally dissolve in the α-phase at these temperatures. We can assume that such increment in the Mg content in the solid solution by increasing ECAP temperature can additionally inhibit the dynamic recovery and, thus, increase the value of driving force for static recrystallization. It can be considered that in the alloy, which contains a solid solution with a fixed Mg content, the driving force for recrystallization will be reduced with increasing the temperature and, hence, after deformation at a higher temperature, recrystallization will occur less readily [14]. However, the absolute value of this force will be always larger in the alloy with a higher solute content. Such dependencies of the driving force, F_d, on T and Mg concentration are represented schematically in Fig 6 by a series of the dashed lines. The solid line in this figure shows, in turn, the change in the driving force in the alloy, in which the Mg content in Al matrix would be increased by increasing the temperature. It can be seen that in this case, there is an additional increment in the F_d value by the transition of the deformation temperature through the solvus point (Fig.5). Note that this schematic representation for F_d may be consistent with the experimental data for hardness in Fig. 4.

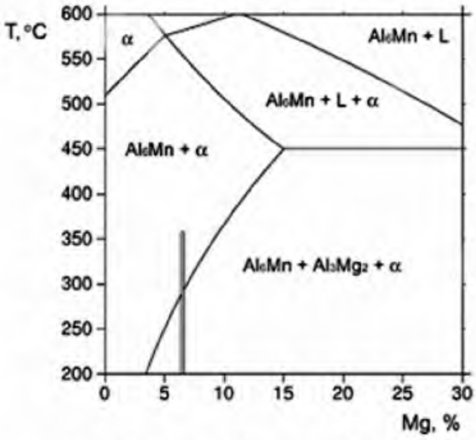

Fig. 5 Phase diagram of Al-Mg-0.4%Mn (THERMOCALC software)

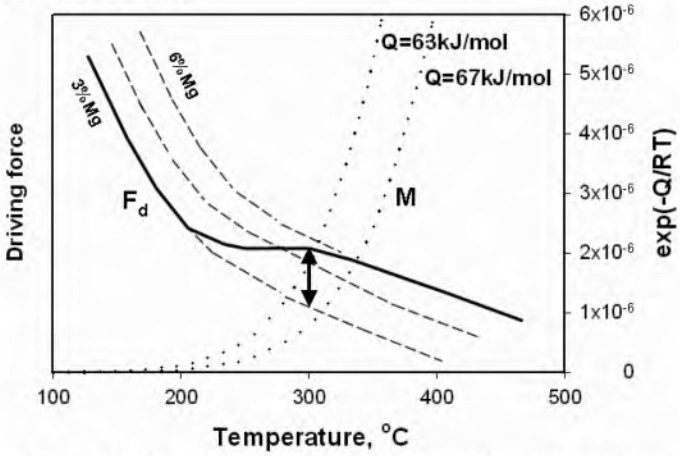

Fig. 6 Schematic representation for the restoration process during ECAP. See explanation in the text.

On the other hand, recrystallization can start to occur under large driving force conditions, when increasing deformation temperature provides a high value of grain-boundary mobility, M. It is known that M obeys an Arrhenius type relationship, i.e. ~ **exp(-Q/RT)**, where **Q** is the activation energy for the grain boundary migration and **R** (=8.31 J/mol K) is the universal gas constant [14]. The dependencies of the **exp(-Q/RT)** vs **T** are represented in Fig. 6 by dotted lines. The activation energy for grain boundary migration in Al, which is ~63-67 kJ/mol [14], was taken into account to evaluate temperature dependencies of M. It can be seen, that M values are relatively low at the temperatures less than 200°C, but start to rapidly grow at T>250°C. Thus, there may be an overlapping of the accelerated grain boundary mobility and enhanced driving force for recrystallization in the vicinity of T~300°C. Hence, a large potential for the grain growth may "suddenly" appear during ECAP at this temperature. In contrast, at T<<300°C or T>>300°C, either the M or F_d values are not large enough to trigger recrystallization and, thus, the microstructural development in the present alloy may occur in accordance with the cDRX mechanism, described above.

Summary

The effect of deformation temperature on microstructural development in an Al-6%Mg-0.3%Sc alloy during ECAP to ε=12 was investigated in the temperature interval from 150 to 450°C. The main results are summarized as follows:

1. ECAP at 150°C is only possible to 5-6 passes due to a strong localization of shear deformation and severe cracking along the shear direction of ECAP die at this temperature. At T $\geq$ 200°C, the ductility of the alloy becomes enough to successfully apply 12 ECAP passes.
2. Grain refinement takes place during ECAP at 200-450°C; the size and the volume fraction of new grains developed at high ECAP strains are continuously increased with increasing temperature in this temperature interval from about 0.4 μm and 0.63 to about 2.5μm and 0.85, respectively. Acceleration of the kinetics of grain refinement and increase in the grain size at larger ECAP temperatures may be related to accelerated rate of dynamic recovery that promote the higher rate of dislocation rearrangement in the heavily-alloyed Al alloy.
3. There is a "processing window" at around 300°C, in which the structural development during ECAP is characterized by the appearance of the coarse-grained structure with the grain size that is much larger than that developed at 450°C. Such microstructural development is resulted from the large potential for the grain boundary migration due to an overlapping of the accelerated grain boundary mobility at high temperature and enhanced driving force for recrystallization, which is caused by a strong inhibition of dynamic recovery in the heavily alloyed Al alloy.

References

[1] R.Z. Valiev, T.G. Langdon: Progr. Mat. Sci. Vol. 51 (2006), p. 881

[2] Y. Iwahashi, Z. Horita, M. Nemoto, T.G. Langdon: Met. Mat. Trans. Vol. 29A (1998), p. 2503

[3] A. Yamashita, D. Yamaguchi, Z. Horita and T.G. Langdon: Mat. Sci. Eng. A Vol. 287 (2000), p.100

[4] C. Pithan, T. Hashimoto, M. Kawazoe, J. Nagahora, K. Higashi: Mat. Sci. Eng. A Vol. 280 (2000), p. 62

[5] I. Mazurina, O. Sitdikov, R. Kaybyshev: Phys. Met. Metall. Vol. 94 (2002), p. 413

[6] Y.C. Chen, Y.Y. Huang, C.P. Chang, P.W. Kao: Acta Mat. Vol. 51 (2003), p. 2005

[7] A. Goloborodko, O. Sitdikov, R. Kaibyshev, H. Miura, T. Sakai: Mat. Sci. Eng. A Vol. 381 (2004), p. 121

[8] M. Popovic, B. Verlinden: Mat. Sci. Tech. Vol. 21 (2005), p. 606

[9] P.J. Apps, M. Berta, P.B. Prangnell: Acta Mat. Vol. 53 (2005), p. 449

[10] O. Sitdikov, T. Sakai, E. Avtokratova, R. Kaibyshev, et al.: Mat. Sci. Eng. A Vol. 444 (2007), p. 18

[11] I. Mazurina, et al., Mater. Sci. Eng. A (2007), doi: 10.1016/j.msea.2007.09.070

[12] O. Sitdikov, T. Sakai, E. Avtokratova, R. Kaibyshev et al.: Acta Mat. Vol. 56 (2008), p. 821

[13] A. Belyakov, T.Sakai, H.Miura, K. Tsuzaki: Phil. Mag. Vol. A81 (2001), p. 2629

[14] F.J. Humphreys, M. Hatherly: *Recrystallization and Related Annealing Phenomena.* (Elsevier 2004).

[15] A. Duckham, R.D. Knutsen, O. Engler: Acta. Mat. Vol. 49 (2001), p. 2739