Magnesium Technology 2013
Edited by: Norbert Hort, Suveen N. Mathaudhu, Neale R. Neelameggham, and Martyn Alderman
TMS (The Minerals, Metals & Materials Society), 2013

Evolution of microstructure during caliber rolling of AZ31 alloy

Alok Singh[1], Hidetoshi Somekawa[1], Tadanobu Inoue[1], Toshiji Mukai[2]

[1]Structural Materials Unit, National Institute for Materials Science, Tsukuba 305-0047, Japan
[2]Department of Mechanical Engineering, 1-1 Rokkodai, Nada, Kobe city, 657-8501, Japan

Keywords: magnesium, deformation microstructure, transmission electron microscopy, caliber rolling

Abstract

An AZ31 alloy has been processed by caliber rolling, in which a repetitive oblique shear strain is applied with a fixed reduction ratio, resulting in increase in strength and ductility by simultaneous grain refinement and texture modification. A total of 18 passes were made at 473K through successive grooves with reduction. A dramatic increase in the tensile YS was achieved with no substantial change in the elongation. Evolution of microstructure by caliber rolling has been studied by TEM. Samples after 4, 6, 8 and 18 passes were examined. After 4 passes the sample showed clear grain boundary structure and grain refinement. After 6 passes, a severely deformed structure is observed, indicating accumulation of strain. Spots with concentration of strain were observed in basal planes. Activation of prismatic slip was observed. After 8 passes the strain appears more severe. After 18 passes, along with concentration of strain, sharp grain and subgrain boundaries appeared, with subgrains of about 100-200nm size. The small grain and subgrain size, along with accumulated strain, is responsible for the high strength.

Introduction

One of the most important microstructural factor in strengthening magnesium alloys is grain refinement [1], which also leads to increased ductility due to enhanced grain boundary plasticity [2]. A crucial factor in ductility is texture, which can be modified by changing the direction of applied shear during severe plastic working [3] and/or by changing recrystallization behavior by dispersion of hard intermetallic particles [4]. Modification of texture, together with grain refinement, has been shown by repetitive shear loading processes such as equal-channel angular extrusion (or pressing) (ECAE or ECAP) [5, 6, 7, 8, 9]. It has also been shown that the yield asymmetry ratio (ratio of compressive yield strength to tensile yield strength) of wrought AZ31 can be reduced by grain refinement to 0.9 [10].

Recently we have reported grain refinement with weakened texture by applying a repetitive oblique shear strain using caliber rolling, which can operate at commercial scale and processing speeds [11, 12]. Caliber rolling can be used to reduce the cross-sectional shape of the products. Graded channels with reducing cross-section are formed in the rolls. For each cross-section the upper and the lower rolls have identical channels. A certain shear strain is created on the material passing through the rolls. This rolling process is described in greater detail in earlier reports [11, 12, 13]. The diameter of a billet is reduced by passing it successively through channels of lower cross-sections. After each pass, the billet was rotated by 90°before the next pass, to make the shear direction opposite to that of the previous pass. After passing the sample through the last channel, it was passed through it one more time after rotating by 90°. By this process, the texture was weakened, while more strain was accumulated in the sample. Starting with a direct extruded AZ31 billet, the tensile yield stress (YS) was nearly doubled while keeping elongation strain nearly constant. This resulted in high fracture toughness [14].

Along with grain refinement, a very high amount of strain accumulation resulted in sub-grain formation at nano-scale. This has to be taken into account for explaining the high YS of the alloy. Formation and structure of these sub-grains has been studied by transmission electron microscopy (TEM) and is reported here.

Experimental Procedure

A billet (42 mm diameter, 100 mm long) of commercially extruded AZ31 alloy (Mg-2.9Al-0.8Zn-0.42Mn, wt%) with an average grain size of 25μm was heated to 473K and then subjected to plastic deformation by caliber rolling. Its grain structure is shown in Fig. 1, which is bi-modal and displaying a number of twins. The billet was rolled through a series of calibers in a single roll, with a reduction in area of -18% for each caliber. A total of 18 passes were made. Details can

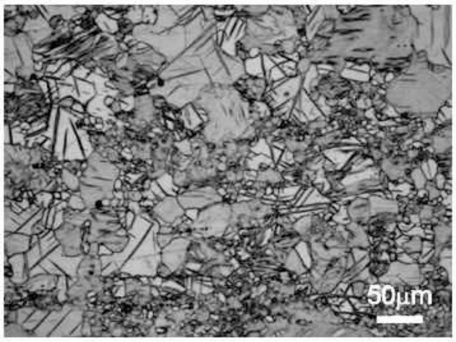

Figure 1: Optical micrograph showing grain structure of the starting material, which is a commercially extruded AZ31 alloy.

be found in earlier reports [13, 11, 12].

For TEM observations, the caliber rolled rod was sectioned longitudinally with a diamond saw to slice a sample of about 1 mm thickness. The sample was mechanically ground down to about 70 μm thickness, and then ion-milled to perforation in a precision ion mill. TEM observations were made in JEOL 2000FX-II microscope operated at 200 kV.

Results

Microstructures after 4 passes (4P sample) are shown in Fig. 2. A grain structure with sharp grain boundaries is observed, with grain size of about 8 μm. Grains with multiple neighbors and curved boundaries indicate that these grains have formed by recrystallization. Fig. 2(b) shows grains oriented along, or nearly along [0001] zone axis, with low angle boundaries in between them. Very little dislocation structures were observed inside the grains, such as the one marked D. However, region D' has a severely deformed structure. Precipitation structure in grain D is shown in Fig. 2(c). The precipitates have nearly uniform size of 20-30 nm.

In the grain in Fig. 3(a), oriented along a $\langle 1\bar{1}00\rangle$ zone axis, dislocation on basal planes are observed prominently. Details in inset show a set of non-basal dislocations too (one marked by an arrow). Besides this, apparent orientation relationships between grains were also observed. An example is shown in Fig. 3(b) (strong contrast of thickness contours are seen in grain M). Grain M is near a $\langle 11\bar{2}0\rangle$ zone axis orientation while grain N is nearly in $\langle 11\bar{2}3\rangle$ zone axis. There is a near planar match of $\{\bar{1}21\}_M \| \{010\}_N$. A stereographic analysis shows that these grains are related by a $\{10\bar{1}1\}$ or $\{10\bar{1}3\}$

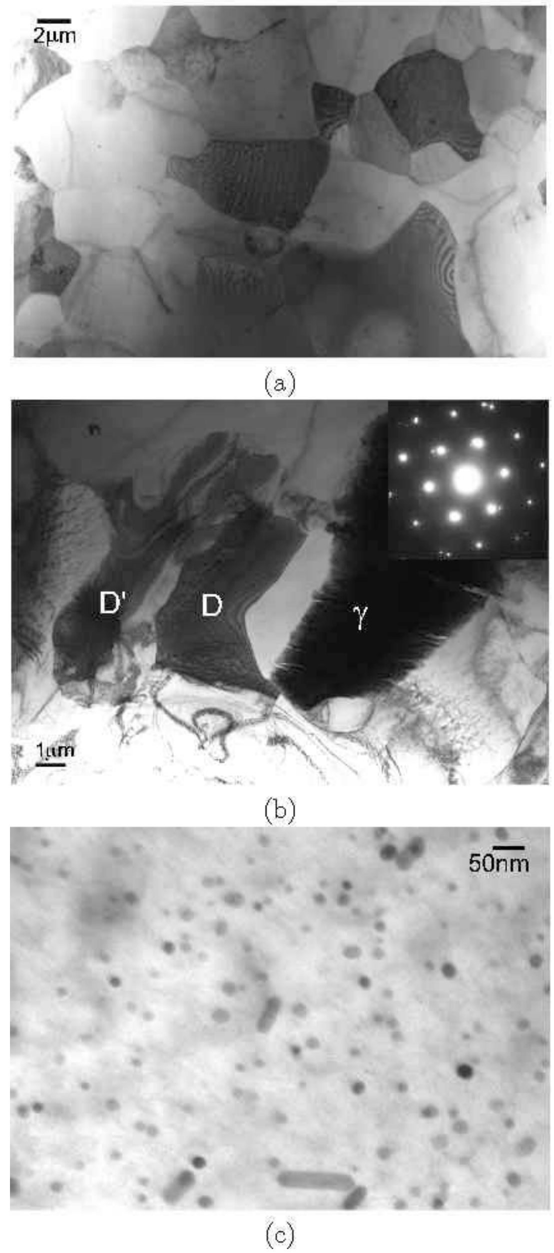

Figure 2: Bright field electron micrographs a 4 pass (4P) caliber rolled sample. (a) Grain structure near. (b) grains along [0001] zone axis (a diffraction patterns is inset) with small and low angle boundaries. γ is the $Al_{17}Mg_{12}$ phase. (c) Precipitation observed near [0001] zone axis.

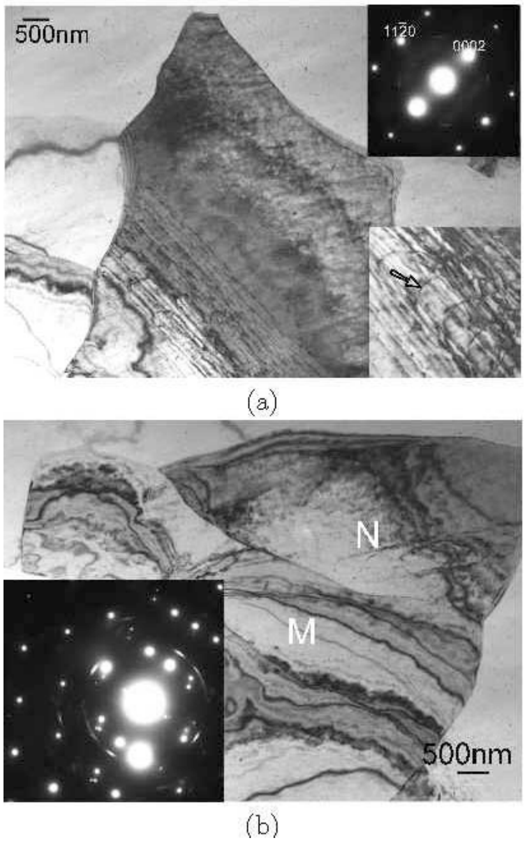

Figure 3: Deformation structures and twin-like orientations in 4P sample. (a) A grain along ⟨1100⟩ zone axis, showing dislocation contrast on basal planes. Inset lower right: detail of a small region in strong diffraction contrast. (b) Grains M in $\langle 11\bar{2}0\rangle$ zone axis and N in $\langle 11\bar{2}3\rangle$ zone axis. The composite diffraction pattern is inset.

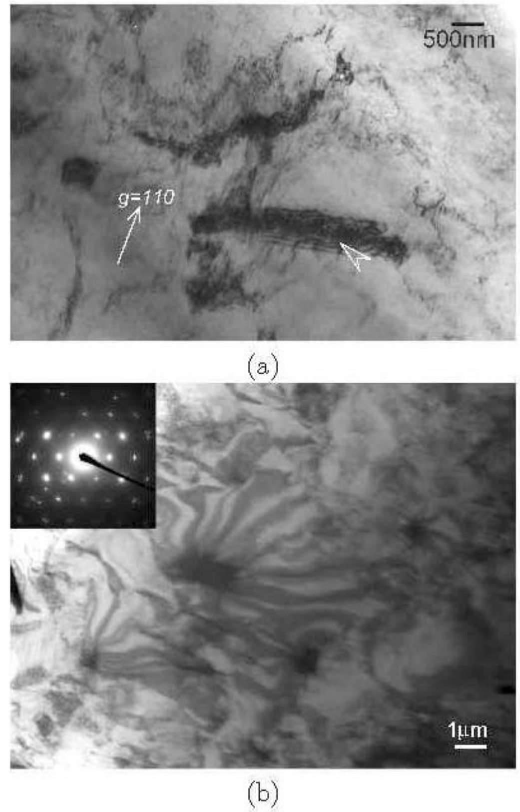

Figure 4: Bright field electron micrographs showing strain after 6 passes in the caliber roll (6P) (a) near zone axis $\langle 1\bar{1}00\rangle$, in a two-beam condition with a $\{11\bar{2}0\}$ spot and (b) along zone axis {0001}.

type twin. This indicates the role of twinning in deformation and evolution of the grain structure.

Fig. 4 from 6P sample shows bright field (BF) TEM micrographs along two zone axes; one in (a) is close to zone axis $\langle 1\bar{1}00\rangle$ in a two beam condition with a $\{11\bar{2}0\}$ reflection. Only a very small area is in contrast, due to the strained condition of the grain. A strong region of contrast is observed, marked by an arrowhead. This contrast is elongated perpendicular to the basal planes. This represents prismatic slip. The region in (b) is oriented along [0001] axis. Very interesting contrast is observed here. There are spots of dark contrast, from which radiate strain contrast. In other words, the strain is concentrated in a few spots and the basal plane is deformed. The corresponding diffraction pattern, inset, displays an asymmetry of spot intensity. The high index spots are weak and in form of arcs because of the matrix strain.

Fig. 5 are TEM BF micrographs showing matrix strain after eight passes (8P sample). (a) shows flow of strain contrast, in form of wavy lines. (b) is observed nearly along zone axis $\langle 11\bar{2}0\rangle$. In the diffraction pattern (inset), high index spots are absent or in form of weak arcs. In the bright field, ring-like contrasts are observed. In some places, like the one marked with an open arrow, the contrast suggests prismatic slip. In Fig. 5(c), a grain in [0001] zone axis is observed. Dislocation structure is observed inside, but the uniform contrast and the diffraction pattern (inset) do not suggest accumulated strain. It possibly is a grain recrystallized during caliber rolling.

Fig. 6 shows microstructures after 18 passes (18P). (a) shows accumulated strain; however, some sharp boundaries are also observed. There is a much finer subgrain structure. When the grain is tilted out of contrast (shown in (b)), distribution of nano-sized precipitates is observed. These precipitates are rounded, typically 10-20 nm or less in size, but occasionally as much as 50 nm. A number of sharp boundaries are observed in (c), which create grains or subgrains as small as 200 nm in size. Strain is observed piled up on these boundaries. The small grain in the center is in [0001] zone axis orientation.

Discussion

The initial material, which is an extruded rod, has a tube texture of basal planes. Thus during caliber rolling the grains, depending on their orientation, experience a compressive stress either along their hexagonal axis, perpendicular to it, or in between. Correspondingly, basal or non-basal slip, and/or twins will be generated. Reported strain simulation and experimental estimation [13] show that the strain becomes larger by increasing number of passes and that it has a distribution with maxima around the corners.

After 4 passes, the grain size is considerably refined, with an average grain size of about 5μm. Absence of deformation inside the grains indicate that they are formed by recrystallization. 4P sample also shows large areas with low angle grain boundaries, such as in Fig. 2(a) along zone axis [0001], which are remnants of initial large size grains.

In this sample, various kinds of lattice defects are observed clearly. In Fig. 2(b), grains D and D' in similar orientations show different levels of strain. Grain D' shows a high level of strain while D does not. Dislocations are observed clearly in Fig. 3(a), which are basal as well as non-basal.

Beyond 4P, the 6P and 8P samples show further accumulation of strain, which lead to the final microstructure in 18P.

Hardness measurements reported by Inoue et al. [13] in a similar experiment show a similar trend in evolution of the microstructure. Partial DRX occurs until 7 passes and complete DRX until 11 passes. Subsequently, continuous DRX occurs until 18P. Similarly, in our samples partial DRX is observed in 4P.

Inoue et al. [13] also report that after 7 passes the microstructure consists of a mixture of fine grains and coarse grains. The coarse grains were elongated along the rolling direction. The fine grains were considered to be generated by DRX during the rolling. The average grain size of the fine grains was estimated to be 3.8μm. From 7 passes to 11 passes the fraction of coarse grains decreases, but the average grain size

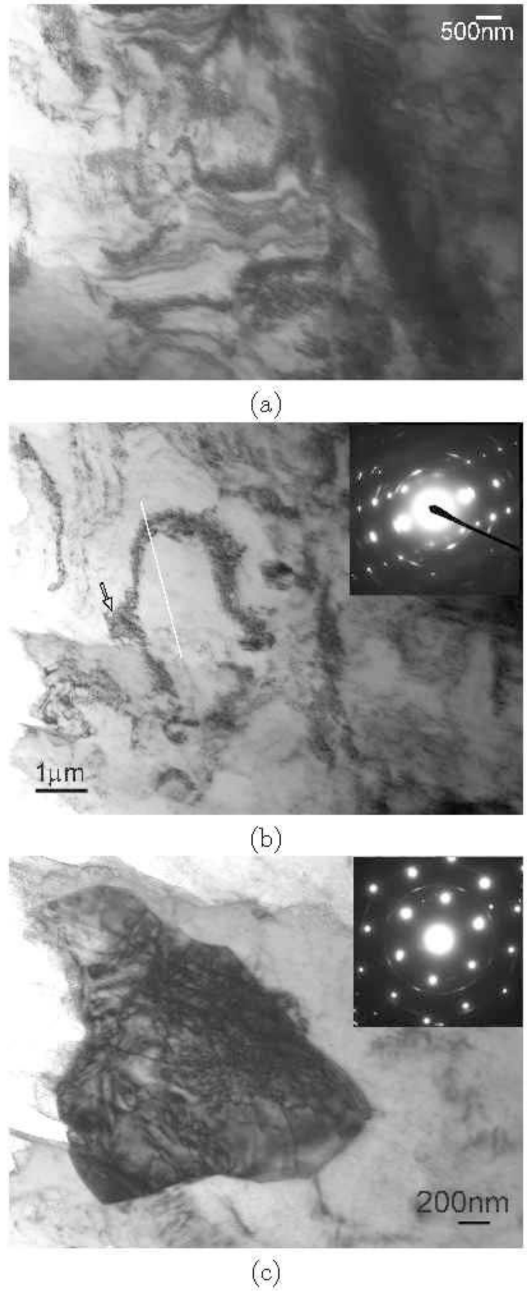

Figure 5: Bright field electron micrographs showing accumulated strain after 8 passes in the caliber roll (8P)(a) severe deformation structures, (b) a region observed along $\langle 11\overline{2}0\rangle$ (white line denotes the direction of the basal planes) and (b) a grain along zone axis [0001].

of the fine grains remained the same. On further rolling to 15 passes, the average grain size decreased to about 2.5μm due to continuous DRX, while the fraction of low angle boundaries increased [14]. Our TEM studies here show that much smaller domains occur because of strain accumulation and sub-grain boundaries formation.

With the evolution of grain structure during the caliber rolling, a change in the strain accumulation mechanism will be expected. This also comes out in the hardness curve reported by Inoue et al. [13]. This is because in the beginning of the rolling process the grain size is considerably large, at 25μm, but once the grain size has become small to about 5μm or less, the stress will generate considerable non-basal slip [15]. Thus in the 4P sample, basal slip is clearly visible (Fig. 3(a)). Much more complex slip is observed in 6P and 8P samples. This is facilitated by change of stress direction by a rotation of the sample after each pass. Accumulation of strain at certain points in basal planes due to this is clearly observed in Fig. 4(b). Such a strain will make basal slip harder. Fig. 4(a) clearly shows strain in basal plane as well prismatic planes. Fig. 5(a) shows wavy patterns due to accumulation of a large amount of multi-directional strain. Fig. 4(b) shows wavy bands of same orientation about 500nm in width.

Inoue et al. [13] also show that the increase in hardness of the bar up to 11 passes is due to strain hardening and grain refinement by DRX, while the subsequent hardness is attributed to sub-grain refinement based on continuous DRX.

It may be assumed that there are cycles of strain accumulation and recrystallization during the caliber rolling. Electron Backscattered diffraction (EBSD) [11] shows sharp boundaries after 15 passes and elongated srained structure after 18 passes. Thus, presumably, the sharp boundaries form after 15 passes followed by further strain accumulation. There is also a marked weakening of texture after 15P [11].

In the final microstructure in 18P, sharp boundaries are observed, which make domains as small as 200nm in size/width (Fig. 6). Accompanying this is the strain in the matrix, which enhances the yield stress. The very high yield stress of the alloy is thus effectively because of the very small size domains with sharp boundaries. These domains can hold a high concentration of strain in them.

It should be noted that a similar microstructure is also shown by multi-directional forging of AZ31 alloy [16].

Precipitation also has a strong contribution to

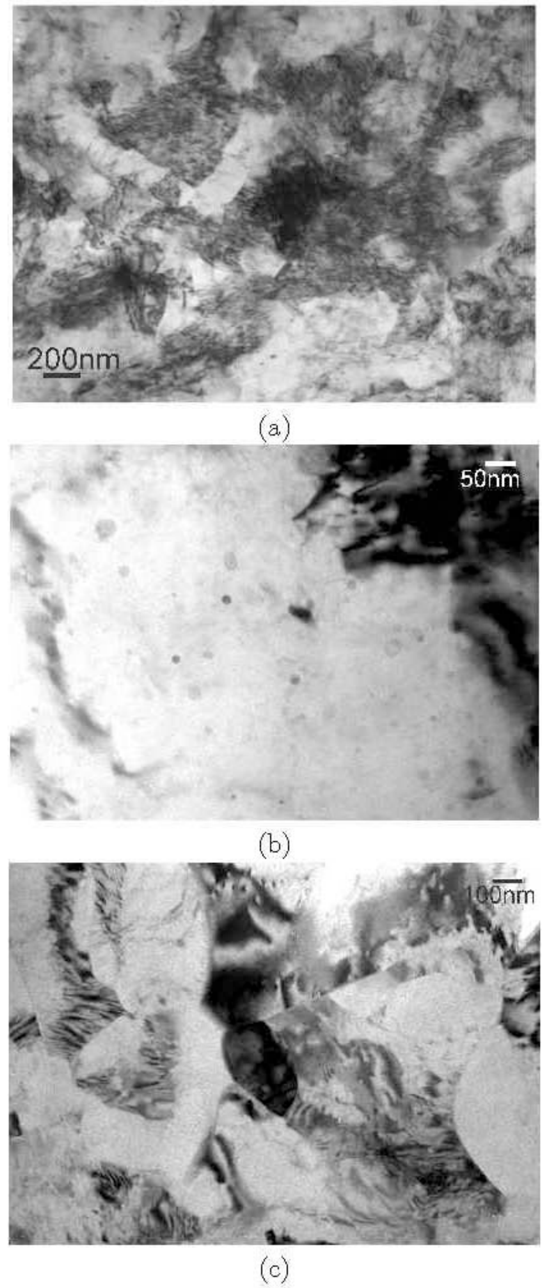

Figure 6: Bright field electron micrographs showing microstructure after 18 passes in the caliber roll (18P)(a) severe deformation structures, (b) precipitation in the matrix and (b) grain and subgrains, the grain in the center is along zone axis [0001].

strength. A modification in the precipitate distribution is observed by caliber rolling. Precipitates observed in 4P sample, Fig. 2(c), are in a narrow size range of about 10 to 20nm. In 18P sample in Fig. 6(b), more of finer size precipitates are observed. A finer distribution of precipitates and possibly a supersaturation of the matrix due to processing effect similar to mechanical alloying will also contribute to the strengthening.

Conclusions

Microstructure development of AZ31 alloy during caliber rolling has been studied by transmission electron microscopy. The following conclusions are made:

1. Well defined basal slip as well as non-basal slips are observed during early passes. Recrystallization occurs by 4 passes, whereby the grain size is reduced to about 5μm.

2. Further accumulations of strain occurs in further passes. Severe localization of strain, basal and prismatic slip are observed after 6 and 8 passes.

3. By 18 passes, sharp boundaries appear, making domains of about 200nm thick, in which are contained severely strained regions.

References

[1] K. Kubota, M. Mabuchi, K. Higashi, "Processing and mechanical properties of fine-grained magnesium alloys", *J.Mater. Sci.* 34 (1999) 2255.

[2] J. Koike, R. Ohyama, T. Kobayashi, M. Suzuki, K. Maruyama, "Grain-Boundary Sliding in AZ31 Magnesium Alloys at Room Temperature to 523K", *Mater. Trans.* 44 (2004) 445.

[3] T. Mukai, M. Yamanoi, H. Watanabe, K. Higashi, "Ductility enhancement in AZ31 magnesium alloy by controlling its grain structure", *Scripta Mater.* 45 (2001) 89.

[4] A. Singh, H. Somekawa, T. Mukai, "Compressive Strength and Yield Asymmetry in Extruded Mg-Zn-Ho Alloys Containing Quasicrystal Phase", *Scripta Mater.* 56 (2007) 935.

[5] A. Yamashita, Z. Horita, T.G. Langdon, "Improving the mechanical properties of magnesium and a magnesium alloy through severe plastic deformation", *Mater. Sci. Eng.A* 300 (2001) 142.

[6] W.J. Kim, C.W. An, Y.S. Kim, S.I. Hong, "Mechanical properties and microstructures of an AZ61 Mg Alloy produced by equal channel angular pressing", *Scripta Mater.* 47 (2002) 39.

[7] Y. Yoshida, L. Cisar, S. Kamado, Y. Kojima, "Effect of Microstructural Factors on Tensile Properties of an ECAE-Processed AZ31 Magnesium Alloy", *Mater. Trans.* 44 (2003) 468.

[8] S.R. Agnew, J.A. Horton, T.M. Lillo, D.W. Brown, "Enhanced ductility in strongly textured magnesium produced by equal channel angular processing", *Scripta Mater.* 50 (2004) 377.

[9] S.R. Agnew, P. Mehrotra, T.M. Lollo, G.M. Stoica, P.K. Liaw, "Texture evolution of five wrought magnesium alloys during route A equal channel angular extrusion: Experiments and simulations", *Acta Mater.* 53 (2005) 3135.

[10] J.T. Wang, D.L. Yin, J.Q. Liu, J. Tao, Y.L Su, X. Zhao, "Effect of grain size on mechanical property of Mg-3Al-1Zn alloy", *Scripta Mater.* 59 (2008) 63.

[11] T. Mukai, H. Somekawa, T. Inoue, A. Singh, "Strengthening Mg.Al.Zn alloy by repetitive oblique shear strain with caliber roll", *Scripta Mater.* 62 (2010) 113.

[12] T. Mukai, H. Somekawa, A. Singh, T. Inoue, "Strengthening Mg-Al-Zn alloy by repetitive oblique shear strain", in *Magnesium Technology 2011* (eds. W.H. Sillekens, S.R. Agnew, N.R. Neelameggham, S.N. Mathaudhu), TMS, 2011, p.211.

[13] T. Inoue, H. Somekawa, T. Mukai, "Hardness Variation and Strain Distribution in Magnesium Alloy AZ31 Processed by Multi-pass Caliber Rolling", *Adv. Eng. Mater.* 11 (2009) 654.

[14] H. Somekawa, A. Singh, T. Inoue, T. Mukai, "Enhancing fracture toughness of magnesium alloy by formation of low angle grain boundary structure", *Adv. Engg. Mater.* 12 (2010) 837.

[15] J. Koike, et al., "The activity of non-basal slip system and dynamic recovery at room temperature in fine-grained AZ31B magnesium alloys", *Acta Mater.* 51 (2003) 2055.

[16] J. Xing, X. Yang, H. Miura, T. Sakai, "Mechanical Properties of Magnesium Alloy AZ31 after Severe Plastic Deformation", *Mater. Trans.* 49 (2008) 69.