Original Article

Evolution in hardness and microstructure of ZK60A magnesium alloy processed by high-pressure torsion☆

Han-Joo Lee a, Byungmin Ahn b, Megumi Kawasaki a,c,* , Terence G. Langdon c,d

a Division of Materials Science and Engineering, Hanyang University, Seoul, South Korea
b Department of Energy Systems Research, Ajou University, Suwon, South Korea
c Departments of Aerospace & Mechanical Engineering and Materials Science, University of Southern California, Los Angeles, USA
d Materials Research Group, Faculty of Engineering and the Environment, University of Southampton, Southampton, UK

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Abstract
Severe plastic deformation is an attractive processing method for refining microstructures of metallic materials to have ultrafine grain sizes within the submicrometer or even the nanometer levels. Especially, it becomes generally known that processing of metals through the application of high-pressure torsion (HPT) provides the potential for achieving exceptional grain refinement in bulk disk metals. In the present study, a ZK60A magnesium alloy was processed by HPT at room temperature for a series of numbers of revolutions under a constant compressive pressure of 6.0 GPa. The change in texture was examined by X-ray diffraction (XRD) analysis and the evolution of hardness was evaluated using Vickers microhardness measurements to provide a comprehensive understanding of microstructural evolution. The XRD analysis showed the texture changed to weak and random in the early stage of HPT. The hardness results demonstrated that the hardness evolution with increasing equivalent strain follows the strain hardening model without microstructural recovery and the degree of hardenability was calculated as ∼0.07 which implies a high degree of strain hardening toward hardness homogeneity.

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* Corresponding author.
E-mail: megumi@hanyang.ac.kr (M. Kawasaki).
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1. Introduction

The processing of metals through the application of severe plastic deformation (SPD) [1] has attracted much attention in the last two decades in order to achieve substantial grain refinement in bulk metal solids. This is due to the potential of the SPD technique as alternative processing methods compared to the general thermo-mechanical processing used in conventional industrial practice which introduces microstructures with submicron grain sizes. The materials produced by the SPD techniques are generally termed ultrafine-grained (UFG) materials. Among a number of possible SPD techniques [2,3], equal-channel angular pressing (ECAP) [4] and high-pressure torsion (HPT) [5] are generally accepted as the major SPD methods [6]. In practice, although the two processing procedures of ECAP and HPT are effective for significant grain refinement, several experiments demonstrated that the grains produced by HPT are generally smaller than those achieved using ECAP [7–9]. Accordingly, HPT processing is advancing rapidly in recent research work in the SPD field and is becoming a significant processing procedure.

Conventional HPT processing uses a thin disk which is placed between two massive anvils and subjected to a high compressive pressure and concurrent torsional straining by rotating one of the anvils. The equivalent von Mises strain, \( \varepsilon_{eq} \), imposed through the rotation of the anvil is given by an expression of the form [10,11]

\[
\varepsilon_{eq} = \frac{2\pi N r}{h \sqrt{3}} \tag{1}
\]

where \( N \) is the number of HPT revolutions and \( r \) and \( h \) are the radius and thickness of the disk, respectively. It is apparent from Eq. (1) that the equivalent strain is a maximum at the edge of the disk sample whereas the strain decreases to zero at the center of the disk where \( r = 0 \). The calculation therefore implies there is a significant microstructural inhomogeneity depending on the location across the diameter of the disk. Nevertheless, a number of experiments demonstrated that, due to the severe deformation under compressive pressure at all times, both sufficiently high numbers of HPT turns and high applied pressure lead to homogeneous microstructures and hardness throughout the disk [12,13].

Magnesium is the most lightweight metal of all practical metallic materials and it has outstanding mechanical properties such as a high strength-to-weight ratio and high elasticity. Therefore, the magnesium alloys have received much attention as lightweight structural materials for promising applications in the automobile and aerospace industries and for portable electrical devices [14,15]. Moreover, magnesium is the 6th most abundant metal on the earth accounting for 2.5% of the crust as a mineral and the recycling properties of magnesium are also satisfactory so that it will become a very important resource in the future.

However, due to the low symmetry in the hexagonal close-packed (h.c.p.) crystal structure of Mg, the slip system at room temperature is limited to basal slip leading to difficulties for adding any forming capability. Accordingly, the microstructural refinement plays a vital role for Mg alloys to provide the capability of achieving both high strength and a reasonable level of ductility. There are a number of experiments available showing successful grain refinement and enhanced mechanical properties through ECAP and special emphasis was placed on high temperature superplastic ductility in many investigations where the data are summarized recently on the Mg alloys processed by ECAP [16,17].

The present study was initiated to examine the microstructural changes in a commercial-quality ZK60A magnesium alloy through the application of HPT at room temperature. Following the changes in microstructure, the evolution of texture and the improvement in hardness were evaluated on the Mg alloy with increasing numbers of HPT turns, thereby increasing the equivalent strains through torsion straining by HPT. In addition, the degree of strain hardening, defined by the hardenability exponent, is calculated in the early stage of HPT on the Mg alloy.

2. Experimental material and procedures

The present experiments used a commercial-quality ZK60A magnesium alloy supplied as a bar after extrusion with a diameter of 10 mm. The bar was cut into billets and they were sliced as disks with thicknesses of \( \sim 1.2 \) mm. A series of disks was then polished to have a final parallel thickness of \( \sim 0.83 \) mm.

The ZK60A disks were processed by HPT at room temperature in quasi-constrained HPT conditions [18,19] following the conventional procedure described earlier [20]. Specifically, the disk was placed between two anvils so as to receive a compressive pressure, and concurrently imposed torsional straining by rotating the lower anvil. The disks were processed under a range of compressive pressures of \( P = 1.0, 3.0 \) and \( 6.0 \) GPa and a rotational speed of 1 rpm for total revolutions, \( N \), of 0 where a disk was compressed without rotation for 1 min and of 1/4, 1/2, 1, 3 and 5 turns.

The microstructure of both as-received material and the disk after HPT for 5 turns at 6.0 GPa were characterized using an equipment of electron backscatter diffraction (EBSD), EDAX Digiview-1, in field emission scanning electron microscopy (FE-SEM), Hitachi S-4300SE. The EBSD specimens were prepared using a broad ion beam polisher, Hitachi IM-3000. The step size of the EBSD map was 1.2 \( \mu \)m at an accelerating voltage of 12 kV. After the raw orientation maps were collected, standard cleanup procedures were conducted to minimize the incorrectly indexed pixels and background noise using TSL OIM software.

Following the microstructural analysis, the texture evolution with increasing pressure and numbers of turns was examined on the processed ZK60A disk. The disk surfaces after HPT were polished slightly and examined by X-ray diffraction (XRD) analysis using a Rigaku UltimaIV XRD with a Cu Kα radiation in a scan speed of 2°/min and a step interval of 0.01°.

The Vickers microhardness measurements were conducted on the processed ZK60A disks with a Shimadzu HMV-2E microhardness tester using a load of 50 gf for a dwelling time of 10 s. Following the procedure described earlier [20], the measurements were conducted along a disk...
diameter on each polished surface of the ZK60A disks after HPT. Each microhardness value, Hv, was calculated from four measurements taken at cruciform directions with a distance of 0.15 mm so that the representative positions for demonstrating the hardness variation lie along a diameter with separations of 0.3 mm from the center of the disk. In order to avoid any influence of the possible hardness gradation through the disk thickness direction [21,22], all measurements were taken on the polished planes at the mid-sections through the disk heights.

3. Experimental results

3.1. Microstructure evolution

Fig. 1 shows an OIM image taken through an EBSD analysis of the ZK60A magnesium alloy in an as-received condition and the orientation of the grains are represented by unique colors denoted by the color key at the bottom of the figure. The as-received material was supplied after extrusion and the microstructure was observed at a cross-section normal to the extrusion direction. Most grain orientations consist of a (1010) plane and this is typical for an extruded h.c.p. structure at normal to the extrusion direction as basal slip on the (0001) plane is parallel to the extrusion direction. Although there are some large grains detectable in Fig. 1, from the wide area of scanning with several OIM photos the average grain size was estimated as ~10 μm in the as-received condition.

![Fig. 1 – An OIM image through an EBSD analysis of the ZK60A magnesium alloy in an as-received condition.](image)

The microstructure of the ZK60A was refined through HPT for 5 turns. An OIM map acquired through the EBSD analysis demonstrated a limited resolution at the center of the disk after 5 turns by HPT. This is due to a complex microstructure with a possibility of the bi-modal formation in the ZK60A by HPT processing. In fact, the formation of the bi-modal structure was reported recently in a ZK60 alloy after HPT at room temperature under 2.0 GPa for 5 turns [23]. Nevertheless, instead of the OIM map, an EBSD image quality (IQ) map is shown in Fig. 2 for the disk center after HPT for 5 turns where an IQ map is also generally used to show detailed microstructures. It is seen in the IQ map that several grains are visible as brighter colors and these are in a range 2–5 μm. The regions shown with darker colors between the grains are anticipated to have a UFG microstructure as reported in earlier literature for the central region of a ZK60 after 5 turns whereas there is a uniform ultrafine grain structure at the peripheral region [23]. The formation of bi-modal microstructure is often observed in Mg alloys after ECAP [24–28] and the unique microstructure tends to be responsible for the superplastic behavior [24,25]. Moreover, it is explained by a suggested model and by microstructural observations that the introduced bi-modal microstructure by severe plastic deformation is observed in a transitional condition and further processing may lead to a homogeneous ultrafine grain distribution [26].

3.2. Texture evolution

The evolution of texture through HPT was examined by an XRD analysis and the results are shown in Fig. 3 where the XRD patterns are shown in (a) for, from the top, the ZK60A alloy in an as-received condition, after compression under 1.0 GPa without torsion straining (0 turn) and after HPT for 1/4, 1/2 and 5 turns under 1.0 GPa and in (b) for the disks processed for 5 turns under, from the bottom, 1.0, 3.0 and 6.0 GPa. In the XRD analysis with a limited angle of 2 theta of 31–40°, relatively high intensities of XRD peaks of pure Mg on the (111) prism plane, (0002) basal plane and (1011) pyramidal plane are visible for all tested disks. Thus, the relative peak intensities
parallel to the extrusion direction and thus the prism plane lies parallel to the disk surface of the ZK60A in an as-received condition. Since the characteristics of the random texture in a general casting microstructure prior to extrusion tends to remain even after extrusion, as shown on the top in Fig. 3(a), the as-received ZK60A disk prior to HPT shows a strong texture with a highest peak intensity on the (1010) prism plane and a second strongest intensity on the (1011) pyramidal plane whereas almost no peak intensity on the (0002) basal plane. These XRD results are consistent with the EBSD results shown in Fig. 1 for the as-received material.

Through uniaxial compression without torsion straining (0 turn), the active slip plane at room temperature of the basal plane tends to lie parallel to the limited outer flow of the material which corresponds to a radial direction within the disk due to the characteristics of the quasi-constrained conditions [18,19]. Moreover, as shown in an earlier paper demonstrating in situ texture analysis on an AZ31 magnesium alloy under uniaxial loading, the rapid texture change in the Mg alloy can be explained by the high twinning activity when grains having their c-axes perpendicular to the loading direction are relatively abundant at the measurement plane [31]. Thus, maintaining the relative peak strength on the pyramidal plane constant, the highest intensity shifts from the prism plane to the basal plane and it is shown in the second upper XRD profile in Fig. 3(a) for the disk under severe compression.

After torsional straining for 1/4 turn, significant grain refinement with increasing high-angle grain boundaries is anticipated within the disk even in the early stage of HPT so that the texture tends to become weaker, thereby demonstrating random texture with the highest peak intensity on the pyramidal plane. Additional torsional straining for 1/2 turn shows a consistent variation of the relative peak intensities on these slip planes with those after 1/4 turn. A consistent trend showing the random texture of Mg was recorded in the ZK60A alloy through HPT for up to 5 turns under a pressure of 1.0 GPa as shown at the bottom in Fig. 3(a). A close inspection of these XRD patterns in Fig. 3(a) shows there is evidence of the occurrence of peak broadening with increasing numbers of HPT turns and it is especially apparent from N=1/2 turn to 5 turns. Thereby, it demonstrates significant grain refinement through HPT up to 5 turns in the ZK60A alloy.

Fig. 3(b) displays the variation of XRD peaks with increasing HPT compressive pressure through 5 revolutions. It is apparent that there is consistency between the relative peak strength on these three major Mg slip planes at all pressures after 5 turns whereas there is a slight increase in peak broadening for each relative XRD peak with increasing compressive pressure from 1.0 GPa to 3.0 GPa and 6.0 GPa. This is also due to the high level of grain refinement in the disks processed under high pressure of 3.0 and 6.0 GPa compared with the disk under 1.0 GPa when processing through 5 turns.

3.3. Hardness evolution

The ZK60A disks after HPT at 6.0 GPa for up to 5 turns were examined to show the hardness variations along the disk diameters and the results are shown in Fig. 4 where the lower
dashed line denotes the microhardness value, Hv, of ~72 of the ZK60A in the as-received condition.

It is apparent that the alloy shows higher Hv values over all of the disk surfaces after HPT. Specifically, higher hardness is shown at the peripheral regions and the hardness values decrease toward the centers of the disks in the early stage of HPT through 1 turn. Thus, there are consistent Hv values of ~75 at r = 0 and ~95 at the edges of the disks after 1/4, 1/2 and 1 turn except for the slightly higher Hv values at some measurement points in the peripheral region in the disk after 1 turn. Additional HPT through 3 turns developed higher Hv values at the peripheral region toward the disk center so that the area showing the higher Hv values becomes wider but there is a consistent low Hv value of ~80 at the disk center. With increasing numbers of revolutions to 5, the hardness values are saturated to Hv ≈ 105–110 through the total disk diameter and thus the ZK60A demonstrates a reasonable hardness homogeneity through 5 turns by HPT. A recent report on ZK60 demonstrated a similar hardness evolution toward homogeneity over the disk surfaces after HPT under 2.0 GPa and the homogeneous distribution of high hardness was achieved also after 5 revolutions [23].

The measured microhardness values in the central region are lower compared with those in the peripheral region in the ZK60A disks in the early stage of HPT and in practice it is consistent with the imposed equivalent strain as calculated in Eq. (1). Thus, the material demonstrates a strain hardening behavior during the hardness evolution toward homogeneously distributed high hardness on the disk surfaces and this type of hardness behavior was predicted earlier by a theoretical approach using strain gradient plasticity modeling [32]. Moreover, a very recent analysis by three-dimensional FEM confirmed the validity of the approach by confirming the increasing strength of the material with increasing distance from the center of the disk and with increasing numbers of HPT turns in strain-hardening materials [33].

4. Discussion

In the present study, a ZK60A was refined successfully from an initial grain size of ~10 μm to 2–5 μm with probable finer grains consisting of a bi-modal structure after HPT for 5 turns at 6.0 GPa. The XRD results shown in Fig. 3(a) demonstrate that there is a significant change in texture toward an ultimate random texture after HPT for 5 turns from the initial strong extrusion texture via a transition texture at a quasi-constrained compression stage without torsion. Similar evolution in an initial extrusion texture toward a random texture was reported earlier in an AZ31 magnesium alloy after ECAP for 8 passes at 473 K [29]. Moreover, the XRD results shown in Fig. 3(b) support the successful grain refinement in the ZK60A when processed through 5 turns by HPT under severe compressive pressures, and this is especially effective at higher pressures such as 3.0 and 6.0 GPa. The significant grain refinement is also demonstrated from the present microhardness measurements where the increasing numbers of HPT turns to 5 led to a saturated high hardness in the ZK60A alloy. The present experimental results are in good agreement with the earlier experiments suggesting that HPT processing conditions of pressures higher than ~6 GPa and more than 5 turns are desirable for producing reasonably homogeneous UFG microstructures in metals [8,13].

In order to fully understand the evolution of microstructure through HPT, an easy and effective approach is to evaluate the measured microhardness values with increasing values of equivalent strain as calculated using Eq. (1). This approach was first demonstrated in an earlier report on an austenitic steel after HPT for a range of turns under a constant pressure of 5.3 GPa [34]. Thereafter, the approach has been applied for a series of metals and alloys so that three different models of evolution toward hardness homogeneity were suggested recently depending upon the nature of recovery during plastic deformation in the material [35-38].

Among the three models, the most common model of hardness evolution shows higher hardness with increasing equivalent strain, thereby exhibiting strain hardening, and the ZK60A is considered to follow this hardness model. Fig. 5 shows a plot of the variations of the measured microhardness values shown in Fig. 4 with the calculated equivalent strains for ZK60A after HPT for 1/4, 1/2, 1, 3 and 5 turns under 6.0 GPa, where the lower dashed line denotes the hardness value in the as-received condition. It is apparent that the hardness increases with increasing equivalent strain and ultimately saturates to a high Hv value of ~110 at a reasonably high strain. It was summarized recently that most commercial purity metals and alloys exhibit this type of plot denoting the material exhibiting strain hardening in the absence of any dynamic recovery [35-38].

The other two models include strain hardening with recovery and strain softening. The former model involves strain hardening in a very early stage of HPT for a short period of time followed by softening due to microstructural recovery so that the hardness values depict a bell-shaped curve with increasing equivalent strain. This behavior was reported in several high-purity metals of Al [20,21,35,38-43], Mg [44] and Zn [45]. The later model reported for two-phase eutectic and eutectoid
alloys having relatively low melting temperature such as Zn-22% Al eutectoid alloy [36,46–48] and Pb-62% Sn eutectic alloy [47]. The unique characteristic of this behavior is that the hardness values after HPT are lower than in the unprocessed condition even though there is a significant grain refinement by HPT processing. The significant softening by HPT in a Zn-22% Al alloy is due to the dynamic change in the precipitation kinetics during the severe deformation by HPT resulting in a loss of hard Zn precipitates in the Al-rich phase [49].

For a better understanding of the hardness evolution in the ZK60A alloy, a quantitative analysis was conducted for estimating the degree of strain hardening by calculating the hardenability exponent which corresponds to the slope in a double-natural logarithmic plot of the Hv values versus equivalent strain. This approach was first demonstrated on a Ti-6% Al-4% V alloy after HPT from two different initial conditions and gave hardenability exponents of 0.031 and 0.052 [50].

The constructed double-natural logarithmic plot of the Hv values versus $\varepsilon_{eq}$ is shown in Fig. 6 for ZK60A after HPT for up to 5 turns under 6.0 GPa in which the Hv values are taken from Fig. 4 up to $\varepsilon_{eq} \approx 20$, where a high degree of strain hardening is observed before reaching the hardness saturation. The solid line in the plot describes a trend of all datum points which denotes the following form:

$$Hv = 84 \varepsilon_{eq}^{0.07}$$

Thus, a hardenability exponent of $\sim 0.07$ is observed for ZK60A where a positive exponent denotes strain hardening. This exponent is consistent with the value of 0.08 reported for an AZ31 magnesium alloy after HPT for 1 and 5 turns at 6.0 GPa [38] and these Mg alloys demonstrate a stronger strain hardening behavior than the processed Ti alloy reported earlier [50]. It is worth noting that this approach is also applicable to materials showing hardness behaviors of strain hardening with microstructural recovery and strain softening [38]. Thus, this approach is promising for acquiring quantitative information of the hardness evolution in metals processed by HPT and now it requires additional data from a series of metals to develop into a practical approach.

5. Summary and conclusions

1. A ZK60A magnesium alloy was processed by HPT at room temperature under a series of pressures up to 6.0 GPa and torsional straining from 1/4 to 5 turns.
2. Microstructural observations demonstrated that there is a significant grain refinement through the HPT processing and it was also apparent from a texture examination by XRD analysis and microhardness measurements.
3. The texture analysis demonstrated apparent changes from a strong extrusion texture to random texture in an early stage of HPT.
4. The ZK60A alloy demonstrated a strain hardening behavior toward hardness homogeneity and a hardenability exponent of $\sim 0.07$ was estimated through the HPT under a pressure of 6.0 GPa.

Conflicts of interest

The authors declare no conflicts of interest.

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