Fabrication and Characterization of Supersaturated Al-Mg Alloys by Severe Plastic Deformation and Their Mechanical Properties

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Excess amount of Mg was intentionally added to fabricate supersaturated ultra-fine grained Al-Mg alloy by high pressure torsion (HPT). Their microstructures and mechanical properties were examined by X-ray diffraction (XRD) analysis, transmission electron microscopy (TEM) and Vickers microhardness test. It was confirmed that the minimum grain size reached ~40 nm and maximum microhardness 292 Hv at the Mg content with 30 mass%. Further increase of the amount of Mg resulted the formation of β-Al3Mg2 and decrease of microhardness.

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1. Introduction

Severe plastic deformation is a promising technique to achieve ultrafine-grained (UFG) metallic materials with high strength according to the Hall-Petch relationship.1–5) One of the SPD processes, high pressure torsion (HPT) introduces intense strain to the sample by way of two anvils with rotation under a high pressure, as illustrated in Fig. 1.1–3) In addition, HPT has a great potential to synthesize densely packed materials from mixed powders without a sintering process.6–9) Furthermore, Senkov et al. showed that it was possible to synthesize the supersaturated solid solution of Fe in the Al matrix using HPT from cast Al-Fe alloy.10) Consequently, there are possibilities to prepare supersaturated Al-Mg alloys by mixing powders of Al and Mg.

Addition of Mg to Al is popular in Al industry because Mg is a typical element to introduce significant hardening of Al through a solid solution effect. Although the equilibrium solid solubility of Mg in Al is about 1 mass% at room temperature,11,12) an extended solid solubility of Mg in Al can be achieved by rapid solidification as well as by mechanical alloying of elemental powders. For example, Luo et al. reported that the solid solubility of Mg in Al can be extended far beyond the equilibrium concentrations up to ~34.6 mass% by rapid quenching.13) A mechanical alloying method has also been employed for synthesis of supersaturated alloys including those in amorphous states. Calka et al. et al. showed that it was possible to extend the solid solubility of Mg in Al up to 16.5 mass% in the case of Al70Mg30 and 42.4 mass% for Al50Mg50 by mechanical alloying.14) Similarly, Zhang et al. and Schoenitz and Dreizin observed the formation of a solid solution containing 21.2 and 19.1 mass% of Mg in Al from Al60Mg40 by mechanically alloying, respectively.15,16)

In this study, fabrication of supersaturated Al-Mg alloys with different contents of Mg is attempted using HPT. The solubility of Mg and related microstructures are characterized by both X-ray diffraction (XRD) analysis and transmission electron microscopy (TEM). Mechanical properties are also examined by Vickers microhardness test.

2. Experimental Methods

The materials used in this study were high-purity (99.99%) Al powders with 75 μm in diameter and high-purity (99.99%) Mg powders with 150 μm in diameter. The Al powders were mixed with various amounts of the Mg powders. Approximately 0.3 g of the powder mixture was placed in a shallow circular hole, 10 mm in diameter and 0.25 mm in depth, located at the center of the lower anvil of the HPT facility. The lower anvil was lifted until it made contact with the upper anvil, which had the same dimensions as the lower anvil, and then rotated with respect to the upper anvil at a rotation speed of 1 rpm. This HPT operation was undertaken at room temperature with an applied pressure of 2.5 GPA and 6.0 GPA for different numbers of revolutions as summarized in Table 1. The rotation was initiated for 5 s after the load application and the temperature rise during the HPT operation was monitored using a thermocouple placed 10 mm above the sample. The thermocouple was guided through a hole made vertically in the center axis of the upper anvil. The measurement indicated that the temperature...
gradually increased up to 353 K after 100 turns. The final sample after the HPT operation was in the shape of a disk with dimensions of 10 mm in diameter and about 0.7 mm in thickness. It should be noted that an extra amount of the mixed powder was deliberately added to prevent the upper and lower anvils from contacting each other directly so that the damage and wear to both anvils would be avoided.6,7) Analysis using XRD was carried out with a Rigaku RINT-2100 using the Co Kα radiation with LaB₆ as a reference material. Regions of 4 mm diameter at the center of the HPT disks were deliberately cut out to minimize the part of specimen where less strain was introduced and thus, the effect of strain was not correctly reflected in the XRD analysis. Then, 3 mm disks were cut from the HPT disks at positions 3 mm away from the center using an electrical spark discharge machine for TEM observations. The 3.0 mm disks were further ground mechanically with abrasive papers to a thickness of 0.15 mm and thinned in a solution of 20% perchloric acid (HClO₄), 10% glycerol (C₃H₈O₃) and 70% ethanol (C₂H₅OH) using a twin-jet electropolishing apparatus. Microstructures were observed using a TECNAI-20 (FEI, Eindhoven, The Netherlands) and H-8100 (Hitachi, Japan) operated at 200 kV. In particular, selected area electron diffraction (SAED) patterns were taken from areas of 2.5 μm diameter for HPT-processed specimens. Mean grain sizes were measured from 100 grains in dark field TEM images. The polished disk samples were also examined for Vickers microhardness and the measurements were undertaken across the diameters of the HPT disks using an Akashi MVKE3. Loads of 50 g for 5 and 10 mass%Mg and 200 g for 20, 30 and 40 mass%Mg were applied for 20 s in such measurements.

3. Results and Discussion

3.1 X-ray diffraction analysis

The XRD patterns of Al-Mg alloys with different contents of Mg are shown in Fig. 2. Except the peaks from LaB₆ used as the reference, only peaks from Al-based fcc crystal structure are present for the alloys up to 30 mass% of Mg. However, a trace amount of β-Al₃Mg₂ was detected from the sample with 40 mass%Mg in addition to the fcc peaks. This clearly indicates that the alloying was achieved from the mixed powders and the supersaturation as well as the new phase formation occurred by the process of HPT for the alloy compositions much higher than the equilibrium composition. Close examination of the XRD patterns reveals that there are appreciable shifts of the fcc peak positions and this shifting occurs invariably to lower angles with increasing addition of Mg. Peak broadening is also apparent with increasing addition of Mg. The peak shifting suggests that the lattice parameter was increased due to the supersaturation of Mg in Al and the peak broadening indicates that large strain was generated in the HPT-processed alloys. Figure 3 plots the lattice parameters as a function of equivalent strain for the different compositions. Here, the lattice parameters were measured using the standard Bragg’s law and the equivalent strain, ε, was calculated through the following equation

\[ \varepsilon = \frac{2\pi r N}{\sqrt{3}t} \] (1)

where \( r \) is the distance from the center of disk, \( N \) is the number of revolutions and \( t \) is the thickness of disk. It should be noted that \( r = 4 \) was used in this study and this use provides the lower limit for each disk because the central region with 4 mm in diameter was cut out from each disk for the XRD measurement.

<table>
<thead>
<tr>
<th>Mg content (mass%Mg)</th>
<th>Applied pressure (GPa)</th>
<th>Number of rotation, ( N )</th>
</tr>
</thead>
<tbody>
<tr>
<td>5</td>
<td>2.5</td>
<td>1  5  10  15  20  30  40  50  60  80  100</td>
</tr>
<tr>
<td>10</td>
<td>2.5</td>
<td>1  5  10  15  20  30  40  50  60  80  100</td>
</tr>
<tr>
<td>20</td>
<td>6</td>
<td>1  5  10  15  20  30  40  50  60  80  100</td>
</tr>
<tr>
<td>30</td>
<td>6</td>
<td>1  5  10  15  20  30  40  50  60  80  100</td>
</tr>
<tr>
<td>40</td>
<td>6</td>
<td>1  5  10  15  20  30  40  50  60  80  100</td>
</tr>
</tbody>
</table>
The lattice parameters increases with straining and saturates to the levels determined by the added amount of Mg. This indicates that Mg is solved in the Al matrix by straining through HPT. The lattice parameters at the saturation levels are documented in Table 2 and they are also plotted in Fig. 4 against the Mg content together with other results reported from rapidly quenched alloys and mechanically milled alloys. At the lower content of Mg, there was a steady expansion of the lattice parameter due to the increased amount of Mg and the full dissolution of Mg with straining. Thus, the lattice parameter increases linearly with the Mg content as shown by the thick dotted line in Fig. 4. It should be noted that there is no longer increase in the lattice parameter for the addition of 40% Mg but the saturation level of the lattice parameter is the same as that for the 30% Mg. This is most likely to be due to the formation of β-Al3Mg2 as detected by the XRD analysis. Comparison with other reports suggests that the lattice parameters obtained in this study are consistent reasonably well with those obtained by rapid quenching and mechanical milling. This indicates that HPT can be used as an alternative process for achieving supersaturation of Mg in Al.

### Table 2 Lattice parameters measured from X-ray peaks for each Mg content and numbers of rotation.

<table>
<thead>
<tr>
<th>Mg content</th>
<th>Lattice parameter ( a_0 ) (nm)</th>
<th>Mean grain size (nm)</th>
</tr>
</thead>
<tbody>
<tr>
<td>0 mass% Mg</td>
<td>( N = 40 )</td>
<td>( N/A )</td>
</tr>
<tr>
<td>5 mass% Mg</td>
<td>( N = 50 )</td>
<td>80</td>
</tr>
<tr>
<td>10 mass% Mg</td>
<td>( N = 60 )</td>
<td>65</td>
</tr>
<tr>
<td>20 mass% Mg</td>
<td>( N = 80 )</td>
<td>45</td>
</tr>
<tr>
<td>30 mass% Mg</td>
<td>( N = 100 )</td>
<td>40</td>
</tr>
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<td>40</td>
</tr>
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### 3.2 Transmission electron microscopy

Figures 5 and 6 show (a) bright-field (BF)-TEM images, (b) dark-field (DF)-TEM images and (c) selected area electron diffraction (SAED) patterns for the powder mixtures of Al-5 mass% Mg and Al-40 mass% Mg alloys after HPT processing, respectively. The microstructures consist of many grains having curved and ill-defined grain boundaries and these microstructural features are similar to the earlier observation of an HPT-processed Al-3 mass% Mg alloy prepared by ingot metallurgy. In the present study, TEM observations were also performed on the other compositions and it was found that the microstructural features are the same as the ones shown in Figs. 5 and 6 except that the grain size decreases with an increase in the Mg content. As listed in Table 3, the average grain sizes are 80, 65, 45, 40 and 40 nm for the alloys with Mg contents of 5, 10, 20, 30 and 40 mass%, respectively. However, these grain sizes are smaller by an order of magnitude than those reported earlier on bulk rods of Al-1 mass% Mg and Al-3 mass% Mg alloys processed by ECAP (Equal-Channel Angular Pressing). The average grain sizes for the ECAP-processed Al-1 mass% Mg and Al-3 mass% Mg alloys were ~450 and
Although the grain size tends to be smaller as the addition of Mg increases, the difference in the Mg content is not totally responsible for the difference in the grain size by an order of magnitude. HPT processing for high purity Al powders led to the grain size of 500 nm whereas the grain size was of 1300 nm when HPT was conducted with a bulk form of Al disks. The difference in the grain size is thus not only the presence of excess Mg but also the presence of oxide dispersion arising from the surface layers of powders. Both Mg and oxide dispersion can be good obstacles for dislocation movement and thus, as discussed earlier, they facilitate the accumulation of dislocations to make grain boundary formation easier.

According to the XRD patterns, the precipitation of $\beta$-$\text{Al}_3\text{Mg}_2$ took place when 40 mass% of Mg was added. However, it should be noted that the diffracted beams associated with precipitates were too weak to be detected in the SAED pattern.

### 3.3 Vickers hardness test

The maximum Vickers microhardness, 4 mm away from the center of the sample disk, is plotted in Fig. 7 with the Mg content.
content. The increase of saturated hardness is obvious for the higher content of Mg as shown in Fig. 7. The maximum Vickers microhardness was 40 Hv for pure Al prepared by HPT from Al powders, but it increased gradually, 202 Hv (at 5 mass%Mg), 252 Hv (at 10 mass%Mg), 288 Hv (at 20 mass%Mg), and 292 Hv (at 30 mass%Mg). However, it decreased to 257 Hv at 40 mass%Mg, possibly due to the formation of $\beta$-$\text{Al}_3\text{Mg}_2$.

Combination of solid solution hardening and Hall-Petch hardening mechanisms are expected to take place as shown in Fig. 8. The Al-Mg alloy increased its hardness up to 292 Hv according to the Vickers microhardness test. The microhardness data are slightly better than the Hall-Petch relationship of pure Al$^{22}$ for specimens with Mg content up to 30 mass%, due to the presence of solid-solution hardening in each sample. Their grain sizes were decreased when the content of Mg were increased because of the increase of the number of rotations during the HPT process, namely the amount of strain.

4. Conclusions

(1) Mixed powders of varying additions of Mg in Al were subjected to HPT processing. Supersaturation was attained for up to the Mg content of 30 mass% but there is a formation of $\beta$-$\text{Al}_3\text{Mg}_2$ for the addition of 40 mass%Mg. The lattice parameter increases linearly with the Mg content up to 30 mass%, but remains the same for the 40% Mg addition.

(2) The average grain size decreases from $\sim$80 nm for the addition of 5%Mg to $\sim$40 nm for the Mg content of 30% or more. These grain sizes are smaller by an order of magnitude than those reported on bulk form of Al-1%Mg and Al-3%Mg alloys.

(3) The maximum Vickers microhardness reached almost 7.5 times higher at 30 mass%, 292 Hv, than pure Al 40 Hv. It is apparent that the effects of solid solution and grain refinement contribute to the hardening of the Al-Mg alloys.

Acknowledgements

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