Achieving Microstructural Refinement in Magnesium Alloys through Severe Plastic Deformation

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Equal-channel angular pressing (ECAP) is an excellent processing tool for achieving exceptional grain refinement, typically to the submicrometer level, in a range of pure metals and metallic alloys. Although processing by ECAP is relatively straight-forward when using soft metals, the processing becomes more difficult when using magnesium-based alloys and other similar difficult-to-work materials. This overview examines the procedures that must be adopted for the successful processing of these materials and then describes some of the advanced properties achieved after the successful processing of two magnesium alloys. [doi:10.2320/matertrans.MD200818]

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1. Introduction

The processing of metals through the application of severe plastic deformation (SPD) is now well established for the production of ultrafine-grained materials having exceptional mechanical and physical properties. Typically, the materials produced in SPD have submicrometer or nanometer grain sizes, they exhibit exceptionally high strength and, if the microstructures are reasonably stable at elevated temperatures, they exhibit a potential for use in superplastic forming operations. Although several SPD processing techniques are now available, the two most promising procedures are Equal-Channel Angular Pressing (ECAP) and High-Pressure Torsion (HPT). Furthermore, processing by ECAP is especially attractive because it has the capability of producing relatively large bulk samples.

Many of the early studies of SPD processing were limited to soft pure metals or solid solution alloys where processing by ECAP is relatively easy. However, more recent attention has focused on the processing of difficult-to-work materials such as magnesium-based alloys. Magnesium alloys are now attracting much attention because of their low density, lightweight, and high potential for recycling. These alloys are excellent candidate materials for use in a wide range of applications in the automotive and other transportation industries and in the manufacturing of commercial products. They also exhibit the optimum damping capacities of any known metal so that the alloys are useful for applications where external vibrations must be damped examples include in helicopters and satellites. This wide range of applications has led to the prediction that we are now on the verge of entering “a new age of magnesium” where magnesium alloys will be widely used for the fabrication of numerous parts and components.

The processing of magnesium alloys by ECAP is challenging. Early experiments showed that ultrafine grain sizes were not produced when pure magnesium and an Mg-9% Al alloy were processed by ECAP and an approach was developed whereby a preliminary extrusion step was introduced prior to ECAP so that a majority of the basal (0001) planes became oriented parallel to the extrusion direction. This procedure of “extrusion plus ECAP” was designated EX-ECAP and experiments have shown it works well in producing significant grain refinement in many magnesium alloys. However, despite this success there remains a problem that many magnesium alloys fail during processing through the development of substantial cracking or segmentation. This overview examines this problem in section 2 and in section 3 there is a description of some of the advanced properties that may be attained when magnesium alloys are successfully processed by ECAP.

2. The Development of Procedures for Successfully Processing Magnesium Alloys

To obtain a detailed understanding of the characteristics of the deformation occurring during ECAP when using difficult-to-work materials such as magnesium-based alloys, it is appropriate to make use of finite element modeling (FEM) as described in an earlier report. Following conventional practice, the FEM simulations incorporated the following relationship to describe the material behavior:

\[
\sigma = A\varepsilon^n\dot{\varepsilon}^m
\]

where \(\sigma\) is the effective stress, \(\varepsilon\) is the accumulated effective strain, \(\dot{\varepsilon}\) is the effective strain rate, the exponents \(n\) and \(m\) are the strain hardening coefficient and the strain rate sensitivity, respectively, and \(A\) is a constant. For a direct evaluation of the validity of the FEM predictions, parallel experiments were also conducted using a commercial extruded ZK60 alloy with a composition, in mass%, of Mg-5.5% Zn-0.5% Zr.

In practice the local strain distributions occurring within the billets during ECAP vary as a function of the strain rate sensitivity of the material. This variation is illustrated in Fig. 1 where the angle between the two parts of the channel in the ECAP die is fixed at 90° and the strain levels are plotted pictorially for strain rate sensitivities of (a) 0, (b) 0.2
and (c) 0.4. This modeling shows directly that the billet with the highest strain rate sensitivity of $m = 0.4$ exhibits the highest strain level of 1.6 and this occurs in a relatively narrow region near the bottom surface of the billet. By contrast, the distribution in the local strain is reasonably uniform across the width of the billet when $m = 0$ although the strain level is lower near the bottom surface, increases rapidly to $\sim 1.0$ within a very short region and thereafter remains constant within the central region and the top region of the billet. Inspection shows that the shapes of the strain level contours are different in the front regions of the billets for materials with high and low strain rate sensitivities. Thus, whereas the simulation with $m = 0.4$ in Fig. 1(c) gives strain contours perpendicular to the billet axis in the front section of the billet with the contours corresponding to strain levels of 0.8 and 1.0 displaying a marked curvature towards the central part of the billet, the simulation with $m = 0$ shows contours lying at approximately 45° to the billet axis in the front section and with a small pocket having the highest strain of 1.12 near the front face.

An important factor that governs the ability to process difficult-to-work alloys is the occurrence of cracking within the billet during the pressing operation. This may be evaluated by employing a macroscopic damage criterion to evaluate the formability of materials during the FEM simulations. To understand this approach, it is convenient to make use of the traditional Cockcroft and Latham\(^{15}\) damage model which is based on the following relationship:

$$C = \int_0^{\tilde{\epsilon}_1} \sigma_{\text{T}} d\tilde{\epsilon}$$

where $C$ is a constant corresponding to a critical condition associated with fracture, $\tilde{\epsilon}$ is the effective strain, $\sigma_{\text{T}}$ is the maximum component of the tensile stress at any point within the billet and the integral is evaluated from zero strain to the final effective strain, $\tilde{\epsilon}_1$. The integral shown in eq. (2) describes the tensile energy per unit volume and this relationship demonstrates that fracture will occur when the value of $C$ reaches a critical value. For convenience, the relationship in eq. (2) is generally modified into a normalized form in which the maximum principal tensile stress is normalized by the equivalent stress to give:\(^{16}\)

$$C_N = \int_0^{\tilde{\epsilon}_1} \frac{\sigma_{\text{T}}}{\sigma} d\tilde{\epsilon}$$

(3)

where $\sigma$ is the effective stress.

The approach with the Cockcroft-Latham damage criterion employs the concept that shearing occurs within the billet when it passes through the theoretical shear plane at the point of intersection of the two parts of the channel. This is demonstrated in Fig. 2 where the three columns relate to channel angles, $\Phi$, of 90°, 110° and 135° and the three rows correspond to strain rate sensitivities of (a) 0, (b) 0.2 and (c) 0.4. Within each set of illustrations, the contour lines delineate consecutive points experiencing constant levels of strain rate where the predicted strain rates are marked on the diagrams in s\(^{-1}\). An important conclusion from Fig. 2 is that the deformation zones and the strain rates depend both upon the channel angle within the die and the value of the strain rate sensitivity. Thus, the deformation zones are wide when pressing a material with a high strain rate sensitivity and these widths are reduced when the value of $m$ is decreased. Furthermore, the strain rates at all angles are lowest in the billets with $m = 0.4$ but become higher as the strain rate sensitivity is reduced.

The normalized Cockcroft-Latham damage distributions are shown in Fig. 3 based on simulations involving strain rate sensitivities of (a) 0, (b) 0.2 and (c) 0.4 for the three different die angles of 90°, 110° and 135°. The contour lines in Fig. 3 correspond to the values predicted for $C_N$ using eq. (3) and they represent the local level of damage occurring in each billet. It is readily apparent from Fig. 3 that the strain rate sensitivity affects not only the precise level of damage occurring within the billet but also the distribution of this damage. For a material having $m = 0$ as shown in Fig. 3(a), there is a constant region of damage along the billet and the highest levels of damage, with values of $C_N \approx 0.28$–0.32, occur between the central regions and the lower edges of the billets. Figures 3(b) and (c) show there are higher damage levels when the strain rate sensitivity is increased and also the region of maximum damage is displaced from the middle of the billet when $m = 0$ to the top surface when $m = 0.4$. The damage levels for the billet with $m = 0.4$ are high and up to a maximum of $\sim 0.49$ for the die with a channel angle of $\Phi = 90°$. Similar damage levels are also visible on the top surface for the die when the channel angle is $\Phi = 110°$ although the damage is significantly lower when $\Phi = 135°$.

It follows from these and other similar calculations that the overall level of damage is reduced when the channel angle is increased. This trend is clearly shown by plotting the maximum value of damage, quantified by the values of $C_N$ along the middle cross-sections of the billets, as a function of the channel angle within the die as given in Fig. 4.

A direct experimental verification of the predictions of the FEM simulation is shown in Fig. 5 where two billets of the ZK60 alloy were annealed to give an initial strain rate sensitivity of $m \approx 0.2$ and then processed by ECAP. The side profiles of the billets are shown in Fig. 5 where both billets were pressed through one pass at a temperature of 473 K.
using dies having channel angles of either 90° for the upper billet or 110° for the lower billet. It is apparent that the upper billet exhibits very regular segmentation which occurs throughout the length of the billet and was initiated at the upper surface of the billet. By contrast, the lower billet demonstrates a successful pressing without the introduction of any significant damage when using a die with a channel angle of 110°. These results are therefore consistent with the FEM predictions and they provide strong support for using this type of approach in evaluating the optimum conditions for achieving successful processing and significant grain refinement by ECAP.

3. Examples of Advanced Properties Achieved in Magnesium Alloys Processed by ECAP

Experiments were conducted on a Mg-0.8 mass% Li two-phase alloy to evaluate the potential for achieving superplastic properties after ECAP. Initial experiments showed that the alloy cracked after a single pass when pressing at room temperature using a die with a channel angle of 90°. Accordingly, the channel angle was increased to 135° where it was possible to press through ten passes at room temperature without experiencing any visible cracking. For an ECAP die with a channel angle of 135° and an arc of curvature of Ψ = 20°, it can be shown that the imposed strain is equal to ~0.5 on each separate pass through the ECAP die where this is one-half of the strain generally imposed in a single pass when using a conventional die with a channel angle of Φ = 90°.

An example of the experimental results with the Mg-8% Li alloy are shown in Fig. 6 where the measured elongation to failure is plotted against the initial strain rate for tests conducted under conditions of a constant rate of cross-head displacement. These results demonstrate the very significant differences that may be recorded in the same alloy when processing in different ways. Thus, the cast alloy having an average phase width of ~60–70 μm exhibits only low elongations up to a maximum of ~200%. By contrast, the alloy which was cast and extruded using an extrusion temperature, T_{ex}, of 373 K and an extrusion speed, r_{ex}, of 1 mm s⁻¹ contained a microstructure with an average phase...
size of \( \sim 3-5 \mu m \) and in this condition the maximum elongation was \( \sim 630\% \) at the lowest strain rate of \( 1.5 \times 10^{-4}\) s\(^{-1}\). Finally, the cast alloy which was extruded and then processed by ECAP through four passes using an ECAP die with a channel angle of 135°, corresponding to an imposed strain of \( \sim 2 \), contained an average phase size of \( \sim 1-3 \mu m \) and in this condition a maximum elongation of \( \sim 1780\% \) was achieved at the slowest experimental strain rate. These results demonstrate the influence of additional grain refinement in promoting higher elongations at the lowest strain rates. Nevertheless, it is also apparent from Fig. 6 that all processing conditions lead to similar low elongations when testing to failure at strain rates above \( \sim 10^{-2}\) s\(^{-1}\), where this corresponds to the conventional transition to a flow region where an intragranular dislocation creep mechanism is dominant\(^{19,20}\).

The results documented in Fig. 6 show that exceptional superplastic properties may be achieved at elevated temperatures even in a magnesium-based alloy. Furthermore, the
recorded maximum elongation in Fig. 6 is very high even by comparison with the many other reports of superplasticity in aluminum and other alloys processed by ECAP: a complete tabulation of these various results is given elsewhere.21) An example of an exceptionally high superplastic ductility is shown in Fig. 7 for a ZK60 alloy containing, in mass%, Mg-5.5% Zn-0.5% Zr.22) This alloy was received in an extruded condition with an initial grain size of $\sim 2.9 \mu m$ and it was then processed by ECAP at 473 K using a die with a channel angle of $\Psi = 90^\circ$ and an arc of curvature of $\Psi \approx 20^\circ$ using processing route BC in which the billets are rotated by $90^\circ$ in the same sense between each consecutive pass.23) The tensile tests were conducted at the same temperature of 473 K using an initial strain rate, $\dot{\varepsilon}$, of $1.0 \times 10^{-4} s^{-1}$. In Fig. 7 the upper specimen is untested and the other specimens were processed through different numbers of passes from 1 to 6 passes and then pulled to failure. It is readily apparent that all of these specimens exhibit elongations larger than 500% which corresponds to the onset of true superplasticity. Furthermore, the specimen processed through 2 passes exhibits an exceptionally high superplastic elongation of 3050%. This elongation is the highest recorded in any magnesium alloy processed under any conditions with or without ECAP. In addition, a comparison with the detailed tabulation of superplastic data presented elsewhere21) shows this ductility is also the highest recorded in any of the many alloys processed by ECAP to date including the highly ductile aluminum-based alloys.

An apparently unusual result in Fig. 7 is that the maximum superplastic elongation is achieved in this alloy after processing through only 2 passes. After 3 and 4 passes the ductilities remain unusually high but they are lower than in the specimen processed through only 2 passes. This result tends to be unexpected because earlier tests on an aluminum alloy demonstrated that higher ductilities are generally achieved after larger numbers of passes through the ECAP die.24) It is appropriate, therefore, to examine the separate flow characteristics of the samples taken through 2 and 6 passes to give elongations of 3050% and 930%, respectively.

The results may be understood by examining the role of strain hardening which controls the overall elongations to failure achieved at this relatively low testing temperature. Strain hardening serves a dual purpose under these experimental conditions because it both balances the reduction in the cross-sectional area and stabilizes the load during the tensile flow. Figure 8 shows the plots of true stress versus true strain for the two specimens taken through $N = 2$ and 6 passes, where $N$ represents the number of passes in ECAP. Following the early analysis by Hart,25) it is assumed that plastic instability occurs at the point at which the rate of variation of the strain rate at the smallest cross-section of the specimen first starts to increase. This condition may be written in explicit form as:

$$\sigma \leq \frac{d\sigma}{d\varepsilon} \left(1 - \frac{1}{m}\right)$$

where $\sigma$ and $\varepsilon$ are the true stress and true strain, respectively. In Fig. 8, the parameter on the right of the Hart relationship in eq. (4) is plotted on the right-hand axis using the experimentally determined values for the strain rate sensitivity of $m = 0.44$ after 2 passes and $m = 0.40$ after 6 passes. The point of instability is then given by the point where these
two curves intersect and these points are delineated by the two vertical lines. It is apparent that the higher flow stress and the lower value of \( m \) leads to a relatively low strain for the onset of plastic instability in the specimen processed through 6 passes. By contrast, the higher flow stress and lower strain in the specimen processed through 2 passes leads to a higher strain for plastic instability in this specimen. After the onset of plastic instability, there is also a larger accrued strain in the specimen processed through 2 passes and this is a direct consequence of the higher strain rate sensitivity in this specimen. Thus, the onset of plastic instability is consistent with the Hart criterion\(^{25} \) given by eq. (4).

4. Discussion

The successful processing of magnesium alloys by ECAP requires a careful consideration of the experimental conditions. A step of extrusion prior to ECAP is beneficial in reducing the grain size to a level that is more amenable to easy refinement to produce a true ultrafine-grained microstructure. But even for extruded magnesium alloys, the occurrence of cracking and segmentation during the processing operation may hinder the ability to achieve successful results. These problems may be overcome by using finite element modeling to evaluate the local strain distributions and the inherent strain rates involved in the processing operation. The incorporation of an appropriate damage factor such as the Cockcroft-Latham criterion\(^{15} \) permits the development of criteria that may be used to successfully process many difficult-to-work alloys. For example, although the present paper deals exclusively with magnesium-based alloys, an increase in the channel angle \( \Phi \) within the ECAP die has been used to successfully press other hard materials such as tungsten\(^{26} \) and titanium.\(^ {27} \)

When magnesium-based alloys are processed successfully by ECAP, mechanical testing may produce evidence for exceptional properties that are not usually achieved in magnesium alloys. Two examples are shown in Figs. 6 and 7 where extremely good tensile ductilities are evident in tests conducted at elevated temperatures. Furthermore, it is known that improved results are often achieved by ECAP when using an experimental facility which imposes a back-pressure during the processing operation,\(^ {28} \) but the present results, including the record tensile elongation shown in Fig. 7, were attained without the imposition of a back-pressure.

5. Summary and Conclusions

(1) There are unusual difficulties associated with the processing of magnesium alloys by equal-channel angular pressing (ECAP) because of the relatively easy development of cracking and/or massive segmentation of the billets.

(2) It is shown that the processing procedure may be optimized by using finite element modeling to delineate the strain distributions and the internal damage introduced during the processing operation. Using this approach, it is possible to achieve excellent properties after successful processing by ECAP.

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