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LETTER

Principles of grain refinement in magnesium alloys processed by equal-channel angular pressing

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Processing through the application of severe plastic deformation (SPD) provides a useful tool for introducing very significant grain refinement in bulk solids [1]. Although several SPD techniques are now available, processing by equal-channel angular pressing (ECAP) is especially attractive because it is easily scaled-up for use with large samples [2]. Numerous experiments have shown that processing by ECAP produces ultrafine-grain structures in a range of fcc metals, including Al and Cu, and this led to the development of a model for grain refinement based on the formation of an elongated array of subgrains or cells in the first pass of ECAP, the further development of elongated arrays in subsequent passes and the evolution of this structure into an array of equiaxed grains separated by high-angle grain boundaries [3]. In practice, there is excellent agreement between this model of grain refinement for fcc metals [3], the shearing patterns predicted in ECAP processing [4] and experimental observations of the microstructural evolution in Al single crystals [5, 6] and polycrystalline Al [7, 8] processed by ECAP.

Despite the success with fcc metals, early experiments on pure magnesium and a magnesium-based alloy revealed significant difficulties in achieving grain refinement by ECAP due to a necessity to press at high temperatures in order to avoid premature cracking [9]. Subsequently, it was shown that ultrafine grains may be achieved by introducing a two-step procedure in which the grain size is initially reduced by extrusion prior to ECAP [10] and this process of extrusion and ECAP, termed EX-ECAP, was used successfully for the production of ultrafine-grained structures in several Mg alloys [11–17]. Other processing procedures were also proposed for reducing the pressing temperature in magnesium alloys including by using a back-pressure [18, 19], increasing the channel angle within the ECAP die [20, 21], reducing the pressing speed [22] and using sequential reductions in the pressing temperature with subsequent passes through the die [23, 24].

The evidence available to date suggests there are very significant differences between the grain refinement process in magnesium alloys and in fcc metals such as aluminum. Whereas the process of grain refinement is insensitive to the pressing speed in pure aluminum and an Al–1% Mg solid solution alloy [25], experiments on the AZ31 magnesium alloy show the grain size is reduced by pressing at a faster speed [24]. Furthermore, there is evidence in the AZ31 and ZK60 alloys for the formation by ECAP of either a homogeneous distribution of equiaxed grains [13, 26] or a bimodal distribution where coarse grains are surrounded by arrays of much smaller grains [19, 26]. The present investigation was initiated to investigate these differences and specifically to develop a model for grain refinement when magnesium alloys are processed by ECAP.

Experiments were conducted using two different commercial magnesium alloys, AZ31 (Mg–3% Al–1% Zr) and ZK60 (Mg–5.5% Zn–0.5% Zr), where these two alloys were selected because extensive information is available on the microstructures produced in these alloys both with and without ECAP processing. The alloys were received in an extruded condition with initial grain sizes of ~9.4 and ~2.9 μm for AZ31 and ZK60, respectively. The alloys

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were processed by ECAP at a temperature of 473 K using a die with an angle of 110° between the two parts of the channel and an outer arc of curvature of ∼20°: these angles produce a strain of ∼0.8 on every pass through the die [27]. The pressing was conducted at a speed of ∼2 mm s^{-1} with the billets rotated by 90° in the same sense between each pass using processing route B_c [28]. Following pressing, small samples were cut from the billets, mounted, ground on abrasive papers, polished with 0.05 μm alumina powder and then etched in a solution of picric acid, acetic acid, distilled water and ethanol to reveal the grain boundaries. Optical microscopy was used to examine the microstructures and the average grain sizes were measured using the mean linear intercept method.

An examination of many samples pressed under different conditions led to the conclusion that a critical grain size, d_c, is needed in order to achieve a homogeneous distribution of equiaxed grains following processing by ECAP. Accordingly, the principles of grain refinement in magnesium alloys may be represented by the schematic illustrations in Fig. 1 where the left column denotes the initial condition for grain sizes larger and smaller than d_c and the right column shows the resultant microstructures after processing through one pass by ECAP.

In Fig. 1a there is an initial relatively coarse structure where $d > d_c$ and processing by ECAP leads to the formation of new grains along the initial grain boundaries in a necklace-like arrangement. However, the initial grain size is now sufficiently large that, as illustrated in Fig. 1b, a bimodal structure is produced wherein the new grains occupy only a fraction of the entire volume of the material and there are areas in the centers of the larger grains which are not consumed by the formation of these smaller grains. Conversely, Fig. 1c shows an initial fine structure where $d < d_c$ and in this situation processing by ECAP leads again to the formation of new grains along the original grain boundaries but this now produces a homogeneous microstructure of fine grains which, as shown in Fig. 1d, occupies the entire volume of the sample.

Figure 2 shows an example for the extruded AZ31 alloy where (a) is the initial grain structure and (b) gives the microstructure after one pass. The initial structure has an average grain size of ∼9.4 μm but it is evident that some relatively coarser grains are also present. After one pass the grain size distribution is bimodal such that there are large areas of new fine grains but there are also discrete and well-defined large grains that appear to correspond to the inner core areas of some of the larger initial grains: the

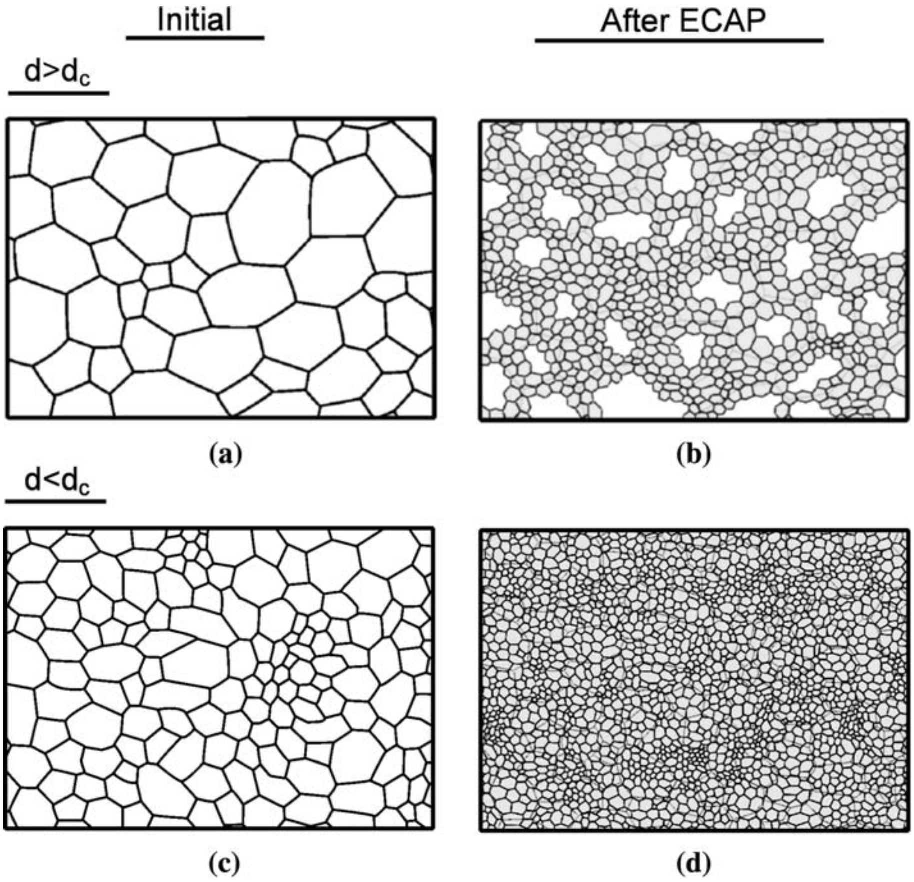

Fig. 1 A model for grain refinement in Mg alloys processed by ECAP from a coarse initial structure with an average grain size larger than the critical value d_c (*upper row*) and a fine initial structure with an average grain size smaller than the critical value d_c (*lower row*): the *left column* shows the initial microstructures prior to ECAP and the *right column* shows the microstructures after one pass

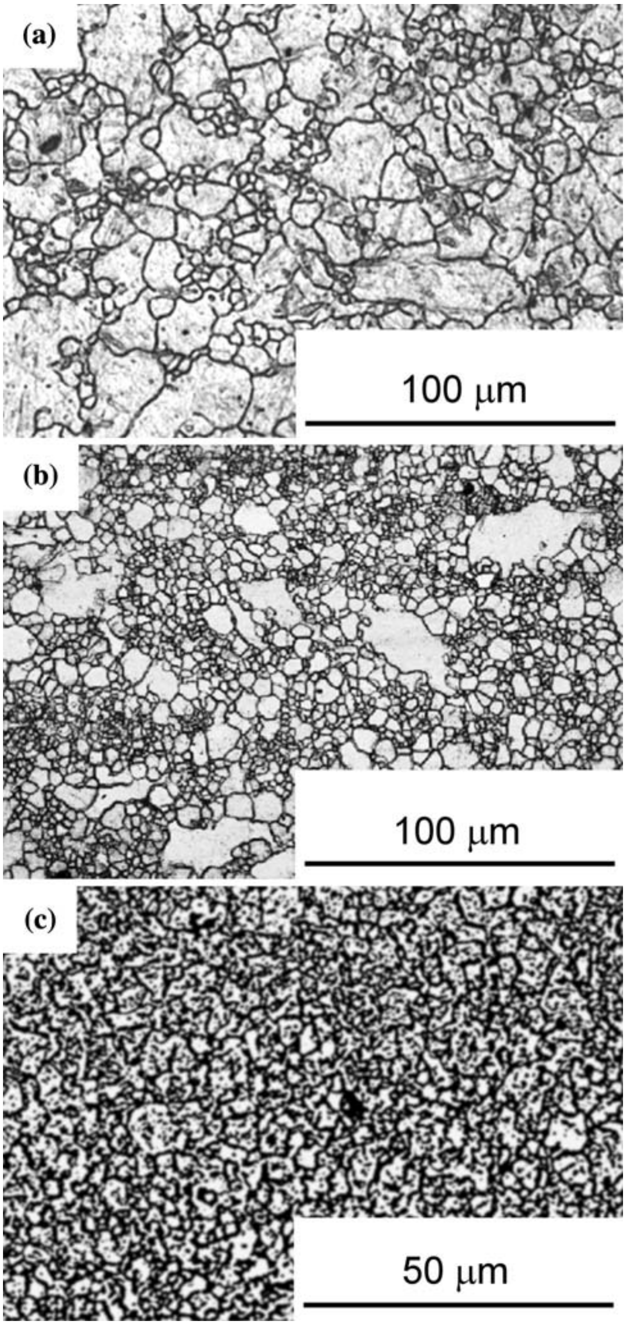

Fig. 2 Representative grain structures in the AZ31 alloy in **a** the extruded condition prior to ECAP, **b** after one pass of ECAP and **c** after six passes of ECAP

microstructures in Fig. 2a, b correspond to Fig. 1a, b, respectively, in the schematic illustration. An important additional observation is given in Fig. 2c which shows the microstructure of the alloy after six passes where the bimodal condition has now evolved to ultimately form a homogeneous distribution of fine grains with a measured average size of ~1.6 μm. These photomicrographs demonstrate that the bimodal grain distributions frequently reported in magnesium alloys processed by ECAP are transitional in nature and they may evolve into regular arrays of fine grains if the processing is continued through a sufficiently large number of passes.

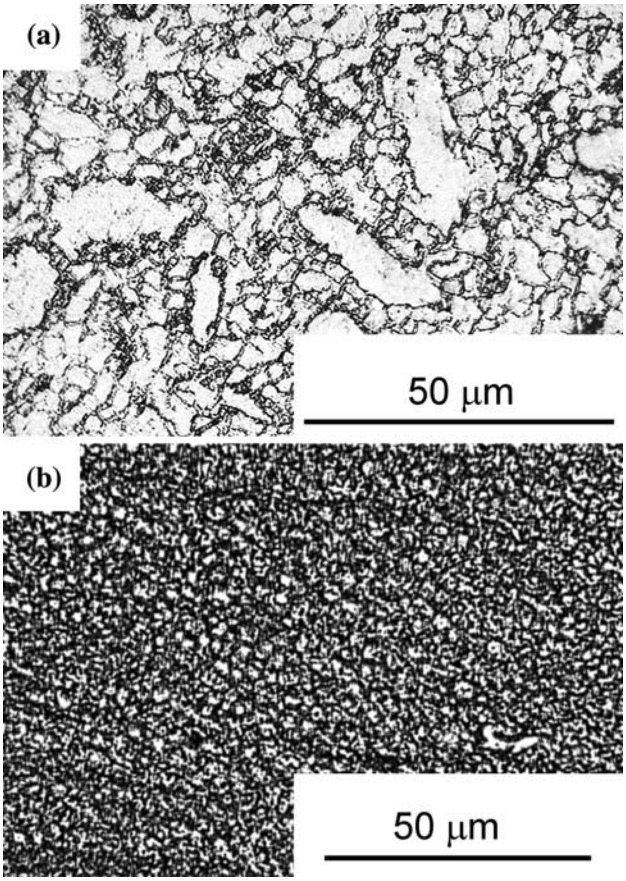

Fig. 3 Representative grain structures in the ZK60 alloy in **a** the extruded condition prior to ECAP and **b** after one pass of ECAP

A second example is given in Fig. 3 for the ZK60 alloy where (a) shows the initial extruded condition where the average grain size is ~2.9 μm and (b) shows the formation after a single pass of a homogeneous distribution of ultrafine grains with an average size of ~0.8 μm: these microstructures correspond to Fig. 1c, d in the schematic illustration. Thus, for the extruded ZK60 alloy the initial grain size produced by extrusion is sufficiently small that it is possible to form a homogeneous distribution of grains in only one pass.

Grain refinement in magnesium alloys is therefore characterized by the nucleation of fine grains along the pre-existing grain boundaries, where this nucleation is attributed to the development of stress concentrations at the boundaries and the subsequent activation of both basal and non-basal slip processes [29]. This is consistent with the conclusion that the production of homogeneous three-dimensional microstructures in magnesium alloys requires the activation of both non-basal and basal slip and with experimental observations showing that non-basal slip is activated more easily in the interiors of the grains of magnesium alloys when the grain size is reduced [30].

The model for grain refinement depicted schematically in Fig. 1 is consistent with a number of experimental observations recorded from the processing of magnesium alloys by ECAP, hot compression and torsion.

First, in order to successfully process magnesium alloys by ECAP under conditions leading to homogeneous ultrafine-grain structures, it has been established that it is necessary to introduce a step of intermediate grain refinement [10] where this is needed to produce conditions that avoid billet failure and lead to the accommodation of deformation throughout the volume of the material. The model depicted in Fig. 1 explains the excellent homogeneous grain refinement achieved with the EX-ECAP process when extrusion is used to produce an average grain size that is smaller than the critical size required for homogeneous refinement by ECAP at the processing strain rate and temperature. The model also explains the good results observed when pressing Mg alloys by ECAP with an initial high temperature and subsequent lower temperatures because the initial processing at high temperatures produces an intermediate grain refinement which is then suitable for processing at lower temperatures.

Second, the model is consistent with the reports of new grains forming along the original grain boundaries in the hot compression testing of the AZ31 [31] and Mg–0.8% Al [32] alloys and with reports of a necklace-like appearance of new grains in several magnesium-based alloys tested in hot compression [33, 34] and hot torsion [35]. For the experiments in torsion [35], it was concluded specifically that, as in the present model, a homogeneous grain structure is attained only if the initial structure is sufficiently fine that the newly formed grains are able to impinge on the new grains developing on the opposite side of the grain.

Third, there are reports of the development of either bimodal or homogeneous grain structures when using ECAP with magnesium alloys but these differences arise because of variations in the initial grain sizes and/or the pressing conditions. For example, an AZ31 alloy with an extruded grain size of 2.5 μm was successfully processed by ECAP at 473 K using a die with a channel angle of 110° and no back-pressure and this produced a homogeneous grain size of ~0.7 μm [13]. By contrast, a cast AZ31 alloy with an initial grain size of 640 μm was pressed at 423 K using a die with a channel angle of 90° and with an imposed back-pressure to produce a bimodal array of coarse (16.5 μm) and fine (0.5 μm) grains. Thus, these two conditions correspond directly to the models shown in Fig. 1. Furthermore, the cast alloy yielded a homogeneous microstructure of fine grains, with an average size of ~2.5 μm, when the temperature of ECAP was increased to 473 K. This result demonstrates that the critical grain size, d_c, is not defined uniquely for any selected alloy but rather it is dependent upon the pressing conditions such that d_c becomes larger, and homogeneous structures are attained more easily, when the pressing temperature is increased to aid in the development of the non-basal slip processes.

Fourth, although earlier experiments on pure Al and an Al–1% Mg alloy demonstrated there was no influence of the pressing speed on the equilibrium grain size produced by ECAP [25], recent experiments on the AZ31 alloy showed that smaller grains are produced when the speed of processing by ECAP is increased [24]. Again, this is consistent with the model because higher processing speeds impose larger stresses and thereby facilitate the occurrence of non-basal slip and the easier development of a more refined microstructure.

Finally, it should be noted that a very recent report described a model for grain refinement in Mg alloys processed by ECAP where shear bands and dislocation entanglements form within the grains and lead directly to a refined grain structure [36]. However, this model fails to account for the formation of bimodal grain structures, it does not satisfactorily address the experimental observations of grain refinement as recorded in Fig. 2 and it is not consistent with the observations of a necklace-like formation of new grains along the original grain boundaries.

In summary, there is a significant difference in the nature of grain refinement by ECAP in fcc metals and magnesium alloys. A model is presented to describe the mechanism of grain refinement in magnesium alloys based on the formation of new grains along the original grain boundaries in a necklace-like manner. There is a critical grain size, d_c, which delineates whether the pressed microstructure is homogeneous or bimodal. In practice, bimodal grain distributions are transitional in nature and they may evolve into homogeneous grain distributions with increasing numbers of passes. The proposed mechanism is consistent with experimental results on the pressing of two magnesium alloys and with extensive data in the literature on the processing of magnesium alloys in hot compression and hot torsion.

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