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The Effect of Warm Equal Channel Angular Extrusion on Ductility and Twinning in Magnesium Alloy ZK60

Rimma Lapovok[1], Peter F. Thomson[1], Ryan Cottam[1,*] and Y. Estrin[2]

[1]*School of Physics and Materials Engineering, Monash University, Clayton, Vic, 3800, Australia*
[2]*IWW, TU Clausthal, Agricolastr. 6, 38678 Clausthal-Zellerfeld, Germany*

Tensile ductility of magnesium alloy ZK60 pre-strained by warm equal channel angular extrusion (ECAE) was investigated at 300°C. It was shown that a significant ductility (262%) is achievable with this technique for a fairly high strain rate of $3 \cdot 10^{-3}\,s^{-1}$. The effect of pre-straining by warm ECAE on twinning under room temperature deformation was also investigated. It was shown that a bi-modal grain structure, whose characteristics are determined by the ECAE route, number of passes and temperature, has a strong effect on the propensity for twinning and mechanical properties, including ductility.

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1. Introduction

It is generally believed that extreme grain refinement that can be achieved by severe plastic deformation may lead to enhanced ductility.[1)] However, room temperature ductility of a number of nanostructured metallic materials was found to be extremely poor - the more so, the smaller the grain size.[2)] It was shown recently[3)] that premature necking in nano-structured fcc materials, such as copper, is, indeed, promoted at room temperature when grain size decreases. The stabilising effect of increased strain rate sensitivity of the flow stress owing to the increased role of the diffusion controlled mechanism of plasticity turns out to be outstripped by a concomitant loss of strain hardening. Zhang *et al.*[4)] found the dependence of the room temperature ductility of Zn (taken here as representative of hcp metals) on grain size d was similar to that found in fcc metals, such as Cu. Ductility at room temperature was shown to increase to 110% as grain size decreased to 238 nm. However, room temperature ductility was shown to deteriorate dramatically with further decrease of the grain size (down to 23 nm) — in accord with the general trend reported by Koch.[2)]

The situation with high temperature ductility is different. Here — mainly due to grain boundary sliding — the effect of grain refinement on ductility, particularly due to severe plastic deformation[5)] is favourable. However, a detailed understanding of the effect of severe plastic deformation on the microstructure of Mg alloys and the resulting tensile ductility is far from having been achieved. The work of Agnew *et al.*[6)] and Chuvil'deev *et al.*[7)] is particularly relevant in this context, as it addresses just that question. Agnew *et al.* found that the microstructure of hot-rolled ZK60 plate was heterogeneous, some larger grains (~50 µm) being embedded in an array of smaller grains (<10 µm). Little change occurred after one pass of equal channel angular extrusion (ECAE) at 300°C, but after two or more passes, the microstructure was more homogeneous, consisting of a matrix of fine grains (~2.5 µm) in which a few larger grains (>10 µm) were embedded. The ductility of ZK60 improved by a factor of 2-3 after two or more passes. The increase in the strain rate sensitivity index, m, with the number of passes of ECAE suggested that grain boundary sliding accommodated by grain boundary diffusion may have occurred, cf. Zhang *et al.*[4)] Chuvil'deev *et al.*[7)] established routes for processing by ECAE to produce optimal superplastic behaviour in analogues of ZK60 and AZ31 Mg alloys. A grain size of 1 µm was associated with low temperature superplastic behaviour of Mg alloys, including ZK60, which was manifested by 810% elongation to failure at 250°C under a strain rate of $3 \times 10^{-3}\,s^{-1}$ and a 2-3 fold increase in room temperature ductility to 45%. Referring to reports that the distribution of basal planes plays an important role in enhancing room temperature ductility of Mg alloys by promoting basal slip, Mukai *et al.*,[8)] annealed extruded ZK60 to produce grain sizes of 4 µm and 120 µm. Quasi-static room temperature ductility was greatly increased for the smaller grain size (from 20 to 35%). Failure in the coarse-grained material occurred by coalescence of cracks along twin and grain boundaries, while no macroscopic twins occurred in the fine-grained material. The same authors[9)] subjected ZK60 to eight passes of ECAE at 160°C and annealed the product to produce equiaxed grains as small as 1.4 µm. The strain rate sensitivity index, m, found from strain rate jump tests at 160°C, was approximately 0.5 at the mid-range of strain rates (about $5 \times 10^{-3}\,s^{-1}$), which they interpreted to indicate grain boundary sliding was the dominant mechanism of deformation, confirmed by SEM, and this remained so throughout the range of strain rates investigated. Under optimal conditions at 160°C, the ductility of ZK60 processed by ECAE reached 600% and 1083% before and after annealing, respectively. It was suggested that equilibrium grain boundaries are necessary in Mg alloys for attaining low temperature superplasticity.

The previous evidence suggests that tensile ductility at elevated temperatures of Mg alloys, and ZK60 in particular, is strongly enhanced by grain refinement down to the micrometer range. Ductility is favoured by basal slip, promoted by suitable mechanical pre-treatment, most notably ECAE, to re-orient basal planes suitably. The absence of

*Graduate Student, School of Physics and Materials Engineering, Monash University, Clayton, Vic, 3800, Australia

twinning in ultra fine grained Mg alloys also favours ductility. The observed stress exponent in the law relating stress and strain rate suggests that the dominant mechanism of deformation in Mg alloys at room temperature and elevated temperatures is dislocation controlled when grain size exceeds $\sim$50 µm, whereas grain boundary sliding becomes increasingly important at grain sizes on the nano-scale.

The literature reveals a lack of quantitative agreement on the effect of grain size and on mechanical properties in hot and cold deformation. Much of this may be traced to differences in the parameters of processing by ECAE, notably the route (orientation between passes) chosen. For this reason, one of the main aims of the present work was a consistent investigation of the effect of the ECAE process parameters (including route and number of passes and the history of temperature and ram velocity) on the tensile ductility of alloy ZK60 at room temperature and moderately elevated temperatures (up to 300°C). Another goal was to study the occurrence of twinning in relation to grain size after warm ECAE, to elucidate its role in the ductility of fine-grained Mg alloys.

2. Experimental

Rectangular specimens of $20 \times 10\,\text{mm}^2$ cross-section and 70 mm length as well as cylindrical specimens with $10\,\text{mm}^2$ diameter and 70 mm length were machined from a continuously cast billet of ZK60 (Mg-4.95%Zn-0.71%Zr). The samples were homogenised for four hours at 460°C in alumina powder, to reduce surface oxidation. The homogenisation conditions were chosen on the basis of previous time-temperature homogenisation experiments to avoid incipient melting and possible grain growth. As a result of homogenisation, grain boundary precipitates were dissolved, leaving behind regions rich in zirconium. The initial microstructure consisted of equiaxed grains with average diameter of 45.5 µm and isolated zinc-zirconium particles dispersed within the grains or occurring in clusters.

The rectangular samples were subjected to 8 passes of ECAE (Routes A and C) with a 120° die at a ram speed of 0.5 mm/s. Cylindrical samples were subjected to 6 passes of ECAE (Route B) with a 90° die at a ram speed of 0.5 mm/s. In some experiments, a ram speed of 50 mm/s was also used. The test temperatures were 200, 250 and 300°C. Ductility measurements on tensile specimens machined from ECAE-processed material were carried out at 300°C at two different strain rates ($3 \times 10^{-3}\,\text{s}^{-1}$ and $3 \times 10^{-2}\,\text{s}^{-1}$) using an Instron 1341 testing machine.

Microstructures formed by warm ECAE were investigated with an Olympus JSM-840 optical microscope and a transmission electron microscope Philips CM20.

3. Experimental Results

3.1 Evolution of microstructure under warm ECAE

For all three routes and all three temperatures employed, warm ECAE with a constant ram velocity of 0.5 mm/s for all passes resulted in a bi-modal microstructure with two populations of grains distinctly different in size and morphology. Large elongated grains were embedded in regions of small, equiaxed recrystallised grains. The microstructures obtained after eight ECAE passes at different temperatures are shown in Fig. 1 (Route A), Fig. 2 (Route B), and Fig. 3 (Route C), along with the initial microstructure (Fig. 1(a)). As Route B deformation was performed with a 90° die (and not the 120° die used in the rest of the extrusions), the comparison of results is based on the accumulated *equivalent strain*, rather than the number of ECAE passes. Thus, six passes through 90° die are comparable to eight passes through 120° die with equivalent strain equal to about 6.9. It should be noted that the amount of strain introduced by one pass has a significant effect, especially as a trigger of recrystallisation during subsequent heating for the next pass.

For all three routes and at all three temperatures, an overall grain refinement was obtained upon ECAE. The grain size distribution was shifted towards a larger fraction of small recrystallised grains when processing temperature was increased. The population of small grains had the biggest grain size after ECAE performed at 300°, especially for Route B for which grain growth was observed, cf. Fig. 2(d).

Due to a pronounced bi-modality in the grain size distribution, the average grain size of large and small grains could be determined separately, using the linear intercept method. The average grain size (Fig. 4) depends on temperature, the ECAE route and the amount of strain introduced in each pass as well as the total strain. A general trend observed is that the area fraction of large grains and their average diameter decrease with temperature and the number of passes for all routes. This tendency was more pronounced for Route A, as for Route C the scatter in the grain size measurement was greater due to grain rotation. The intensive grain refinement for Route B is explained by more severe deformation introduced in each pass of ECAE through the 90° die. It was observed that dynamic recystallisation set in earlier, *i.e.* required a smaller number of ECAE passes, at higher temperatures or larger strain per pass. The volume fraction of dynamically recrystallised grains also increased with temperature.

The degree of grain refinement increased with the number of ECAE passes and/or with the amount of strain introduced per pass. Due to severe deformation, the mean size of large grains (in the transverse cross-section) dropped from 45.5 µm to 26.7 ± 2.5 µm for Route A, to 19.3 ± 7.5 µm for Route B and to 29.7 ± 4.1 µm for Route C. Figure 4 shows a weak dependence of the average grain size on the temperature of warm ECAE, except for Route B. A new population of small recrystallised grains with the average diameter ranging from 2 µm for ECAE at 200°C to 4.8 µm for ECAE at 300°C was also observed. The area fraction of large grains was found to be smaller for higher ECAE temperatures, a very small number of large grains being retained after pressing at 300°C.

We have also experimented with the ECAE schedules using *two-stage* Route B ECAE. After a certain number (usually two) of ECAE passes, the temperature of the process, and in most cases also the ram velocity, were changed. Various combinations of the process parameters in the two stages were tried out in order to find the conditions leading to the smallest average grain size. The experimental schedules and the resulting average grain size for the fine

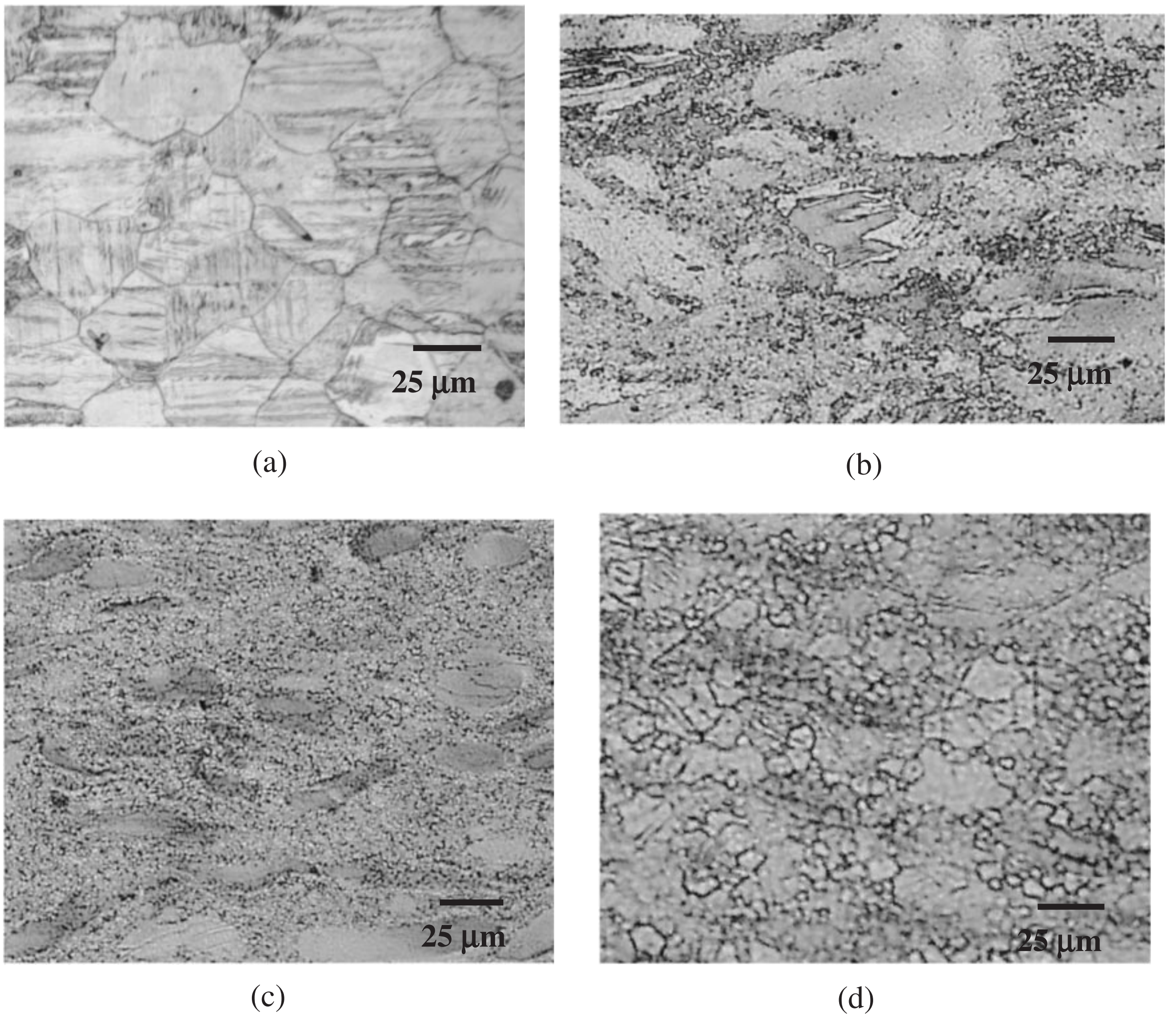

Fig. 1 Microstructures of specimens deformed by warm ECAE vis-à-vis the initial one (Route A, 120° die) (a) initial, (b) after 8 passes at 200°C, (c) after 8 passes at 250°C, (d) after 8 passes at 300°C.

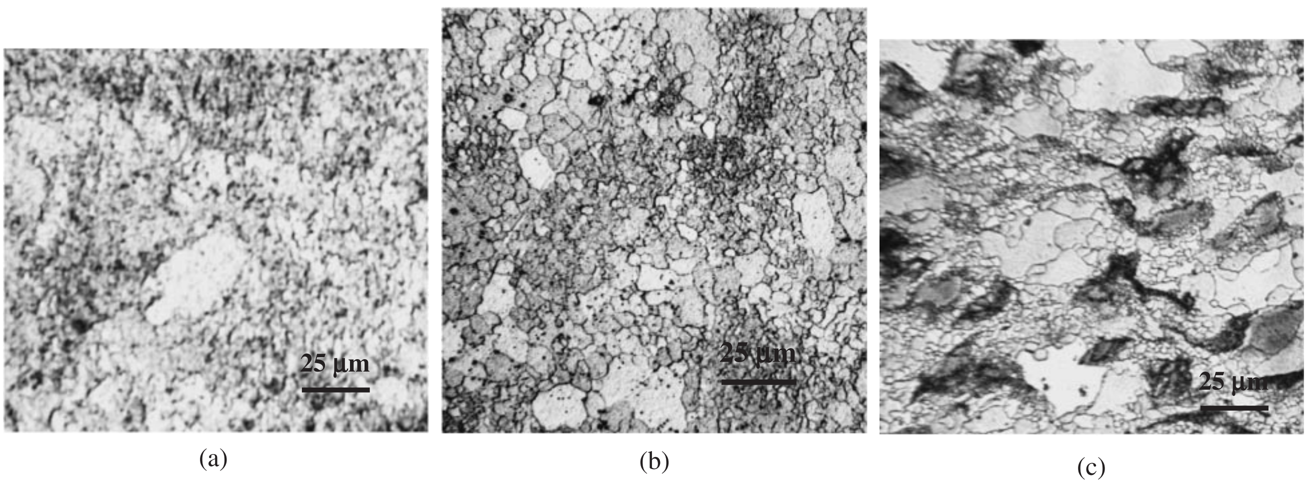

Fig. 2 Microstructures of specimens deformed by warm ECAE (Route B, 90° die) (a) after 6 passes at 200°C, (b) after 6 passes at 250°C, (c) after 6 passes at 300°C.

grain fraction of the grain population are presented in Table 1.

In the first two experiments, the first stage (2 passes) of ECAE was performed at 350°C. A higher density of twinning was found for the slower ram velocity, Fig. 5, which promotes the subsequent recrystallisation of fine grains. Further ECAE (6 passes) at 200°C produced microstructures that were very similar. The area fraction of large grains was smaller for the case where twinning density was substantially

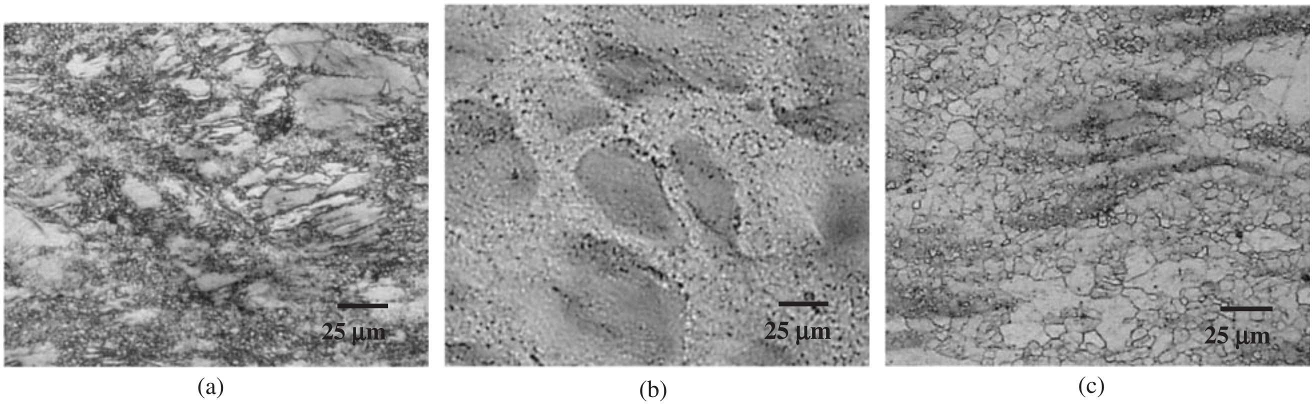

Fig. 3 Microstructures of specimens deformed by warm ECAE (Route C, 120° die) (a) after 8 passes at 200°C, (b) after 8 passes at 250°C, (c) after 8 passes at 300°C.

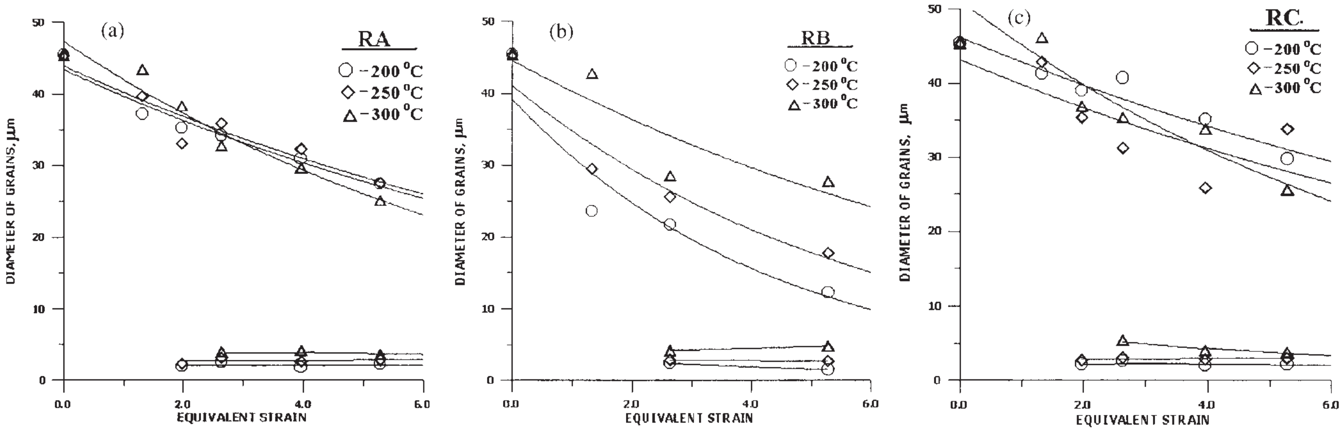

Fig. 4 Average grain size as a function of equivalent strain introduced by ECAE. The average diameter of large and small grains is shown for three ECAE temperatures. (a) Route A, (b) Route B, (c) Route C.

Table 1 The experimental schedules and the resulting average grain size for the fine grain population.

	ECAE (Route B) — Stage I			ECAE (Route B) — Stage II			
No. of exp.	Number of passes	Temperature, °C	Ram velocity, mm/s	Number of passes	Temperature, °C	Ram velocity, mm/s	Average grain size, μm
1	2	350	50	6	200	0.5	2.1
2	2	350	0.001	6	200	0.5	2.1
3	2	200	50	6	350	0.5	1.8
4	2	200	0.001	6	350	0.5	2.8
5	4	200	0.5	4	350	0.5	2.4

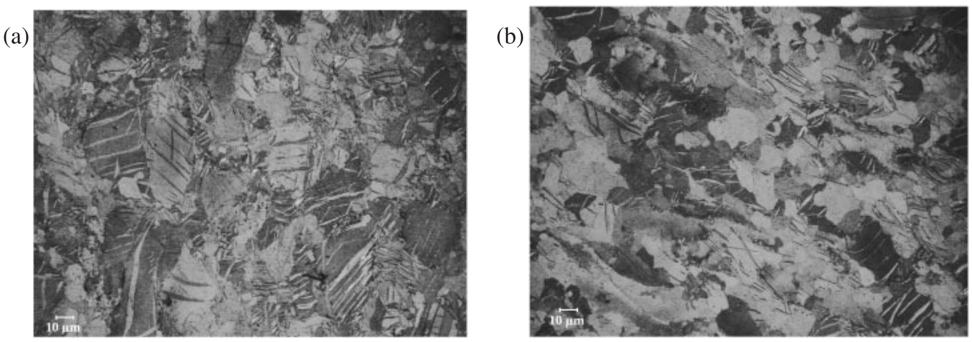

Fig. 5 Microstructures of specimens processes by ECAE (Route B, 90° die) at 300°C with the ram velocity: (a) 0.001 mm/s, (b) 50 mm/s.

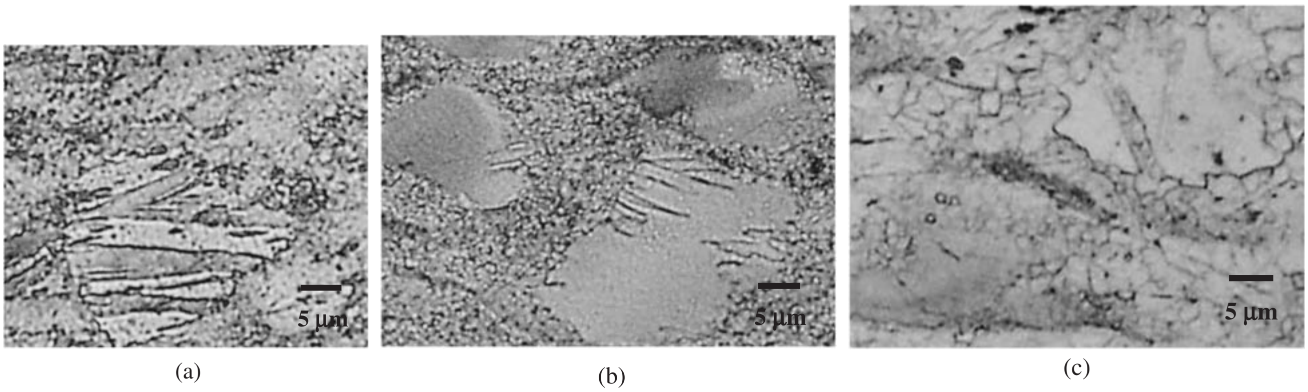

Fig. 6 Microstructures of specimens deformed by warm ECAE (8 passes, Route A, 120° die) showing twins in large grains in the transversal section (a) −200°C, (b) −250°C, (c) −300°C.

higher. However, in both cases the area of large grains was reduced to only about 1%. The fine grain size produced as a result of dynamic recrystallisation was the same for both samples.

When the first stage of ECAE was performed at 200°C, the density of twinning was also higher for the slower ram velocity, which resulted in an earlier start of dynamic recrystallisation in the second stage that was conducted at 350°C. Grain growth usually occurred in the final ECAE passes with the area fraction of large grains increasing to 14%. The lowest average grain size (1.8 μm) corresponds to the case when dynamic recrystallisation was delayed and grain growth did not occur.

3.2 Twinning under warm ECAE

Evidence for twinning under warm ECAE is seen in Fig. 6 that shows microstructures of specimens deformed by 8 passes of ECAE under Route A at three different temperatures. Deformation twins in large grains are clearly seen at all three temperatures. As could be expected,[10] the occurrence of twins decreases with increasing ECAE temperature. The number of twins per unit area of large grain regions decreases almost to zero as the temperature of warm ECAE increases from 200 to 300°C, cf. Fig. 7.

The evolution of the density of twins in large grains with the number of warm ECAE passes was also studied. Measurements were taken after each even pass number. It was found that the twin density acquired within the first two passes either stayed constant, as in the case of 200°C ECAE, or exhibited an apparent decrease with the number of passes, as for the other two temperatures. This decrease can be attributed to dynamic recrystallisation at higher temperatures that is believed to be enhanced in the presence of twins,[11] as the twins can act as recrystallisation sites for new grains. The evidence for this is seen in Fig. 8.

3.3 Tensile ductility at elevated temperature

The results of uniaxial tensile tests on specimens pre-strained by warm ECAE with a ram speed of 0.5 mm/s are presented in Table 2.

An obvious improvement of ductility by warm ECAE pre-treatment was obtained. In particular, pre-straining by ECAE (Route C with the ram velocity of 0.5 mm/s) produced a

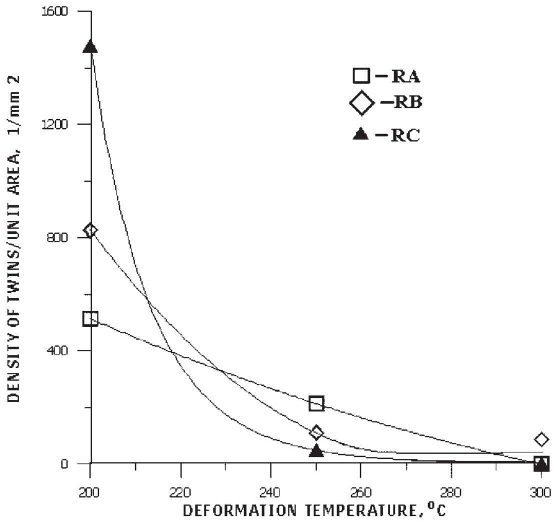

Fig. 7 Density of twins in large grains at equivalent strain of 6.9 as a function of the ECAE temperature (Squares - Route A, diamonds - Route B, triangles - Route C).

Fig. 8 The microstructure after warm ECAE (300°C) showing a large grain (∼40 μm) containing a twin and small grains (indicated by arrows) nucleated at the twin boundary.

Table 2 The results of uniaxial tensile tests on specimens pre-strained by warm ECAE.

ECAE				Tensile Test			
Die angle, °	Temperature, °C	Route	No. of passes	Temperature, °C	Strain rate, s^{-1}	Strength, MPa	Elongation, %
120	200	A	6	300	0.003	33	137
120	200	A	8	300	0.003	30	88
120	200	C	6	300	0.003	37	80
120	200	C	8	300	0.003	31	—
90	200	B	6	300	0.003	28	138
120	250	A	6	300	0.003	26	204
120	250	A	8	300	0.003	27	147
120	250	C	6	300	0.003	31	154
120	250	C	8	300	0.003	35	142
90	250	B	6	300	0.003	24	110
120	300	A	6	300	0.003	26	180
120	300	A	8	300	0.003	27	86
120	300	C	6	300	0.003	28	237
120	300	C	8	300	0.003	21	262
90	300	B	6	300	0.003	18	211
120	200	A	6	300	0.03	62	92
120	200	C	6	300	0.03	56	—
90	200	B	6	300	0.03	43	155
120	250	A	6	300	0.03	65	94
120	250	C	6	300	0.03	60	91
90	250	B	6	300	0.03		
120	300	A	6	300	0.03	58	79
120	300	C	6	300	0.03	54	71
90	300	B	6	300	0.03		

tensile elongation of 262% at 300°C and the strain rate of $3 \cdot 10^{-3}\,s^{-1}$. This is a reasonably good result, especially as no attempt was made in the present work to define strain rate and temperature range for maximum tensile ductility.

The stress-strain curves obtained by tensile testing at the temperature of 300°C and strain rate of $3 \cdot 10^{-3}\,s^{-1}$ are shown in Fig. 9. Pre-straining was done by employing one of the three ECAE routes (A, B, or C) up to 6 or 8 passes using 120° die and up to 4 or 6 passes using 90° die, the corresponding equivalent strain being 4.6 or 6.9. The character of flow curves is seen to change for pre-straining performed at 300°C. After this treatment the microstructure, as shown, is no longer bi-modal, but rather consists of small recystallised grains with diameter less than 4 µm. Pre-straining performed at 300°C, Route C results in a long portion of the deformation curve with a stable flow stress and delayed necking, Fig. 9, suggesting superplastic behaviour at the chosen parameters of the tensile test, namely temperature of 300°C and strain rate of $3 \cdot 10^{-3}\,s^{-1}$. Tensile tests performed at a strain rate ten times higher, returned lower values of ductility, cf. Table 2.

3.4 Strain rate jump test

Strain rate jump tests were performed for homogenised material at 300°C and for ECAE processed materials at 275°C, 300°C (Fig. 10) and 325°C. Typically, the applied strain rate was changed from 10^{-5} to $10^{-4}\,s^{-1}$, then to $10^{-3}\,s^{-1}$ and finally to $10^{-2}\,s^{-1}$. The strain rate sensitivity index, m, defined as

$$m = \frac{\ln\left(\dfrac{\sigma_2}{\sigma_1}\right)}{\ln\left(\dfrac{\dot{\varepsilon}_2}{\dot{\varepsilon}_1}\right)} = \frac{1}{2.3}\ln\left(\frac{\sigma_2}{\sigma_1}\right) \cong \frac{1}{2.3}\,\frac{\sigma_2 - \sigma_1}{\sigma_1}$$

was determined from these measurements.

The results, Fig. 11, show that m is much larger for material processed by ECAE than for the homogenised material used as a reference. In fact, after ECAE processing the value of m reaches the level of 0.5 for the temperature of 325°C, which already signifies the superplastic range. Thus, the requirements of superplastic deformation, such as small grain size in the micrometer range and a sufficiently high value of m approaching 0.5, resulting in delayed necking and a stable plateau on the stress-strain curve, are fulfilled. One can conclude that the significant ductility of 262% achieved at 300°C by virtue of ECAE pre-straining is a result of superplasticity or a forerunner of superplasticity to set in at somewhat more elevated temperature.

4. Conclusions

The experimental observations made in this work lead us to conclude that:

(1) Pre-straining of alloy ZK60 by warm ECAE leads to a significant grain refinement, down to about 2 µm resulting in improved ductility and even superplasticity.
(2) The density of twinning in warm ECAE increases as a temperature of ECAE decreases and is greatest for

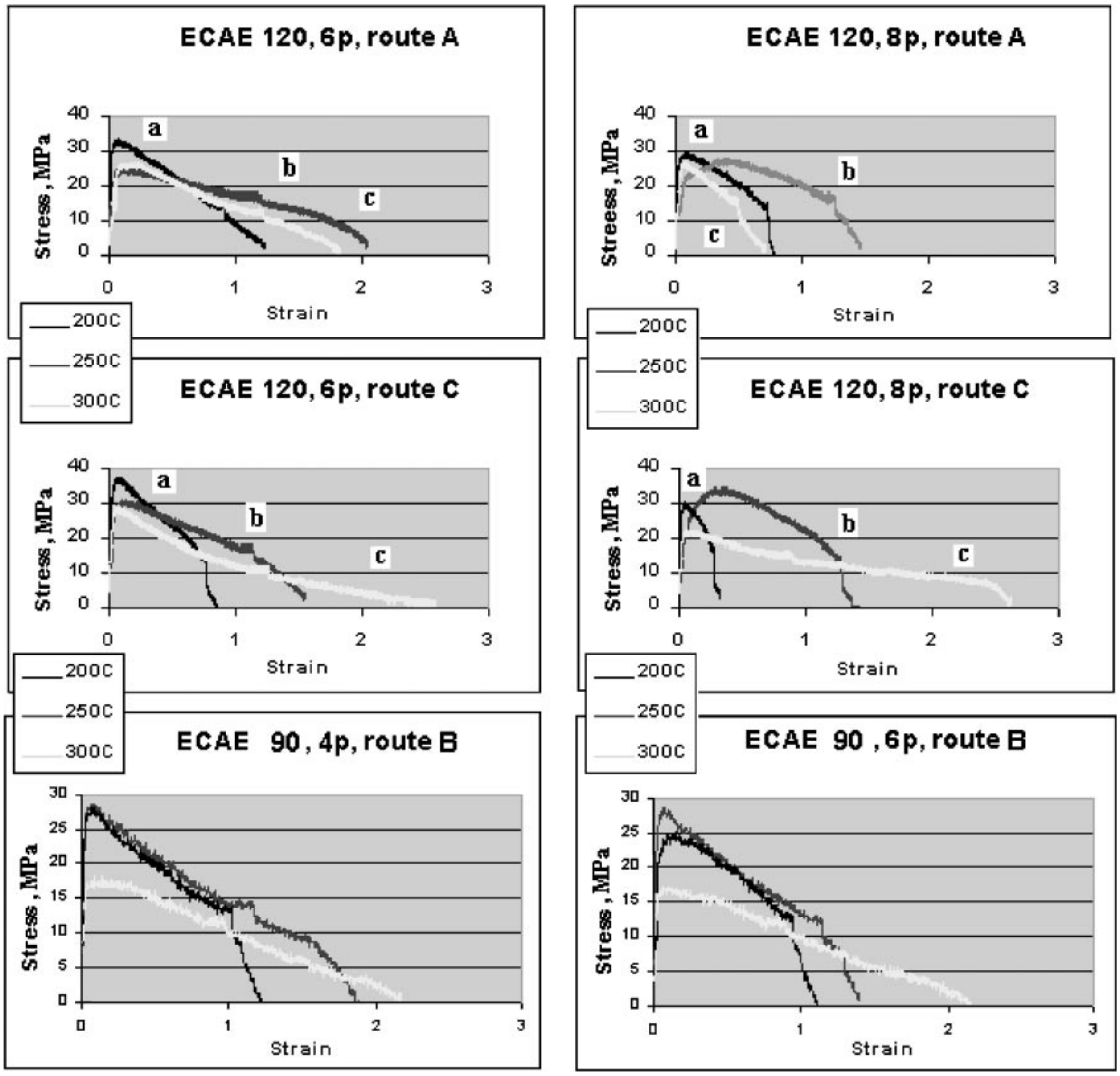

Fig. 9 Stress-strain curves from tensile tests for specimens with different pre-straining histories.

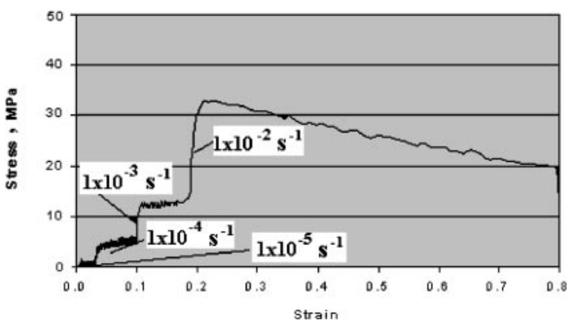

Fig. 10 Strain rate jump tests for ZK60 processed by ECAE (Route B, 120° die) at 300°C.

Route C.

(3) Because twinning influences the propensity for dynamic recrystallisation, an avenue for obtaining a desired fine grain structure appears to be the use of a two-stage ECAE schedule introducing controlled twinning in the first ECAE stage.

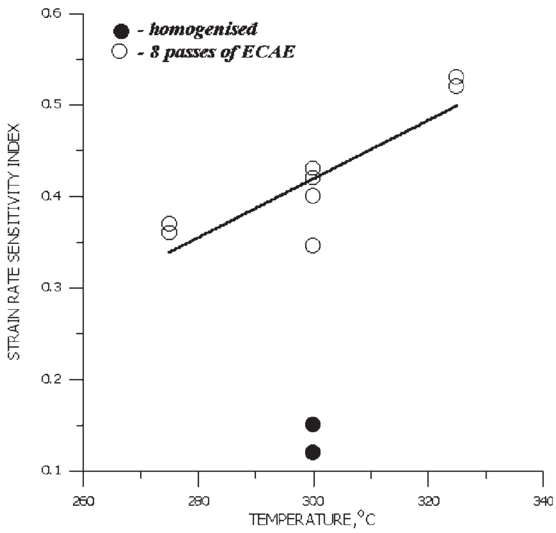

Fig. 11 Strain rate sensitivity *vs.* temperature.

(4) Further fine-tuning of the process parameters of the ECAE pre-treatment for optimising the microstructure and the resulting ductility of the alloy appears to have the potential for significant further improvement in ductility, especially in low temperature superplasticity.

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