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ULTRAFINE-GRAINED MATERIALS

Enhanced superplasticity of magnesium alloy AZ31 obtained through equal-channel angular pressing with back-pressure

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Abstract Excellent superplastic elongations (in excess of 1,200%) were achieved in a commercial cast AZ31 alloy processed by low temperature equal-channel angular pressing (ECAP) with a back-pressure to produce a bimodal grain structure. In contrast, AZ31 alloy processed by ECAP at temperatures higher than 200 °C showed a reasonably uniform grain structure and relatively low ductility. It is suggested that a bimodal grain structure is advantageous because the larger grains contribute to strain hardening thus delaying the onset of necking, while grain boundary sliding associated with small grains provides a stabilizing effect due to enhanced strain rate sensitivity.

Introduction

Magnesium alloys have a great potential as structural materials for aerospace, automotive and electronics applications owing to their low density and high strength-to-weight ratio. However, the fabrication of magnesium parts (including forging and sheet rolling) is limited due to the relatively low ductility of magnesium alloys owing to their hcp crystal structure. The development of thermo-mechanical processing leading to significantly enhanced superplastic behaviour in magnesium alloys would open up new avenues for industrial applications of superplastically formed magnesium parts.

In order to enhance ductility and to establish superplastic properties in magnesium alloys, various methods of thermo-mechanical processing, such as hot-rolling and extrusion, have been used [1–6]. In the last decade, significant efforts to use severe plastic deformation methods, such as equal-channel angular pressing (ECAP), to produce materials with extremely fine grain structure have been made [7, 8]. Combining ECAP with conventional forming techniques was also considered as a possible processing route [9, 10]. However, even for widely investigated magnesium alloys, such as AZ31, there is no consensus concerning the optimum thermo-mechanical processing route leading to enhanced tensile ductility. At the present time there is no agreement on the type of microstructures most favourable for increasing the elongation-to-failure. A summary of results for AZ31 published to date is given in Table 1.

Analysis of the results presented in Table 1 shows that severe hot extrusion or hot extrusion followed by the ECAP leads to extreme grain refinement and significantly enhanced superplastic properties with a maximum strain of 900% obtained at 300 °C [4] and 460% obtained at 150 °C [9]. Most of the published papers report microstructures with uniform grain distributions varying in size depending on the particular processing route from 0.7 to 250 μm.

In this study, straight ECAP without any further processing steps was used for grain refinement of magnesium alloy AZ31. The effect of the ECAP parameters, such as

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Table 1 Summary of publications on superplasticity of AZ31

References	Processing history	Grain size (μm)	Temperature of tensile test (°C)	Strain rate in tensile test (s^{-1})	Eng. strain at fracture (%)
[1]	Hot-rolling	130	350	6×10^{-5}	190
[2]	Hot-rolling	250	500	1×10^{-2}	189
[3]	Hot-rolling	130	375	3×10^{-5}	196
[4]	Hot-extrusion	3	200	1×10^{-4}	600
			300	1×10^{-4}	900
[5]	Hot-extrusion	15	177	1×10^{-5}	120
[6]	Hot-extrusion	5	325	1×10^{-4}	608
[7]	ECAP	1	250	3×10^{-3}	400
[8]	ECAP + annealing	15	RT	1×10^{-3}	45
[9]	Extrusion + ECAP	0.7	150	1×10^{-4}	460
[10]	Rolling + ECAP	25	450	5×10^{-3}	80
[11]	Hot-rolling	150	375	1×10^{-2}	130
[12]	Hot-extrusion	60/4	250	5×10^{-4}	149
[13]	Hot-rolling	6	450	2×10^{-4}	265
[14]	Rolling + extrusion + ann.	5	400	1.4×10^{-3}	360

temperature and back-pressure, was investigated with respect to possible bi-modality of the structure and enhanced superplastic properties of this alloy. The findings of this study are reported below. The report expands on our recent results presented in Ref. [15].

Experimental material and procedures

The as-received material was obtained in the form of continuously cast billet. Before ECAP processing, the cast alloy was homogenized at the conditions defined in section "Solution heat treatment and initial microstructure", followed by water quenching. In order to avoid oxidation during heat treatment, the cast AZ31 billets were packed in alumina powder.

Route B_C ECAP was performed in a 90° die with sharp corners at the ram velocity of 15 mm/s. Graphite spray was used for die lubrication. Multi-pass ECAP was carried out in the temperature range of 150–250 °C, using a heated die with temperature stability during pressing within 2 °C. At the ram velocity used, the temperature rise due to plastic deformation was negligible. After each pass the billet was quenched in water to retain the microstructure obtained. The billets were pressed 1, 2, 4, 6 or 8 times at 150 °C under a back-pressure of 260 MPa to investigate the influence of the number of ECAP passes on the maximum elongation-to-failure in the post-ECAP tensile tests. A back-pressure (BP) was applied in the exit channel of the die by a backward punch. The level of back-pressure was varied from 44 MPa at 200 °C to the maximum value of 260 MPa at the lowest ECAP temperature of 150 °C.

Tensile specimens with a gauge length of 8 mm and diameter of 4 mm were machined from the ECAP processed samples. Tensile tests were carried out using an electro-mechanical INSTRON®4505 machine with Series IX software. Elevated temperature tensile tests were performed in an environmental chamber.

For optical microscopy (OM), samples were cut, and then mounted in a transparent epoxy with a subsequent polishing using a Struers RotoPol-21auto-polisher. The microstructure of the samples was analysed with an Olympus® PMG3 optical microscope under 5, 10, 20, 50 and 100 times magnifications before and after etching. An etching solution of the following composition was used: 6 g picric acid dissolved in 40 mL water, 40 mL acetic acid and 100 mL ethanol. The images were obtained with the help of a digital camera attached to the microscope and evaluated using version 3.5.0 SPOT software.

The specimens for transmission electron microscopy (TEM) were examined using a Philips CM20 microscope operated at 200 kV. The TEM samples were prepared by electropolishing using Tenupol 3 in a solution of 33.48 g magnesium perchlorate and 15.9 g lithium chloride in 300 mL of butoxy ethanol and 1,500 mL methanol at 100 V and −50 °C with a current of ~150 mA. Bright field images and selected area diffraction patterns with aperture diameters of 5.8 and 1.1 μm were taken on a transverse section of specimen using a double-tilt specimen holder.

The fracture surfaces of tensile specimens were studied by scanning electron microscopy (SEM) using a JEOL JSM-840A microscope operated at 20 kV.

Results and discussion

Solution heat treatment and initial microstructure

The solution heat treatment prior to ECAP influences the tensile ductility in subsequent tensile test quite substantially (cf. Table 2). The as-cast microstructure had large particles and precipitate clusters (Fig. 1a), which can play a significant role in reduction of tensile ductility. An investigation of the effect of different solution heat treatments was carried out and the distribution and volume fraction of precipitates were analysed. The optical micrographs of AZ31 in as-cast state and after solution treatment (ST) are shown in Fig. 1, representing ST at one of the temperatures studied (440 °C) for 20 min and at 420 °C for 4 h. As listed in Table 2, a long homogenization time was required to eliminate precipitate clusters (Fig. 1c), but the overall volume fraction of precipitates decreased only for temperatures above 500 °C, the size of precipitates remaining practically unchanged. It can be seen from Fig. 1d that the microstructure of the as-cast material after heat treatment consists of very large grains of the size in the range of 200 μm–1.2 mm and contains incoherent $Mg_{17}Al_{12}$ constituent particles precipitated from the supersaturated solid solution. Various particle morphologies were observed, including needle-shaped, globular and polyhedron-shaped

Table 2 Solution heat treatments used prior to ECAP

N	Solution heat treatment	Engineering strain at failure (%)	Volume fraction of precipitates (%)	Comments
M1	As-cast	16	2.00	Large particles and numerous precipitate clusters
M2	20 min at 420 °C	25	1.88	A precipitate dispersion with a size distribution (diameter up to ∼40 μm); precipitate clusters
M2	20 min at 440 °C	31	1.76	A precipitate dispersion with a size distribution (diameter up to ∼20 μm); precipitate clusters of ∼20 μm diameter
M3	20 min at 480 °C	32	1.41	A precipitate dispersion with a size distribution (diameter up to ∼10 μm); precipitate clusters of ∼20 μm diameter
M4	20 min at 550 °C	29	1.13	The precipitates in both clusters and matrix had similar diameters of up to ∼20 μm
M5	4 h at 420 °C	37	1.06	A precipitate dispersion with a size distribution (diameter up to ∼20 μm). No precipitate clusters observed

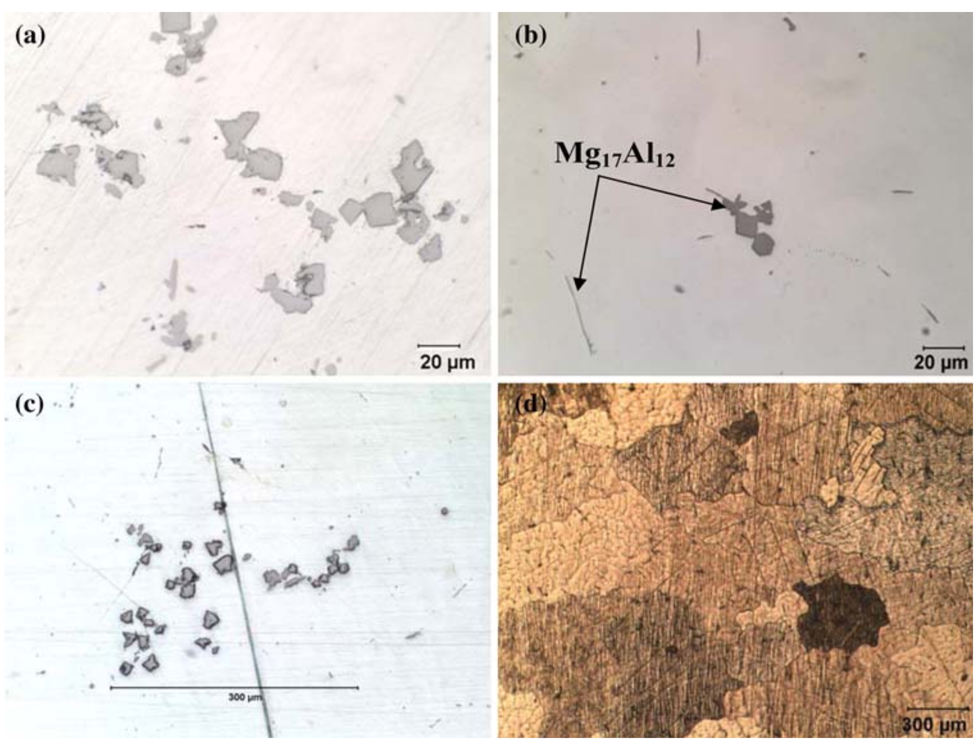

Fig. 1 Microstructure of AZ31 (OM) of as-cast billet before and after heat treatment. (**a**) Unetched as-cast; (**b**) unetched after heat treatment at 420 °C for 4 h; (**c**) unetched after heat treatment at 440 °C for 20 min; (**d**) etched after heat treatment at 420 °C for 20 min

particles (cf. Fig. 1b). An additional SEM investigation revealed the presence of uniformly distributed round Mn_5Al_8 precipitates of size 1–2 μm in diameter.

Microstructure evolution during ECAP

The microstructure of samples processed by ECAP at a temperature of 200 °C and above tended to exhibit a more uniform grain size with the number of ECAP passes increasing. A significant grain refinement was observed, with the grain size ranging between 1.3 and 7.5 μm (Fig. 2a). No bi-modal structure was observed for these temperatures, and the large variation in the grain size was attributed to the effect of non-uniformity of initial grain structure and dynamic recrystallization (DRX). The constituent particles were not refined any further. In contrast, in samples processed at 150 °C with a back-pressure of 218 MPa, a typical bi-modal grain structure was observed (Fig. 2b).

Our previous studies of microstructure formation during ECAP of Mg alloys have shown [16] that grain refinement appears to result from two concurrent effects: formation of a large population of small recrystallized grains at old grain boundaries and fragmentation of large grains by twins serving as sites at which new recrystallized grains formed. Route B_C ECAP conducted at the low end of the temperature interval studied resulted in profuse twinning in the first two passes triggering DRX at twin boundaries. The twinning activity then decreased over the subsequent four passes. Although aspects of DRX were not studied in the present work, a general trend observed for alloy ZK60 [16] appears to be relevant for AZ31 as well: the area fraction of surviving large grains and their average diameter, as well as the diameter of small recrystallized grains, were the smallest for the lowest possible temperature of ECAP.

Tensile ductility as a function of back-pressure and ECAP temperature

A decrease in the ECAP temperature at a fixed level of back-pressure leads to increased tensile ductility (cf. Fig. 3). A simultaneous decrease in ECAP temperature and an increase in back-pressure results in further enhancement of tensile ductility (Fig. 3) with the highest ductility being achieved at the strain rate of 10^{-4} s^{-1} at the temperature of 350 °C. It should be noted that those results were obtained for samples with relatively uniform fine-grained microstructure.

As the tensile ductility obtained for these values of temperature and strain rate already exceeded most known results (cf. Table 1), the effort was concentrated on further

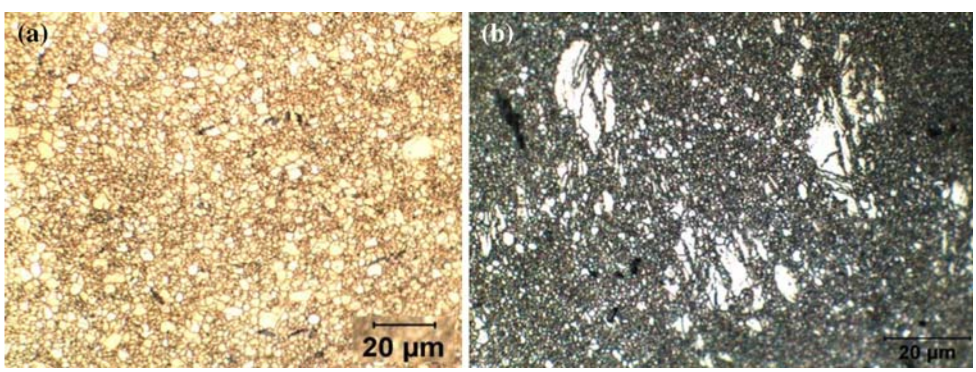

Fig. 2 Microstructure of AZ31 (OM) after ECAP. (**a**) Six ECAP passes at 200 °C, BP = 43 MPa; (**b**) six ECAP passes at 150 °C, BP = 262 MPa

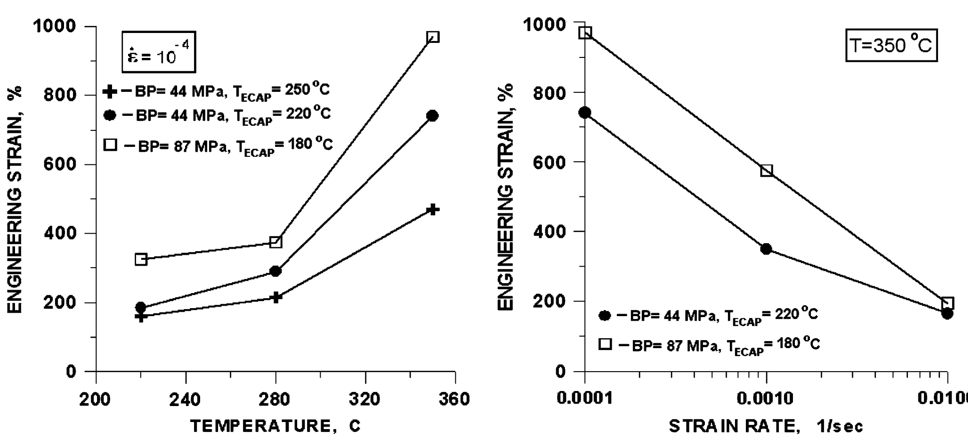

Fig. 3 Engineering strain-to-failure for samples processed by six ECAP passes at different levels of temperature and back-pressure as a function of (**a**) temperature; (**b**) strain rate

lowering the ECAP temperature (down to 150 °C) while increasing back-pressure (up to 260 MPa) to obtain the bi-modal microstructure. It was shown in our previous work on ZK60 magnesium alloy that bi-modality of grain structure in combination with continuous DRX during a tensile test is a key to improve superplastic behaviour [16, 17].

An important characteristic of superplasticity is the strain-rate sensitivity of the flow stress. The strain-rate sensitivity parameter (m) was estimated from the stress–strain curves obtained at different strain rates. Using the peak stress values for corresponding strain rates, a value of $m = 0.45$ was calculated, which is well within the strain-rate sensitivity range typical of superplastic deformation.

Tensile ductility as a function of the number of ECAP passes

The influence of the number of ECAP passes on ductility of AZ31 was investigated on samples processed by ECAP at the temperature of 150 °C with a back-pressure of 260 MPa (Fig. 4) and solution-treated according to M3 schedule (see Table 2). The tensile tests were performed at the temperature of 350 °C with the strain rate of 10^{-4} s^{-1}. The strain-to-failure increased with the number of ECAP passes from 345% after one pass to 1,090% after six passes.

Six ECAP passes were chosen as a maximum, as a number of passes larger than six lead to the development of internal cracks initiated at the second-phase particles even under increased back-pressure (Fig. 5). It will be shown below that the temperature of ECAP for enhanced ductility should be kept as low as possible. Therefore the optimum number of passes is defined from the condition that the ductility of the matrix is not exhausted and that rotation of constituent particles does not generate defects on particle/matrix interface leading to crack initiation. As the presence of the particles and large precipitates seems to be the main reason for limited ductility, the M5 solution treatment (see Table 2) was chosen for further optimization of superplastic ductility.

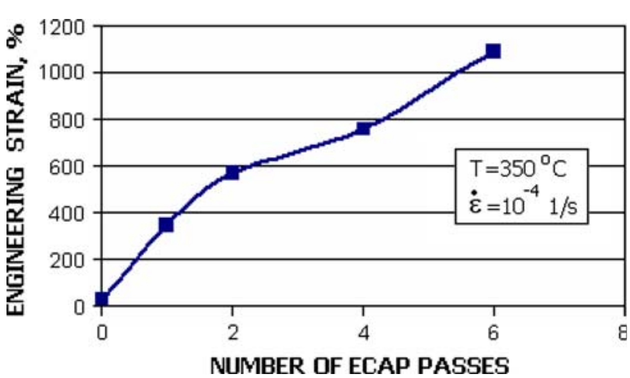

Fig. 4 Engineering strain-to-failure for samples processed by 1, 2, 4 and 6 ECAP passes at 150 °C with a back-pressure of 260 MPa after M3 solution treatment (tensile test performed at 350 °C with the strain rate of 10^{-4} s^{-1})

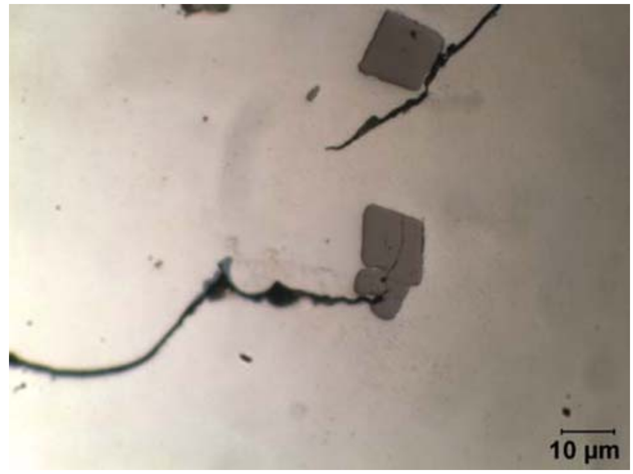

Fig. 5 Development of cracks from the constituent particles after more than six ECAP passes. The example shows cracks formed during the seventh ECAP pass at 200 °C and back-pressure of 44 MPa

Microstructures developed during ECAP at low temperature

Further TEM investigation of microstructure developed after six ECAP passes at 150 °C with a back pressure of 260 MPa confirmed the aforementioned bi-modality of grain-size distribution. The two grain populations had distinctly different grain sizes: in the range of 0.5 ± 0.25 μm for small grains and about 16.5 ± 3.5 μm for large grains, as seen in the light microscopy picture (Fig. 2b) and in TEM micrographs (Fig. 6).

Unlike in alloy ZK60, where a "tri-modal" grain structure was found [16], no similar trend to tri-modality was observed in AZ31, although the occurrence of small grains of about 100 nm in diameter ("a third population") has been observed. Some singular small grains were present, but they did not form a population stable at low temperatures. This can be interpreted in terms of the absence of very fine precipitates pinning the grain boundaries and also acting as nucleation sites for new grains.

Enhanced superplastic behaviour

The samples processed by six ECAP passes at 150 °C and back-pressure of 260 MPa exhibited enhanced superplastic behaviour with a nearly-record strain-to-failure of 1,215% (cf. Fig. 7a, b), which is significantly higher than the strain-to-failure of the samples processed at higher temperatures. It should be noted that ductility of original samples before ECAP at the same testing conditions was only 37%.

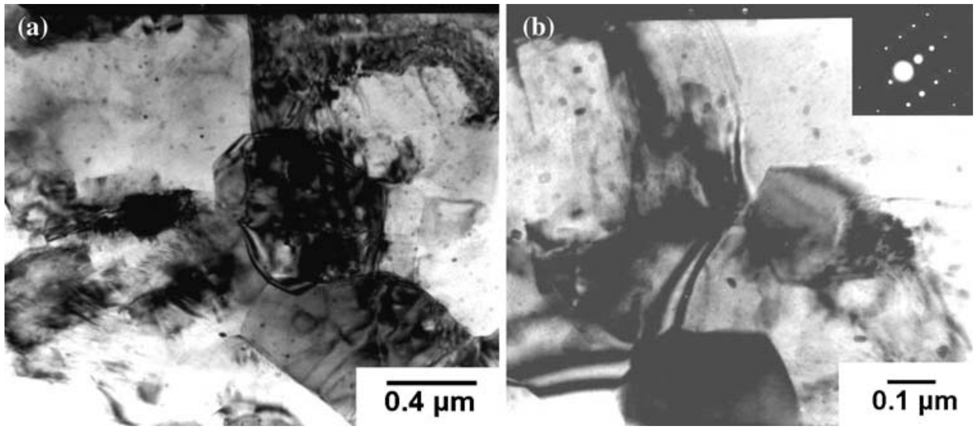

Fig. 6 TEM images of microstructure developed after six ECAP passes (route B_C) at 150 °C and back-pressure of 260 MPa. (**a**) Recrystallized small grain at a triple-junction; (**b**) freshly recrystallized small grains and formation of dislocation cells in bigger grains

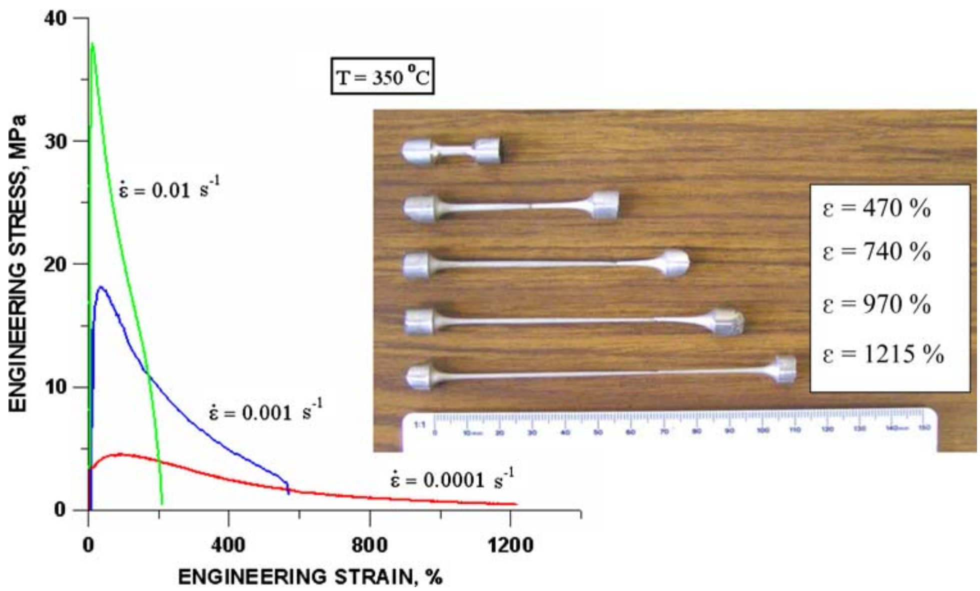

Fig. 7 Engineering stress–strain curves for 350 °C and different strain rates for samples processed by six ECAP passes at 150 °C and back-pressure of 260 MPa. Also shown are an original specimen and tensile specimens machined from ECAP bars processed at 250, 220, 180 and 150 °C and tested at 350 °C under strain rate of 10^{-4} s^{-1} (from top to bottom)

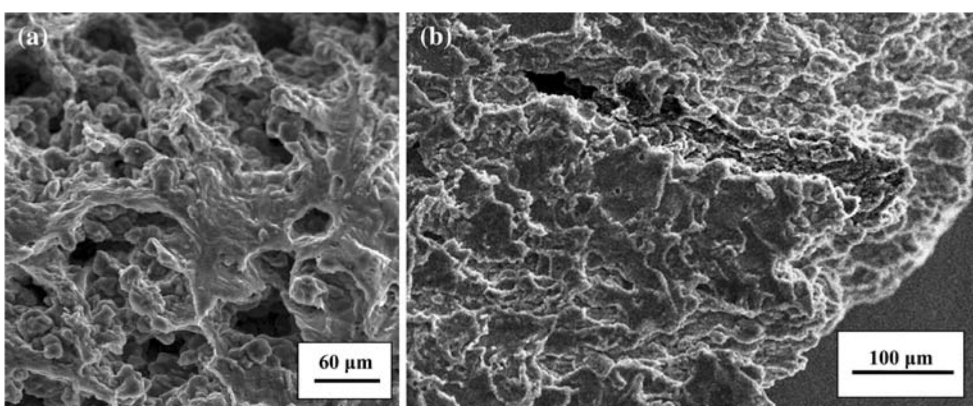

Fig. 8 SEM images of the tip of a tensile specimen. (**a**) Fracture surface of a tensile sample tested at 350 °C at a strain rate of 10^{-4} s^{-1}; (**b**) longitudinal section of the tip made by wire-cuting (the specimen was prepared from the material processed by six ECAP passes at 150 °C and back-pressure of 260 MPa)

Fractography

The mechanism of significantly enhanced ductility was studied by examination of fracture surface in SEM (Fig. 8a). Unusually deep elongated dimples (Fig. 8b) were observed on the micrographs. This indicates that the dimples were formed at an early stage of the tensile test, perhaps due to ripping of relatively large precipitates as shown in Figs. 1 and 4, as the size of the dimples is comparable with the precipitate size. However, the loadbearing capacity of the intact filaments between the dimples was apparently high enough to carry the load during the continued tensile test, as the dimples were elongated together with the sample. The ductility of the material in the filaments seems very high, which suggests that there are still some unused reserves of ductility. It can be conjectured that—should large precipitates be eliminated by heat treatment—the elongation-to-failure could be raised even further.

Conclusions

Enhanced superplastic behaviour of magnesium alloy AZ31 was studied as a function of the initial heat treatment prior to ECAP, the number of ECAP passes and the level of back-pressure. It was shown that material with a rather uniform microstructure can reach 980% ductility in tensile test performed at 350 °C at the strain rate of 10^{-4} s^{-1}. The processing by ECAP at temperature as low as 150 °C with a back-pressure of 260 MPa leads to microstructures more favourable for enhanced superplasticity, notably bi-modal grain structures. Stability against tensile necking is controlled by two factors: strain hardening and strain-rate sensitivity of the flow stress. In a bi-modal structure, the large grain fraction gives rise to strain hardening, while the small grain size one provides enhanced strain-rate sensitivity due to diffusion-controlled processes there. It was shown that a decrease in the temperature of ECAP processing and concurrent increase in the back-pressure result in corresponding increase in elongation-to-failure to a record level of 1,215%, which is by more than 300% higher than the best results published on AZ31 superplasticity to date.

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