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Grain refinement in aluminium and the aluminium Al–Cu–Mg–Mn alloy by hydrostatic extrusion

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Hydrostatic extrusion was used as a method for grain refinement in technically pure aluminium and in an aluminium alloy. Both materials were deformed up to a true strain of ~4. Such a deformation resulted in substantial grain size refinement to below 1 μm in aluminium and below 100 nm in the aluminium alloy. In pure aluminium, microstructure evolution proceeds by a continuous increase in the grain boundary misorientation, without changing the grain size. In the aluminium alloy, which has lower stacking fault energy, grains continuously decrease in size, down to the nanometre scale. As a consequence of such microstructure evolutions, the mechanical properties of pure aluminium remain almost constant within a wide range of strains, whereas the mechanical properties of the aluminium alloy are significantly improved. From the present study, one can conclude that hydrostatic extrusion can offer an alternative way to produce nano-metallic elements made of aluminium alloys for light-weight applications.

Key words: aluminium, aluminium alloys, ultra-fine grained microstructure, severe plastic deformation, hydrostatic extrusion

1. Introduction

Ultra-fine grained and nanocrystalline materials arouse great interest due to their attractive properties. In particular, nano-metals exhibit a high strength combined with sufficiently good ductility. The fabrication methods of bulk ultra-fine and nanocrystalline metals can roughly be classified into the three groups: (i) nano-crystallization of amorphous materials, (ii) severe plastic deformation (SPD), and (iii) consolidation

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of nano-powders. In the last decade, various SPD methods have been developed to produce a wide range of ultra-fine and nano-scale grained metals and alloys [1–5]. Among these methods, those most frequently reported include equal channel angular pressing (extrusion) (ECAP or ECAE) [6], high-pressure torsion (HPT) [7], accumulative roll bonding (ARB) [8], cyclic extrusion–compression (CEC) [9], repetitive corrugation and straightening [10], cold rolling [11] and hydrostatic extrusion (HE) [12, 13]. Among these methods, HE offers the potential for producing homogeneous, fully dense bulk materials in a variety of forms. Rods, wires of complex cross-sections, and also small tubes can be produced by HE [14]. These are features often unattainable by other SPD techniques. In the present work, HE was used to refine the grains in technically pure aluminium and in an aluminium alloy. Attention was focused on the possibility of obtaining a homogenous nanocrystalline structure in aluminium alloys intended for light-weight constructions.

2. Experimental methods

Technically pure 1050 (99,5%) aluminium and a 2017 Al–4Cu–0.5Mg–1Mn aluminium alloy were used in this study. The as-received aluminium was work-hardened and its microstructure contained elongated subgrains of about 1 μm equivalent diameter. The aluminium alloy was solution heat-treated at 505 °C and water quenched to obtain a single-phase material. Its microstructure consisted of nearly equiaxed subgrains, about 2 μm in diameter. The initial microstructures of aluminium and aluminium alloy are shown in Fig. 1.

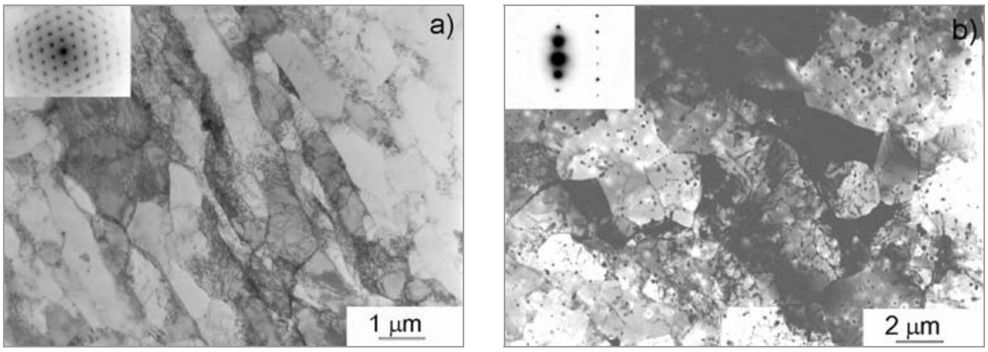

Fig. 1. TEM micrographs of aluminium (a) and Al–Cu–Mg–Mn alloy (b) in their initial state

The rods, 20 mm in diameter, were subject to HE, which resulted in a total cross -section reduction of 44.4. The HE was run in a press, designed and constructed at the Institute of High Pressure Physics, Warsaw. The cross section reduction obtained during a multipass HE operation corresponds to an accumulated true strain of 3.79. The strain rate during HE exceeded 3.0×10^2 s^{-1} (for the smallest wire diameters). The extrusion products were cooled with water at the die exit.

The microstructure and mechanical properties of the materials in their initial state and after HE were investigated. The microstructure was evaluated by transmission electron microscopy (TEM). The observations were carried out with thin foils cut perpendicularly to the extrusion direction. The microstructures were also quantitatively described using computer-aided image analysis. Mechanical properties were characterized by microhardness, measured on transverse sections of the rods and wires. Tensile tests were conducted at room temperature at a strain rate of 8.33×10^{-4} s^{-1}. Both microstructure and mechanical properties were investigated seven days after the extrusion process.

3. Results

TEM images and corresponding selected area electron diffraction (SAED) patterns for an aluminium sample subjected to multipass HE are shown in Figs. 2a, b. After the true strain of 1.39, the microstructure consists of small equiaxed grains (Fig. 2a). Dislocations of high density are visible within the grains. The SAED pattern (the insert in Fig. 2a) is typical of a mosaic structure built-up of grains with small misorientations (diffracted beams are scattered by a few degrees). After the true strain of 3.79, the microstructure changes profoundly (Fig. 2b). Small equiaxed grains are still characteristic features of the microstructure, but the grains are free of dislocations and the diffracted beams visible in the diffraction patterns are scattered into rings. A diffraction image of this kind indicates that the microstructure contains grains separated by both low- and high-angle grain boundaries. Misorientation measurements of the grain boundaries in HE-processed aluminium are in progress.

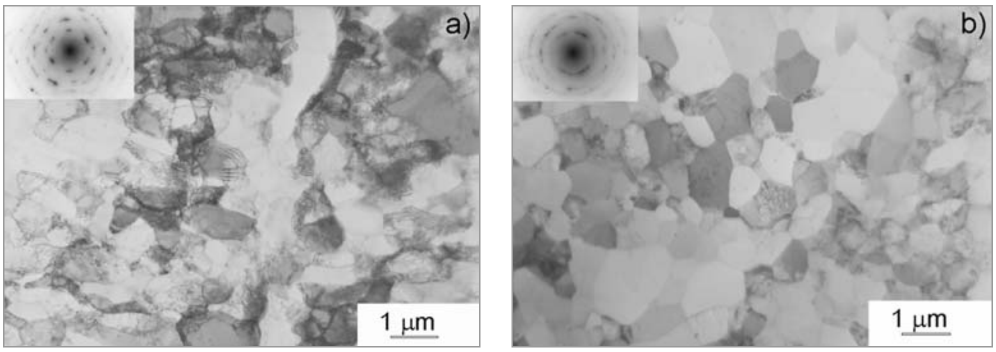

Fig. 2. TEM micrographs and corresponding SAED patterns for aluminium:
(a) after HE to a true strain of 1.39, (b) after HE to a cumulative true strain of 3.79

The microstructures of technically pure aluminium processed by HE were also evaluated quantitatively in terms of their grain size and shape. To this end, all types of grain boundaries (with low and high misorientation angles) were taken into account. The size of grains was determined in terms of the mean value of the equivalent diameter $E(d_{eq})$ and the variation coefficient *CV*, defined as the ratio $\mathrm{E}(d_{eq})/SD(d_{eq})$, where

$SD(d_{eq})$ is the standard deviation of d_{eq}. The grain elongation factor α, defined as the ratio of the maximum diameter to the equivalent diameter of a given grain, d_{max}/d_{eq}, was used to describe the shape of grains. The measured values of $E(d_{eq})$, $CV(d_{eq})$, and α are given in Table 1. It can be seen that, in technically pure aluminium processed by HE, the average grain size does not depend on the accumulated plastic strain. On the other hand, the grain elongation decreases continuously with the imposed strain.

Table 1. Values of microstructural parameters (mean equivalent diameter of grains $E(d_{eq})$, variation coefficient $CV(d_{eq})$, and grain elongation factor α) for aluminium in the initial state and after HE

True strain	$E(d_{eq})$ [μm]	$CV(d_{eq})$	α	Spread of diffraction spots
0	0.92	0.41	1.48	9°
1.39	0.71	0.37	1.37	15°
2.77	0.68	0.47	1.33	33°
3.79	0.70	0.31	1.30	not measurable

Compared to the microstructure of technically pure aluminium, the microstructure of the aluminium alloy evolves in a different way. After a true strain of 1.39, the TEM observations reveal subgrains with dislocations of high density (Fig. 3a). The subgrain size is smaller and the dislocation density is much higher than those in technically pure aluminium. The SAED patterns exhibit a scatter of the diffracted beams by a few degrees. This suggests that the misorientation angles between the subgrains are small. After the final step of HE, the microstructure consists of small equiaxed grains (see Fig. 3b) with grain boundaries with various (low and high) misorientation angles (see the SAED pattern presented as an insert). Compared to the results for technically pure aluminium, grain refinement in the aluminium alloy is much more effective.

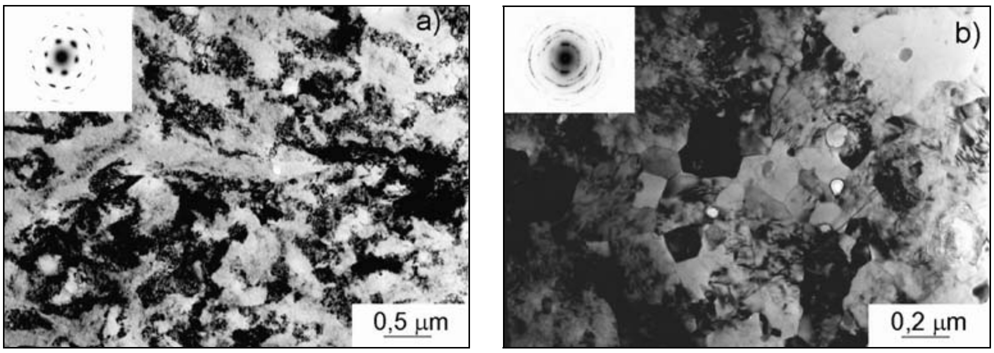

Fig. 3. TEM micrographs and corresponding SAED patterns for Al–Cu–Mg–Mn alloy: (a) after HE to a true strain of 1.39, (b) after HE to a cumulative true strain of 3.79

Grain size measurements in the aluminium alloy processed by HE show that the mean equivalent diameter of the grains sharply decreases with increasing strain, from a few micrometers in the initial state to about 100 nm after applying a cumulative true strain of 3.79. At the same time, grain elongation remains unchanged and the misorientation between neighbouring grains increases, as seen in the SAED pattern. The microstructural parameters for the aluminium alloy are given in Table 2.

Table 2. Values of microstructural parameters (mean equivalent diameter of grains E (d_{eq}), variation coefficient $CV(d_{eq})$, and grain elongation factor α) for aluminium alloy in the initial state and after HE

True strain	$E(d_{eq})$ [μm]	$CV(d_{eq})$	α	Spread of diffraction spots
0	2.180	0.23	1.38	0°
1.39	~0.500	-	-	15°
2.77	0.170	0.29	1.29	38°
3.79	0.095	0.42	1.3	not measurable

One of the main factors that decide about the performance of engineering materials is the homogeneity of their microstructures. In order to analyse the homogeneity of extruded wires in relation to their mechanical properties, microhardness was measured on transverse-sections. The results of these measurements are given in Fig. 4. In both materials, irrespective of the strain applied, microhardness was evenly distributed across the diameter of wires. The results exhibit a relatively small scatter, with the variation coefficient being smaller than 0.02. No visible gradient of the microhardness distribution at the circumference of the wire or near its axis was observed. This confirms that the materials processed by HE are highly homogeneous in terms of their microstructural features.

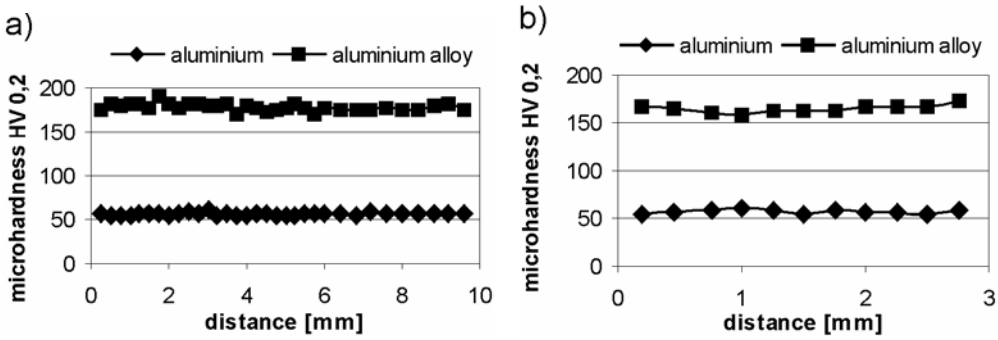

Fig. 4. Microhardness distributions measured on the transverse-sections of hydrostatically extruded rods: (a) to a strain of 1.39 (b) to a cumulative strain of 3.79

In order to determine mechanical properties, static tensile tests were performed at room temperature. The yield stress and elongation for both aluminium and the aluminium alloy are plotted in Fig. 5. In both materials, the yield stress increases with the imposed extrusion strain. The increase in yield stress, however, is greater in the aluminium alloy, amounting to 100%. The highest increase in the yield stress occurs after the first pass.

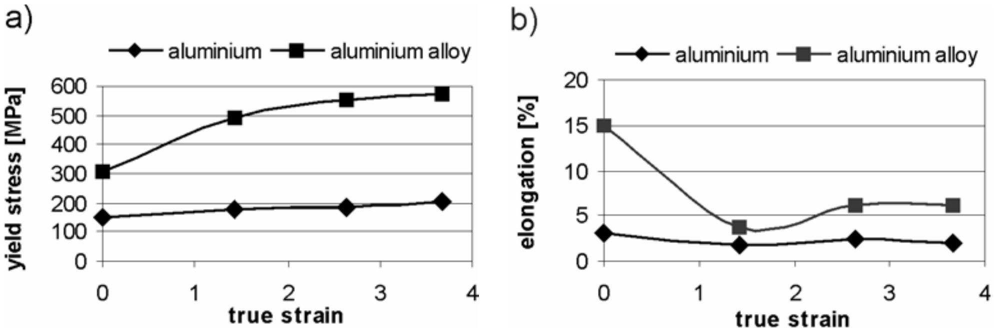

Fig. 5. The yield stress (a) and elongation (b) as functions of the imposed strain during HE of pure aluminium and Al–Cu–Mg–Mn alloy

In subsequent passes, the process saturates and the yield stress approaches a “plateau”. It should be noted that in the aluminium alloy the yield stress achieves the value of 570 MPa, compared to 350 MPa for the same alloy subjected to traditional heat treatment. Here, too, the greatest reduction in elongation is observed after the first extrusion. In the next passes, it remains constant (aluminium) or even increases (aluminium alloy), as seen in Fig. 5b. The change in ductility is more pronounced in the aluminium alloy, where it reaches a useful level of 5% after three passes.

4. Discussion

In literature, various SPD methods have been used for grain refinement in aluminium and its alloys. The reported grain sizes obtained in technically pure aluminium by ECAP vary between 0.5 and 1.5 μm [e.g. 6; 15–16]. As to the influence of the deformation on microstructure, it is commonly observed that ECAP microstructure evolves from subgrain bands (after a single pressing) into a system of high angle boundaries (during subsequent passes) [6]. At high strains, the average grain size is only slightly refined, whereas the average boundary misorientation increases more rapidly [16].

In CEC, the microstructure evolution in technically pure aluminium proceeds in a similar way [9]. The microstructure transforms from a cellblock type into an almost equiaxed one. The spacing between the boundaries that subdivide the structure is almost unaffected by the strain, and the misorientation across these boundaries increases with increasing strain.

The present experiments with HE indicate that the mechanism of microstructure evolution is similar in this case. The misorientation angle of the grain boundaries increases as a function of the accumulated strain, but with grain size remaining almost constant. This type of microstructure evolution has been confirmed by previous results [12], where a true strain of ~6.2 was achieved. Aluminium is a metal with a high stacking fault energy, in which a cross slip occurs very easily and thus, during the consecutive steps of deformation, dislocations can move easily towards the grain boundaries and be absorbed into them. As a result, the grain interiors undergo recovery, while the misorientation angles of the grain boundaries increase. These two processes may take place at a constant grain size, as observed in the present work.

The SPD methods differ in their efficiency of generating high angle boundaries [15]. It has been observed that, in terms of grain boundary misorientations, ECAP with a strain of ~8 gives similar results to CEC conducted at a strain of 60. From this point of view, HE seems to be almost as efficient as ECAP. The results of diffracted beam scattering measurements (Table 1) are very similar to those obtained in aluminium processed by ECAP [6].

In literature, microstructure evolution in aluminium alloys subjected to SPD has been studied primarily for ECAP. It has been found that in Al–Mg [17, 18] and Al–Mn [19] alloys grain refinement is greater than that in technically pure aluminium. This was attributed to the lower rate of recovery in aluminium alloys, which results in a smaller final grain size.

In the Al–Cu–Mg–Mn alloy investigated here, the lower recovery rate is especially visible during the first HE pass, where the deformed microstructure is characterized by a high density of the dislocation tangles, and the grain structure is not well established. During consecutive extrusions, dislocations cannot easily move to the grain boundaries, and in order to minimize the stored energy they tend to divide the grains into blocks with different slip system combinations [20]. These blocks can rotate to achieve different final orientations, causing a significant increase of grain boundary misorientations. This is accompanied by a continuous decrease of the final grain size. The mechanism described above can explain the drop in the average grain size down to 95 nm, which leads to a substantial improvement of the mechanical properties of the alloy. It should be noted that the grain size reduction observed in the present work is similar to that reported for Al–1.7 at. Cu subjected to 8 passes of ECAP [21]. This confirms that HE is an effective process, which leads to a similar grain size reduction to that achieved by other SPD methods.

5. Conclusions

The following conclusions may be drawn from the results obtained in this study:

- HE can be used for grain refinement in aluminium and aluminium alloys. Grain size below 1 μm in technically pure aluminium and below 100 nm in the aluminium alloy can be obtained at a final strain smaller than 4.
- Due to the high ability of pure aluminium to recover, microstructure evolution proceeds through an increase of grain boundary misorientation without changing grain

size. As a consequence, mechanical properties remain almost constant within a wide range of strains.

• In the aluminium alloy with low stacking fault energy, the grains continuously decrease in size during consecutive extrusions, which leads to a grain size reduction much more effective than that observed in technically pure aluminium. As a consequence, mechanical properties are significantly improved.

• Taking into account the technological features of HE, one can conclude from the present study that this technique offers an alternative way of producing nano-metallic tubes and rods of aluminium alloys intended for light-weight applications.

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