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Microstructure and deformation behaviour of the ECAP Al-Mn-Sc-Zr alloy

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Abstract

The Al-Mn alloy stabilized by the addition of Sc and Zr was subjected to equal-channel angular pressing (ECAP) at room temperature. The microstructure evolution as a function of the number of ECAP passes was studied by transmission electron microscopy. The room temperature strength was characterized by microhardness measurements and tensile tests. The stability of the very fine-grained microstructure and strength characteristics were studied at elevated temperatures. Due to a good microstructure stability even after annealing at 500 °C the possibility of superplastic behaviour was tested. The value of the strain rate sensitivity parameter m exceeding the bottom limit of superplastic behaviour was found.

Key words: Al-alloys, ECAP, microstructure, mechanical properties, superplasticity

1. Introduction

The deformation behaviour of metallic materials depends significantly on their microstructure. Especially the microstructure refinement has been very popular during the last three decades and numerous entirely new processing routes have been developed. Some of them are based on rapid solidification (e.g., melt spinning and atomization) [1] and they produce generally materials far from the thermodynamic equilibrium state – supersaturated solid solutions [2], very fine grain size [3], etc. From the technological point of view, these methods are accompanied by a very important drawback – only a limited amount of material can be produced. In the sheet industry, there is one producing route representing a compromise between the microstructure refinement and mass production – twin-roll casting (TRC) [4, 5]. The resulting materials exhibit extended solid solution, finer grain size, and very small and finely distributed primary phases.

Another group of processing routes producing fine-grained microstructures relies on severe plastic deformation (e.g., ECAP, high pressure torsion, accumulative roll-bonding) [6–9]. These methods introduce a large amount of deformation energy into the material and result in an enormous grain refinement and also a reduction in the size of present particles. The effort of material scientists is directed to the improvement of parameters of these processing methods in order to achieve the microstructures as fine as possible and to ensure the stability of the refined microstructures when they are exposed to elevated temperatures. TRC materials can be a more suitable precursor for further processing by ECAP in comparison with direct chill-cast (DC) materials because of finer initial grain size and absence of coarse primary particles which could serve as preferential sites for crack initiation during ECAP.

Aluminium-manganese alloys are frequently used functional materials. Their mechanical properties are affected by adequate additions of Fe, Si or Cu which form coarse primary particles during solidification or secondary particles during precipitation treatment. Recovery and recrystallization processes can be significantly influenced by the addition of small amounts of Zr, Sc or both elements together. These elements form a fine dispersion of Al_3Zr, Al_3Sc or $Al_3(Zr_xSc_{1-x})$ particles [10], which influence significantly especially high-temperature mechanical properties of Al-Mn alloys. A complex research starts at the Department

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Table 1. Chemical composition of the investigated material

Element	Mn	Sc	Zr	Fe	Si	Al
Composition (wt.%)	1.35	0.27	0.23	0.072	0.034	balance

of Physics of Materials of the Charles University in Prague on the Al-Mn-based alloys prepared using various combinations of DC, TRC and ECAP. As the first part of this research, the results obtained on the Al-Mn-Sc-Zr alloy produced by DC casting followed by ECAP are presented.

2. Experimental

The chemical composition of the studied alloy is given in Table 1. The casting was annealed at 300 ℃ for 2 h in order to support the precipitation of the $Al_3(Sc_xZr_{1-x})$ particles and then pressed at room temperature and pressing speed of 10 mm min^{-1} through a die consisting of 2 channels (cross section 8×8 mm^2) intersecting at an angle of 90°. Samples with 1 to 8 passes were produced using the B_c rotation between subsequent passes [7].

The influence of the number of ECAP passes on the room temperature strength was studied using Vickers microhardness measurements (load 10 g) and using tensile tests with the strain rate of 10^{-3} s^{-1}. The tensile samples with the cross section of 1×5 mm^2 and gauge length of 17 mm were cut using a diamond saw parallel to the ECAP direction.

The stability of the material during annealing at elevated temperatures was tested using Vickers microhardness measurements. The samples were annealed in the temperature range between 20 and 400 ℃ for 30 min. The tensile tests performed in the same temperature range were used for the investigation of the plastic deformation and for the testing of possible superplastic behaviour at elevated temperatures. The strain rate change method was used for the evaluation of the strain rate sensitivity parameter m defined as $\partial \log \sigma / \partial \log \dot{\varepsilon}$, where σ represents the true stress and $\dot{\varepsilon}$ the true strain rate. After a pre-strain to 10 % of elongation at 10^{-3} s^{-1}, the strain rate was reduced to 10^{-4} s^{-1} and then gradually increased in steps up to the 3×10^{-2} s^{-1}. The parameter m was evaluated from individual steps as $\log(\sigma_2/\sigma_1)/\log(\dot{\varepsilon}_2/\dot{\varepsilon}_1)$, where σ_1 and σ_2 are the stress values corresponding to strain rates $\dot{\varepsilon}_1$ and $\dot{\varepsilon}_2$, respectively.

The microstructure was studied using transmission electron microscopy (TEM) both in the state after ECAP and after annealing at elevated temperatures. The TEM specimens were prepared by electropolishing of thin discs in the solution of 33 % of nitric acid in methanol at –20 ℃. The observation of thin foils, selected area electron diffraction (SAED), and energy dispersion analysis were performed in JEOL JEM 2000FX analytical electron microscope.

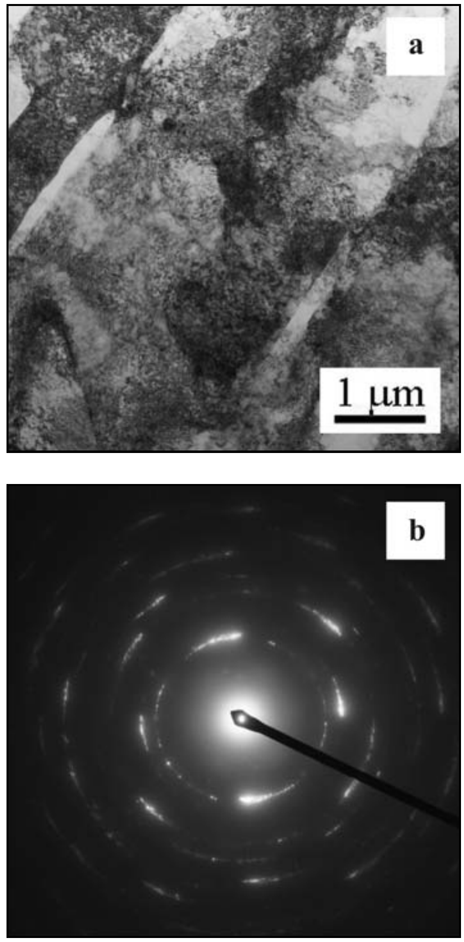

Fig. 1. The microstructure of the material subjected to 1 pass of ECAP (a) and corresponding SAED micrograph (b).

3. Experimental results

Figures 1–3 document the evolution of the microstructure with increasing number of ECAP passes. The microstructure of the material subjected to 1 pass (Fig. 1) is heavily deformed with partially fragmented original grains containing numerous subgrains with dense dislocation network. The electron diffraction reveals a strong texture. The fragmentation is much more intensive after 4 passes (Fig. 2), and the microstructure after 8 passes (Fig. 3) is very fine with the grain size in the sub-micrometer range. The distinctive character of diffraction spots and the splitting of diffraction segments confirm the increasing fraction

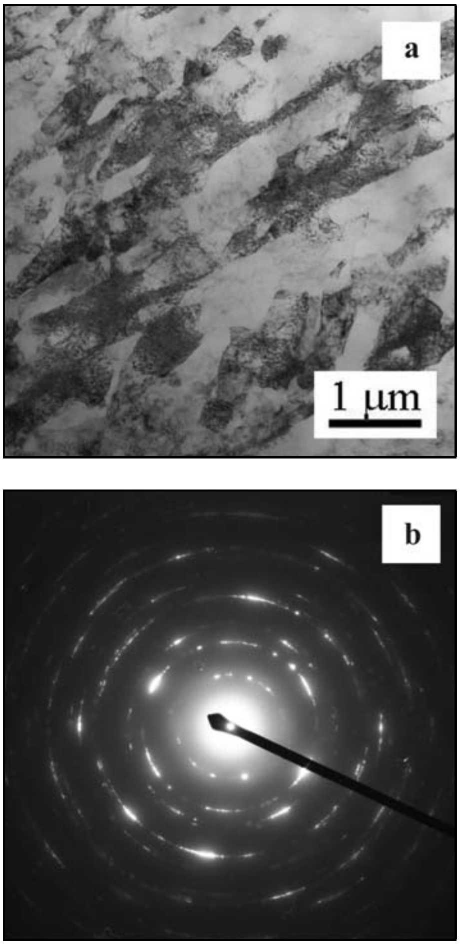

Fig. 2. The microstructure of the material subjected to 4 passes of ECAP (a) and corresponding SAED micrograph (b).

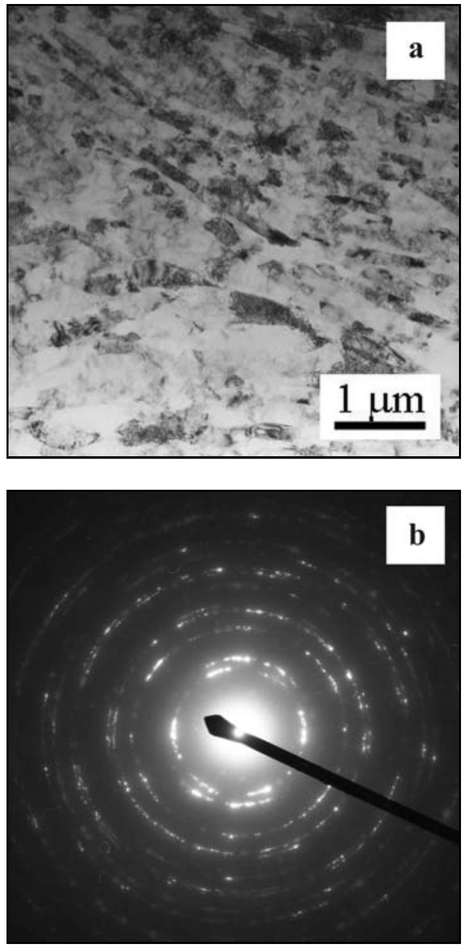

Fig. 3. The microstructure of the material subjected to 8 passes of ECAP (a) and corresponding SAED micrograph (b).

of high-angle boundaries. These results were verified by EBSD experiments [11]. It was found that strongly deformed regions without any visible grain boundaries and numerous low-angle boundaries were still present even after 8 passes.

The Vickers microhardness HV, the yield stress $R_{p0.2}$, the ultimate tensile strength R_m, and the ductility A values after various numbers of ECAP passes are summarized in Table 2. For comparison, the HV value of the material prior to ECAP was 54 MPa. It is evident that the strongest strengthening occurs already during the first ECAP pass.

The stability of the material subjected to various numbers of ECAP passes at elevated temperatures was verified using microhardness measurements. Table 3 shows the HV values after annealing at various temperatures for 30 min. Whereas the material after the 1st pass retains its microhardness up to 300 ℃, the HV of materials subjected to more ECAP passes starts to decrease already between 200 and 300 ℃. Consequently, the microhardness values decrease with

Table 2. Room temperature deformation characteristics of the material subjected to various numbers of ECAP passes

Number of passes	1	4	8
HV (MPa)	96	99	103
$R_{p0.2}$ (MPa)	262	296	308
R_m (MPa)	267	310	319
A (%)	8	5.8	5.8

increasing number of ECAP passes in materials annealed at 300 and 400 ℃.

The microstructure changes occurring during annealing at 400 ℃/30 min are documented in Figs. 4, 5. In comparison with the as-pressed state, the high angle boundaries are better developed and the grain size is close to 1 μm in the material subjected to 4 passes (Fig. 4) and slightly lower in the material sub-

Table 3. Microhardness values HV in MPa for the material subjected to various numbers of ECAP passes and annealed at elevated temperatures

Number of passes/ annealing temperature	1	4	8
20 ℃	96	99	103
100 ℃	100	103	101
200 ℃	100	109	106
300 ℃	98	95	89
400 ℃	88	76	70

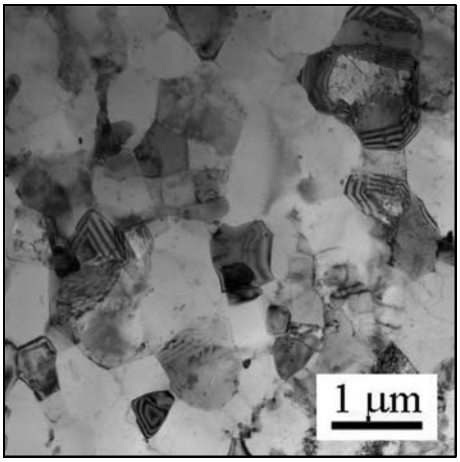

Fig. 4. The microstructure after annealing at 400 ℃/30 min of the material after 4 ECAP passes.

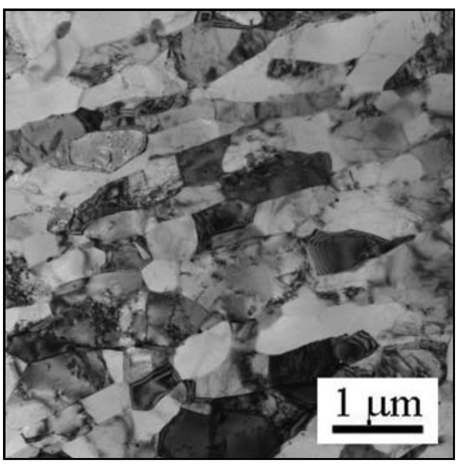

Fig. 5. The microstructure after annealing at 400 ℃/30 min of the material after 8 ECAP passes.

jected to 8 passes (Fig. 5). The micrographs taken at higher magnification revealed the presence of numerous low angle boundaries. A similar microstructure was surprisingly observed even after annealing 500 ℃/60 min (Fig. 6). The size of some grains ex-

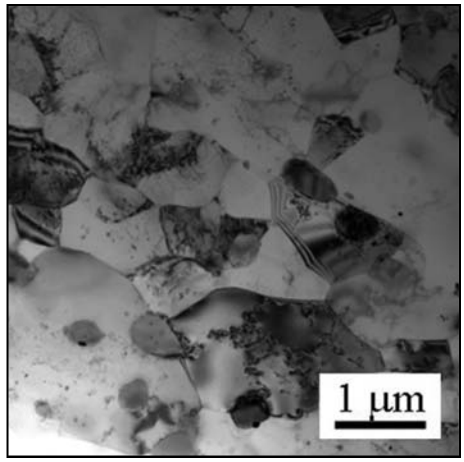

Fig. 6. The microstructure after annealing at 500 ℃/60 min of the material after 8 ECAP passes.

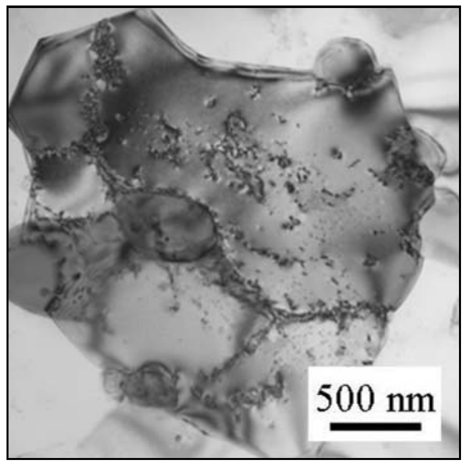

Fig. 7. Low-angle boundaries in the microstructure after annealing at 500 ℃/60 min of the material after 8 ECAP passes.

ceeds slightly 1 µm, however, finer grains with submicrometer size are still present. The material is not fully recrystallized and contains only sparsely distributed dislocations and numerous low angle boundaries in the grain interiors (Fig. 7). These results are in agreement with EBSD measurements revealing a relatively high fraction of low-angle boundaries [11].

The surviving fine-grained microstructure should be a good prerequisite of superplastic behaviour at high straining temperatures. Two main characteristics of superplasticity were measured – the strain rate sensitivity parameter m and ductility A. Table 4 summarizes the maximum values of the parameter m, the strain rates corresponding to this maximum of m, and ductility values. The values of $m = 0.3$ and ductility of 200 % are usually considered as the bottom limit of superplastic behaviour. The observed parameter m values reach this limit in materials subjected to 4 and

T a b l e 4. Deformation characteristics at various straining conditions

Number of passes	Temperature (℃)	Strain rate (s^{-1})	Maximum of m	Ductility A (%)
1	300	7×10^{-5}	0.1	19
1	400	10^{-4}	0.21	25
1	500	3×10^{-3}	0.25	43
4	400	10^{-2}	0.27	103
4	500	10^{-2}	0.34	135
8	450	10^{-2}	0.31	165

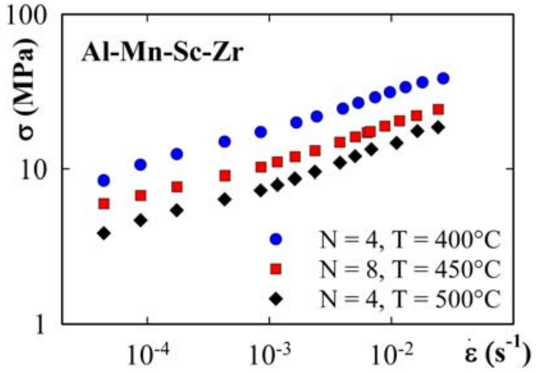

Fig. 8. The influence of the straining temperature on the strain rate dependence of the stress.

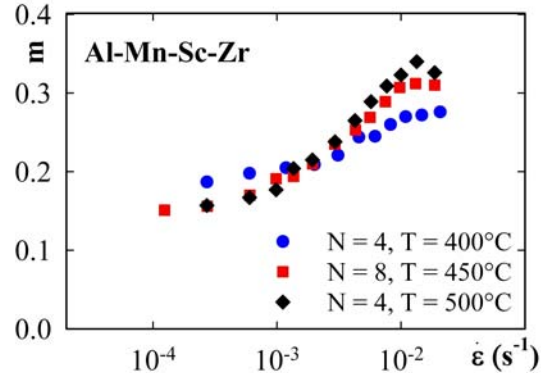

Fig. 9. The influence of the straining temperature on the strain rate dependence of the parameter m.

8 passes at the straining temperatures above 450 ℃. On the other hand, the ductility values remain below the superplastic limit. Nevertheless, it is necessary to mention that the ductility was measured as an integral value during a complicated tensile test with multiple strain rate changes.

Figure 8 shows the influence of straining temperature on the strain rate dependences of stress for the materials subjected to 4 or 8 ECAP passes. The maximum stress values observed at each strain rate during the strain rate change experiment were plotted. As can be seen on this figure the curves shift to lower stresses with increasing temperature. Figure 9 shows the corresponding strain rate dependences of the parameter m. All curves exhibit maxima of m at the strain rate of the order of $10^{-2}\,s^{-1}$. The maximum m values increase with increasing straining temperature.

4. Discussion

The Al-Mn-based alloys are used in automotive industry frequently at high temperatures or after high temperature exposure. The research of these alloys has been, therefore, performed in two directions:

a) the improvement of strength predominantly by strain hardening,

b) the strength and microstructure stability at high temperatures determined predominantly by the presence of particle forming elements and their influence on recovery and recrystallization processes.

Despite of the importance of these materials for technical applications the results concerning the influence of progressive processing methods are relatively scarce. Karlík et al. [12] investigated several direct-chill cast Al-Mn-based alloys with addition of Zr, Fe, and Si. The cold rolling with 89 % reduction (the equivalent strain $\varepsilon \sim 2.5$) resulted in the Vickers hardness values HV = 80 MPa and HV = 67 MPa in the Al-Mn-Zr-Si-Fe and Al-Mn-Zr alloys, respectively. For the twin-roll cast Al-Mn-Si-Fe alloy, microhardness values HV = 80 and 100 MPa were reported after cold rolling to the equivalent strain $\varepsilon = 2.1$ and 3.5, respectively [13]. The Al-Mn alloy with high content of Si and Fe processed by accumulative roll bonding (ARB) exhibited the increase of tensile strength from 200 MPa in the initial state to about 300 MPa after 5 ARB cycles (the equivalent strain $\varepsilon \sim 4$) [14]. ECAP (the angle between channels equal to 90°) of the commercial Al-Mn-Si 3103 alloy resulted in the maximum microhardness close to 70 MPa after 6 passes [15]. Similar maximum microhardness value of 76 MPa was reported after 6 ECAP passes with the angle between channels equal to 120° [16]. Finally, the same alloy processed by 12 cycles of continuous confined strip shearing (the equivalent strain $\varepsilon \sim 7$) exhibited the maximum microhardness of 67 MPa [17].

The comparison of our results with literature data indicates that the ECAP processing route used in our experiment is very effective and yields the highest microhardness and tensile strength values. It is evident that the main changes occur already during the first ECAP pass. High dislocation density is responsible for the strength increase. Subsequent passes result only in a moderate increase in strength in agreement with results reported in [14, 16, 17]. The TEM investigation revealed that recovery processes resulting in the rearrangement of dislocations and formation of both low- and high-angle boundaries occurred during these ECAP passes. This conclusion is supported by the EBSD experiments showing fragmentation of initial grains and formation of high-angle grain boundaries [11].

A reduction in strength characteristics after annealing at elevated temperatures is a typical feature of Al-Mn-based alloys. Cieslar et al. [13] observed a drop in microhardness of the cold-rolled Al-Mn-Si-Fe alloy starting at temperatures above 200 °C and occurring in two stages – the border between both stages was close to 400 °C. TEM investigation revealed that recovery of dislocation structure was the dominant softening process in the "low-temperature" stage where the HV values decreased to about 70 MPa. Further reduction of HV to values close to 40 MPa in the "high-temperature" stage (above 400 °C) was attributed to recrystallization and grain growth. A very rapid drop of HV to values below 40 MPa was reported in the AA3103 alloy after severe plastic deformation using continuous confined strip shearing and annealing already at 350 °C [17]. EBSD experiments revealed abnormal grain growth and grains of the size exceeding 100 μm were observed after 300 s of annealing.

Table 3 shows only a moderate drop in HV to values above 70 MPa at temperatures up to 400 °C. This behaviour corresponds to the first stage of softening reported in [13] and similar explanation can be accepted. The onset of this drop shifts to lower temperatures with increasing number of ECAP passes. This phenomenon can be explained assuming that a large energy stored during ECAP creates a strong driving force for restoration processes at elevated temperatures. The increasing number of ECAP passes should thus enhance this driving force and accelerate the restoration processes. Nevertheless, contrary to AA3103 alloy investigated in [17], no abnormal grain growth was observed and the grain size in the range of 1 μm is retained even at 500 °C (Fig. 6). Similarly to other microcrystalline Al-based alloys [18, 19], the presence of $Al_3(Sc,Zr)$ phase particles has a positive stabilizing effect on the microcrystalline structure.

High temperature tensile tests were performed in order to test the possibility of superplastic behaviour. Table 4 shows that the maximum ductility is below the lower limit of superplasticity (200 %). On the other hand, the maximum value of the strain rate sensitivity parameter $m = 0.34$ exceeds the lower limit of superplasticity ($m = 0.3$) [20]. Such deformation behaviour can result either from the operation of grain boundary sliding typical for superplastic deformation [20] or from the mechanism of viscous glide of dislocations [21]. The sigmoidal character of the strain rate dependence of the parameter m (Fig. 8) supports clearly the first mechanism. An unambiguous evidence for the operation of grain boundary sliding was obtained using atom force microscopy measurements reported in [11]. It can be thus concluded that the material behaves superplastically and low values of ductility result from rather complicated course of tensile tests where straining occurred at variable strain rates and not at the strain rate corresponding to the maximum of the parameter m. The experiment performed at the optimum strain rate revealed ductility exceeding 300 % [11].

5. Conclusions

1. The microstructure of the Al-Mn alloy stabilized by the addition of Sc and Zr can be refined to the sub-micrometer range using ECAP. The microstructure evolution as a function of the number of ECAP passes is relatively slow and even after 8 passes the microstructure contains a large fraction of low angle boundaries.

2. The application of ECAP results in an increase of room temperature strength characteristic. The main increase occurs already during the first pass when the largest increase in dislocation density occurs. Further ECAP passes result in an only moderate strength increase. Dislocations are rearranged and fragmentation of original grains through the formation of low- and high-angle boundaries occurs.

3. The microstructure is extremely stable at elevated temperatures. The grain size after annealing at 400 °C remains mostly below 1 μm and even annealing at 500 °C does not result in a significant grain growth. The microstructure after such annealing is not fully recrystallized and contains both sparse individual dislocations and dislocations arranged into low-angle boundaries.

4. The measurement of the strain rate sensitivity parameter m reveals the ability of the material after at least 4 ECAP passes to behave superplastically at temperatures above 450 °C.

Acknowledgement

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