SUPERPLASTIC BEHAVIOUR OF THE Al-Mg-Sc-Zr ALLOY PROCESSED BY ECAP

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A very fine-grained Al-1.5wt.%Mg alloy stabilised by small amounts of Sc and Zr was prepared using ECAP. The values of ductility and strain rate sensitivity parameter m corresponding to superplastic behaviour were found in tensile tests at temperatures above 573 K. The optimum superplastic behaviour was observed at strain rates of the order of 10^{-2} s^{-1}. The surface examination of slightly strained samples by atom force microscopy revealed that grain boundary sliding did not occur at all interfaces. The diversity of misorientation angles is probably responsible for this effect.

K e y w o r d s: Al-Mg-Sc-Zr alloy, ECAP, submicrocrystalline microstructure, superplasticity, grain boundary sliding

1. Introduction

The grain size is the most important structural parameter for superplastic materials. Its reduction results in an improvement of superplastic characteristics and in a shift of superplastic behaviour to higher strain rates [1]. This shift is of great importance for the prospective utilisation of superplasticity in industrial forming processes as it reduces the forming time of individual components to reasonable values. The minimum grain size achievable in superplastic Al-based alloys produced by conventional processing routes is usually close to 10 μm and this leads to the best superplastic characteristics at strain rates of the order of 10^{-4} to 10^{-3} s^{-1} (e.g. [2–4]). To shift superplasticity to strain rates above 10^{-2} s^{-1} the grain size has to be reduced to about 1 μm. Such grain size may be produced by a powder metallurgical route (e.g. [5–7]), which is followed by a suitable powder consolidation.

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However, this processing is rather expensive and brings serious difficulties (porosity, internal oxidation) that make such materials hardly applicable in industrial processes.

At present, a new processing route – equal channel angular pressing (ECAP) is available for the production of materials with grain sizes in submicrometer range. During ECAP, the ingot is pressed through a special die consisting of two channels intersecting usually at an angle of 90° [8]. Although a large shear deformation occurs during each pass of the ingot through a die the shape of the pressed ingot is the same as the initial one. Pressing can be performed repeatedly, so that the shear stress cumulates. Very fine subgrains are usually formed during the first pass, following passages result into an increase in the boundary misorientation and a creation of fine-grained structure [9]. The grain size produced by ECAP is usually between 0.1 and 1 μm and depends on the parameters of pressing (die geometry, rotation between passes, pressing temperature) and on the chemical and phase composition of pressed materials [10, 11].

The minimum grain size in ECAP pure Al is usually slightly above 1 μm [12]. The addition of Mg reduces the rate of recovery and leads to further grain refinement down to about 0.2 μm [13]. Unfortunately, a significant grain coarsening starts above 400 K and the grain size exceeds 100 μm at temperatures where superplasticity is usually observed in Al-based materials [12, 13]. The fine-grained structure has to be, therefore, stabilised by the addition of some dispersoid forming elements. Small particles of the metastable β' Al_3Zr phase ensure a good stability of fine-grained superplastic Al-based alloys up to temperatures of 750 K [14]. However, a transformation of this metastable phase to the stable modification with much larger particle size occurs at higher temperatures and results in a decrease in the retarding force for grain growth. Alternatively, scandium may be used for similar purposes. It forms a stable Al_3Sc phase of the same structure as the β' Al_3Zr phase [15]. The Al_3Sc phase does not undergo any phase transformation but fine Al_3Sc particles coarsen at temperatures close to 773 K [16] what also results in a significant grain growth. The best stability of the fine-grained structure may be reached by the simultaneous addition of small amounts of Zr and Sc [16]. This composition was selected in our experiments.

It is usually believed that interfaces have a high-angle character in most Al-based alloys after a sufficient number of ECAP passes [17]. Increasing number of studies using electron back scattered diffraction (EBSD) have nevertheless revealed that the fraction of high-angle boundaries is saturated in the range of 70–85 % [10], i.e. low-angle boundaries are still present. Grain boundary sliding as the main deformation mechanism of superplasticity [1] depends on the misorientation angle and is much higher at general high-angle boundaries than at low-angle ones. In case of materials containing both types of interfaces grain boundary sliding can be distributed inhomogeneously and does not occur at all interfaces. This inhomogeneity may significantly influence the phenomenological description of superplastic deformation.

The main objective of the present paper is to verify the possibility of low temperature or high strain rate superplasticity of an ECAP Al-Mg-Sc-Zr alloy. The attention was also paid to the microstructure of samples after straining on superplastic conditions.

2. Experimental material and procedure

The experiments were performed at an Al-1.5Mg-0.21Sc-0.18Zr (in wt.%) alloy. The billets having a square cross-section of 14×14 mm^2 were repeatedly pressed through a die consisting of a single channel bent through the angle of 90°. Totally 6 passes were performed with B_c rotation (90° in the same sense between each pass) at the temperature of about 423 K.

Following ECAP, tensile specimens with the gauge length of 18 mm and the cross-section of 1×8 mm^2 were cut parallel to the Y plane (side plane perpendicular to the shear plane). A special type of tensile tests was chosen. Specimens were pre-strained to the elongation of 10 % at the initial strain rate of 10^{-3} s^{-1}. After achieving a quasi-steady state at this elongation, the strain rate was reduced to 2×10^{-5} s^{-1} and then gradually enhanced up to 2×10^{-2} s^{-1} (route I). At other samples, the strain rate was after the same pre-strain only gradually enhanced up to 7×10^{-2} s^{-1} (route II). The ratio of subsequent strain rates was between 1.4 and 1.66. The strain rate sensitivity parameter m was evaluated from individual strain rate changes (not as a slope of the $\log \sigma / \log \dot{\varepsilon}$ dependence) according to formula (1)

$$m = \log(\sigma_2/\sigma_1)/\log(\dot{\varepsilon}_2/\dot{\varepsilon}_1), \qquad (1)$$

where σ_2 and σ_1 are stress values corresponding to strain rates $\dot{\varepsilon}_2$ and $\dot{\varepsilon}_1$, respectively. Both types of tests were performed in the temperature range between 573 and 773 K. The method of strain rate changes where the whole $\log \sigma / \log \dot{\varepsilon}$ dependence is obtained on only one sample has several advantages in comparison with straining of a set of individual samples at constant strain rates:

– it needs much smaller amount of experimental material,

– it is less sensitive to prospective microstructure inhomogeneities as these defects are common for the whole strain rate dependence,

– it needs much shorter time of straining.

The last item is very important in case of materials the microstructure of which is not fully stable at the deformation temperature used and the time of straining influences the microstructure and, consequently, mechanical behaviour of materials tested. The time of straining in our experiments was about 1 hour for route I and below 10 minutes for route II.

The internal microstructure was investigated on thin foils using TEM. The surface relief of strained samples was studied qualitatively using light and scanning electron microscopy (SEM) and quantitatively using atom force microscopy (AFM).

3. Experimental results

3.1 Deformation tests

Figure 1 shows the true stress vs. true strain rate dependences determined from strain rate change tests (route I) for various straining temperatures. All curves may be divided in two regions – the region of decreasing slope with decreasing strain rate (non-superplastic region I) at low strain rates and the region of maximum slope (superplastic region II) at strain rates above 10^{-3} s^{-1}. The non-superplastic region III observed usually at high strain rates was not reached in our material within the range of strain rates used. Figure 2 brings the dependence of the parameter m on the true strain rate for selected temperatures. Considering the value of $m = 0.3$ as a bottom limit of superplasticity it may be concluded that the material exhibits superplasticity even at the temperature of 573 K. The curves obtained at temperatures above 673 K are very similar and the maximum values of m over 0.5 were observed at true strain rates of the order of 10^{-2} s^{-1}.

The strain rate change method yields the best results in case of materials the microstructure of which remains constant during the whole test. In case of microstructure instability during the test the parameter m values might be influenced by the strain history. The comparison of $\log\sigma/\log\dot{\varepsilon}$ dependences obtained using different routes I and II corresponding to different times of straining (Fig. 3) documents a slight influence of strain history – route I including straining at low strain rates

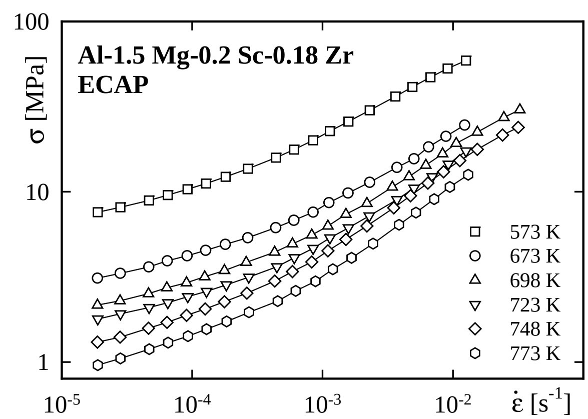

Fig. 1. The $\log\sigma/\log\dot{\varepsilon}$ dependences for various temperatures.

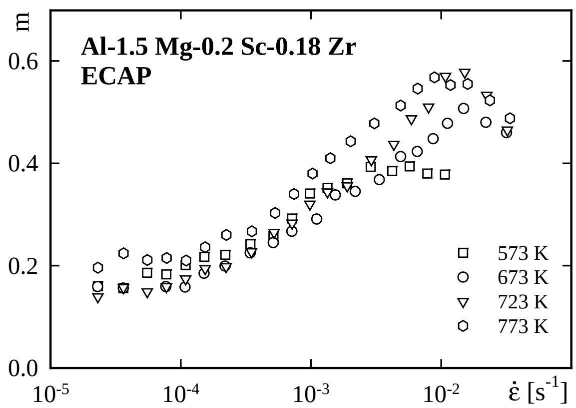

Fig. 2. The strain rate dependence of the strain rate sensitivity parameter m.

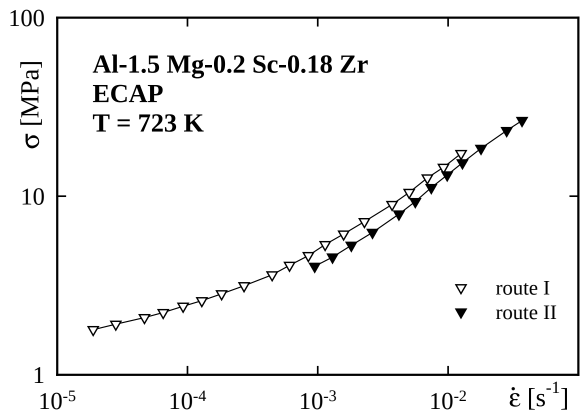

Fig. 3. The influence of strain history on the $\log \sigma / \log \dot{\varepsilon}$ dependence.

results into slightly higher stress, probably due to a small grain coarsening during straining at low strain rates. The difference in the parameter m obtained from both routes is nevertheless negligible (Fig. 4) so that the method of strain rate changes may be considered as a reliable method for the evaluation of the parameter m.

Despite of a complicated strain history the samples strained at 723 and 773 K exhibited elongation over 900 %. The tests had to be interrupted at this elongation because of a limited length of the constant temperature zone.

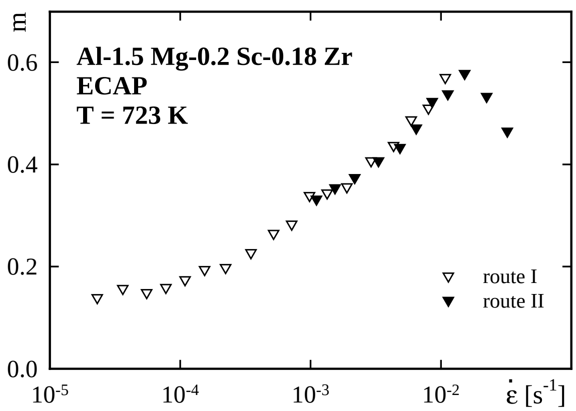

Fig. 4. The influence of strain history on the parameter m.

3.2 Microstructure investigation

The microstructure of the material after ECAP (Fig. 5) is very fine, composed mostly of deformed and recovered parts. Many interfaces have a low-angle char-

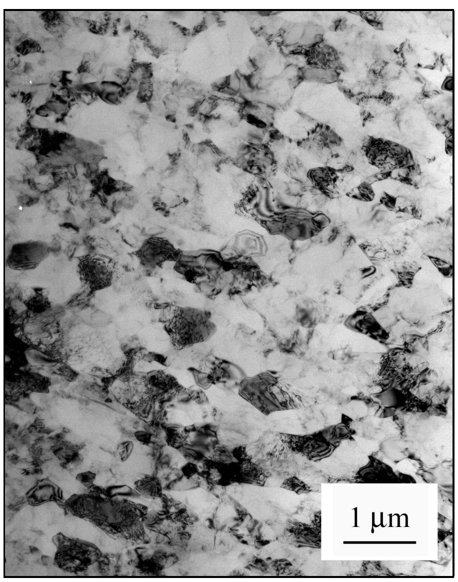

Fig. 5. TEM micrograph of the alloy after ECAP.

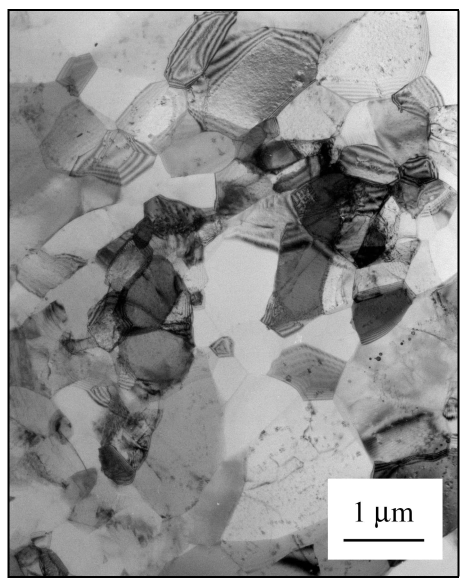

Fig. 6. TEM micrograph of the alloy after annealing at 723 K for 1 hour.

acter and the typical size of subgrains is between 0.2 and 0.3 μm. Some larger recrystallised grains of the size below 1 μm are also present.

Annealing at temperatures where the alloy exhibits superplastic characteristics modifies its microstructure. The microstructure of the sample annealed at 723 K for 1 hour (Fig. 6) consists predominantly of recrystallised grains of the size close to 1.5 μm, however, a part of microstructure remains still only recovered with subgrains of the typical size of 0.5 μm. Very fine particles (50 nm) pinning the lattice dislocations were found inside some grains. These particles contain Zr and Sc and are formed probably by the $Al_3(Zr, Sc)$ phase.

In order to elucidate the deformation mechanism the polished samples were strained to elongations of 10 or 20 %. Their surface was then investigated using several microscopic methods at various magnifications. A general feature of microstructure at all magnification levels is a remarkable inhomogeneity. Figure 7 shows deformation bands on the surface of the sample strained to 20 % at superplastic conditions ($T = 700$ K, $\dot{\varepsilon} = 10^{-2}$ s^{-1}). The bands inclined about 45° to the tensile axis and parallel to the shear plane were identified as elongated regions of different height with the vertical difference between "valleys" and "ridges" exceeding usually 1 μm. The typical distance between them is close to 100 μm.

The surface relief formed during superplastic straining results predominantly from grain boundary sliding. In a hypothetical case when each interface is involved in the sliding process all boundaries should be visualised already after a small strain and the distance between them should thus determine the grain size. The SEM

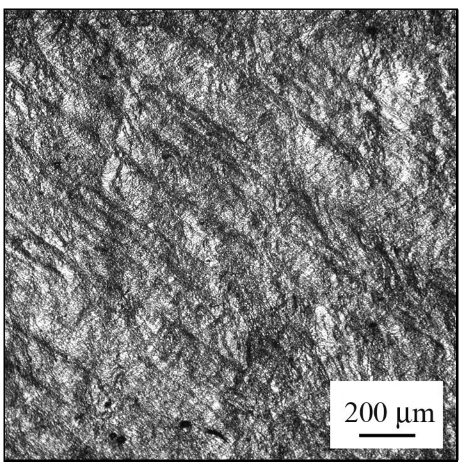

Fig. 7. The deformation bands formed during straining to the elongation of 20 %, light microscopy, tensile axis horizontal.

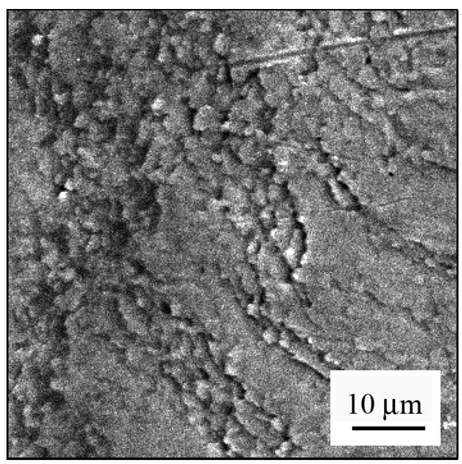

Fig. 8. The structure inhomogeneity after straining to the elongation of 10 %, SEM, tensile axis horizontal.

micrograph of the sample strained to 10 % ($T = 700$ K, $\dot{\varepsilon} = 10^{-2}$ s^{-1}) documents that the surface consists both of regions where the boundaries are clearly exposed (the distance between exposed boundaries is mostly between 1 and 2 μm) and regions that remain flat (Fig. 8). There are two possible explanations for these flat regions:

– The microstructure is inhomogeneous and contains both fine grains of the size close to 1 μm and coarse grains of the order of 10 μm.

– The microstructure is homogeneous but grain boundary sliding occurs inhomogeneously, so that not all boundaries are exposed and the distance between exposed boundaries does not represent the grain size.

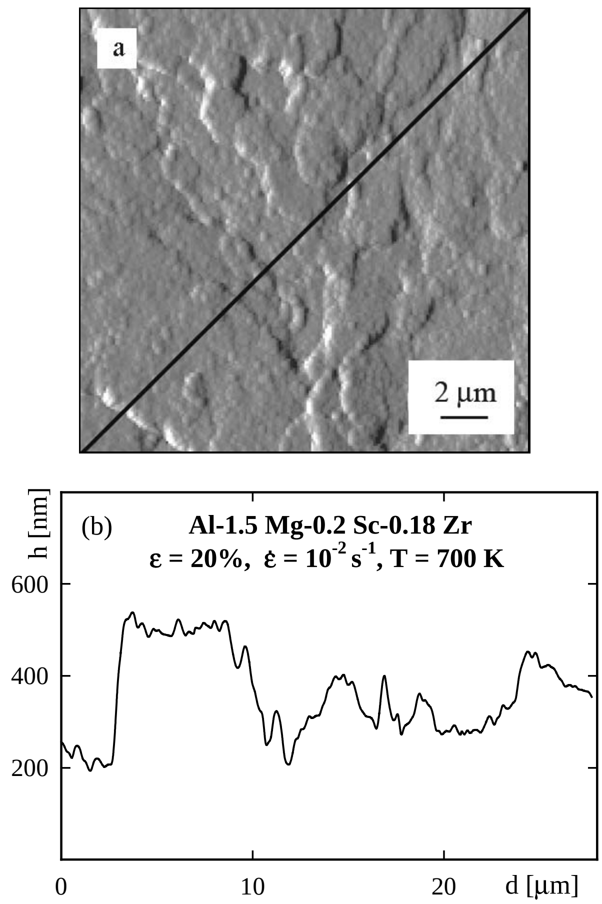

Fig. 9. The structure after straining to the elongation of 20 %, AFM, tensile axis horizontal, a) reconstructed surface relief, b) profile along the line in Fig. 9a.

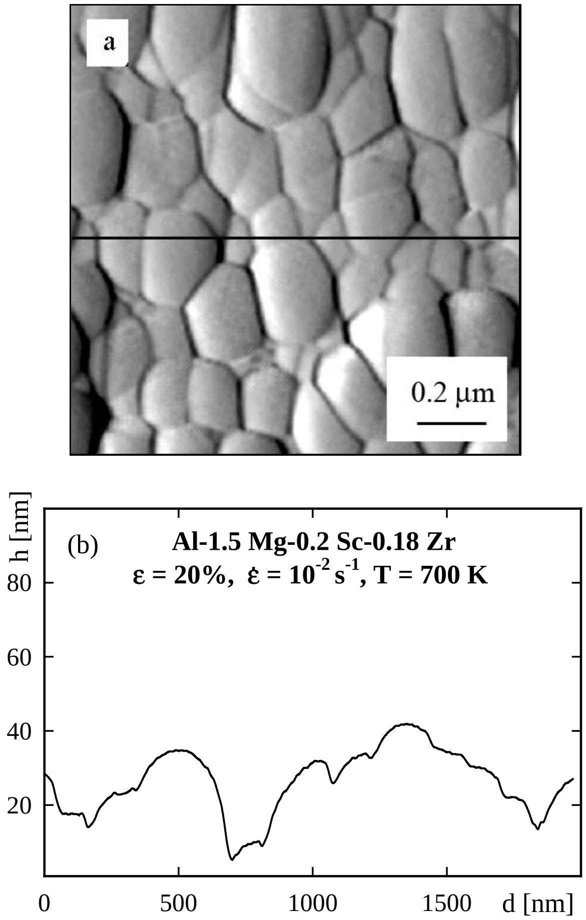

Fig. 10. The structure detail after straining to the elongation of 20 %, AFM, tensile axis horizontal, a) reconstructed surface relief, b) profile along the line in Fig. 10a.

In order to distinguish between both hypotheses atom force microscopy (AFM) experiments were performed at the sample strained to 20 % ($T = 700$ K, $\dot{\varepsilon} = 10^{-2}$ s^{-1}). Selecting the 20 × 20 μm area the AFM pictures show the presence of both fine and coarse "grains" (Fig. 9a). The mutual shifts of neighbouring "grains" along the direction perpendicular to the sample surface are mostly between 100 and 300 nm (Fig. 9b). However, the detailed inspection of surface profiles in Fig. 9b suggests that smaller objects exist within exposed coarse "grains". The AFM picture of the 2 × 2 μm area clearly shows the existence of fine objects of the size about 0.3 μm (Fig. 10a) with mutual shifts in the direction perpendicular to the sample surface typically between 10 and 20 nm (Fig. 10b).

4. Discussion

The experimental results obtained at the Al-1.5Mg-0.21Sc-0.18Zr alloy document unambiguously that the anticipated grain refinement due to ECAP has a positive influence on superplastic behaviour – superplastic characteristics are shifted both to lower temperatures and higher strain rates in comparison with superplastic Al-based alloys prepared through classical thermomechanical treatment [2–4]. The results obtained have to be compared with those obtained at similar Al-Mg-based alloys prepared using ECAP. Furukawa et al. [18] studied Al-0.2wt.%Sc alloys with variable content of Mg from 0 to 5 wt.% and found the best superplastic characteristics for 3 wt.% Mg. Maximum values of ductility above 2000 % and parameter m close to 0.5 were found at 673 K and 10^{-2} s^{-1}. A lower ductility was observed at the Al-3Mg-0.2Sc alloy by Komura et al. [19]. A dramatic drop of ductility with increasing deformation temperature is a typical feature of these ternary alloys and results from grain coarsening occurring at high temperatures [18, 19]. The maintenance of very high values of ductility (maximum of 1680 %) and parameter m (close to 0.5) even at very high temperature (773 K) was on the other hand found at a quaternary Al-3Mg-0.2Sc-0.12Zr alloy [16], i.e. at the alloy with higher Mg and lower Zr content than in our material. Ductility data presented in [16, 18, 19] might be rather overestimated as very short samples were used in all these experiments and it is difficult to determine exactly the initial gauge length. Our experiments using samples with the gauge length of 18 mm yield much more reliable data. The maximum elongation of 900 % achieved without any necking suggests that our material might exhibit also a very high ductility.

Our experiments aimed at the evaluation of the strain rate sensitivity parameter m are much more precise than those presented in [16, 18, 19]. The strain rate change method yields more experimental data and enables a better determination both of the position of superplastic region II and the value of m. Additionally, this method reduces significantly the influence of grain coarsening during straining at high temperatures. In case that individual samples are used for each strain rate [16, 18, 19] the values of σ used for the construction of $\log\sigma/\log\dot{\varepsilon}$ plots correspond to different times of straining and, therefore, to different values of grain size. This reduces the effective strain rate sensitivity especially in the region of low strain rates. The comparison of our results obtained using different routes I and II shows that even a small difference in strain history influences slightly the values of σ, however, the parameter m values are not modified.

The quaternary ECAP Al-Mg-Sc-Zr alloy retains high superplastic characteristics up to 773 K. Such outstanding properties reflect the best microstructure stability among ECAP Al-Mg-based alloys resulting from the presence of mixed $Al_3(Zr_xSc_{1-x})$ particles [15], which pin grain boundaries and retard grain coarsening. In order to retain fine-grained structure up to very high temperatures the pinning particles have to be small and stable, i.e. resistant to growth and dissolution. Recent investigation of the stability of the $Al_3(Zr_xTi_{1-x})$ particles revealed that a partial substitution of Ti for Zr atoms in this phase resulted in a lower lattice mismatch to the matrix and thus in a lower rate of particle growth at high temperatures [20, 21]. Although no detailed experiments were performed at the Al-Zr-Sc system a similar influence of substitution of Sc for Zr atoms might slow down the rate of ripening of complex $Al_3(Zr_xSc_{1-x})$ particles in comparison with Al_3Zr and Al_3Sc particles. The small size of $Al_3(Zr_xSc_{1-x})$ particles and, consequently, the high retarding force for grain growth should be retained up to high temperatures.

Grain size is the only structure parameter appearing in the phenomenological equation describing superplastic deformation. Its importance follows from the hypothesis that grain boundaries are the main "carriers" of superplastic deformation and grain boundary sliding is the deformation mechanism with the greatest contribution to overall strain. However, it can be hardly expected that all interfaces are involved in grain boundary sliding process at a given moment and that the individual grain shears occur independently. According to some topological models of superplasticity grain boundary sliding is localised at a limited number of favourable boundaries and groups of grains (rather than individual grains) slide as entities [22, 23]. In this concept the grain size loses its importance and should be replaced in phenomenological equations by the size of sliding units.

The main objective of the surface relief investigation was to estimate the homogeneity of grain boundary sliding distribution and to verify the concept of co-operative sliding. The samples were strained to strains 10 to 20 % that are relatively low in comparison with ductility of this material. The results of these investigations thus describe the development of deformation at the initial part of straining. Significantly higher strains cannot be nevertheless used in this investigation because of a limited depth of focus of light microscopy and a limited vertical shift of the tip in AFM. Additionally, the estimate of mutual shifts of neighbouring grains is possible only at low strains where individual grains retain their neighbours and no rearrangement of grains occur over a larger distance.

All experimental methods used in the surface relief investigation show clearly that remarkable mutual shifts (up to 200 nm) occur between objects of the size slightly above 1 μm. This size corresponds well to the size of recrystallised grains observed using TEM and represents thus the structural parameter controlling deformation behaviour. On the other hand, the mutual shifts of smaller objects observed using AFM inside these units are about one order lower. It might be supposed that these smaller objects are subgrains and the low-angle character of boundaries dividing them results into a much lower sliding. Nevertheless, further experiments under protective atmosphere are needed to exclude the possibility that the smaller objects result from surface contamination during straining at high temperatures.

5. Conclusions

1. The Al-1.5Mg-0.21Sc-0.18Zr (in wt.%) alloy was prepared using ECAP with a submicrocrystalline microstructure.

2. The alloy exhibits superplastic behaviour already at 573 K. The best characteristics were found at temperatures between 673 and 773 K and strain rates of the order of 10^{-2} s^{-1}. The maximum of the parameter m reaches the value of 0.6.

3. The microstructure of the as ECAP material is not recrystallised and contains predominantly low angle boundaries. The subgrain size is deeply below 1 μm. Annealing at 723 K results into a predominantly recrystallised microstructure with the grain size slightly above 1 μm.

4. The strain distribution after a small elongation at superplastic conditions is inhomogeneous. Deformation bands were observed using light microscopy and grain boundary sliding occurring only at some preferential interfaces was established in SEM experiments.

5. Atom force microscopy revealed that objects of the size slightly above 1 μm slide as units and their mutual shifts are typically of the order of 0.1 μm. Mutual shifts of smaller objects (probably subgrains) of the size of about 0.3 μm are about one order lower. Different structure of various interfaces is believed to be a reason for this inhomogeneity.

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