Rev. Adv. Mater. Sci. 31 (2012) 138-144

CREEP IN Al SINGLE CRYSTAL PROCESSED BY EQUAL-CHANNEL ANGULAR PRESSING

P. Král[1], J. Dvořák[1], P. Šedá[2], A. Jäger[2] and V. Sklenička[1]

[1]Institute of Physics of Materials, Academy of Sciences of the Czech Republic,
Žižkova 22, CZ-616 62 Brno, Czech Republic
[2]Institute of Physics, Academy of Sciences of the Czech Republic, Na Slovance 2, CZ-182 21 Praha 1,
Czech Republic

Received: April 20, 2012

Abstract. In this work the creep behavior and microstructure changes in aluminum single crystal during equal-channel angular pressing (ECAP) were investigated in order to understand the relationships between microstructure and creep behavior. The investigated crystal was oriented with {111} parallel to the shear plane and <110> was parallel to the direction of shear. During ECAP by route A at room temperature single-crystalline sample was subsequently transformed into the polycrystalline material. Microstructure of samples processed by 1 and 4 ECAP passes was characterized by transmission electron microscope (TEM) and scanning electron microscope (SEM) equipped with the electron backscatter diffraction unit (EBSD). Microstructure analyses showed inhomogeneity of microstructure after ECAP and the increase in the fraction of HAGB's (θ>15°) with increasing number of ECAP passes. Tensile creep tests at temperature of 373K and at applied stress of 50 MPa were performed on the Al single crystal after one and four ECAP passes. It was found that the creep behavior is influenced by cooperative grain boundary sliding along mesoscopic shear bands.

1. INTRODUCTION

Many different experiments were performed on polycrystals processed by severe plastic deformation (SPD). However, only few reports [1-5] investigated evolution of microstructure of single crystals processed by ECAP. The most of works have studied single crystals, however, only after one ECAP pass. Miyamoto *et al.* [3] processed a series of Cu single crystals by 1 ECAP pass. They found that the end orientation, macroscopic heterogeneity and dislocation structures of studied Cu single crystals can be divided into three groups. Their results suggested that the evolution of microstructure relates to the degree of concentration of activated slip systems with respect to the shear plane. Han *et al.* [6] studied a single crystal with special orientation of the (111) plane and the [110] direction with respect to the intersection plane of ECAP die. They suggested that the shear deformations both parallel and perpendicular to intersection plane play important roles in the ECAP process of single crystals.

Creep behavior probably belongs to the fewest examined properties of the ECAP materials. Creep of materials after SPD was investigated on pure metals [7-10], alloys [11-15] and Cu-Al_2O_3 composite [8,10] but the influence of SPD on creep behavior of metals is still unclear.

The most of creep experiments have been performed on the polycrystalline materials [7-15] describing creep behavior of materials subjected only certain number of ECAP passes. Thus, there are only a few reports [7,11,12,16] documenting the creep properties and creep mechanisms of the

Corresponding author: P. Král, e-mail: pkral@ipm.cz

ultrafine-grained (UFG) materials processed by different number of ECAP passes. Our previous experiments conducted on pure polycrystalline Al revealed that the creep resistance of pure aluminum at 473K is considerably improved after one ECAP pass in comparison with coarse grained material, but repetitive pressing leads to a noticeable decrease in the creep properties of the ECAP material. Nevertheless, the creep resistance of pure Al after ECAP was higher than unpressed material even after 12 ECAP passes [17]. The decrease of creep resistance with increasing number of ECAP passes could be explained by increasing contribution of grain boundary sliding to the total creep strain. However, from the stress dependences of the minimum creep rates it was proposed that the operating creep mechanism is intragranular dislocation process. Kawasaki *et al.* [15] examined pure Al after 4 ECAP passes. Based on the texture measurements they demonstrated that creep occurs through an intragranular dislocation process with no significant contribution of diffusion creep. The creep behavior of ECAPed metals can be influenced by microstructure stability (grain size), presence of precipitates in the microstructure and their coherency, presence of the non-equilibrium grain boundaries, history of initial material before ECAP *etc*. The creep mechanisms in metals processed by ECAP are not well understood so far. From this reason it is useful to study the creep behavior on single crystals of pure metals where all grain boundaries are created only by ECAP.

2. EXPERIMENTAL MATERIAL AND TECHNIQUES

Randomly oriented Al single crystal was grown by modified Bridgeman technique in floating zone furnace from 99.5% polycrystalline Al. The starting polycrystalline Al was placed in graphite crucible and pulled with a speed of 18.8 mm/h under argon atmosphere. A single crystalline sample for ECAP was machined by electric discharge machining. The crystallographic orientation of single crystal was determined using Laue backscatter reflection technique. Slip plane of the single crystal (111) was parallel to the theoretical shear plane of the ECAP die and the slip direction [110] was parallel to the shear direction of ECAP process. The single crystal was divided in two parts. The billets were processed by ECAP at room temperature using a die that had an internal angle of 90° between the two parts of the channel and an outer arc of curvature of ~ 20°, where these two parts intersect. The pressing speed was 10 mm/min. The billets were subsequently pressed by route A by 1 and 4 ECAP passes. In the route A the sample is pressed repetitively without rotation. The route A was selected because the investigated section XZ (X parallel with the last pressing direction and Z perpendicular to the bottom of the channel) of samples (see [18]) is in the same position after 1 and 4 ECAP passes with respect to the position of the sides of ECAP die. The constant load creep tests in tension were performed at 373K and under applied stress of 50 MPa. The tensile samples, having gauge lengths of 10 mm and cross-sectional areas of 8 x 3.2 mm, were machined from billets parallel to the section XZ. The creep testing was conducted in an environment of purified argon with the testing temperatures maintained to within ± 0.5K of the desired value. All of the tests were continued until fracture. The microstructure was examined by TEM and scanning electron microscope Jeol 6460 equipped by electron back scatter diffraction (EBSD). The surface of sample processed by 4 ECAP passes was investigated by laser confocal microscope Olympus Lext. The TEM foils were prepared by twin-jet electropolishing facility and by ion polishing using FEG/ SEM Tescan equipped by focused ion beam (FIB).

3. RESULTS AND DISCUSSION

3.1. Creep behaviour of Al single crystal processed by ECAP

Representative creep curves for Al single crystal processed by 1 and 4 ECAP passes by route A are shown in Figs. 1a and 1b. Standard ε versus t creep curves in Fig. 1a can be easily replotted in the form of the instantaneous strain rate $d\varepsilon/dt$ versus t as shown in Fig. 1b. The creep tests were performed at 373K and run up to the final rupture of the creep specimens. The creep curves show that the creep resistance of the sample after 1 ECAP pass is higher and the minimum creep rate is lower in comparison with Al single crystal processed by 4 ECAP passes. Similar behavior was usually observed in the creep of metals processed by ECAP [7,11,16]. It is generally believed that grain boundary sliding is more important process and its activity is influenced by number of ECAP passes.

3.2. Microstructure observations after ECAP

Microstructure after 1 ECAP pass (Fig. 2a) contained the (sub)grains with the mean size of about 3 μm. EBSD analysis found very low number of high-

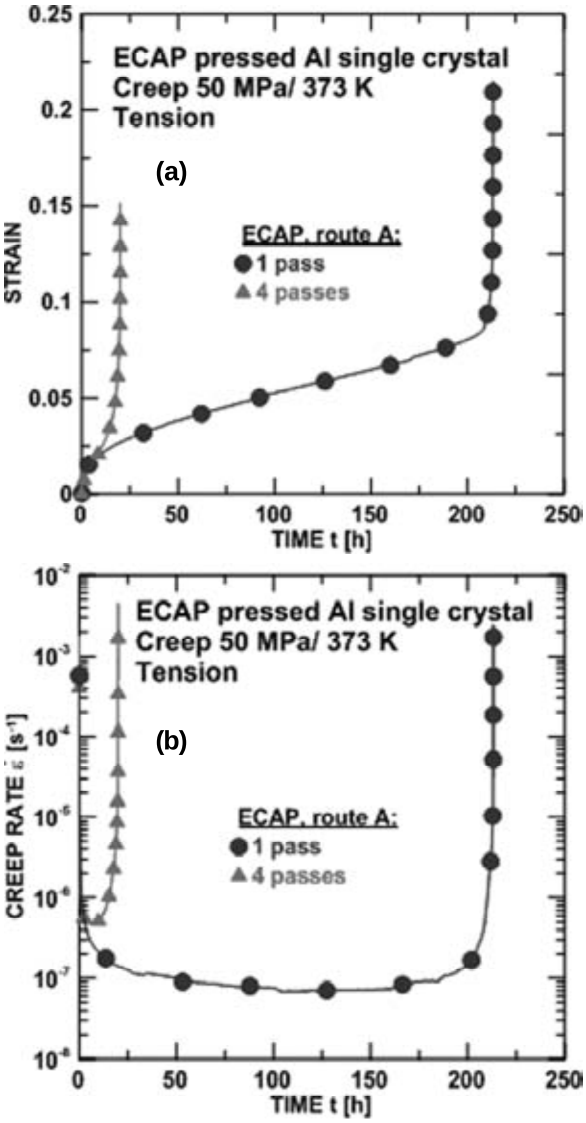

Fig. 1. Creep results for Al single crystal processed by different number of ECAP passes showing: (a) creep curves of strain versus time, (b) creep rate versus time.

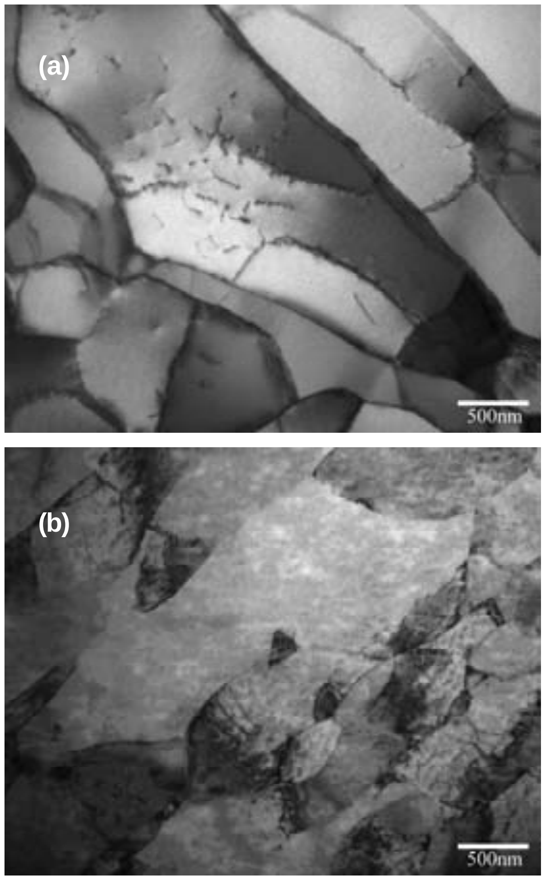

Fig. 2. Microstructure of Al single crystal processed by: (a) 1, (b) 4 ECAP passes at room temperature.

angle grain boundaries with misorientation θ>15° (HAGB's) in the structure. The number of HAGB's was lower than 1%. By contrast, microstructure after 4 ECAP passes (Fig. 2b) contained grains with the mean size of 1.6 μm and about 44% of HAGB's. Inhomogeneity of microstructure can be qualified by the coefficient of profile area variation CV*a* [19]: the higher is its values the more pronounced is the inhomogeneity. In homogeneous systems 0.55 ≤ CV*a* < 1, in mildly inhomogeneous systems is CV*a* lower then 2 and higher values are typical for systems with multimodal grain size distributions. Inspection of Fig. 2b showed that the microstructure after 4 ECAP passes contains mixture of larger elongated and smaller more equiaxed grains. The coefficient of profile area variation CV*a* of microstructure was determined about 1.7 which represents mildly inhomogeneous systems with bimodal character.

From initial crystallographic orientation of single crystal used in this investigation can be predicted that shear system with shear factor 1 is distributed mainly on one primary system with shear factor [1]. The evolution of heterogeneous distribution of HAGB's can be influenced by low number and high value of shear factors of activated slip systems with respect to shear plane during ECAP deformation. Heterogeneity created during ECAP can influence creep behavior of samples and caused non-uniformity of creep strain. Inhomogeneous structures with bimodal character can be used for optimization of strength and ductility in nanocrystalline and UFG materials at room temperature [20].

3.3. Microstructure observation after creep

The microstructure of sample after 4 ECAP passes and creep is shown in Fig. 3. The inspection of sample after 4 ECAP passes and subsequent creep exposure at 373K showed the grain growth and the

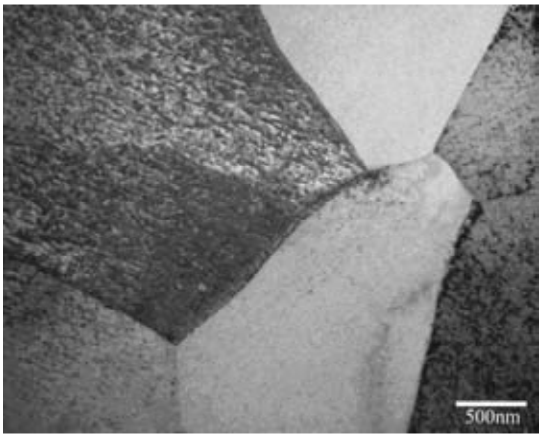

Fig. 3. Microstructure of Al single crystal processed by 4 ECAP passes and creep test at 373K.

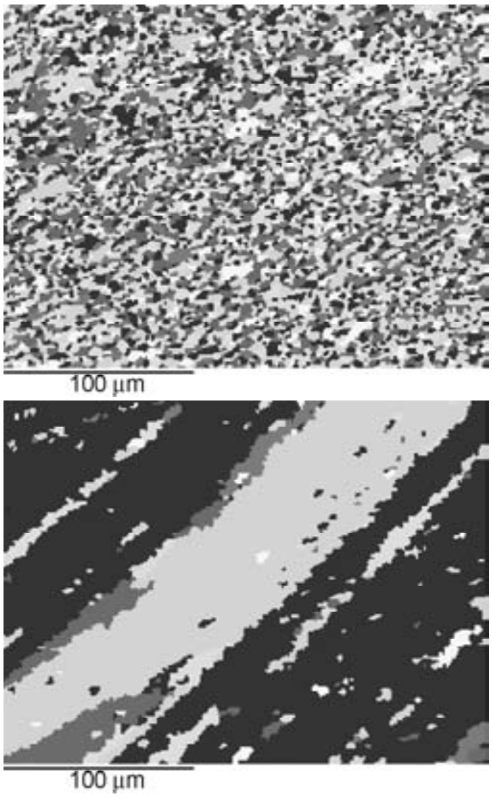

Fig. 4. EBSD image of Al single crystal processed by 4 ECAP passes and creep test at 373K observed in the section XZ: (a) misorientation > 2, (b) misorientation > 15.

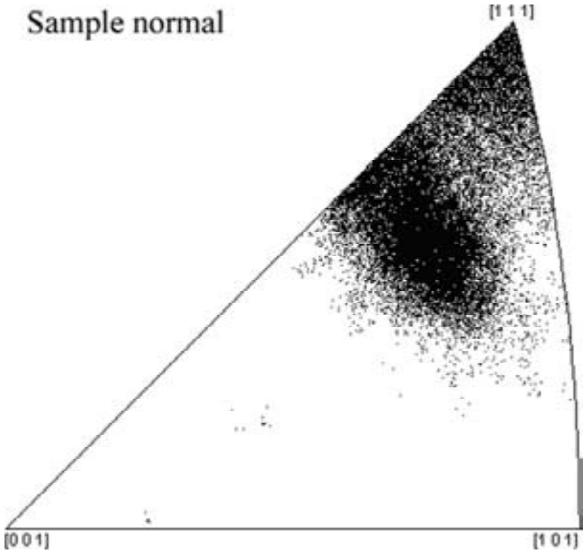

Fig. 5. Inverse pole figure of microstructure of Al single crystal processed by 4 ECAP passes and creep test at 373K.

mean grain size was about 3.4 μm. EBSD analyses of Al single crystal processed by 4 ECAP passes and creep at 373K showed that microstructure contains about 21% of HAGB's. The mean grain size was ~3.8 μm when the grain is characterized as the regions having an orientation range within 2° (Fig. 4a). The mean grain size is similar to the value measured by TEM (Fig. 3). We can consider that the regions having an orientation range within 2° are grains separated by grain boundaries with low angle of misorientation. The assumption is influenced by relatively strong texture which was observed after creep exposure (Fig. 5). It is known that the grain size of sample with weak texture measured by EBSD is usually similar to the grain size measured by SEM [21]. However the grain size of samples with strong texture which was measured by EBSD is larger in comparison with the grain size measured by SEM. In the case that the EBSD method involves all boundaries the grain size is similar as the grain size measured by SEM.

In the Fig. 4b is shown the microstructure, when the grain is characterized as the regions surrounded by boundaries with misorientation θ>15°. The microstructure contains very long shear bands exceeding the picture window and parallel with the theoretical shear plain of the last ECAP pass. The mean interboundary spacing measured parallel with the stress axis (horizontal axis) is approximately 30 μm.

The investigation of the surface of sample after 4 ECAP passes showed the formation of mesoscopic shear bands (MSB's) lying near the shear plain of the last ECAP pass (Fig. 6a). The MSBs were observed near the fracture after creep testing at 373K. The appearance of MSB's was observed in Al-0.2Sc and Cu-0.2Zr alloy after ECAP and subsequent creep exposure and was connected with the decrease of creep resistance [22]. The MSB's can influence the strain and fracture process in creep.

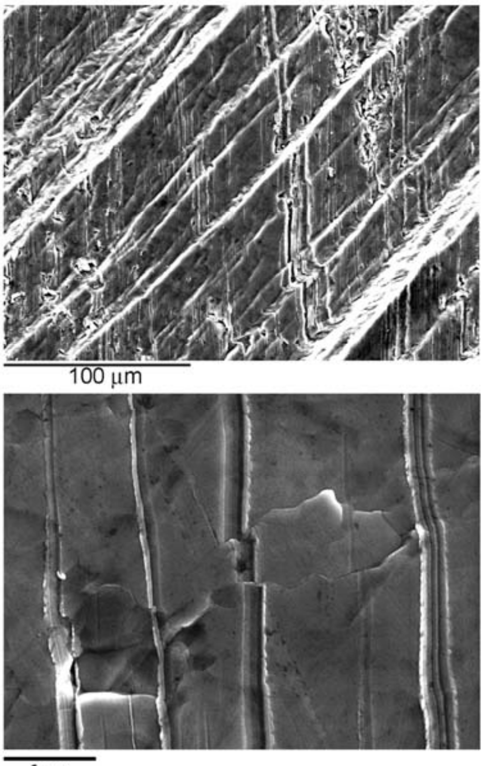

Fig. 6. Appearance of shear bands on the surface of single crystal processed by 4 ECAP passes and creep test at 373K.

On the surface of Al single crystal processed by 4 ECAP passes the two categories of MSB's were observed - the main and minor MSB. The characteristics are shown in the Tab. 1. The mean vertical relief $\bar{v}$ of main MSB's was ~7.3 µm and the mean interband spacing $\bar{w}$ about 135 µm. The displacement of marker lines $\bar{u}$ transversal to the stress axis at the interfaces of main MSB's was about 20 µm. The displacement of marker lines exceeded more than 5 times the mean grain size. The mean vertical relief of minor MSB's was ~1.8 µm and the mean interband spacing ~21 µm. The displacement of marker lines at interfaces of minor MSB's was approximately 2.3 µm. The minor MSB's were situated between main MSB's (Fig. 6a). The formation of main and minor MSB's was observed near the fracture. It was found that the main MSB's were formed at first and with decreasing distance from the fracture the appearance of the minor MSB's were observed. The formation of MSB's can be related to the heterogeneous distribution of HAGB's which was observed after creep exposure (Fig. 4b). The different interband spacing, displacements of marker lines at the interfaces and its occurrence near the fracture lead to the opinion that bands are probably generated during creep exposure.

Table 1. The characteristics of MSB's on the surface of Al single crystal processed by 4 ECAP passes and creep test at 373K.

MSB's	$\bar{v}$ [µm]	$\bar{w}$[µm]	$\bar{u}$ [µm]
Main MSB's	7.3	135	20.5
Minor MSB's	1.8	21	2.3
All MSB's	4	69	12.5

Fig. 6b shows the surface of sample after 4 ECAP passes. This place is situated far from the fracture where the occurrence of MSB's did not observe. We can assume that it can be the place of nucleation of MSB's. The sliding of individual grains is documented by displacement of marker lines. The amount of GBS was determined by measuring the surface offsets produced at the intersections of GBS with marker lines transversal to the stress axis. The mean displacement of marker lines at individual grains was about 0.52 µm.

The strain component of GBS can be expressed as [17,23,24]:

$$\varepsilon_{gb} = \left(1+\varepsilon\right)\cdot\kappa_s\cdot\bar{u}/\bar{L}, \qquad (1)$$

where ε is the true strain, $\bar{L}$ is mean grain size, $\bar{u}$ is mean displacement of marker lines at individual grains. The GBS is not observed at all grain boundaries, that is why the relative frequency of sliding boundaries κ_s was determined. In this work the contribution of GBS can be overestimated because we assumed that all HAGB's measured by EBSD after creep exposure participate in GBS. The contribution of GBS to the total creep strain in Al single crystal processed by 4 ECAP passes was estimated as $\gamma = \varepsilon_{gb}/\varepsilon$. The result of measurement is in the Table 2.

In the interfaces of mesoscopic shear bands the high mean displacement of marker lines was observed. From this reason the TEM foils perpendicular to the interfaces of mesoscopic shear bands on the surface of samples after 4 ECAP passes and subsequent creep exposure were prepared. Microstructure analyses in the interfaces of mesoscopic shear bands revealed the formation of high local concentrations of dislocations at the triple junctions (Fig. 7) which can be caused by GBS.

The microstructure results showed that creep behavior of Al single crystal processed by 4 ECAP passes is influenced by GBS at individual grains and cooperative GBS at the interfaces of MSBs.

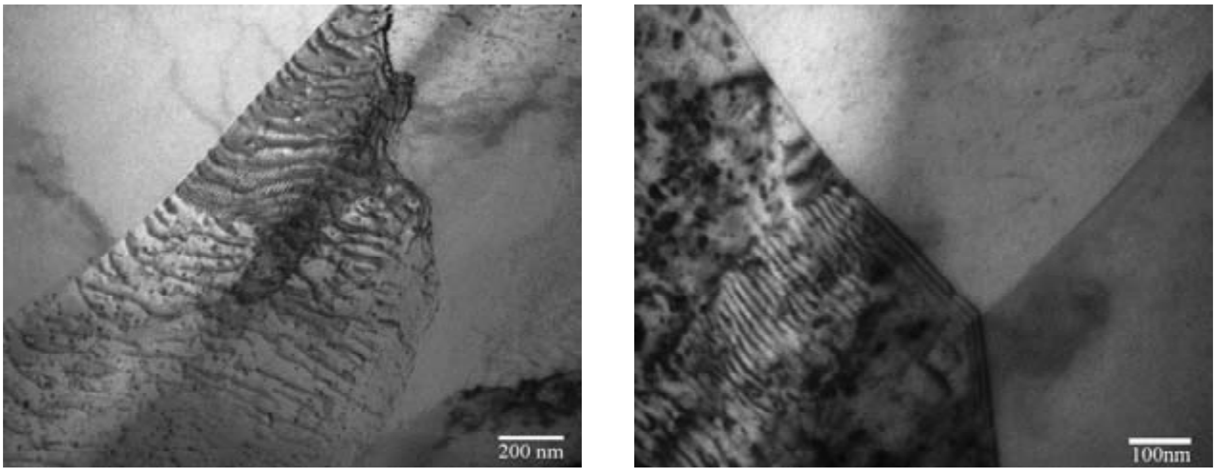

Fig. 7. Triple junctions at the interfaces of MSB's of Al single crystal processed by 4 ECAP passes and creep test at 373K.

Table 2. The measurement of contribution of GBS to the total creep strain of Al single crystal processed by 4 ECAP passes and creep test at 373K (ε~0.15).

Al single crystal	$\bar{u}$ [μm]	$\bar{L}$ [μm]	κ_s	$\varepsilon_{gb}\times10^2$ [%]	$\varepsilon_{gb}/\varepsilon\times10^2$ [%]
4 passes	0.52	3.4	0.21	3.67	24.5

These mechanisms were predicted in the experiments conducted at the polycrystalline material processed by ECAP. Nevertheless the value of the stress exponent n was usually ~ 6 for material processed by 8 ECAP passes whereas the value of stress exponent n for GBS is ~2. The results in this work support the idea that creep behavior of materials processed by ECAP is influenced by contribution of GBS depending on the number of HAGB's in the microstructure. Generally the GBS in the UFG materials can be accommodated by diffusion, local deformation of grains and cavitation. The accommodation of GBS by diffusion can be expected because in the microstructure after ECAP the non-equilibrium grain boundaries can be found. The large concentration of extrinsic non-equilibrium defects in non-equilibrium grain boundaries accelerates the grain boundary diffusivity because the accumulated free volume and energy of grain boundary increase [25]. Divinsky *et al.* [26,27] revealed ultra-fast diffusivity along some paths in materials processed by SPD. In the work [26] was showed that ECAP introduces additional free volume in the Cu-1wt.%Pb alloy in the form of non-equilibrium interfaces, vacancy clusters, nanovoids and micropores. These defects probably provide pathways for ultra-fast atomic transport. In the present work the temperature of creep was relatively low and the minimum creep rate relatively fast. In these conditions the diffusion is probably slow and cannot accommodate GBS. From this reason the observed GBS was probably accommodated by high local concentration of dislocations at the triple junctions.

4. SUMMARY AND CONCLUSIONS

The Al single crystal was processed by equal-channel angular pressing (ECAP) through 1 and 4 passes and then tested under constant tensile load of 50 MPa at 373K. The main results can be summarized as follows:

- The fraction of HAGB's in the microstructure increases with increasing number of ECAP passes and their inhomogeneous distribution influence creep behavior.
- Creep resistance of ECAP processed Al single crystal decreases with increasing number of ECAP passes.
- The creep behavior of ECAP processed Al single crystal is influenced by cooperative grain boundary sliding along mesoscopic shear bands.
- The GBS in Al single crystal processed by 4 ECAP passes can be probably accommodated by dislocation glide and migration of triple junctions.

ACKNOWLEDGEMENTS

This work was financially supported by the Czech Science Foundation under Grant P108/10/P469 and by the Academy of Sciences of the Czech Republic under the Institutional Research Plan AV0Z20410507 and project KAN300100801.

REFERENCES

[1] Y. Fukuda, K. Oh-ishi, M. Furukawa, Z. Horita and T.G. Langdon // *Acta Mater*. **52** (2004) 1387.

[2] M. Hafok and R. Pippan // *Mater. Sci. Forum* **550** (2007) 277.

[3] H. Miyamoto, J. Fushimi, T. Mimaki, A. Vinogradov and S. Hashimoto // *Mater. Sci. Eng. A* **405** (2005) 221.

[4] T. Grosdidier, J.-J. Fundenberger, D. Goran, E. Bouzy, S. Suwas, W. Skrotzki and L.S. Tóth // *Scripta Mater.* **59** (2008) 1087.

[5] P. Šedá, A. Jäger and P. Lejček // *Mater. Sci. Forum* **667-669** (2011) 355.

[6] W. Z. Han, Z.F. Zhang, S.D. Wu and S.X.Li // *Acta Mater*. **55** (2007) 5889.

[7] J. Dvorak, V. Sklenička, P. Kral, M. Svoboda and I. Saxl // *Rev. Adv. Mater. Sci.* **25** (2010) 225.

[8] G. P. Grabovetskaya, K.V. Ivanov and Y.R. Kolobov // *Ann. Chim. Sci. Mat*. **27** (2002) 89.

[9] W. Blum and Y.J. Li // *Mater. Sci. and Technology* (2005) 65.

[10] G. P. Grabovetskaya, Y.R. Kolobov, K.V. Ivanov and N.V. Girsova // *Physics of Metals and Metallography* **94** (2002) 37.

[11] V. Sklenicka, J. Dvorak, M. Svoboda, P. Kral and B. Vlach // *Mater. Sci. Eng. A***482** (2005) 83.

[12] P. Král, J. Dvořák and V. Sklenička // *Mater. Sci. Forum* **584-586** (2008) 846.

[13] A. Kostka, K.-G. Tak and G. Eggeler // *Mater. Sci. Eng. A* **481-482** (2008) 723.

[14] V. Sklenicka, J. Dvorak, M. Kvapilova, M. Svoboda, P. Kral, I. Saxl and Z. Horita // *Mater. Sci. Forum* **539-543** (2007) 2904.

[15] M. Kawasaki, I.J. Beyerlein, S.C. Vogel and T.G. Langdon // *Acta Mater*. **56** (2008) 2307.

[16] V. Sklenička, J. Dvořák, P. Král, M. Svoboda and I. Saxl // *int. J. Mat. Res. (formely Z. Metallkd.)* **100** (2009) 775.

[17] V. Sklenicka, J. Dvorak, P. Kral, Z. Stonawska and M. Svoboda // *Mater. Sci. Eng.* ***A*410-411** (2005) 408.

[18] P. Kral, J. Dvorak, M. Kvapilova, M. Svoboda, V. Benes, P. Ponizil, O. Sedivy and V. Sklenicka // *Mater. Sci. Forum* **667-669** (2011) 235.

[19] I. Saxl, V. Sklenicka, L. Ilucova, M. Svoboda, P. Kral and J. Dvorak // *Rev. Adv. Mater. Sci.* **25** (2010) 233.

[20] E. Ma // *Scripta Mater*. **49** (2003) 663.

[21] F. J. Humphreys // *Journal of microscopy* **195** (1999) 170.

[22] V. Sklenicka, P. Kral, J. Dvorak, M. Kvapilova, M. Kawasaki and T.G. Langdon // *Mater. Sci. Forum* **667-669** (2011) 897.

[23] V. Sklenicka and J. Cadek // *Z.Metallkd*. **61** (1970) 575.

[24] J. Cadek, *Creep in metallic materials* (Prague, Academia, 1988).

[25] A.A. Nazarov, A.E. Romanov and R.Z. Valiev // *Nanostructured Mater*. **6** (1995) p. 775.

[26] S.V. Divinski, J. Ribbe, D. Baither, G. Schmitz, G. Reglitz, H. Rösner, K. Sato, Y. Estrin and G. Wilde // *Acta Mater*. **57** (2009) 5706.

[27] Y. Amouyal, S. V. Divinski, Y. Estrin and E. Rabkin // *Acta Mater*. **55** (2007) 5968.