

# Quantitative characteristics of inhomogeneous microstructure in UFG copper

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**Abstract.** The ultrafine-grained microstructure of pure copper processed by equal-channel angular pressing and its temperature-induced changes were evaluated in order to characterize heterogeneous distribution of fine- and larger-sized grains in the microstructure. ECAP was conducted at room temperature with a die that had an internal angle of 90° between the two parts of the channel. The subsequent extrusion passes were performed by route Bc up to 8 ECAP passes and tested under constant load. Creep test was performed on the samples processed by 8 ECAP passes in tension at 373 K under an applied stress 260 MPa. The analyses of microstructure were performed by 3 dimensional electron-back scatter diffraction (3D EBSD) technique. The volume characteristics of microstructure and its inhomogeneity were evaluated and the relationships microstructure/creep behaviour of UFG copper was discussed.

## 1. Introduction

It is generally accepted that methods of severe plastic deformation (SPD) reduce the grain size to the submicrometer or even nanometer level [1,2]. Prominent among SPD techniques are equal-channel angular pressing (ECAP) and high pressure torsion [3]. The homogeneity of deformed microstructure depends on the amount of imposed deformation [1,2]. The homogeneous ultrafine-grained (UFG) structure is formed subsequently with increasing number of ECAP passes or rotations especially when the deformation is performed at room temperature.

It was found that SPD microstructure can be still heterogeneous after relatively high number of ECAP passes when the initial state is extremely coarse-grained [4,5]. The investigation of the microstructure formation in single crystals during SPD revealed that heterogeneity of microstructure can also be influenced by initial crystallographic orientation of single crystal before plastic deformation [6,7]. It was observed that SPD materials are unstable and the thermal exposure can lead to the formation of bimodal or multimodal structures. The co-existence of larger recrystallized grains in the bimodal structures can improve deformation behaviour and thereby a ductility of UFG materials by relaxation of the stress concentration created by grain boundary sliding through plastic deformation inside of larger grains [8,9].

In our previous works [4,5,10] the standard stereological approaches were used to obtain the main characteristics of the specimens, in particular the profile and the intersection counts estimating the profile and the intercept intensities  $N_A$  (the mean number of profiles per unit area of the section plane),  $N_L$  (the mean number of chords per unit length of the test line) in the sections planes. The value of  $N_L$  was determined as the arithmetic mean of results measured in six systematically selected directions (0°, 30°, 60°, 90°, 120° and 150°). The values  $E_l$  (mean grain chord) and  $E_a$  (mean profile area) are the reciprocal values of  $N_L$  and  $N_A$ .

The homogeneity of structure can be characterized by the coefficients of profile area variation  $CV_a$  [4,5]: the higher are their values the more pronounced is the inhomogeneity. In homogeneous systems  $0.55 \leq CV_a < 1$ , in mildly inhomogeneous systems is  $CV_a$  lower then 2 and higher values are typical for systems with multimodal grain size distributions.



The plane and volume microstructure characteristics in our previous works [4,5] were based on the investigations of three mutually perpendicular planar sections by 2D EBSD. The application of 3D EBSD characterization method, which combines ion-based micromilling using Focused Ion Beam with EBSD analysis, are going to be more and more popular. The application of modern imaging method enables the quantitative examination of microstructure characteristics such as real size and shape of grains in UFG materials processed by ECAP [11].

The aim of this work is the quantitative 3D investigation of inhomogeneous microstructures in pure copper after 8 ECAP passes and subsequent thermal exposure.

## 2. Experimental Material and Procedures

The experimental material used in this investigation was cast state of pure copper (99.99 %) with the mean grain size  $\sim 1.2$  mm. The billets were processed by ECAP at room temperature using a die that had an internal angle of  $90^\circ$  between the two parts of the channel and an outer arc of curvature of  $\sim 20^\circ$  where these two parts intersect. From first principles it can be shown that these angles lead to an imposed strain of  $\sim 1$  in each pass of the sample [1]. The pressing speed was 10 mm/min. The billets were subsequently pressed by 8 ECAP passes using route Bc.

The microstructure characteristics were investigated in pure copper after 8 ECAP passes and UFG state was tested at 373 K under constant load. The creep testing was conducted in argon atmosphere with the testing temperature maintained to within  $\pm 0.5$  K of the set value. The creep test was run up to fracture. Microstructure was examined by FIB/SEM Tescan Lyra 3 equipped with electron back scatter diffraction (EBSD) by Oxford Instrument. The 3D investigation was performed by fully automated acquisition of FIB for milling and EBSD for microstructure analyses. The configuration of FIB, electron beam and EBSD enable EBSD acquisition without a necessity of specimen repositioning – static set-up [11,12]. The 3D EBSD analyses were performed in the XZ section [13]. The milling was conducted at 30 KeV and the beam current was about 800 pA. At first the surrounding of the micro block on the edges of specimens was milled by FIB procedure. The FIB procedure ensured sufficient space around the block which is necessary for good-quality of EBSD patterns from the depth of the investigated volume of block. Before FIB milling the surface of block was covered by platinum protected layer to minimize the surface damage by Ga ions. The step size and depth for 3D EBSD maps were 80 nm for UFG Cu and 150nm for UFG Cu after creep testing at 373 K. EBSD maps of individual slices were assembled to form 3D maps (Fig. 1). The reconstruction of 3D structure from individual slices and determination of (sub)grain or grain characteristics was performed

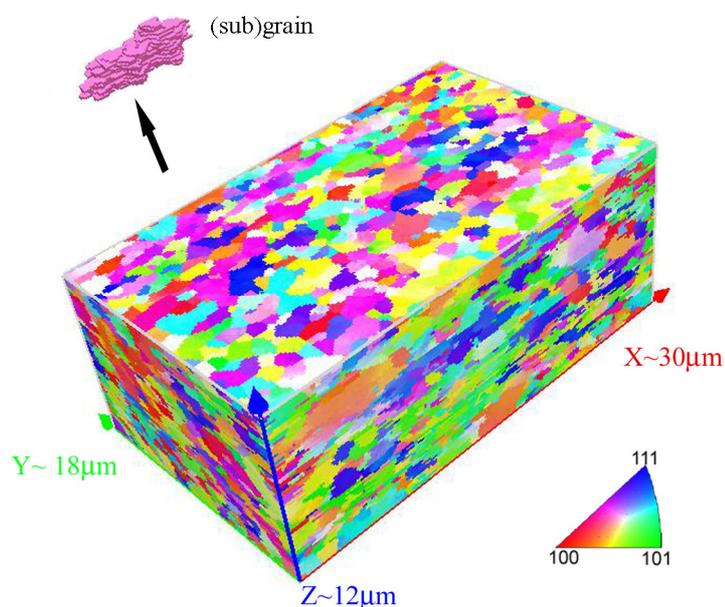


Fig. 1 The 3D microstructure reconstruction of UFG Cu after creep at 373 K

using 3D Viewer software developed by Oxford Instruments. The (sub)grain was characterized as closed volume (area in 2D) surrounded by boundary with misorientation angle  $\theta > 2^\circ$ . The grain was characterized as closed volume (area in 2D section) surrounded by boundary with misorientation angle  $\theta > 15^\circ$ .

### 3. Experimental results

The microstructure of copper processed by 8 ECAP passes contained the (sub)grains with mean volume about  $0.1 \mu\text{m}^3$ . The detailed inspection of 3D (sub)grain reconstruction revealed that about 89% of (sub)grains was up to  $0.1 \mu\text{m}^3$  (Fig. 2a). Nevertheless in the microstructure approximately  $\sim 1.4\%$  of (sub)grain number exceeded the volume of  $0.9 \mu\text{m}^3$ . The volume of  $0.9 \mu\text{m}^3$  was selected as a limiting value because the sphere equivalent diameter of these areas was about 1-1.2  $\mu\text{m}$ . From this reason the value of  $0.9 \mu\text{m}^3$  was assumed as an upper boundary for ultrafine volumes. The (sub)grains with the volume larger than  $0.9 \mu\text{m}^3$  occupied about 63% of whole volume in UFG Cu.

The creep exposure at 373 K caused the coarsening of (sub)grains and their mean volume was  $\sim 0.28 \mu\text{m}^3$ . The distribution of (sub)grains (Fig. 2a) showed that approximately 60% of (sub)grains had the volume up to  $0.1 \mu\text{m}^3$ . In the microstructure was situated about 6.4% of

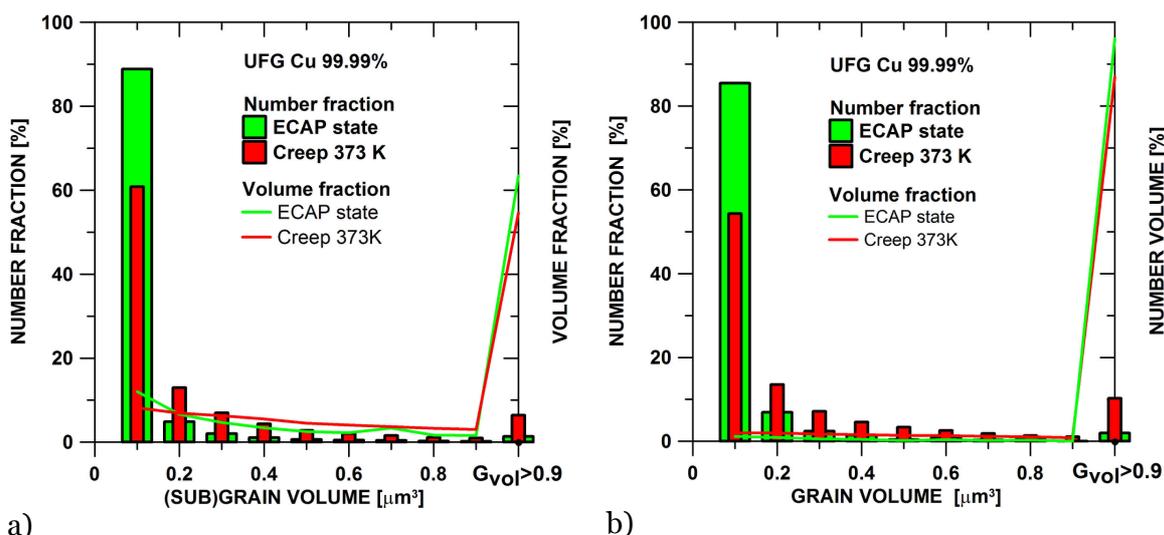


Fig. 2 The distribution of a) subgrains and b) grains in the microstructure of pure copper after 8 ECAP passes and creep testing at 373 K

(sub)grains with the volume larger than  $0.9 \mu\text{m}^3$ . The (sub)grains with the volume larger than  $0.9 \mu\text{m}^3$  occupied about 54% of whole volume in the microstructure after creep testing. The investigation of the 3D grain reconstruction revealed that the mean volume of grains in the microstructure of UFG copper was about  $1.04 \mu\text{m}^3$ . The distribution of the grain volume showed that  $\sim 85\%$  of grains had the volume up to  $0.1 \mu\text{m}^3$  (Fig. 2b) and approximately  $\sim 1.9\%$  of grain number was larger than  $0.9 \mu\text{m}^3$ . The grains with the volume larger than  $0.9 \mu\text{m}^3$  occupied about 95% of whole volume in the microstructure after 8 ECAP passes. The creep exposure at 373 K led to the grain growth (Fig. 2b). The inspection of grain volume distribution showed the decrease of grains with volume up to  $0.1 \mu\text{m}^3$  and the increase of larger grains in comparison with UFG state. In the microstructure was found  $\sim 10.3\%$  of grain with volume larger than  $0.9 \mu\text{m}^3$ . The grains with the volume larger than  $0.9 \mu\text{m}^3$  occupied about 86% of whole volume in the microstructure after 8 ECAP passes

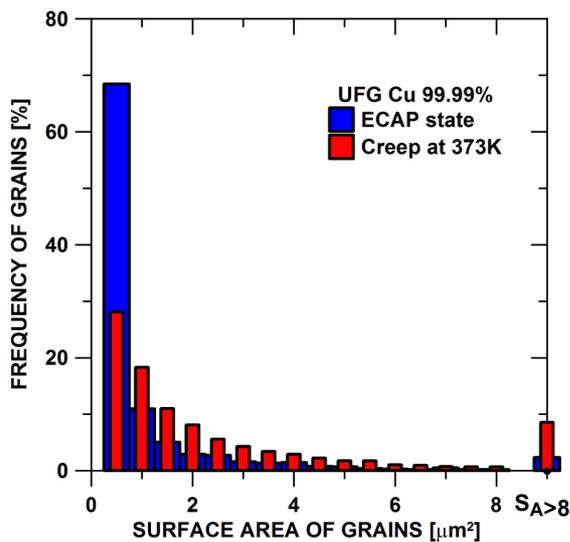


Fig. 3 The distribution of grain surface area in the microstructure of Cu after 8 ECAP passes and creep testing at 373 K

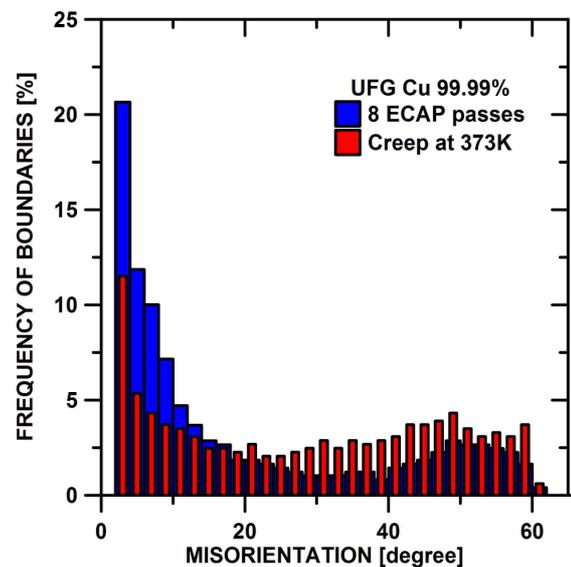


Fig. 4 The distribution of misorientation angle in the microstructure of copper after 8 ECAP passes and creep testing at 373 K

The distribution of grain surfaces showed that  $\sim 68\%$  grains had the surface area up to  $0.5 \mu\text{m}^2$  after 8 ECAP passes (Fig. 3). However in the microstructure one can find  $\sim 2\%$  of grains with surface area larger than  $8 \mu\text{m}^2$ . The surface of  $8 \mu\text{m}^2$  was selected as a limiting value because grains with the maximal volume of  $0.9 \mu\text{m}^3$  occupied maximal surface area about  $8 \mu\text{m}^2$ .

The creep exposure at 373 K led to the growth of grain surface area and approximately 28% of grain surface area had still the surface up to  $0.5 \mu\text{m}^2$  (Fig. 3). In the microstructure one can find more than  $\sim 6.5\%$  of grains with surfaces larger than  $8 \mu\text{m}^2$ .

**Table 1.** The values of CVa for microstructure after 8 passes and creep at 373K

|                                     | CVa_2D     |       | CVa_3D     |       |
|-------------------------------------|------------|-------|------------|-------|
|                                     | (sub)grain | grain | (sub)grain | grain |
| 8 ECAP passes                       | 2.0        | 5.8   | 17.2       | 29.3  |
| 8ECAP passes and creep test at 373K | 1.6        | 2.5   | 3.5        | 30.9  |

The microstructure of UFG copper contained approximately 42% of HAGBs and larger portion of boundaries exhibited the misorientation lower than  $15^\circ$ . However, after creep exposure relatively high number of HAGBs ( $\sim 68\%$ ) was observed (Fig. 4). The HAGBs in the microstructure after creep exposure were formed mostly by random and special boundaries ( $\Sigma 3, \Sigma 9$ ). The values of CVa\_3D determined for (sub)grain and grain volumes of UFG copper and state after creep exposure at 373 K showed extremely high values of CVa (Tab. 1). From comparison reasons the CVa\_2D based on the 2D section was also determined.

#### 4. Discussion

The investigation of (sub)grains and grains volumes in pure copper after 8 ECAP passes showed that majority of number of these areas is in the ultrafine-grained region. However the volume of several (sub)grains or grains was about two or three orders of magnitudes higher than mean grain volume determined for UFG copper and UFG copper after creep testing. The presence of individual large areas in the UFG microstructure processed by 8 ECAP passes can be related to the local heterogeneity of deformation influencing processes which form UFG microstructure.

In our recent works [4,5,10] the inhomogeneity of microstructure was estimated by coefficient of variation  $CV_a$  determined from the planar sections. The microstructure of different materials processed by 8 ECAP passes exhibited bimodal or slightly multimodal character with the value of  $CV_a$  about 2-6 [4,5,10]. Saxl et al. [4] also observed that annealing and creep exposure of ECAP copper led to the decrease of inhomogeneity ( $CV_a \sim 2-3$ ). Similar tendencies in homogeneity of microstructure were also found in present work when the homogeneity was evaluated on the basis of planar sections. However the values of  $CV_a$  determined from (sub)grain or grain volumes showed unexpected high estimations of inhomogeneity. Such extremely high values have never been observed in 2D sections even not for microstructure after 1 ECAP pass. It can be suggested that in the planar sections one can see only different parts of large (sub)grains or grains not their real size which reduce their influence on the inhomogeneity. From this reason planar sections of microstructure may seem to be more homogeneous in comparison with the microstructure volume.

The presence of large (sub)grains or grains which occupy the majority of microstructure volume can influence creep behavior by the formation of mesoscopic shear bands. In our recent works [14-16] was observed the occurrence of mesoscopic shear band on surface of crept specimens. The mesoscopic shear bands, exceeding considerably mean grain size, were laying near to the shear plane of the last ECAP pass and in the vicinity of mesoscopic shear bands high heterogeneity in the distribution of HAGBs was observed. The volumes of large grains also took up large surface areas. The interlinking of large grain surfaces in the volume can influence the nucleation and propagation of cavities and microcracks during creep. In the work [17] was observed that cavities nucleate and propagate along inhomogeneous structure at the interface of the mesoscopic shear bands.

Blum et al. [18] observed the dynamic coarsening of the grains in microstructure of ECAP copper after creep. It was suggested that creep behaviour is controlled by storage and dynamic recovery of dislocations at high-angle boundaries [18-20]. The presence of large grains in the microstructure of UFG copper after creep test, observed in present work, can be also the result some kind of dynamic recrystallization. The occurrence of twin boundaries after creep may support this suggestion. However from the present result it is not clear whether extremely large grains were formed by recrystallization and/or were inherited from ECAP microstructure.

#### 5. Conclusions

The 3D EBSD analyses of UFG copper found that mean (sub)grain and grain volumes are still in the ultrafine-grained region even after creep exposure at 373 K under 260 MPa. However, UFG microstructure may contain several extremely large (sub)grains and grains. The extremely large (sub)grains and grains may occupy the majority of volume and influence significantly microstructure inhomogeneity and creep fracture behavior on the mesoscopic level.

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