Materials Science Forum Vols. 584-586 (2008) pp 440-445
online at http://www.scientific.net
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Online available since 2008/Jun/17

Mechanical properties and microstructure development of ultrafine-grained Cu processed by ECAP

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Keywords: Ultrafine-grained copper, ECAP, mechanical properties, microstructure development

Abstract. Technical purity Cu (99.95 wt%) polycrystals have been processed at room temperature by equal channel angular pressing. The results of mechanical tests and the microstructure characterization by various experimental techniques are presented. The yield stress as well as the strength were shown to increase with increasing strain and exceed the respective values of a coarse-grained material. The microstructure development and its fragmentation after ECAP was investigated by the TEM and EBSD. The proportion of high angle grain boundaries was found to increase with increasing strain reaching the value of 90% after 8 ECAP passes. Two kinds of defects were identified in ECAP specimens by positron annihilation spectrometry (PAS): (a) dislocations which represent the dominant kind of defects, and (b) small vacancy clusters (so called microvoids). The main increase of defect density was found to occur during the first ECAP pass. PAS analysis indicated that in the specimens subjected to one ECAP pass the mean dislocation density ρ_D and the concentration of microvoids c_v exceeded the values of 10^{14} m^{-2} and 10^{-4} at.$^{-1}$, respectively. After 4 passes, the number of defects becomes saturated and practically does not change with increasing strain.

Introduction

The research activity in the area of severe plastic deformation (SPD) has increased extremely in the last years due to many interesting properties which can be achieved in bulk materials by SPD [1]. Compared to classical deformation processes, the big advantage of SPD techniques (represented in particular by equal channel angular pressing (ECAP)) is the lack of shape-change deformation and the consequent possibility to impart extremely large strain. SPD has received enormous interest over the last two decades as a method capable of producing fully dense and bulk submicrocrystalline and nanocrystalline materials. Significant grain refinement obtained by SPD leads to improvement of mechanical, microstructural and physical properties [2].

Copper represents an ideal model material to study the processes of deformation and microstructure development due to its low cost, simple FCC structure, medium stacking fault energy and long history of research of this material prepared by conventional techniques as e.g. rolling, extrusion, compression, etc. which were capable of imparting large strains to the workpiece. This investigation represents the basis of our knowledge and understanding of the properties and associated microstructure changes in SPD copper [3].

Numerous experimental data reporting various properties of ultrafine-grained Cu prepared by SPD are now available, see e.g. [4] for an excellent review and many literature references. On the other hand, there are still many inconsistencies and ambiguities in literature data which motivate further investigation of this material.

This paper summarizes the results of the investigation of microstructural changes of ultrafine-grained Cu polycrystals due to ECAP processing obtained by various experimental techniques and of the post-ECAP behaviour of the material in mechanical testing with the aim to smooth some existing discrepancies in current research findings.

Experimental procedures

Technical purity (99.95 %) Cu was severely deformed by equal channel angular pressing (ECAP) to a maximum equivalent strain of 8 (1, 2, 4 and 8 passes) at room temperature following route B_c. Prior to ECAP processing the specimens were annealed for 2 hours at 450 °C in a protective inert atmosphere. The initial specimen dimensions were 10 mm x 10 mm x 60 mm. ECAP pressing was carried out using a split design die manufactured from tool steel X38CrMoV 51. The details of the die design as well as the ECAP pressing are given elsewhere [5].

Mechanical properties were determined by testing tensile specimens in a conventional universal screw-driven Instron 5882 machine at the initial strain rate of 4×10^{-4} s^{-1} at room temperature (RT).

The microstructure of the initial state was examined by s*canning electron microscopy* using a Tesla BS434 SEM operated at 15 kV.

For *transmission electron microscopy* (TEM) investigations on specimens prepared by mechanical and electrolytic polishing from middle sections of the ECAP processed billets, a Jeol 200FX electron microscope operating at 200 kV was used. Electrolytic polishing was carried out at 10°C using 50% H_3PO_4 in a Tenupol 5 (Struers) jet polishing unit.

EBSD measurements were performed using the high-resolution scanning electron microscope LEO-1530 equipped with a field emission cathode and Nordlys II EBSD detector (HKL Technology). The measurements were carried out at the acceleration voltage of 20 kV with the step size varying from 50 to 500 nm, depending on grain size (i.e. number of ECAP cycles). For identification of the Kikuchi patterns, and measured data evaluation, the software package CHANNEL 5 was employed. The scanning was performed in the transversal plane perpendicular to the direction of ECAP pressing (plane X of the billet [1]).

A $^{22}Na_2CO_3$ positron source (~1.5 MBq) deposited on a 2 μm thick Mylar foil was used in *positron annihilation spectroscopy* (PAS) measurements. This source was always forming a sandwich with two identically treated Cu specimens.

Positron lifetime (PL) measurements were performed using a fast-fast spectrometer [6] with a timing resolution of 160 ps (FWHM ^{22}Na). At least 10^7 positron annihilation events were accumulated at each PT spectrum which was subsequently decomposed using a maximum likelihood procedure [7].

Results and discussion

The true stress true strain curves for the initial coarse grained (CG) material and the specimens after ECAP (1, 2, 4 and 8P) are presented elsewhere [8]. Table 1 shows a quantitative summary of tensile test data after 0, 1, 2, 4 and 8 passes represented in terms of the yield $\sigma_{0.2}$ and ultimate tensile strength σ_{max} (YS and UTS) and the total elongation (ε_{tot}).

Table 1 Summary of experimental data obtained from mechanical testing

No. of passes/ n_p	0	1	2	4	8
$\sigma_{0,2}$ (MPa)	78	293	250	330	258
σ_{max} (MPa)	215	314	270	455	371
ε_{tot} (%)	40	9,5	10,6	8,7	12,7

ECAP Cu specimens show significantly higher yield strength as compared to the CG specimen. Both YS and UTS increase up to $n_p = 4$ followed with a slight decline in the specimen $n_p = 8$. Significantly reduced total elongation $\varepsilon_{tot} < 10\%$ as compared to the CG Cu was observed for all passes. Slightly larger elongation ($\varepsilon_{tot} \approx 13\%$) was found only in the 8P specimen. These values are in agreement with the data reported by other authors on UFG Cu [4].

A *SEM* micrograph of the initial state before ECAP pressing is shown in Fig. 1. The microstructure consists of fully recrystallized grains. Extensive twinning associated with annealing at 450 °C is also seen. The average grain size excluding twins is approximately 50 µm. The microstructure evolution due to ECAP is in detail described elsewhere [5, 8]. Significant grain size reduction (by a factor of 100 approximately) was observed in the specimen already after one pass. The microstructure similar to rolling comprising elongated cells and/or subgrain boundaries oriented in the <011> along the trace of <111> plane dominated in the specimens after 1 and 2 passes. After 4 passes the microstructure was inhomogeneous with enhanced fraction of equiaxed subgrains/grains (approximately 40-50% of the transparent area) and the rest of elongated cells/subgrains. The microstructure of the specimen after 8 passes is presented in Fig. 2. It shows an almost homogeneous microstructure with equiaxed grains separated mostly by high angle grain boundaries. The individual boundaries are straight with sharp contrast and very few dislocations in the grain interior. The grain boundaries are obviously closer to the equilibrium state than grain boundaries in the specimens that underwent a smaller number of ECAP passes. The average grain size ranged between 200-300 nm. In some areas bigger grains having the average size of about 500 nm were also found.

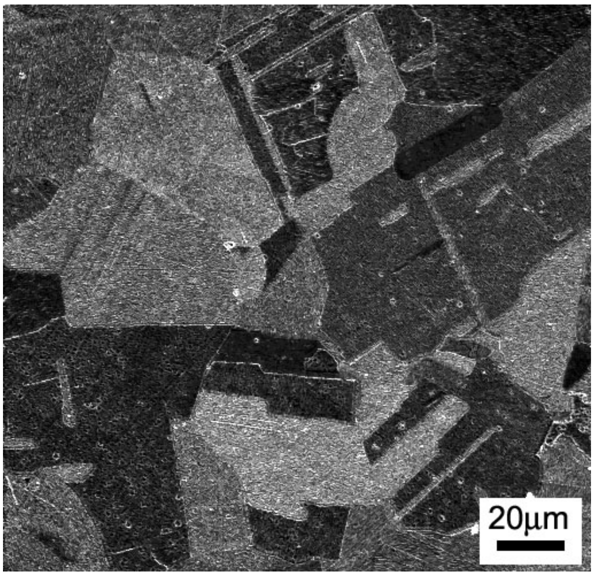

Fig.1 Initial CG structure – SEM micrograph

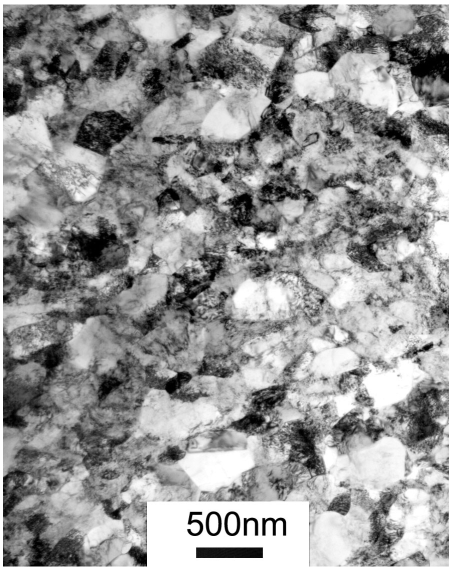

Fig.2 TEM bright field image – ECAP 8P

Results of *EBSD measurements* presented in the form of inverse pole figures are shown in Fig. 3. The results confirm the local image obtained by TEM. In the specimens after 1 and 2 passes, relatively large areas with low misorientations as indicated by slight variations of the colour code dominate in the microstructure. They correspond to bands of elongated subgrains observed by TEM [5]. With further ECAP straining these zones decompose into much smaller subgrains and grains having larger misorientations. Detail analysis of misorientation and grain boundary character distributions is given elsewhere [9]. It is important to note, however, that our EBSD measurements revealed a high density of twins (Σ 3) and multiple twin boundaries (Σ 9, Σ 27), in particular in specimens after 4 and 8 passes (see Fig.4). This somewhat surprising fact was also reported by other authors and assumed to occur only after substantial strain hardening due to ECAP once a certain critical dislocation density is reached [10, 11]. EBSD also confirmed a great presence of low angle grain boundaries having the misorientation angle $\theta < 15°$ (LAGBs) in the specimen after 1P. With further ECAP straining the LAGBs were continuously transformed into high angle grain boundaries (HAGBs; $\theta > 15°$). In the specimen after 8P almost 90% of all grain boundaries had the high angle character (see Fig. 5 and also [9] for details).

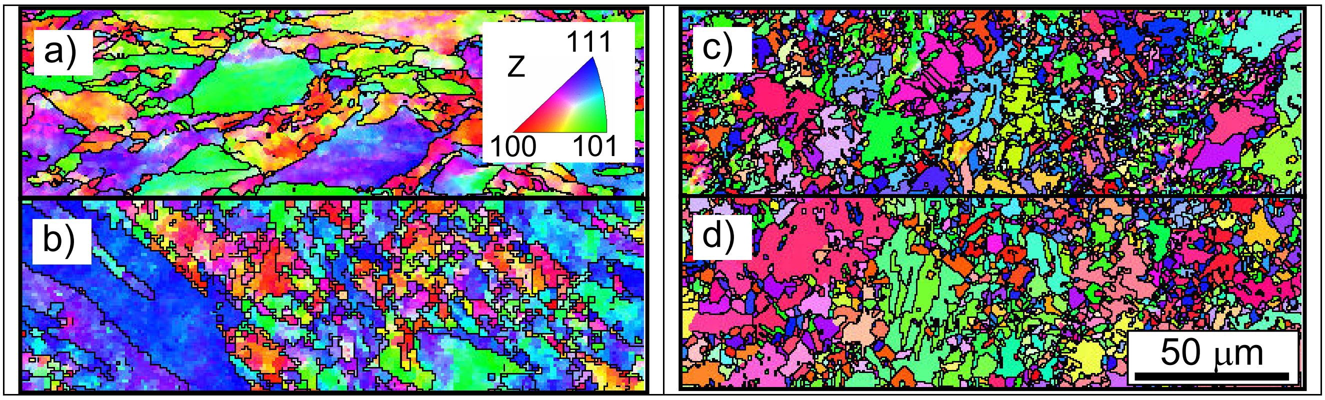

Fig. 3 IPF map of ECAP specimens a) 1P, b) 2P, c) 4P, d) 8P (plane X)

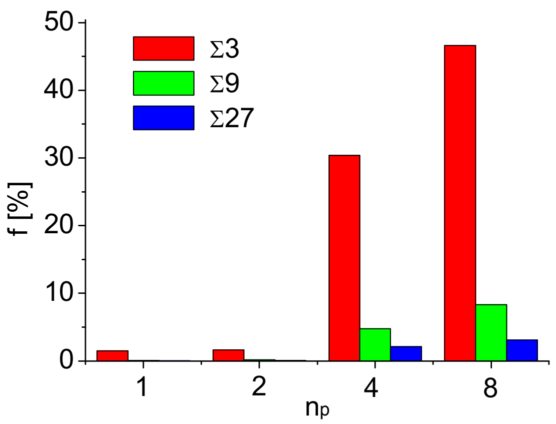

Fig. 4 Twinning and multiple twinning in ECAP Cu

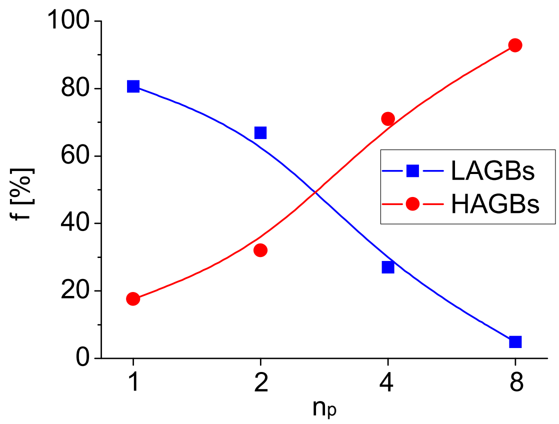

Fig. 5 GB character evolution in ECAP Cu

Results of *PAS* measurements on Cu specimens subjected to increasing number of ECAP passes are shown in Fig. 6. In general, three different components can be observed in PL spectra measured on these specimens: (i) a short lived free positron component with lifetime τ_1 and relative intensity I_1, which comes from free positrons delocalized in the lattice as a Bloch-like wave, (ii) a component with intensity I_2 and lifetime $\tau_2 \approx 164$ ps, which is known to represent a contribution of positrons trapped at dislocations [12], and (iii) the longest component with lifetime τ_3 and intensity I_3, which can be attributed to positrons trapped at small vacancy clusters called microvoids [13], which are often detected in ultra fine grained metals prepared by SPD. It is assumed that microvoids were formed by clustering of vacancies created during ECAP deformation.

The well annealed Cu specimen (0 P) exhibits a single component PL spectrum with lifetime $\tau_1 = 114$ ps, $I_1 = 100\%$, i.e. virtually all positrons in this specimen annihilate from the free state. Note that the lifetime τ_1 agrees well with the calculated lifetime of free positrons in Cu [14].

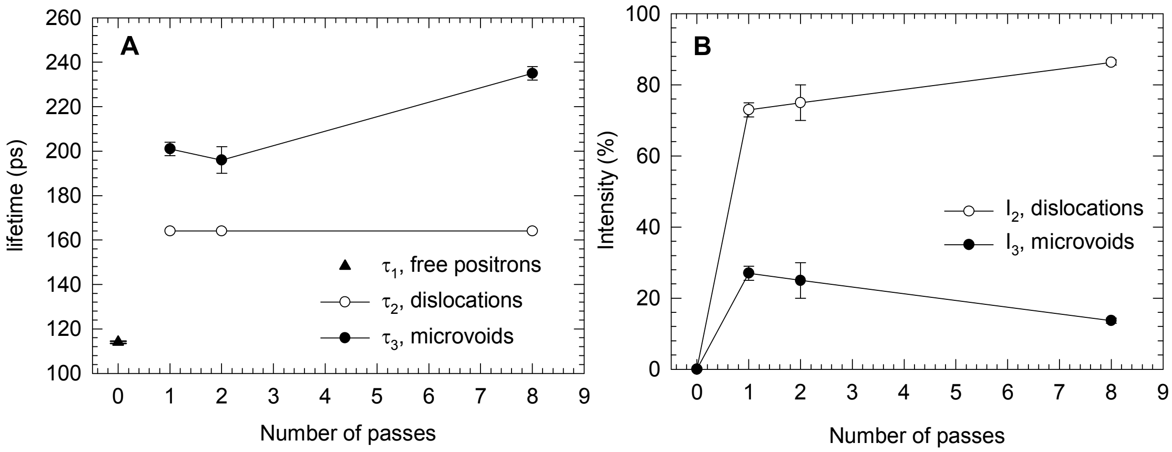

Fig. 6 PAS results for Cu specimens subjected to various number of ECAP passes: (A) lifetimes of the components resolved in PL spectra, (B) intensities of the components arising from positrons trapped at defects.

On the other hand, no free positron component was observed in the specimens deformed by ECAP, i.e. I_1 = 0%. Thus, virtually all positrons in the ECAP deformed specimens are trapped at defects (saturated trapping). This testifies a high density of defects in Cu specimens deformed by ECAP. One can estimate that the mean dislocation density in specimens deformed by ECAP and the concentration of microvoids being $\rho_D \geq 10^{14}$ m^{-2} and $c_v \geq 10^{-4}$ at.$^{-1}$, respectively. Fig. 6B shows that the contribution of positrons trapped at dislocations increases with increasing number of passes, while the intensity of positrons trapped at microvoids reached a maximum already after 1 pass and gradually decreases with further increase in the number of passes.
Because of saturated positron trapping in defects (I_1 = 0%) the ratio of intensity of positrons trapped at dislocations and positrons trapped at microvoids I_2/I_3 equals to the ratio of positron trapping rates to these defects, which is directly proportional to the ratio of corresponding defect densities, i.e.

$$\frac{I_2}{I_3} = \frac{K_D}{K_v} \sim \frac{\rho_D}{c_v}.$$

The ratio K_D/K_v is plotted in Fig. 7A as a function of the number of passes. It is clearly seen that the dislocation density in the specimen deformed by ECAP increases faster than the concentration of microvoids.

By comparing the lifetime t_3 of the component arising from microvoids with theoretical calculations performed in [13], one can determine the size of these defects. The results of these calculations are displayed in Fig. 7B where microvoid diameter is plotted as a function of the

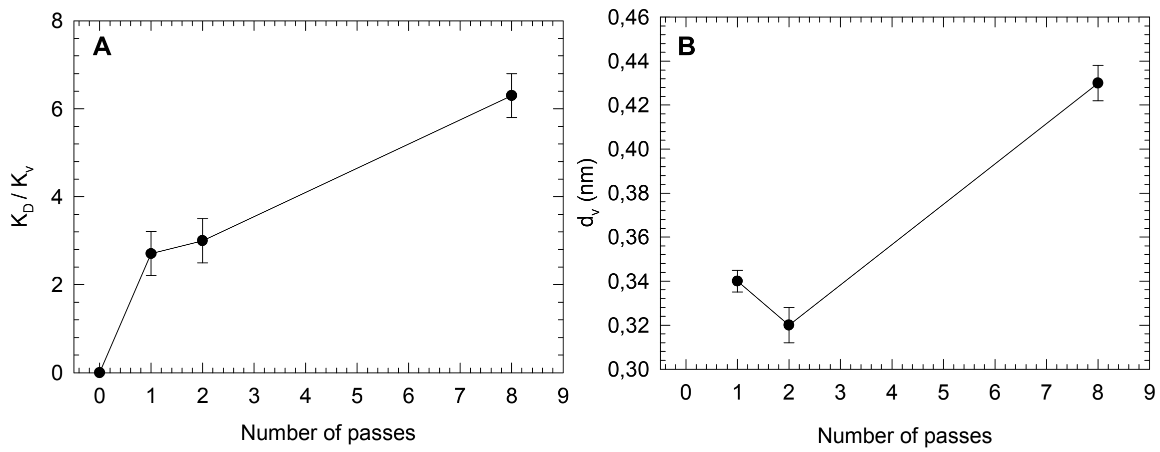

Fig. 7 (A) ratio K_D/K_v of positron trapping rate to dislocations and microvoids, (B) diameter of microvoids calculated from PAS results.

number of ECAP passes. Microvoids are very small defects having the size of 2-4 monovacancies. The difference between microvoid diameter in the specimens subjected to 1 and 2 passes seems to be statistically insignificant, while the sample subjected to 8 passes contains remarkably larger microvoids, see Fig. 7B.

Conclusions

The mechanical properties and microstructure evolution in technical purity Cu processed by ECAP have been studied by various experimental techniques. The following conclusions may be drawn from this investigation:

- severe plastic deformation imposed by ECAP resulted in significant grain refinement (factor 100),
- significantly enhanced mechanical properties (both the yield and the tensile strength) were found in fine grained Cu as compared to the CG material, while the ductility declined in the UFG Cu,
- the initial CG microstructure evolves from elongated dislocation cells/subgrains separated by low angle grain boundaries to more equiaxed subgrains/grains separated by high angle grain boundaries, while the accompanying average grain size changes only slightly with further ECAP processing,
- extensive twinning occurred during ECAP straining,
- besides dislocations which are the dominant defects PAS identified the presence of small vacancy clusters (so called microvoids) in specimens after ECAP.

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Acknowledgements:

The authors acknowledge funding of the Academy of Sciences of the Czech Republic through the grants IAA101120803 and KAN300100801 (J.C.). One of authors M.D. acknowledges the financial support of this work through the German Research Council (DFG) under the project # KL 1274/4-1. Technical assistance of Mr. Torbjoern Lamark by ECAP pressing is also gratefully acknowledged.