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PROCESSING OF COPPER BY HYDROSTATIC EXTRUSION – STUDIES OF MICROSTRUCTURE AND PROPERTIES

The present study attempts to apply HE to 99.99% pure copper. The microstructure of the samples was investigated by both light microscopy and scanning transmission electron microscopy (STEM). Additionally, the microhardness was measured, the tensile test was made, and statistical analysis of the grains and subgrains was performed. Based on Kikuchi diffraction patterns, misorientation was determined. The obtained results show that microstructure of copper deformed by hydrostatic extrusion (HE) is rather inhomogeneous. The regions strongly deformed with high dislocation density exist near cells and grains/subgrains free of dislocations. The measurements of the grain size have revealed that the sample with an initial in annealed-state grain size of about 250 μm had this grain size reduced to below 0.35μm when it was deformed by HE to the strain ε=2.91. The microhardness and UTS are stable within the whole investigated range of deformation.

Keywords: HE, ultra-fine grained copper, microstructure characterization, mechanical properties

1. Introduction

Nano and ultrafine-grained (UFG) materials with the grain size below 1μm contain a large fraction of grain boundaries and, therefore, exhibit a wide spectrum of unique properties and property combinations. For example, the exceptional grain refinement generally leads to very significant strengthening at ambient temperature [1-2]. A possible way for microstructure refinement in metals is the use of severe plastic deformation (SPD) [3]. Severe plastic deformation may be defined as a metal forming process in which a large plastic strain is introduced into a bulk metal in order to create ultra-fine grained metals without any significant change in the overall dimensions of the workpiece [4,5]. The most popular SPD methods are equal-channel angular pressing (ECAP) [6], cyclic extrusion compression (CEC) [7,8] and high pressure torsion (HPT) [9]. Recently, the hydrostatic extrusion method (HE) has been used to produce UFG metals [4, 10,11]. The HE was invented over a 100 years ago and was patented by James Robertson in1893, however, the first experiments with this method were carried out by P. W. Bridgman in the mid of previous century. In HE, the extruded billet is located in a container and surrounded with a pressure transmitting medium. The ram compresses the pressure transmitting medium until the billet starts being extruded through the die. From the technological point of view, HE gives the possibility to obtain bulk materials in a variety of forms, e.g. wires, tubes and others [11].

The aim of this research was to determine the influence of Hydrostatic Extrusion (HE) process on changes in the microstructure and properties of polycrystalline 99.99% pure copper.

2. Test materials and methods

Studies were carried out on the polycrystalline Cu99.99 copper having an average grain size of 250 μm. The chemical composition of the deformed copper is given in Table 1.

TABLE 1

Chemical composition of polycrystalline Cu99.99, % ppm

Zn	Fe	Ni	Sb	Bi	S	Ag	Sn	Te	Se	Pb	As	oxygen	Cu
1.7	4.4	2.2	1	0.5	6	12	0.4	0.1	0.1	0.5	0.3	50	balance

TABLE 2

Parameters of the hydrostatic extrusion process

No.	d_0, mm	d_f, mm	Reduction, %	Linear velocity, mm/s	Strain rate, s^{-1}	True strain
1	5.92	2.96	75	658	3.68×10^2	1.39
2	7.3	2.96	83.6	658		1.81
3	9	2.94	89.3	667		2.24
4	12.7	2.96	94.6	658		2.91

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Before deformation, samples were annealed in a sillite furnace at a temperature of T = 850°C for 2 hours and then were cooled in water. The examined samples were subjected to Hydrostatic Extrusion (HE). The process was carried out at a constant strain rate of 3.68×10^{2} s^{-1} in the range of true strains $\varepsilon = 1.39 – 2.91$. The HE process was performed in the Institute of High Pressure Physics of the Polish Academy of Sciences (UNIPRESS) in Celestynów.

Detailed information about the deformation process is given in Table 2.

Although the process was performed at room temperature, sample heating induced by the high strain rates was possible. Therefore samples were water cooled at the exit from the die in order to minimize the effect of temperature on properties and microstructure.

After the deformation, samples were examined using both Olympus GX51 optical microscope (MO) and Hitachi SU-70 scanning transmission electron microscope (STEM) equipped with a transmission electron detector. Samples for examinations by optical microscopy were mechanically ground, polished with diamond pastes and a colloidal suspension of SiO_2 and then etched in a reagent, which was composition of 30 ml HCl +10g $FeCl_3$ + 120 ml CH_3OH. The thin foils for STEM examinations were prepared by the standard technique of electrolytic polishing using Struers apparatus. The statistical analysis of the grain size diameter was performed manually using mean chord perimeter. In each case approximately 100 – 120 grains/subgrains were measured. It is important to mention the fact that the grain size measurements covered both grains and sub-grains because diffraction contrast did not allow distinguishing between low and high angle boundaries.

The tensile test was performed at room temperature using INSTRON 1115 testing machine with constant cross-head velocity corresponding to an initial strain rate of 10^{-3} s^{-1}. For each variant of deformation at least three specimens were tested. Based on obtained results, the 0.2% yield strength ($\sigma_{0.2}$), ultimate tensile strength (σ_{UTS}) and elongation to failure (ε_f) were determined.

The Vickers microhardness (HV) was measured on mirror-shine polished sample surfaces using standard microhardness tester equipped with a Vickers indenter. The hardness measurements used a load of 100 g. The measurements were performed on longitudinal sections of the samples perpendicular to the sample axis.

3. Results and discussion

In order to avoid misunderstanding, for the needs of this article, the definitions of “bands” and “shear bands” should be introduced. Thus, the word “band” refers to a band which is limited to a single grain only, whereas “shear bands” may run through a considerable distance, cross grain boundaries and sometimes form distinct jogs at the intersected boundaries.

The microstructure of the HE polycrystalline copper looks similar in all ranges of the deformation. A characteristic feature are bands and grains elongated in the extrusion direction (Fig. 1). At high magnification it can be seen that the bands propagate over a long distance, sometimes forming bundles (Fig.2). Bands have different inclinations, but it was also observed that in some areas they maintain the same direction while crossing several grains. In some regions mutually crossing bands were observed (Fig. 4a). The additional characteristic feature of the examined microstructure are short bands placed inside the grains elongated in the extrusion direction. Bands appear as thick lines (Fig. 1,2). Another typical feature is the “wave-like” microstructure (Fig. 1). A similar type of the microstructure is described in publications dealing with the high strain rates, e.g. [12,13].

Fig. 1. Microstructure of the HE copper, ε = 2.24; longitudinal section of the sample

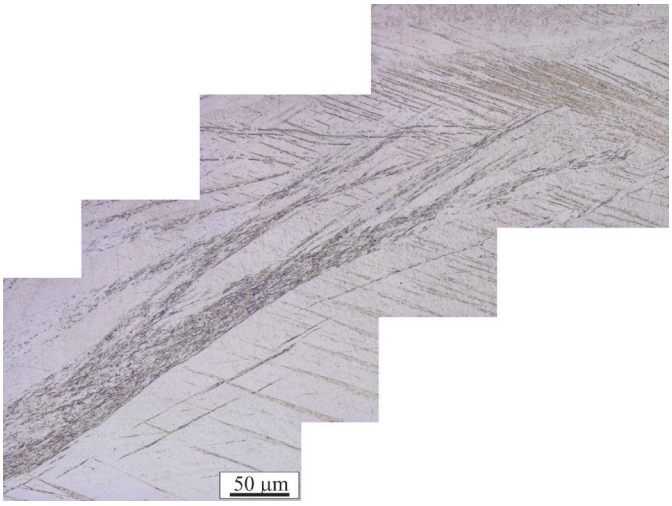

Fig. 2. Boundles of bands; ε = 1.81

In the substructure, microbands were observed in the background of dislocation cells microstructure. They were grouped into characteristic “bundles”, which propagated over considerable distances (Fig. 3f). Some of the microbands were free of the dislocations occurring inside them, while others showed a large number of these dislocations. Inside some of microbands, there were also small cells (Fig. 2a) and some of them were composed of submicron sized grains (Fig. 4b) probably resulting from the dynamic recrystallization. From the literature data it is the phenomenon very well known that the shear bands are privileged sites for the nucleation of new grains and nanograins formation [14].

The boundaries of microbands were built from dense tangles of dislocations. Also characteristic were intersections of microbands, particularly well visible in Figures 3b and 4b. Another feature of the HE copper microstructure was the occurrence of small grains and subgrains formed near the

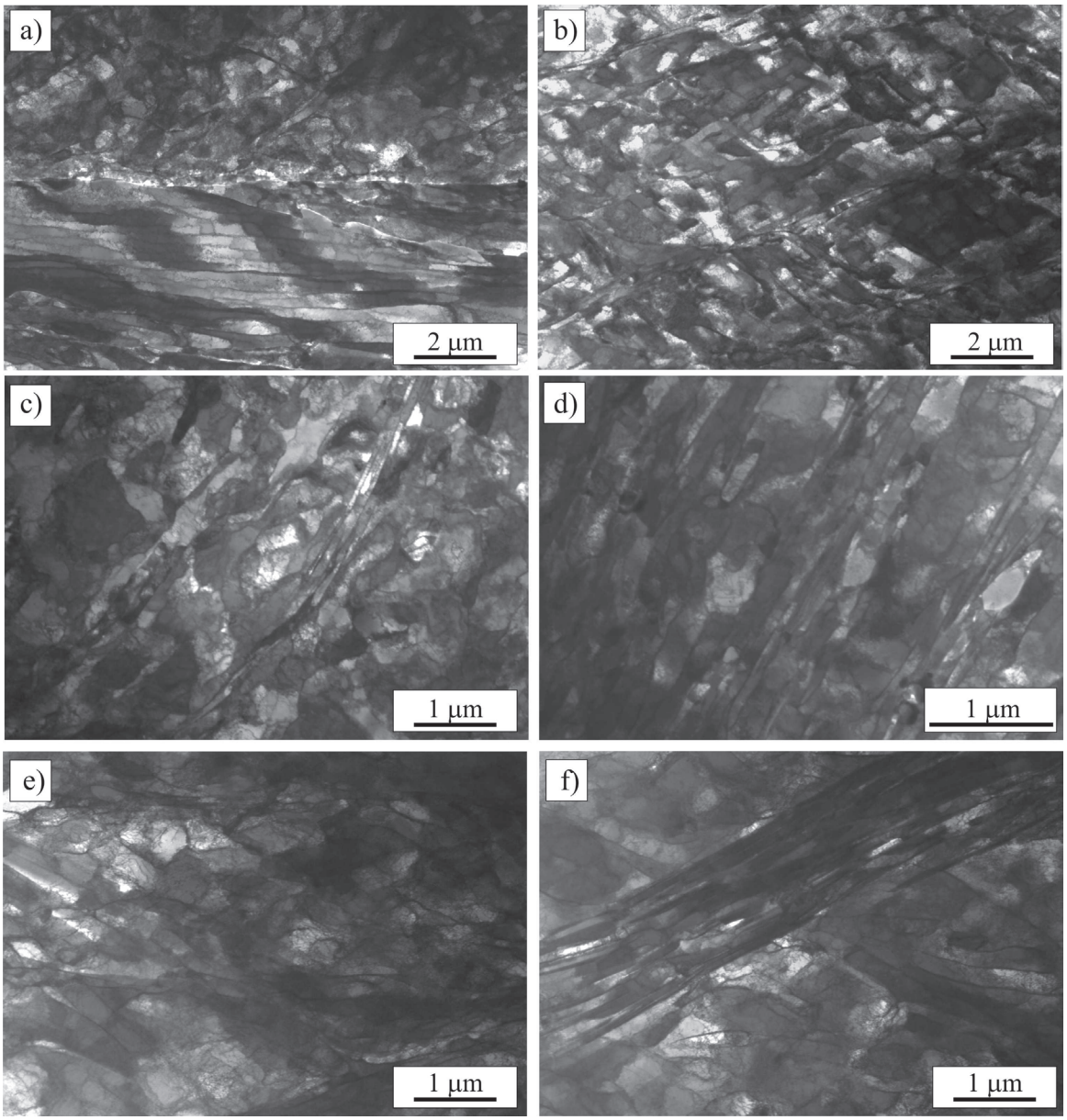

Fig. 3. Selected microstructures of HE Cu99.99 coper; a, b) ε = 1.39 (longitudinal section), c, d) ε = 1.81 (cross section), e, f) ε = 2.24 (longitudinal section); STEM

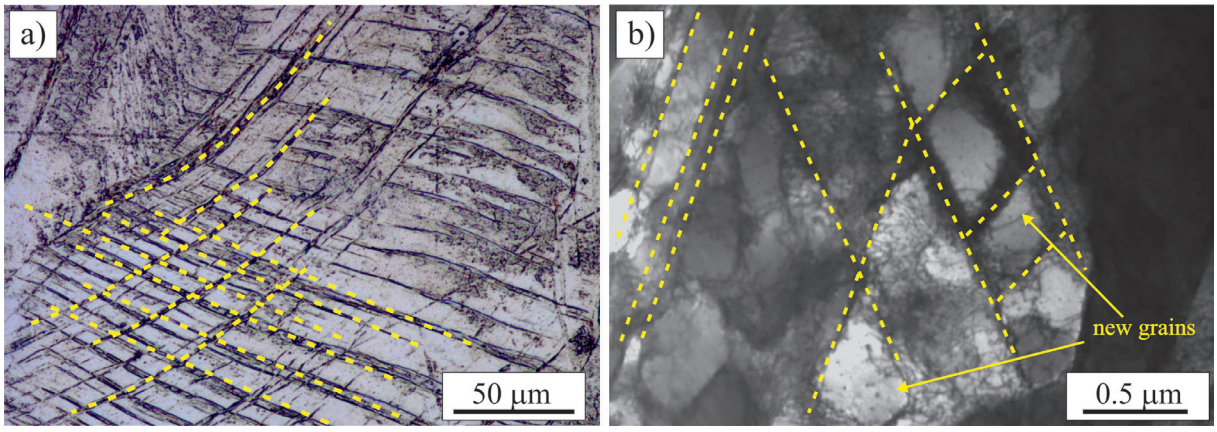

Fig. 4. Mechanism of the new grains formation; a) as a result of bands intersection, ε = 1.81; b) as a result of microbands intersection, ε = 2.24

regions of mutually crossing microbands (Fig. 4b). Some of them were observed to have dislocations inside, while others were free of these dislocations.

The obtained results show that the microstructure of copper deformed by hydrostatic extrusion is rather inhomogeneous, there are regions strongly deformed with high density of dislocations in the neighbourhood of cells which are free of dislocations (Fig. 3). The new recrystallized grains near microbands are also observed (Fig. 3d). Examinations have revealed that at first the new grains were spreading along the direction of bands propagation. Probably microstructure recovery occurred during the deformation under the effect of large strains or just after the deformation as a result of dynamic or post dynamic recrystallization process.

As was noted above, a characteristic feature of HE copper was the intersection of bands and microbands. The new grains can form as a result of microbands intersection and the microstructure renewal process, as seen in Figure 4b. A similar situation takes place in the samples deformed with enormous strains by the cyclic extrusion compression method (CEC) [8]. This phenomenon is also connected with considerable energy storage in the vicinity of newly created grain boundaries. Under the conditions of high strain rates this can lead to a considerable temperature rise in the whole system and the processes of structure renewal are intensified.

The results of grain/subgrain measurements presented in Table 3 show that after hydrostatic extrusion the microstructure refinement occurs. An average grain size decreases from the initial value of ~250 μm to ~0.55 μm after ε = 1.39. In the range of higher deformations, i.e. ε = 1.81 – 2.91, further reduction in the grain/subgrain size occurs and the measured grain/subgrain size is about ~0.35 – 0.39. These results allow qualifying the examined materials as ultrafine grained.

The EBSD analysis of the copper deformed to ε = 2.24 shows the domination of the small misorientation angles. The fraction of large misorientation angles was about 40%. The large misorientation angles were found among others between small grains formed inside shear bands. The inverse pole figures (IPF) corresponding to the EBSD maps (inserts in Fig. 5) show fibre-like texture with the maximum close to [112].

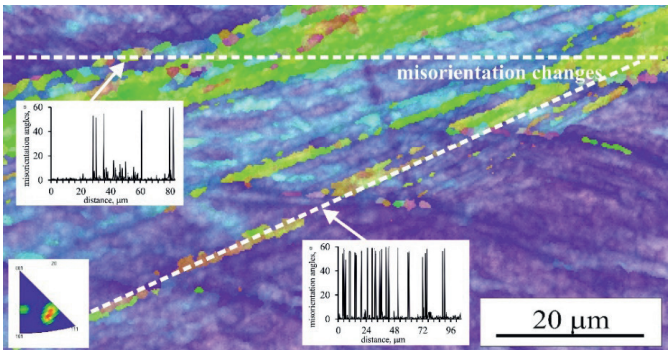

Fig. 5. EBSD map of the microstructure of a HE Cu99.99 copper and misorientation changes along the dashed lines; ε = 2.24

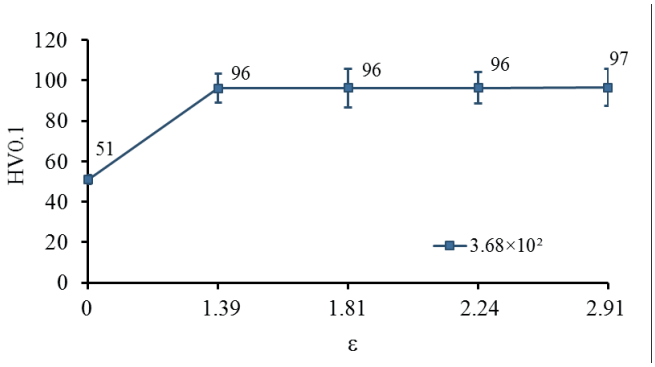

Fig. 6. Influence of deformation value on microhardness of HE copper Cu99.99

Figure 5 shows the results of microhardness measurements taken after the HE process. Compared to the initial state, a nearly double increase in microhardness values after the deformation was observed, i.e. from 45 HV0.1 to about 96 HV0.1. An almost stable microhardness level was found in all ranges of the deformation. The "plateau" of properties indicates that a state of equilibrium has been achieved between the hardening and renewal processes.

Likewise, no serious changes have been observed in the UTS and YS values in the investigated ranges of deformation. UTS increased nearly two times, YS increased more than five times compared to the initial state, a threefold to fivefold decrease in elongation was observed (Table 3). The UTS and YS values were in the range of 411-427 MPa and 370 – 420 MPa, respectively.

TABLE 3

Mechanical properties and the corresponding average grain size of HE Cu99.99 copper subjected to different degrees of deformation at a constant strain rate of 3.68x102s^{-1}

ε	UTS, MPa	YS, MPa	Af, %	Average grain/subgrain size
0	207	75	36	250 μm
1.39	427	420	7	0.55 μm
1.81	411	370	11	0.39 μm
2.24	423	399	8	0.38 μm
2.91	426	410	10	0.35 μm

The results obtained and presented in this paper indicate a strong possibility of production by the hydrostatic extrusion process of UFG copper with the grain size below 0.5 μm.

4. Conclusions

The results obtained in the present study lead to the following conclusions:

- An increase in deformation level under the conditions of constant strain rate ($3.68x10^{2}s^{-1}$) was observed to have no major effect on changes in the microstructure and properties - microhardness level and the UTS nearly doubled compared to the initial state.
- The mutually crossing microbands led to the formation of micro– and nanovolumes, which could transform into new grains.
- The microstructure of copper deformed by hydrostatic extrusion is rather inhomogeneous, there are regions strongly deformed with high dislocation density in the neighbourhood of cells which are free of dislocations.

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