A R C H I V E S O F M E T A L L U R G Y A N D M A T E R I A L S
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MICROSTRUCTURE AND TEXTURE DEVELOPMENT IN Ag-Cu ALLOYS SUBJECTED TO SEVERE PLASTIC DEFORMATION

ROZWÓJ MIKROSTRUKTURY I TEKSTURY W STOPACH Ag-Cu PODDANYCH DUŻEMU ODKSZTAŁCENIU PLASTYCZNEMU

In this work, modification of microstructure and texture development in Ag and Ag-Cu system alloys (Ag – 4% wt. Cu, Ag – 12% wt. Cu) subjected to Severe Plastic Deformation (SPD) was analysed. Rods of 15 mm diameter were the starting material for the experiment, they were obtained with the extrusion-torsion (ET) method. Next, the rods were subject to deformation by hydrostatic extrusion (HE) (ε = 3.2) and 3 times subjected to Equal Channel Angular Pressing (ECAP) – ε = 3.4.

As a result of the tests performed, it was found out that the alloys examined, subjected to SPD, displayed a high refinement of the microstructure, which led to a significant improvement of mechanical properties. The microstructure of materials produced by SPD strongly depends on the technological parameters of deformation.

In the initial state (ET deformation) all of the alloys exhibited a fibrous character of texture. A similar fibrous texture characteristic was also found after HE, whereas after the ECAP the initial texture was completely changed. In all methods of deformation, the Ag sample shows a very strong texture comparison to Ag – 4% wt. Cu and Ag – 12% wt. Cu.

Keywords: Severe plastic deformation, microstructure, texture, Ag – Cu alloys

W niniejszej pracy dokonano zmian mikrostruktury i tekstury w stopach z układu Ag – Cu (Ag – 4% wg. Cu, Ag – 12% wg. Cu) oraz w czystym srebrze po dużym odkształceniu plastycznym. Pręty w stanie wyjściowym o średnicy 15 mm uzyskano poprzez zastosowanie wyciskania ze skręcaniem wlewków o średnicy 50 mm. Następnie poddano dużemu odkształceniu plastycznemu przez zagięty kanał kątowy (ε = 3.4).

Przeprowadzone badania wykazały znaczne rozdrobnienie mikrostruktury stopów poddanych SPD w odniesieniu do stanu wyjściowego. Charakter otrzymanej mikrostruktury jest silnie uzależniony od rodzaju zastosowanej metody odkształcenia.

W stanie wyjściowym wszystkie badane materiały wykazywały osiowy charakter tekstury. Podobnie sytuacja wyglądała w przypadku wyciskania hydrostatycznego, jednakże metoda ECAP spowodowała całkowitą zmianę charakteru tekstury w odniesieniu do stanu wyjściowego. Po wszystkich metodach odkształcania stwierdzono ponadto, że próbki wykonane z czystego srebra wykazują silniejszą teksturę w odniesieniu do próbek Ag – 4% wg. Cu oraz Ag – 12% wg. Cu.

1. Introduction

The last two decades may be described as dominated by nanomaterials, owing to the huge interest in R&D efforts targeted at techniques used to produce materials with ultrafine- and nano-grained structures.

One of the popular techniques applied in the production of massive nano-crystalline materials is the SPD. This technique involves subjecting material to plastic deformations ($\varepsilon > 1$) with a simultaneous application of high pressure reaching several GPa at relatively low temperatures [1]. One of the SPD techniques is a hydrostatic extrusion process, where the charge is surrounded with pressure medium, whereas the piston moving inwards the working chamber builds up hydrostatic pressure that impacts the sample. During the extrusion process, a high degree of microstructure homogeneity and relatively high volumes of ultrafine-grained and nano-crystalline materials are obtained [2]. A unique feature of the HE process is the fact that the material subject to extrusion in a working chamber is surrounded with the so-called pressure medium. Therefore, in the deformation zone (die), the material is subject to triaxial compression, which significantly hampers the micro-crack generation and propagation processes.

Forcing the material through a folded angular channel-die is yet another SPD technique that allows to produce ultrafine-grained and even nano-crystalline

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structures of materials [3]. During this technique, a sample is extruded through a die containing two channels, equal in cross section, intersecting at an angle. A sample with an almost identical cross section is inserted into one of the channels, and then by means of a punch is extracted into the other channel. Depending on the angle applied, various shapes of grains are achieved respectively in the material deformed [4].

Another technique, applied in particular to hardly deformable materials, is the Extrusion Torsion (ET) process. It allows to fragment the structure to ultra – and nanometric dimensions. [5]. In this case, in addition to the conventional setup that brings axial thrust on the extruded material, a reverse motion of a die is introduced i.e. torsion at a certain angle and frequency, that decidedly facilitates the extrusion process in a highly effective manner.

It should also be noted that the materials subjected to SPD are characterised by strong crystallographic texture that leads to high anisotropy of mechanical properties. [6, 7]. In literature [8, 9], the texture that has originated during ECAP is frequently compared to that obtained as a result of pure shearing. Meanwhile, while applying the HE process to material deformation, one can expect a texture similar in nature to that obtained during conventional extrusion [10]. The aim of the current work is to analyse the development of microstructure and texture in the silver -copper alloys and pure silver processed by HE and ECAP.

2. Experimental procedure

For testing purposes, the Ag-Cu alloys and pure silver material with the chemical constitution specified in Table 1 were used.

TABLE 1

Chemical composition of the investigated alloys (wt. %)

Alloy	Composition [wt. %]	
	Ag	Cu
Ag	99,9	–
Ag4Cu	96	4
Ag12Cu	88	12

The starting material for the experiment were 15 mm diameter rods obtained with the use of the Extrusion-Torsion (ET) technique. The application of HE reduced the rod diameter down to 3 mm, which gives deformation equal to $\varepsilon = 3.2$. Prior to the deformation of samples using the ECAP technique, they were prepared by annealing of input rods at 450°C for 1.5 hours in the case of Ag4Cu and Ag12Cu alloy rods, and at temperature of 350°C for 1 hour in the case of Ag rods. Then, the samples were subjected three times to Equal Channel Angular Pressing (ECAP) ($\varepsilon = 3.4$).

In order to make observations using Transmission Electron Microscope (TEM), the tested material was made thin mechanically, using the Dimple Grynder Gatan Model 656. To obtain TEM thin film samples, the 691 Precision Ion Polishing System (PIPS^{TM}) by Gatan Inc was used. The microstructure was examined using a TEM made by Philips, while applying the accelerating voltage equal to 100kV.

The following parameters were measured for each grain section: d_{max}, grain maximum diameter, d_{eq} – grain equivalent diameter and α defined as d_{max}/d_{eq}. The shape factor α is a sensitive measure of grain elongation. It is equal to 1 for circles and exceeds 1.5 for elongated grains.

The texture was analysed on the grounds of incomplete pole figures (111), (200), (311). The measurements were made using the Brucker D8 Discover X-ray diffractometer, at planes perpendicular to the extrusion direction, applying filtered radiation Cu Kα (Kα_1 = 0.154 nm). On the basis of measured pole figures, the Orientation Distribution Functions (ODF) were computed for each sample, and then the quantitative assessment of the shares of major texture constituents was made.

3. Results and discussion

The microstructure and texture of investigated alloys before and after SPD were studied. For texture analysis several pole figures were measured by standard X-ray diffraction techniques and used to compute the three dimensional orientation distribution functions. All measurements were done on the plane perpendicular to the extrusion direction.

4. Microstructure and texture of the starting materials

Figure 1 shows the microstructures of tested materials following the Extrusion-Torsion (ET) process. Stereological analysis clearly indicated that the average grain size in the tested materials decreases in line with the amount of copper added. For the pure silver sample, the equiaxial grains were characterised by the largest size of all the alloys and their states tested (1.15 μm). Other materials were characterised by a microstructure fragmented to a much greater extent, with grains having a shape close to the equiaxial (Fig. 2) and medium-size of approximately 0.36 μm (Ag4Cu) and 0.23 μm (Ag12Cu).

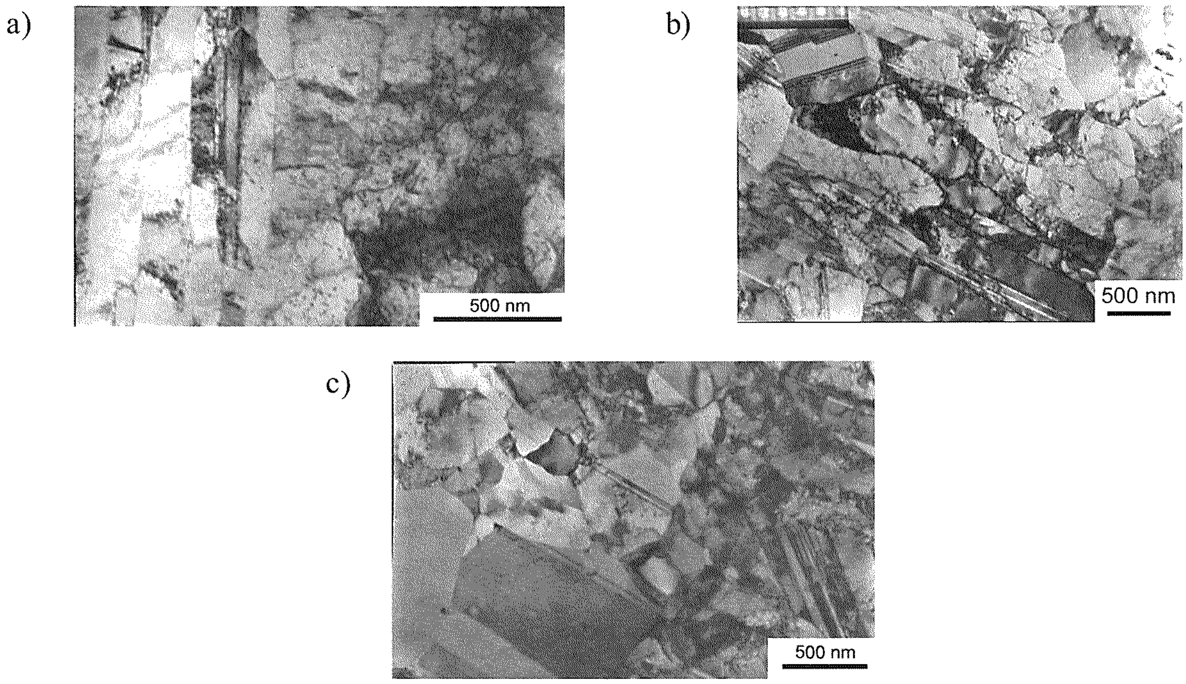

Fig. 1. Microstructure of the investigated alloys in an initial state: Ag (a), Ag4Cu (b) and Ag12Cu (c) (cross – sections perpendicular to the extrusion direction)

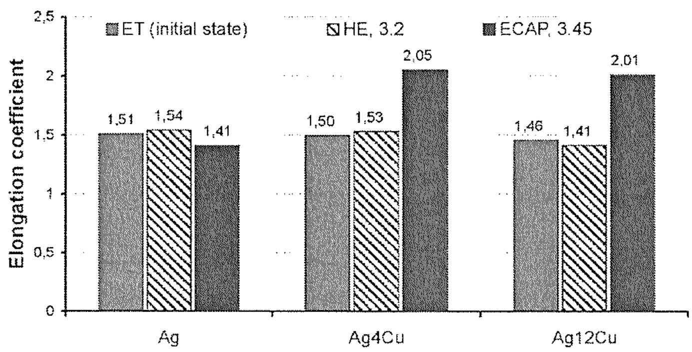

Fig. 2. Grain elongation coefficient (α) of the investigated alloys after different methods of SPD

In the pure silver sample subjected to the Extrusion-Torsion (ET) process, a strong (maximum FRO value = 27) fibrous texture of <111> type was formed – its volume fraction exceeded 42%. Moreover, the texture component <221> was formed equally strong, and it accounted for more than 20% of volume. Figure 3 shows FRO for the pure silver sample in the initial state.

With regard to the Ag4Cu and Ag12Cu alloys in their initial state, it can be stated that in both cases the axial texture can be described using the {h11} and {h21} fibres. In the sample with a lower copper content, the share of the {h11} fibre accounts for more than 30% of the sample volume, whereas the share of h21 fibre – for 36%. Within the limits of the {h11} fibre it is possible to distinguish the following components: <311>, <411>, <511>, whose volume fractions are specified in Table 2. In the Ag12Cu alloy the texture looks similar, however, the difference is that the volume fraction of the {h11} fibre is much lower – 24%, whereas the volume fraction of the {h21} fibre amounts to 23%. The share of the <100> fibre should be also noted – in the Ag4Cu alloy material it accounts for 8%, whereas in the Ag12Cu sample it is almost three times lower and amounts to less than 3%. Meanwhile the share of the {111} fibre is at a similar level and accounts for approximately 4%. It should be

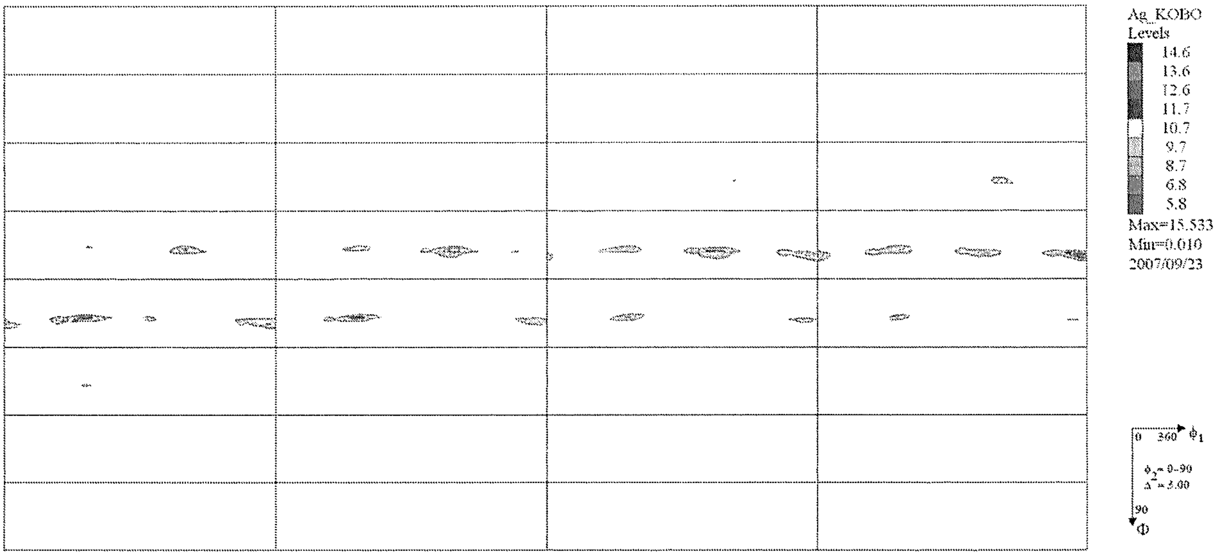

Fig. 3. ODF's for constant ϕ_2 value in range of 0-90°/5° for Ag alloy in an initial state

also noted that textures formed are not too sharp, as evidenced by the low FRO values. Moreover, the computed volume fractions of individual texture components and the FRO values give grounds to state that the texture of the Ag4Cu sample is formed more strongly than that of the Ag12Cu sample.

TABLE 2

Volume fraction of the principal texture components in the investigated alloys in an initial state

Material	Texture component {hkl}		Volume fraction of the main texture components V [%]	
Ag	<111>		42.6	
	<221>		22.3	
Ag4Cu	h11	<311>	31.3	7.8
		<511>		12.7
		<411>		10.3
		<011>		0.5
	h21	<521>	35.5	14.1
		<821>		21.4
	<221>		8.9	
	<111>		4	
	<100>		8.4	
Ag12Cu	h11	<311>	24.5	5.7
		<511>		8.4
		<411>		8.6
		<011>		1.8
	h21	<521>	22.9	9.3
		<821>		13.6
	<221>		21.8	
	<111>		3.8	
	<100>		2.7	

After the HE processing

The HE application led to the reduction of the average grain size from approximately 0.36 μm in the initial state, down to 0.19 μm for the Ag4Cu alloy material (Figure 4). In the case of an alloy with a higher copper content, the Hydrostatic Extrusion process resulted in fragmentation of the microstructure, and consequently the average output grain size equal to 90 nm was obtained. In the pure silver sample, on the other hand, the value of the equivalent diameter was reduced twofold after the application of HE technique. It should be noted that in all cases the shape of grains has practically remained unchanged as compared to the initial state – this is evidenced by the values of α coefficient (Fig. 2).

The pure silver sample subjected to the HE technique was also characterised by an axial texture, however, the {111} fibre, which was predominant prior to the SPD application, has undergone a significant broadening and weakening. At the same time other texture components were formed; they are presented in Table 3. The hydrostatic extrusion of a sample with a lower content of copper led to a decline of volume fractions of almost all identified fibres (Table 3) as compared to the state prior to the deformation. An exception here is the {221} fibre, the share of which increased more than twofold. Figure 5 shows FRO for the Ag4Cu sample after the hydrostatic extrusion process.

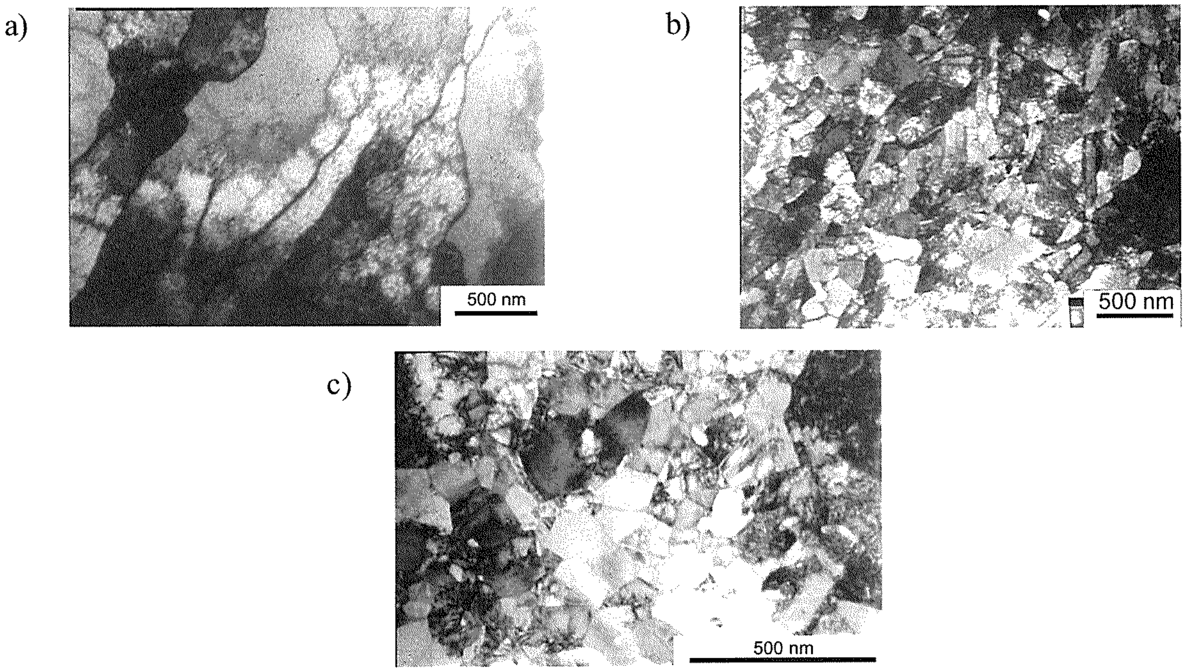

Fig. 4. Microstructure of the investigated alloys after HE: Ag (a), Ag4Cu (b) and Ag12Cu (c) (cross – sections perpendicular to the extrusion direction)

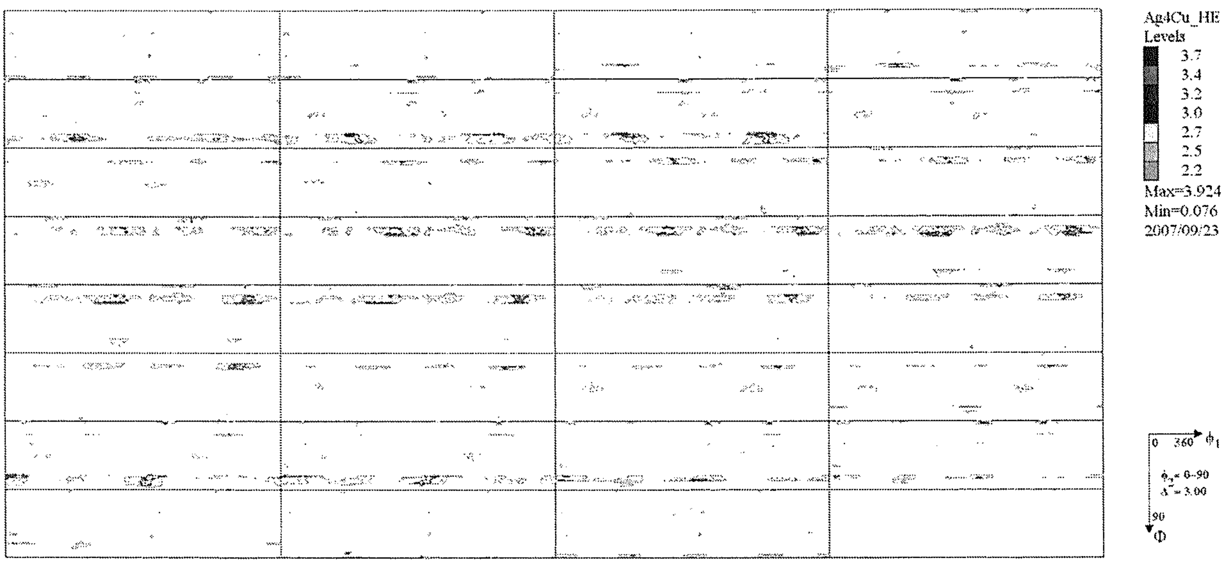

Fig. 5. ODF's for constant ϕ_2 value in range of 0-90°/5° for Ag4Cu alloy after HE

In the Ag12Cu sample, the Hydrostatic Extrusion process has not led to such significant changes in volume fraction of individual texture components as in the Ag4Cu sample. It should be noted that following SPD the share of the same components is higher for the sample with a higher copper content.

TABLE 3
Volume fraction of the principal texture components in the investigated alloys after HE

Material	Texture component {hkl}		Volume fraction of the main components V [%]	
Ag	<417>		13.6	
	<135>		14.02	
Ag4Cu	h11	<311>	23.6	5.5
		<511>		7.0
		<411>		8.2
		<011>		2.9
	h21	<521>	20.1	9.2
		<821>		10.9
	<221>		21.4	
	<111>		1.0	
	<100>		1.8	
Ag12Cu	h11	<311>	29.9	7.3
		<511>		8.8
		<411>		10.3
		<011>		3.5
	h21	<521>	22.7	11.0
		<821>		11.7
	<221>		26.2	
	<111>		1.4	
	<100>		2.3	

After the ECAP processing

Figure 6 shows the microstructure of the tested materials after the ECAP processing.

Application of the ECAP technique led to the formation of a microstructure in the Ag4Cu and Ag12Cu alloy materials, one that is characteristic for the materials subjected to severe plastic deformation. The average grain size for these two alloy materials was 80 and 115 nm respectively. Moreover, the deformation led to the apparent fragmentation of original grains and their elongation. The determined α coefficient for this technique is presented in Figure 2. Such high values indicate high grain elongation. In the case of the pure silver sample, the grains assumed a shape close to the equiaxial shape (α = 1.4), and the grain average size was 80 nm.

The ECAP technique resulted in the formation of a different material texture than that in the initial state – the tested materials were not characterised by the existence of an axial texture. Similarly to the previous cases, the nature of the texture of the pure silver sample differs from the texture of the samples with copper added. In the pure silver sample, the maximum FRO values (above 30), indicate a sharp texture with the following predominant components: {122} <221> and {110} <1-11>. In the

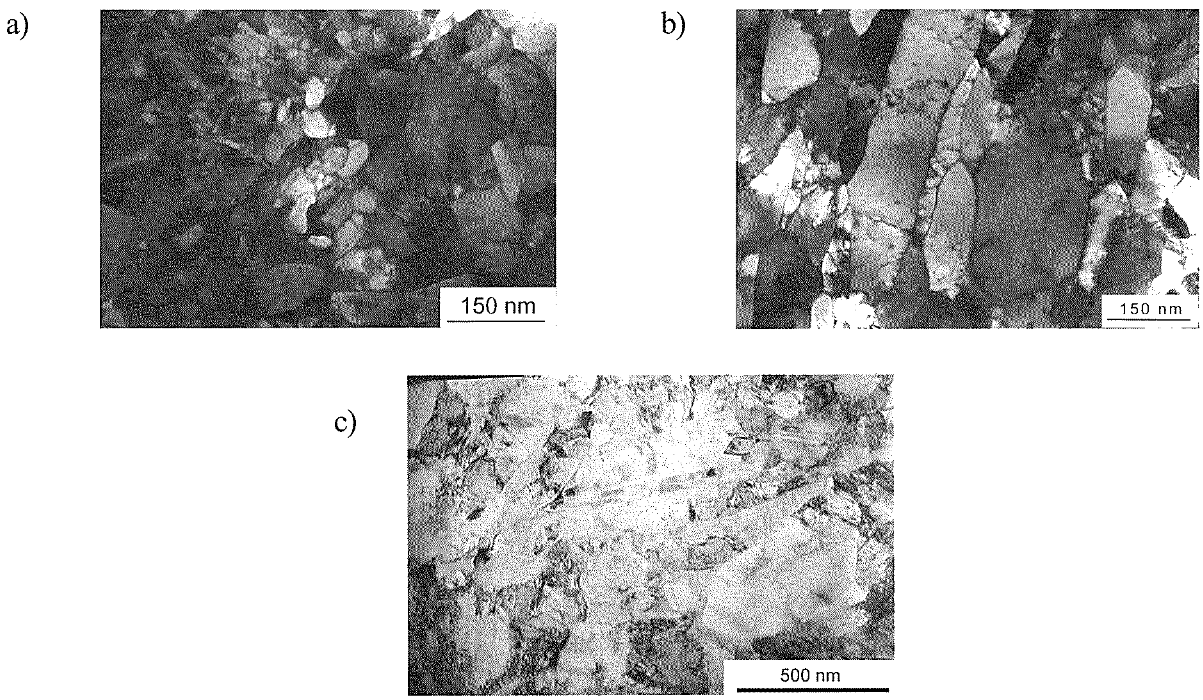

Fig. 6. Microstructure of the investigated alloys after ECAP: Ag (a), Ag4Cu (b) and Ag12Cu (c) (cross – sections perpendicular to the extrusion direction)

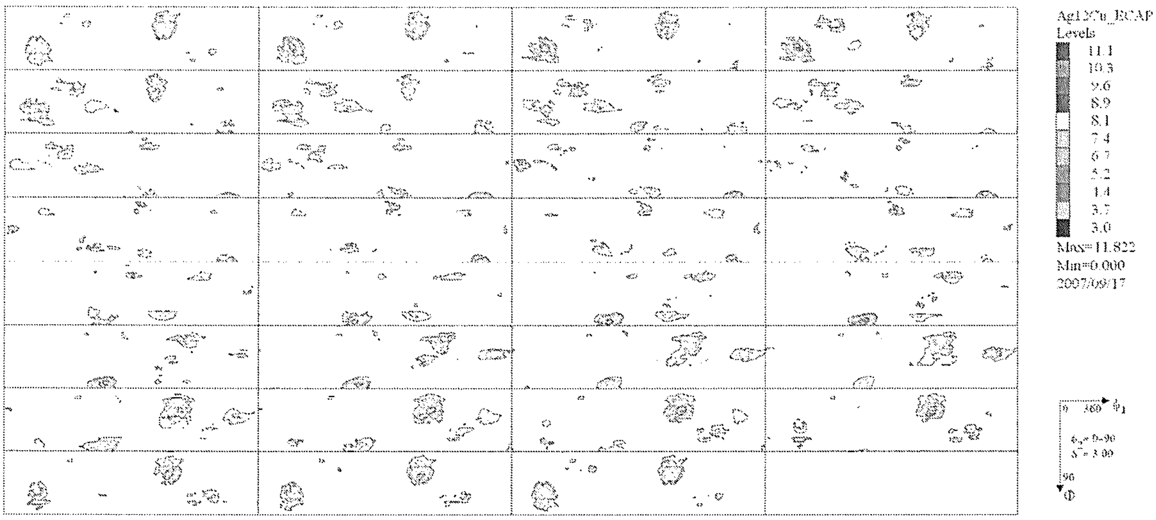

Fig. 7. ODF's for constant ϕ_2 value in range of 0-90°/5° for Ag12Cu alloy after ECAP

the case of other samples, the FRO values slightly exceeded 12. Figure 7 shows representative FRO for the Ag12Cu sample, whereas Table 4 shows the final collated results for the samples deformed using the ECAP technique.

TABLE 4

Volume fraction of the principal texture components in the investigated alloys after ECAP

Material	Texture component {hkl}	Volume fraction of the main components [%]
Ag	{122} <221>	6.8
	{110} <1-11>	5.0
	{454} <-44-1>	3.5
	{586} <4-1-2>	4.1
	{14 1 13} <-2-11 3>	3.0
Ag4Cu	{9 1 2} <1 5 -7>	5.5
	{1 1 5} <10 -15 1>	5.9
	{1 2 11} <5 -8 1>	5.3
	{1 1 7} <6 -13 1>	4.6
	{1 1 7} <8 -15 1>	5.1
	{1 1 6} <5 -11 1>	4.8
	{3 2 13} <2 -3 0>	5.6
	{1 1 3} <1 -1 0>	2.8
Ag12Cu	{8 5 1} < -3 4 4 >	6.78
	{10 2 15} <5 5 -4>	6.6
	{9 2 14} <10 11 -8>	6.78
	{1 6 4} <6 -5 6>	6.47
	{3 1 5} < 9 8 -7>	6.17
	{1 10 5} <5 -3 5>	6.42
	{0 2 1} <2 -1 2>	6.48
	{1 11 15} <-3 3 -2>	3.68

5. Conclusions

The results of the investigations indicate a strong refinement of the microstructure of examined alloys as a result of their HE and ECAP deformation. Evaluated texture of examined alloys after the HE process is relatively weak and can be described by a <11X> and <12X> fibres, while the texture after the ECAP deformation is much less distinct. This can be interpreted as being a result of its destruction during the deformation process.

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