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, TEXTURE AND MECHANICAL PROPERTIES OF COPPER UNDER ARB PROCESSING

MIKROSTRUKTURA, TEKSTURA I WŁASNOŚCI MECHANICZNE MIEDZI PODDANEJ PROCESOWI ARB

The accumulative roll-bonding (ARB) process, invented not long ago, is a promising mode for introducing severe plastic deformation into industrial practice to produce bulk ultra-fine grained materials. The ARB process consists in rolling of the pack of two sheets up to 50% reduction. Then, the rolled material is sectioned into two halves, stacked and the procedure of roll-bonding is repeated. The orientation distribution of ARB processed Cu 99.95% up to ε ~ 4 is analyzed in the paper. The evolution of crystallographic texture has been discussed in relation with changes of mechanical properties and structure.
Keywords: accumulative roll-bonding, copper, microstructure, texture, mechanical properties

Metoda wielokrotnego, połączonego ze spajaniem podczas następujących po sobie przepustów, walcowania pakietowego (Acumulative roll-bonding – ARB) leży u podstaw wzrastającego zainteresowania wprowadzeniem do praktyki przemysłowej intensywnego odkształcania plastycznego (severe plastic deformation – SPD) w celu wytwarzania materiałów ultradrobnokrystalicznych. Polega ta metoda na walcowaniu pakietu, złożonego z dwóch fragmentów blachy, do ich grubości początkowej, czyli z 50% redukcją przekroju poprzecznego, następnie przepołowieniu tak otrzymanej blachy i ponownym przeprowadzeniu opisanego postępowania. Tak pomyślany proces może być kontynuowany teoretycznie bez ograniczeń.

W pracy przedstawiono wyniki badań miedzi Cu 99.95%, odkształcanej metodą spajania w procesie walcowania pakietowego aż do ε ~ 4. Dyskutowane są zmiany tekstury krystalograficznej w odniesieniu do zmian własności mechanicznych i mikrostruktury.

1. Introduction

The increased interest in materials of the ultra-fine grained (UFG) structure, with the grain size of sub-micrometers order, known as nanocrystalline materials when the grain diameter is below 100 nm, is due to the evident advantages resulting from the development of such a structure and manifested by increased strength properties and greater hardness at higher ductility. Moreover the low temperature superplastic flow occurring in these materials permits to reduce the drawing temperature of complicated products [1, 2]. In our experiments of UFG cold rolled up to 96% copper sheets, the tensile elongations in rolling direction RD was significantly greater than those of cold rolled sheets recrystallized to obtain small or coarse grains. Severely pre-deformed copper demonstrated higher yield point as well as the ultimate tensile strength in RD direction. The macroscopic shear banding during cold rolling was highly reduced in sheets preliminarily severely deformed by equal-channel angular pressing (ECAP) processing [3, 4]. The improvement of mechanical properties of cold rolled copper sheets is promising for industrial practice as it extends the range of their workability. These improvements stimulate the research on influence of grain size on copper properties in the range of UFG structure [5, 6]. Severe Plastic deformation (SPD) methods, like ECAP method [1, 2, 7] or high pressure torsion (HPT) [1] should have been widely applied to produce bulk ultra-fine grained materials if these had not required special equipment.

The roll-bonding, applied generally to perform a special purposes cladding, seems to be the most efficient and economical procedure for manufacturing the ultra-fine grained materials by severe plastic deformation.

The accumulative roll-bonding (ARB) consists in rolling to 50% reduction of the two sheets pack, of which the stacked surfaces were initially cleaned [8]

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(some recent studies, dedicated ARB processing are presented in papers [9–11]). Then, the rolled material is sectioned into two halves, stacked again and the procedure of roll-bonding repeated. Generally, the severe plastic deformation methods like the ARB or the ECAP can produce bulk samples of relatively large dimensions and without pores compared with the powder metallurgy methods, in which the pores are difficult to remove. The ARB method, additionally, can supply semi-final products in the form of sheets, which can be directly used for further work forming operation.

The paper is aimed at presenting the changes of mechanical properties during accumulative roll-bonding of Cu 99.95% strips. The evolution of crystallographic texture and grain size in dependence on number of passes is also discussed.

2. Accumulative roll-bonding

Fig. 1 presents the scheme of accumulative roll-bonding (ARB) procedure. Two pieces of strip with clean and degreased surfaces are put together and after annealing joined by means of 50% consecutive rolling. The obtained strip which is composed of two bond layers is then sectioned in two pieces and after cleaning of the surfaces which are to be fixed together, consecutively heated and rolled 50% to perform roll-bonding. Due to 50% rolling reduction followed by stacking 2 halves and consecutive 50% rolling reduction, the procedure can be continued practically without limits [8, 9].

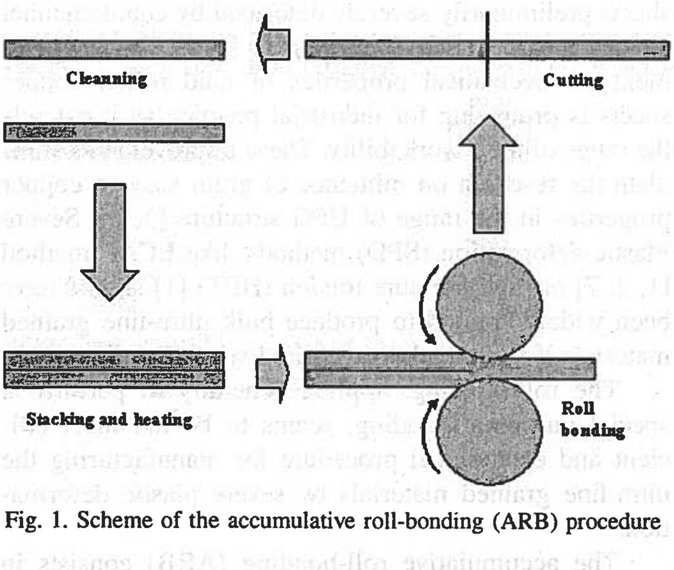


Fig. 1. Scheme of the accumulative roll-bonding (ARB) procedure

Suppose the strip with initial thickness t_o, was rolled with 50% reduction in each cycle, so the thickness t_n of individual layer, after n cycles, can be calculated according to the formula

$$t_n = t_o / 2^n. \tag{1}$$

The total reduction z_n after n cycles is

$$z_n = 1 - t_n / t_o = 1 - 1/2^n \tag{2}$$

and with the Huber-Hencky-v. Mises yield criterion of plasticity [8] and plane strain state of deformation under rolling the equivalent plastic strain ε is equal to

$$\varepsilon_n = \left\{ \frac{2}{\sqrt{3}} \ln\left(\frac{1}{2}\right) \right\} * n = 0.8 \text{ n}. \tag{3}$$

Assuming initial thickness t_o = 1 mm and performing exemplary n = 5 cycle roll-bonding, the thickness of layer equal to t_5 = 1/32 mm = 31.25 μm, total reduction z_5 = 96.875% and total equivalent deformation ε_5 = 4; in the case of n =10 cycles of roll-bonding the final thickness of layer is equal to t_{10} = 1/1024 mm = 0.98 μm, total reduction z_{10} = 99.9% and total equivalent deformation ε_{10} = 8 can be obtained.

Although relatively large deformation can be arrived, in fact the end parts of strip have to be prepared each time after roll-bonding and so, with increasing number of cycles, the length of roll-bonded strip decreases.

3. Material and experimental technique

Experiments were carried out on copper Cu 99.95 recrystallized at 773 K, with the initial grain size diameter d = 30 μm [11]. Tensile test were performed by means of INSTRON 6025 machine and specimens with the heads and gauge dimensions 20×4 mm^2 were cut out in RD direction of roll-bonded sheets.

The changes of crystallographic orientations were studied by the analysis of pole figures. The measurements of pole figures in the middle layer of the rolled strip were performed using Philips X-ray diffractometer and CoKa characteristic radiation and using neutron diffraction method at Leon Brillouin Laboratory of CEA/Saclay, France for the average global texture of stocked sheets of 1 cm^3. For local texture, the measurements of pole figures were performed using scanning electron microscopy (SEM) equipped with electron back scattering diffraction (EBSD) technique and using orientation imaging microscopy (OIM™) software.

Microstructure observations (TEM) were carried out using Philips CM20 electron microscope; thin foils, parallel to sheet plane, were prepared by electrolytic thinning.

4. Results and discussion

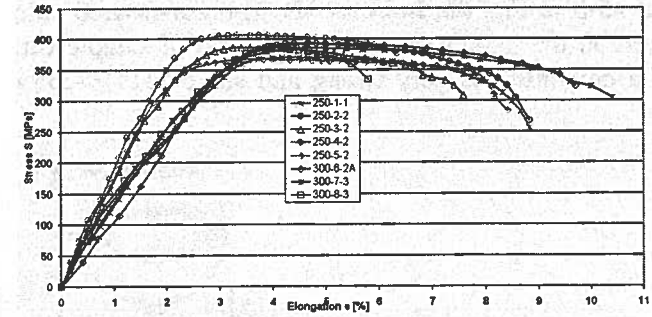
4.1. Tensile tests

The experiments of roll-bonding at ambient temperature usually do not give satisfactory good final product as was discussed in the case of aluminium [9, 10]. Then an initial heating of copper strips was executed before the roll-bonding pass. The results of tensile tests in dependence on temperature and number of passes are presented in Table. Temperature 250°C÷350°C of initial heating and 5 passes (ε = 4.0) seems to be appropriate for copper as optimal roll-bonding conditions [9]. Similar number of passes was stated in the case of aluminium [9, 10]; an increased number of passes resulted in the worse tensile properties of final product. Exemplary tension curves visualizing the changes of strength and elongation, following the increasing number of passes from 1 to 5, are presented in Fig. 2a. Similarly as in the case of aluminium [9] the strength of copper increased twice and elongation decreased down to 8%. The drop of tensile strength in Fig. 2b illustrates that small inaccuracy in roll-bonding procedure can lead to a drastic change in the strength of processed strips (microstructure and texture changes will be discussed later), suggesting that the saturation state of dislocations was reached in the case of aluminium [9, 10] and copper [11].

TABLE

Tensile properties of Cu 99.95% processed by accumulative roll-bonding; Temperature and number of passes are marked i.e. Cu250-1 is for sample processed by 1 pass at 250°C, r – recrystallized.

No	Sample	YP $R_{0.2}$ [MPa]	UTS R_m [MPa]	Elongation A [%]
1	Cu0-1	365,0	374,5	5,6
2	Cu250-1	357,7	370,0	7,7
3	Cu250-2	363,3	391,3	7,7
4	Cu250-3	366,5	386,5	7,6
5	Cu250-4	370,3	406,4	5,7
6	Cu250-5p	326,0	369,0	7,7
7	Cu250-5a	340,0	416,0	8,9
7	Cu300-1	340,5	348,4	6,4
8	Cu300-5	350,0	369,4	6,5
9	Cu300-6	373,3	392,8	9,5
10	Cu300-7	343,3	372,9	8,5
11	Cu300-8	325,0	370,9	6,5
10	Cu350-5	377,0	392,5	7,9
11	Cu400-1	329,0	347,0	6,3
12	Cu400-5	341,0	354,4	5,8
13	Cu-r	112,5	218,1	48,2



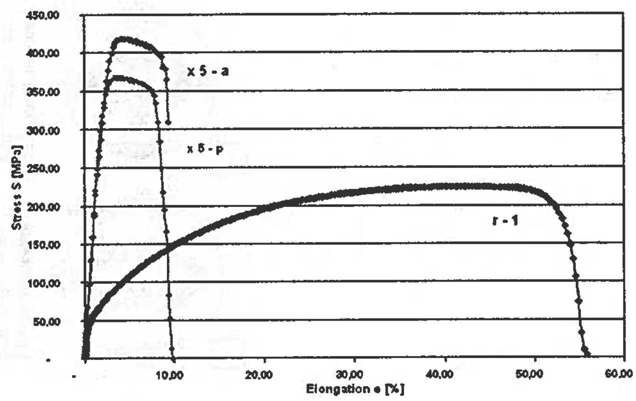


Fig. 2. Tensile stress versus elongation curves for samples a) after roll-bonding at 250°C: 1 pass (250-1-1), 2 passes (250-2-2), 3 passes (250-3-2), 4 passes (250-4-2), 5 passes (250-5-2), and 6 passes at 300°C (300-6-2A), 7 passes at 300°C (300-7-3) and 8 passes at 300°C(300-8-3); (b) recrystallized r-1 and after roll-bonding at 250°C and 5 passes: x 5 – a and x 5 – p

4.2. Crystallographic texture measurement

Figure 3 presents pole figures of roll-bonded strips from 1 to 5 passes measured by the neutron diffraction technique. One can observe the "copper" type rolling texture (the strong {112}<111> component was observed) after the first and fifth passes; in-between passes an additional shear strain texture component {001}<110> was created. In aluminium under roll-bonding conditions however, the distribution of texture component was very inhomogeneous across the thickness of ARB processed strip [12]. The drop of tensile strength after fifth ARB pass observed in Fig. 2a has been confirmed by changes in deformation textures (Fig. 3e, pole figure of 5Cu). It suggests a strong sensitivity of copper to ARB rolling conditions during fifth pass at pre-heating to 250°C. Another two ARB experiments performed with five passes revealed a difference of tensile strength (Fig. 2b). In the sample with lower tensile properties (Cu: x5-p in Fig. 2b) the effects of structure recovery or even early stages of recrystallization were observed (see microscopy observations) which was demonstrated by the X-ray measured pole figure in the middle layer of rolled strip of sample

Cu: x5-p in Fig. 4b; however, the X-ray measured pole figure in the middle layer of rolled strip of sample Cu: x5-a demonstrates very strong and sharp {311}<-233> texture component (Fig. 4a). The shear {001}<110> texture component is absent in both five passes processed samples.

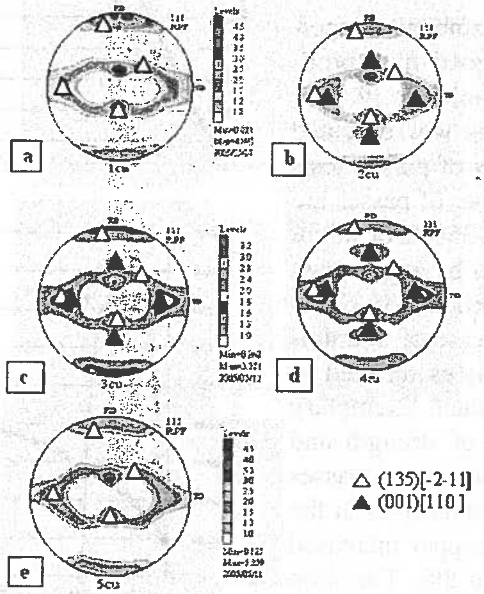


Fig. 3. {111} pole figures measured with the neutron diffraction of ARB processed samples: a) 1 pass, b) 2 passes, c) 3 passes, d) 4 passes and e) 5 passes

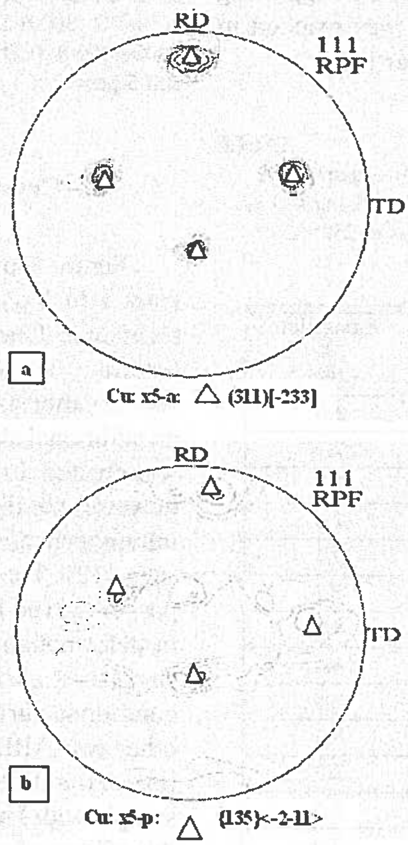


Fig. 4. {111} pole figures measured with the X-ray diffraction of ARB processed samples at 250°C and 5 passes: a) Cu: x 5 – a and b) Cu: x 5 – p

4.3. Scanning electron microscopy observations

Scanning electron microscopy (SEM) examination was performed only for samples processed 5 times (Fig. 5a and 5b). The scans of cross-sections perpendicular to the TD of roll-bonded strips demonstrated still existing traces due to the last roll-bonding. Calculated thickness of individual layer after 5 passes, according to the formula (1), is equal to t_5 = 1/32 mm = 31.25 μm, as can be observed in Fig. 5. The {111} pole figures, reproduced from the scans presented in Fig. 5 and transformed to external reference system with axis: RD, ND and TD, are presented in Fig. 6. Pole figures are presented as equal area projections thus with deformed angular relations. One can recognize "copper" rolling textures (Fig. 3 and Fig. 4), although rotated about the TD direction. The EBSD scan for sample Cu: x 5 – p (Fig. 5b and 6b) reveals greater number of grains, more recovered or even recrystallized in comparison with the scan for Cu: x5 – a (Fig. 5a and 6a) with no relaxed grains. These conclusions are also confirmed by measurement of misorientation angles (Fig. 7) in which about 4 times more grains resolved in the case of Cu: x5 – p (Fig. 7b) in comparison with sample Cu: x5 – a (Fig. 7a) were observed; in both cases the major part of grains was with low angle grain boundaries. It is worth to underline that due to greater grain size and grains with recovered structure, the number of grains resolved by EBSD technique in sample Cu: x5 – p is predominant; in both cases the misorientation for random orientation (MacKenzie plot) is also presented.

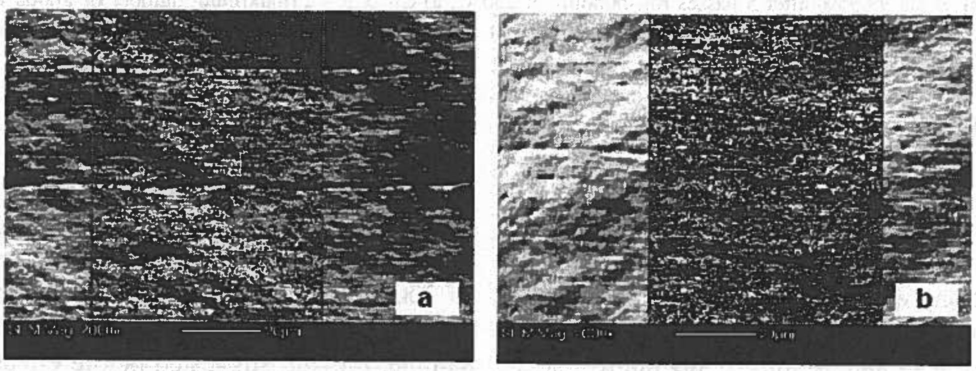


Fig. 5. EBSD scan of ARB processed samples at 250°C and 5 passes: a) Cu: x 5 – a and b) Cu: x 5 – p

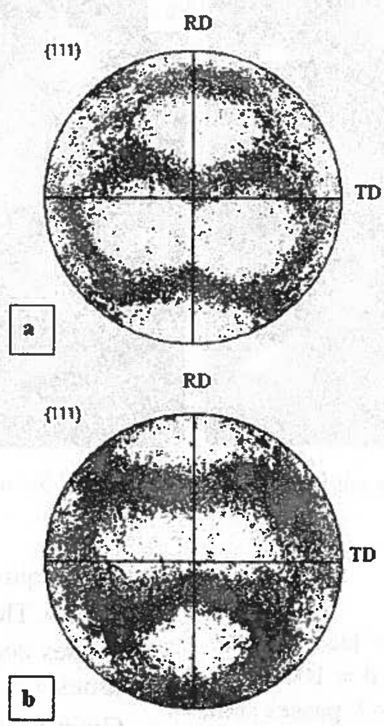


Fig. 6. {111} pole figures, reproduced from EBSD scans presented in Fig. 5 and transformed to external reference system with axis: RD, ND and TD: a) Cu: x 5 – a and b) Cu: x 5 – p

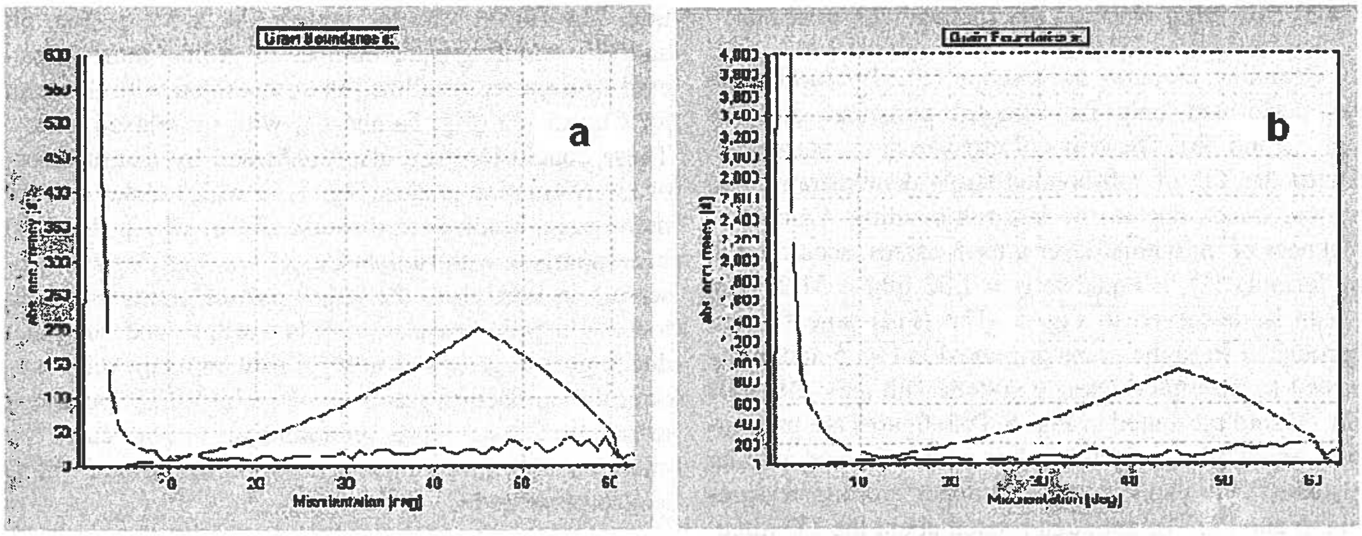


Fig. 7. Misorientation of Cu 99.95% after 5 passes roll-bonding at 250°C: a) Cu: x 5 – a (maximum number of grains with low angle grain boundaries was equal to 3200) and b) Cu: x 5 – p (maximum number of grains with low angle grain boundaries was equal to 13 000)

4.4. Transmission electron microscopy observations

Transmission electron microscopy (TEM) study, similarly as in the case of scanning electron microscopy (SEM) examination, was performed only for samples processed 5 times (Fig. 8a and 8b). Five passes of roll-bonding (ε = 4.0) and heating temperature 250°C of Cu 99.95% strip led to considerable grain refinement. Numerous dislocation cells of 100–200 nm diameter for sample Cu: x5 – a (Fig. 8a) were observed. As was discussed before in sample Cu: x5 – p grains more recovered or even recrystallized (Fig. 5b and 6b) during SEM study were noticed. Greater dislocation cells of 200–300 nm diameter, free of dislocations are documented by TEM photographs in the case of Cu: x5 – p sample, proving suggested before effects of recovery or recrystallization.

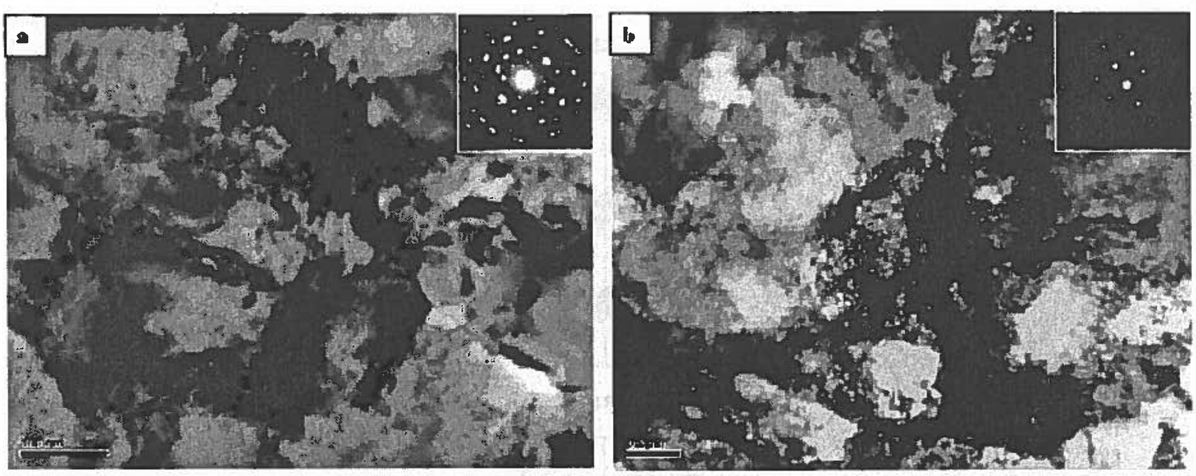


Fig. 8. Microstructure of Cu 99.95% after 5 passes roll-bonding at 250°C: a) Cu: x 5 – a and b) Cu: x 5 – p

5. Concluding remarks

The ARB procedure of Cu 99.95% leads to the considerable grain refinement (grain size d = 100÷300) at deformation ε = 4.0÷5.6 i.e. after 5÷7 passes and pre-heating to 250°C÷350°C before each roll-bonding.

The maximum of strength was noted after 3 passes (ε = 2.4) whereas the maximum of elongation was observed at equivalent deformation ε = 4.0÷5.6 i.e. after 5÷7 passes. The continuation of ARB processing beyond 5÷7 passes does not lead to the increase of mechanical properties.

Grain refinement depends strongly on ARB conditions; small deviation can decrease the mechanical properties inducing the recovery or recrystallization effects.

The number of grains resolved by EBSD technique under SEM examination is predominant in the sample with recovered microstructure.

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