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ON STABILITY OF MICROSTRUCTURE, TEXTURE AND MECHANICAL PROPERTIES OF COPPER DURING ARB PROCESSING

O STABILNOŚCI MIKROSTRUKTURY, TEKSTURY I WŁASNOŚCI MECHANICZNYCH MIEDZI ODKSZTAŁCANEJ METODĄ ARB

The increased interest in materials of the ultra-fine grained (UFG) structure is due to the evident advantages resulting from increased strength properties and greater hardness at higher ductility. The accumulative roll-bonding (ARB) process is a promising mode for introducing severe plastic deformation into industrial practice to produce bulk UFG materials. The ARB process consists in rolling the pack of two sheets up to 50% reduction, which is then sectioned into two halves, stacked and theoretically un-limitedly repeated; total equivalent deformation is equal to $\varepsilon = n^*0.8$ where n is the number of rolling passes.

The orientation distribution of ARB processed Cu 99.95% up to $\varepsilon \sim 8$ is analyzed in the paper. The stability of the mechanical properties and structure have been discussed in relation to changes of crystallographic texture.

Keywords: accumulative roll bonding, ultra-fine grain structure, copper, mechanical properties, microstructure, EBSD

Wzrost zainteresowania materiałami o ultra-drobnym ziarnie (materiał o UDZ) wynika z praktycznych korzyści związanych ze wzrostem wytrzymałości i twardości przy większej ciągliwości tych materiałów. Intensywne odkształcanie sposobem spajania podczas walcowania pakietowego (Accumulative Roll-Bonding ARB) stwarza perspektywiczne możliwości wytwarzania materiałów o UDZ na skalę przemysłową. Technika ARB polega na spajaniu pakietu złożonego z dwóch blach podczas walcowania z 50% redukcją przekroju. Pakiet po takim walcowaniu jest cięty na dwie połowy i po oczyszczeniu składanych powierzchni walcowany ponownie. Teoretycznie, taka procedura może być prowadzona nieograniczenie, przy czym zastępcze odkształcenie po n przepustach wynosi $\varepsilon = n^*0.8$.

W pracy, proces walcowania pakietowego ze spajaniem jest analizowany dla miedzi Cu 99.95% odkształcanej do $\varepsilon \sim 8$. Stabilność mikrostruktury i własności mechanicznych podczas walcowania techniką ARB rozpatrywana jest w odniesieniu do wytworzonej tekstury krystalograficznej.

1. Introduction

Intensive studies on the mechanical properties of grain refined materials (ultra-fine grained (UFG) structure materials) have been carried out in the last years. The increased interest is due to the evident advantages resulting from the development of such a structure in these materials. Increased strength, greater hardness and higher ductility in such materials are reported; also the lower temperature of superplastic flow occurring in these materials permits to reduce the drawing temperature of complicated products [1, 2]. Severe Plastic deformation (SPD) methods, like ECAP method [3, 4] or high pressure torsion (HPT) [1] are frequently applied to prepare UFG materials. The accumulative roll-bonding (ARB) method [5] as a special purpose cladding by rolling is generally applied to perform the manufacturing of ultra-fine grained materials by severe plastic deformation. It seems to be the most efficient and economic procedure for the production of such materials on industrial scale.

In our experiments of UFG cold rolled up to 96% copper sheets the tensile elongation in rolling direction RD was significantly greater than that 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) pro-

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cessing [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 the influence of grain size on copper properties in the range of UFG structure.

The orientation distribution of ARB processed Cu 99.95% up to $\varepsilon \sim 8$ is analyzed in the present paper which follows a recent study of ARB processed copper where some instabilities of structure were encountered [6]. The evolution and stability of the mechanical properties and structure have been discussed in relation to changes in the crystallographic texture.

2. Accumulative roll-bonding

Figure 1 presents the scheme of accumulative roll-bonding (ARB) procedure. It is worth to remember that in ARB mode of deformation two pieces of strip with clean and degreased surfaces are put together and after annealing joined by means of 50% consecutive rolling.

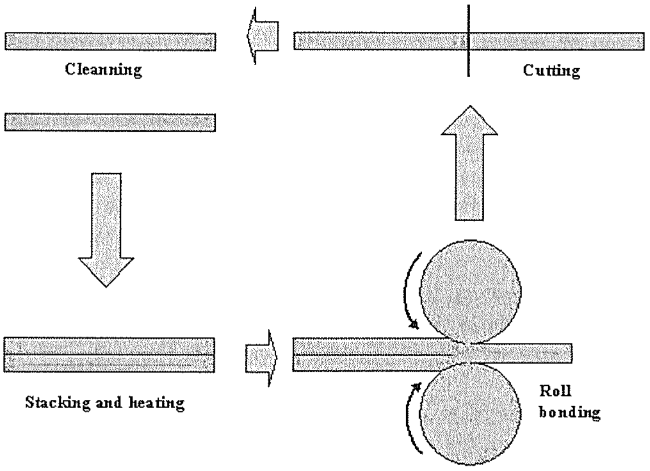

Fig. 1. Scheme of the accumulative roll-bonding (ARB) procedure

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 [5]. The thickness t_n of individual layer, after n cycles, can be calculated according to the formula $t_n = t_o/2^n$, where t_o the is initial thickness. The total reduction z_n after n cycles is

$$z_n = 1 - t_n/t_o = 1 - 1/2^n \qquad (1)$$

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

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

To make the changing of the sheet layer correspond to the initial sheet thickness after n cycles of ARB deformation we can assume for simplicity that the initial thickness is equal to $t_o = 1$ mm and after 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 $\varepsilon_5 = 4$; 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 $\varepsilon_{10} = 8$ can be obtained. Although relatively large deformation can be obtained, in fact the end parts of the strip have to be prepared each time after roll-bonding and thus, with increasing number of cycles, the length of roll-bonded strip decreases.

3. Experimental results

Experiments were carried out on copper Cu 99.95 recrystallized at 773 K, with the initial grain size diameter $d = 30$ μm. The accumulative roll-bonding (ARB) procedure of copper was described in detail in a previous paper where a particular stress was put on certain instabilities during ARB deformation [6]. In order to understand the unpredicted instabilities observed in texture evolution during cycling under ARB processing it is necessary to remember some results as below.

3.1. Tensile tests

The experiments of roll-bonding at ambient temperature usually do not give a satisfactory good final product as it was discussed earlier [7–9]. Then an initial heating of copper strips was executed before the roll-bonding pass. The temperature 250°C ÷ 350°C of initial heating and 5 passes ($\varepsilon = 4.0$) seem to be appropriate for copper as optimal roll-bonding conditions. Similar number of passes was found in the case of aluminium; an increased number of passes resulted in worse tensile properties of the final product. Exemplary tension curves visualizing the changes of strength and elongation, following the increasing number of passes from 1 to 8, are presented in Fig. 2.

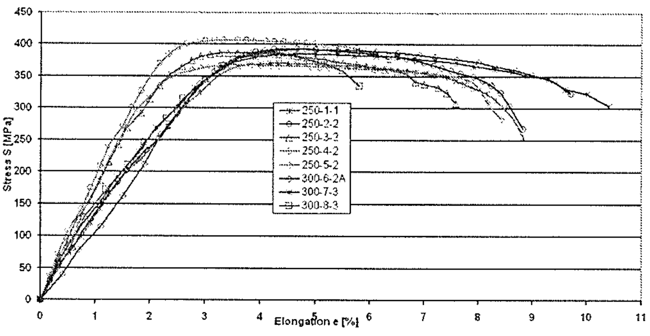

Fig. 2. Tensile stress versus elongation for samples after roll-bonding at 250°C [6]: 1 pass (250-1-1), 2 passes (250-2-2), 3 passes (250-3-2), 4 passes (250-4-2), 5 passes (250-5-2), and 6 passes at 300°C (300-6-2A), 7 passes at 300°C (300-7-3) and 8 passes at 300°C(300-8-3)

3.2. Crystallographic texture measurement

Figure 3 presents pole figures of recrystallized and roll-bonded strips from 1 to 7 cycles measured by the neutron diffraction technique. In recrystallized copper sheet (Fig. 3, pole figure Cu-0) we find that the cubic recrystallization component is accompanied by the rest of the rolling texture component. One can observe the strong {135}<-2-11> component (near the "copper" type {112}<111> rolling texture component) after the first and the fifth passes; in-between the passes an additional shear strain texture component {001}<110> was created. In a previous experiment of ARB processed Cu [6], the shear {001}<110> texture component was absent in five passes processed sample, whereas the strong {135}<-2-11> component was observed. In the present experiment, the shear {001}<110> texture component is strong after 4 and 5 passes which suggests high sensitivity of ARB processed Cu to the experimental conditions during the 5th pass (compare in Table 1 the intensity of texture components derived from ODF, calculated using neutron diffraction measurements).

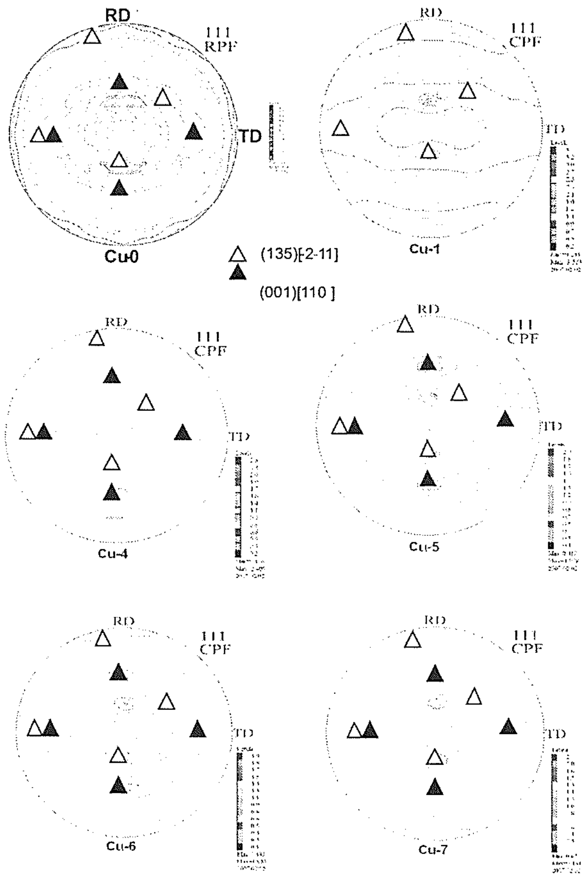

Fig. 3. {111} pole figures measured with the neutron diffraction of ARB processed samples: Cu-0: recrystallized, Cu-1: 1 pass, Cu-4: 4 passes, Cu-5: 5 passes, Cu-6: 6 passes and Cu-7: 7 passes. Orientations {135}<-2-11> and {001}<110> are marked

TABLE 1

Neutron measured crystallographic texture

{111} pole figure of Cu	Equivalent deformation ε	Sample	Ideal orientation (135)[-2-11] ODF f/fmax	Ideal orientation (001)[110] ODF f/fmax
Cu-0	0	Cu0-0	3.5/4.0	2.5/4.0
Cu-1	0.8	Cu250-1	6.0/6.8	1.0/6.8
Cu-4	3.2	Cu250-4	2.0/5.7	5.0/5.7
Cu-5	4.0	Cu250-5	3.0/5.5	5.0/5.5
Cu-6	4.8	Cu300-6	2.0/4.0	3.5/4.0
Cu-7	6.4	Cu300-7	2.0/3.6	3.5/3.6

3.3. Scanning electron microscopy observations

Examination by means of scanning electron microscopy (SEM) equipped with field emission gun (FEG) was performed only for samples processed 4 times (Fig. 4) and 5 times (Fig. 5), where the instability of texture was observed before in the cross-sectional plane [6]. These scans were performed in the rolling plane section, parallel to the roll-bonded strip planes and they demonstrated differences in the grain size approving the differences encountered in the cross-sectional scans. Estimated from SEM with FEG scan grain size is equal to ~ 300 nm in the case of ARB processed Cu 4 times (better resolution for <uvw> scan in Fig. 4). In the case of a sample ARB processed 5 times one can observe also grains with the diameter d ~ 300 nm, but the number of grains with the diameter d ~ 1 μm is considerable.

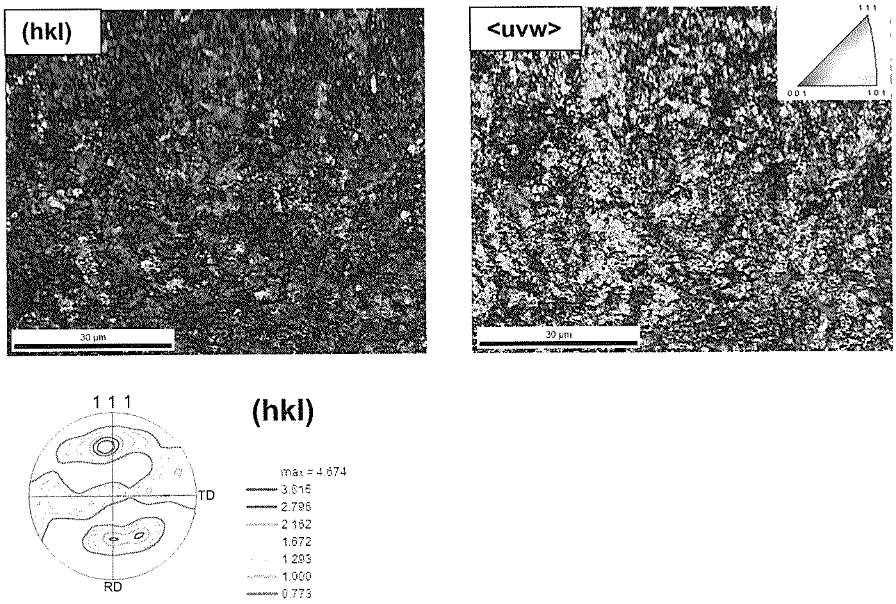

Fig. 4. EBSD scan of SEM with FEG in the rolling plane of sample ARB processed 4 times

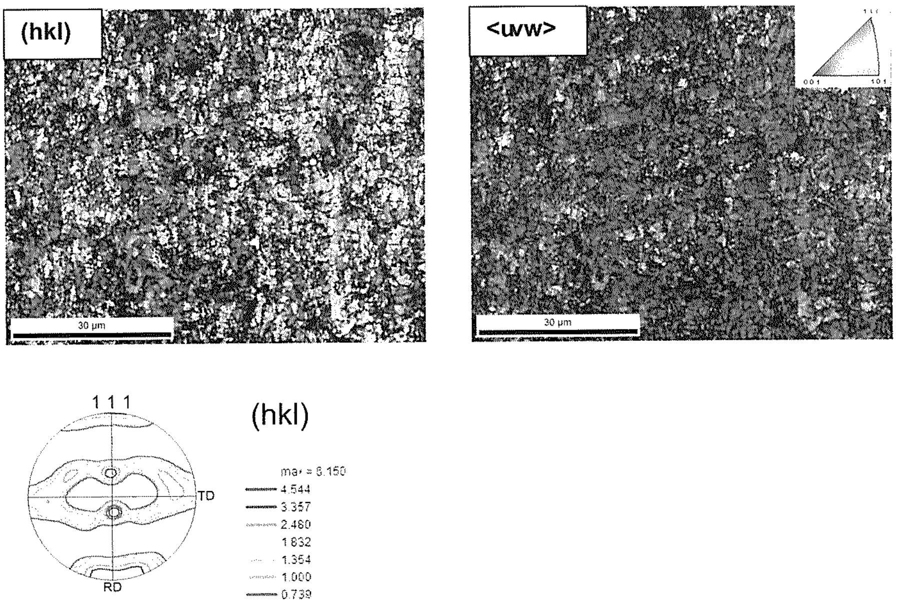

Fig. 5. EBSD scan of SEM with FEG in the rolling plane of sample ARB processed 5 times

4. Discussion

Considering the tensile properties of ARB processed Cu one can conclude finally that: 1) initial heating of copper strips before the roll-bonding pass is necessary and 2) the temperature 250°C ÷ 350°C of initial heating and 5 passes (ε = 4.0) seem to be appropriate for copper as optimal roll-bonding conditions as it was stated before [6]. Similar number of passes was found in the case of aluminium; increased number of passes resulted in worse tensile properties of the final product.

Neutron measured crystallographic textures demonstrate the strong {135}<-2-11> component after the first and the fifth passes; in-between the passes an additional shear strain texture component {001}<110> was created. The drop of tensile strength after the fifth ARB pass observed in Cu after the 5th pass has been confirmed by changes in the deformation textures [6]. It suggests a strong sensitivity of copper to ARB rolling conditions during the fifth pass at pre-heating to 250°C. Other ARB experiments, performed anew with four and five passes, did not reveal a difference in texture (Fig. 3). It was found in a previous experiment that in a sample with lower tensile properties (Cu-5-p [6]) the effects of structure recovery or even early stages of recrystallization were observed in electron microscopy photographs and are also observed in Fig. 5 where more grains seem to be recovered or recrystallized.

5. Concluding remarks

The ARB procedure of Cu 99.95% leads to considerable grain refinement (grain size $d = 100 \div 300$ nm) at deformation $\varepsilon = 4.0 \div 5.6$ 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 ($\varepsilon = 2.4$), whereas the maximum of elongation was observed at equivalent deformation $\varepsilon = 4.0 \div 5.6$ 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 may decrease the mechanical properties inducing the recovery or recrystallization effects.

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

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REFERENCES

[1] R. Z. Valiev, R. K. Ismagaliev, I. V. Alexandrov, Progress in Materials Science **45**, 103-189 (2000).

[2] Y. T. Zhu, T. C. Lowe, T. G. Langdon, Scripta Materialia **50**, 825-830 (2004).

[3] J. Kuśnierz, Archives of Metallurgy **46**, 375-384 (2001).

[4] J. Kuśnierz, J. Bogucka, Archives of Metallurgy **48**, 173-182 (2003).

[5] Y. Saito, H. Utsunomiya, N. Tsuji, T. Sakai, Acta mater. **47**, 579-583 (1999).

[6] J. Kuśnierz, M-H. Mathon, J. Bogucka, M. Faryna, Z. Jasieński, R. Penelle, T. Baudin, Archives of Metallurgy and Materials **51**, 237-243 (2006).

[7] J. Kusnierz, J. Bogucka, Archives of Metallurgy and Materials **50**, 219-230 (2005).

[8] J. Kusnierz, J. Bogucka, Mat. Scie. Forum **495-497**, 797-802 (2005).

[9] J. Kusnierz, M. Kurowski, J. Bogucka, Seminarium poświęcone 70. rocznicy urodzin prof. Z. Jasieńskiego 37-47 (2005).

[10] X. Huang, N. Tsuji, N. Hansen, Y. Minanimo, Mat. Scie. Forum **408-412**, 715-720 (2002).