



Microstructure characterization of deformed copper by XRD line broadening

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ABSTRACT

Purpose: The aim of the present study was to determine the changes in microstructure taking place during deformation with methods like compression, compression with oscillatory torsion as well as compression and subsequent compression with oscillatory torsion (combined method).

Design/methodology/approach: The study was conducted on M1E grade Cu. Several methods were used in this study, such as: transmission electron microscopy and X-ray diffraction. The X-ray diffraction line profile analysis was applied to determine crystallite size and density of dislocations.

Findings: Application of compression and oscillatory torsion without previous compression resulted in decrease of grain size and crystallite size and increase of lattice distortion caused by dislocation compared to the combined method of deformation. In specimen with higher microstrain and smaller grain size a higher fraction of edge dislocations was observed.

Research limitations/implications: Obtained results can be useful to modify the process and design deformation parameters.

Practical implications: The knowledge of the characteristic features of unconventionally deformed materials will provide the usability of the method employed to produce materials with desired functional properties.

Originality/value: Compression with oscillatory compression is a deformation procedure applied to achieve large strains. However there is no studies on evolution of the microstructures during deformation obtained on the way of the methods mentioned above. This paper provides such an information.

Keywords: Nanomaterials; Microstructure; Transmission Electron Microscopy; X-ray diffraction

MATERIALS

1. Introduction

Significant grain refinement may be achieved in bulk polycrystalline metals through the application of severe plastic deformation (SPD) [1-4]. Several SPD processing techniques lead to microstructure refinement with various grain size and various contributions of high-angle boundaries [5-8]. A combined method of equal channel angular pressing (ECAP) + cold rolling and ECAP + cold rolling + high pressure torsion (HPT) for refining the grain size was employed for Ni. This processing allows to obtain equal grains sizes of a 330 nm and 120 nm respectively [9]. In [10] a compression

subsequent the deformation by ECAP induces an increase of the most frequent grain size by about 10% and the size distribution becomes much broader. However, the combined torsion-compression following the ECAP yields a 50 % increase of the most frequent grain size whereas the width of size distributions does not change. In both cases the number of adjacent grains with large misorientations is higher if compared to ECA deformation only. There is discussed in [10] that compression can be used for breaking down an initial coarse structure, refine grain size, and increase the strength of the material. The experimental results presented in [11] demonstrate very clearly the advantage of processing materials by a combination of ECAP followed by HPT.

The aim of the present study was to determine changes in microstructure during the methods of deformation like:

- compression with oscillatory torsion
- compression and subsequent compression with oscillatory torsion (combined method).

For the deformation methods presented, a structural characterization was performed by transmission electron microscope (TEM) and X-ray diffraction. Assuming that a deformation of metals is caused by dislocations the X-ray diffraction profile analysis is a useful method to characterize microstructure of plastically deformed materials.

2. Experimental procedures

M1E grade Cu was used for the experiment. The material was homogenized at 500°C for 2h and then cooled slowly. Sample No. 1 was deformed by using compression with oscillatory torsion methods at: torsion frequency (f) = 0.4 Hz, torsion angle (α) $\approx 6^\circ$ and compression speed (v)=0.1mm/s. The sample No 2 was compressed in room temperature for reach strain $\varepsilon = 0.7$ and the next sample was compressed with oscillatory torsion at: torsion frequency (f) = 1.6 Hz, torsion angle (α) $\approx 6^\circ$ and compression speed (v)=0.1mm/s. A transmission electron microscope (TEM) was used to examine the microstructure. For TEM, longitudinal sections were taken from the 0.8 value of sample radius. Foils were prepared using conventional techniques by electropolishing using: 600 ml CH₃OH, 340 ml C₄H₉OH, 60 ml 60% HClO₄. TEM observations were carried out at 100 kV.

The diffraction profiles were measured by a X-Pert Philips diffractometer equipped with graphite monochromator na diffracted beam and with the following slits (in sequence from Cu tube to counter): Soller (2°), divergence (1/2°), antiscatter (1/2°), Soller (2°) and receiving (0.15 mm). All the diffraction profiles were obtained at step size of 0.02° and were fitted a pseudo-Voigt function. The NIST SRM660a (LaB₆) was used as a line profile standard to determine instrumental broadening. X-ray diffraction results were analyzed according to the model proposed by Ungar et al. [12,13]. Due to the equipment used in this study, the raw data are not of high quality to perform a correct Fourier analysis. Thus the so-called “double-Voigt” method was used to determine the size and the strain Fourier coefficients necessary to perform a Warren-Averbach line profile analysis.

3. Results and discussion

3.1. Electron microscopic results

During deformation at low strain (low value of Hz), fine dislocation cells with a high dislocation density and less sharp grain boundaries were formed by an operation of multi-directional slip (Fig.1). Combined methods (compression followed by compression with oscillatory torsion) yields specimens of uniform structure (Fig.2a), where dislocation cells were developed into ultrafine subgrains, and the microstructural change was dominated by a conversion of low-angled subboundaries to high-angled boundaries, rather than grain refinement. The dislocation density

after processing with combined methods, both at grain boundaries and inside the grains is high enough to lead to dislocation rearrangement into a cell-like structure, which may be responsible for further grain refinement (Fig. 2b). More information about microstructure characterization will be presented elsewhere.

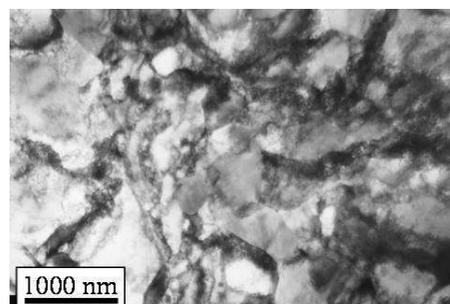


Fig. 1. Microstructure of copper specimen No 1. Transverse section

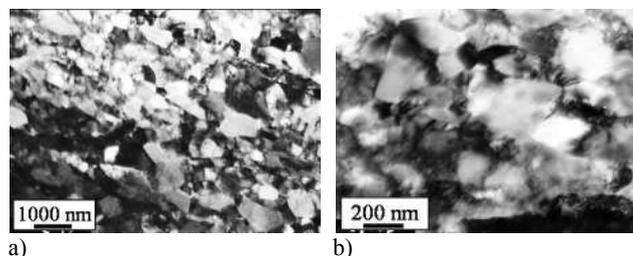


Fig. 2. Microstructure of copper specimen No 2. Transverse section

3.2. X-ray diffraction results

A simple qualitative information regarding the nature of the coherently domain (i.e. morphology, size-strain information) prior to any detailed analysis may be obtained on the basis of Williamson-Hall plot (Fig. 3).

For the copper after compression and oscillatory torsion with frequency $f = 0.4$ Hz slope of approximation line is more positive than for a sample after compression and compression with oscillatory torsion (combined method). This indicates that there are higher values of lattice distortion caused by lattice defects in specimen No. 1, yet the correlation is very low and this method is not effective for determination of lattice distortion and crystallites size. The non-monotonous variations of X-ray line broadening with crystal indices hkl are observed for each sample. Thus, a strong strain anisotropy caused by dislocations can be clearly observed.

Assuming that strain broadening of diffraction lines is due to the creation of dislocations, the results of X-ray diffraction were analyzed according to the model proposed by Ungar et al. [12÷14]. This model is based on the modification of the Williamson-Hall plot:

$$\Delta K = \frac{0.9}{D} + \alpha' (K\bar{C}^{-1/2})^2 + O(K\bar{C}^{-1/2})^4 \quad (1)$$

where D is the apparent size parameter corresponding to the FWHM, α' is the constant depending on the effective outer cut-off radius of dislocations, on the Burgers vector and on the density of dislocations, O stands for higher orders terms in $K\bar{C}$. \bar{C} is the average contrast factor of dislocations and can be calculated using the following formula:

$$\bar{C} = \bar{C}_{h00} (1 - qH^2) \tag{2}$$

where \bar{C}_{h00} is average dislocations contrast factor for the $h00$, and this value as well as the values of q for pure screw and pure edge dislocation can be determined by theoretical calculation [16], $H^2 = (h^2k^2 + h^2l^2 + k^2l^2)/(h^2 + k^2 + l^2)^2$. In the paper [16] the values of q have been calculated for the most common dislocation slip system in copper with the Burgers vector of $b = a/2\langle 110 \rangle$. It was found that the values of q for pure screw or pure edge dislocations in this slip system of copper are 2.37 and 1.68, respectively.

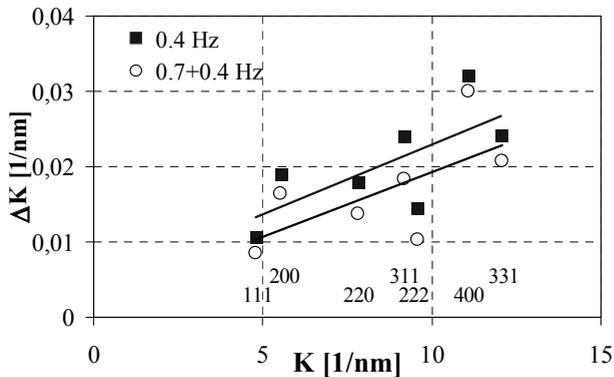


Fig. 3. The classical Williamson-Hall plot of the FWHM for the investigated copper specimens; $\Delta K = \text{FWHM} \cos \theta / \lambda$, $K = 2 \sin \theta / \lambda$

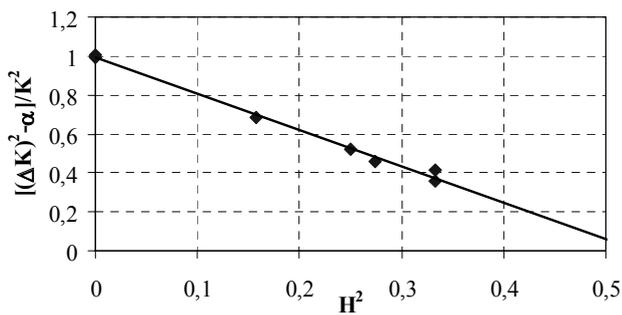


Fig. 4. Determination of the parameter q for the specimen after compression and oscillatory torsion at 0.4Hz

For the specimens analyzed, q can be deduced directly from the line profile analysis of the diffraction pattern. From the linear regression of expression $((\Delta K)^2 - \alpha) / K^2$ versus H^2 the q parameter ($\alpha = 0.9/D$) was determined for each sample as illustrated in Fig. 4. The intercept of H^2 axis gives the value of $1/q$. As shown in

Table 1, values of this parameter vary between 1.98 and 1.87 for the sample after combined method and sample after compression with oscillatory torsion, respectively. In the first case the value of q is close to value of 2.02 and this indicates that the character of the prevailing dislocations is almost half edge, half screw. In the second case edge dislocations are dominant (~70% of total dislocation population).

The modified Williamson-Hall plot of the FWHM for sample after compression and oscillatory torsion is shown in Fig. 5. It can be seen that the values of FWHM (ΔK) follow a smooth curve. This curve is parabolic in nature, thus broadening in analyzed specimens is mainly due to dislocation induced anisotropic strain broadening. From the intercept of ΔK axis the apparent size (D) corresponding to the volume weighted mean columns-lengths of the crystallites is evaluated (Tab. 1). The intersection of the best linear regression at $K=0$ gives apparent size of $D = 143$ nm and $D = 256$ nm for sample No. 1 and sample No. 2, respectively. From Table 1 it is clear that the apparent size determined from modified Williamson-Hall procedure for both samples is smaller than the apparent size obtained from classical method. The dislocation density can not be determined from modified W-H plot as the constant in the coefficient of $KC^{1/2}$, because it is related to the effective outer cut-off radius for dislocations which depends on the tail of the profile.

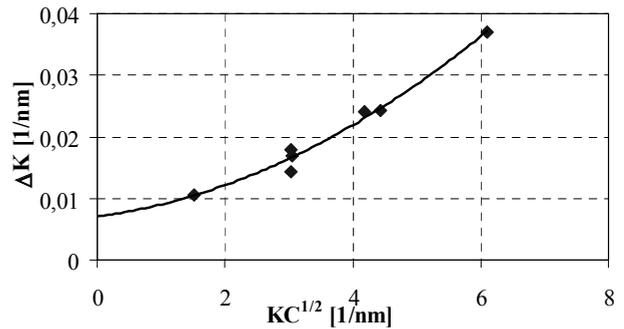


Fig. 5. The modified Williamson-Hall plot of the FWHM for Cu specimen after compression and oscillatory torsion at 0.4Hz

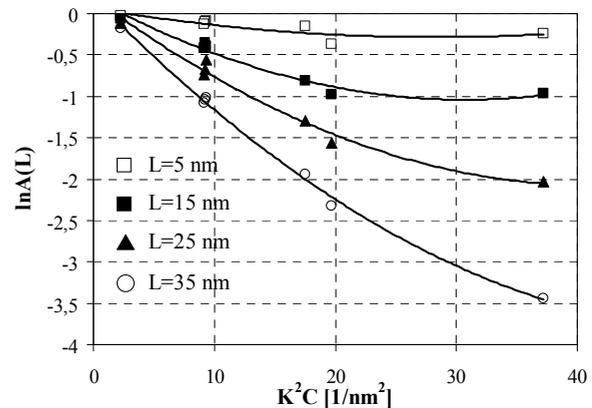


Fig. 6. The modified Warren-Averbach plot of the FWHM for Cu specimen after compression and oscillatory torsion at 0.4Hz

Table 1.

Microstructural parameters calculated from classical and modified Williamson-Hall method and modified Warren-Averbach method

Specimen	q	ρ [10^{15} m^{-2}]	D, cW-H [nm]	D, mW-H [nm]
No. 1	1.87	1.63	227	143
No. 2	1.98	1.02	476	256

In order to quantify the dislocations density the modified Warren-Averbach method was used [12÷14]:

$$\ln A(L) = \ln A^S(L) - \rho B L^2 \ln \left(\frac{R_e}{L} \right) (K^2 \bar{C}) + O(K^4 \bar{C}^2) \quad (3)$$

where ρ is the density of dislocations, $B = \pi b^2/2$ with b the Burgers vector of dislocations, L the Fourier length defined as: $L = n a_3$, where $a_3 = \lambda/2(\sin\theta_2 - \sin\theta_1)$, n are integers starting from zero and $(\theta_2 - \theta_1)$ is the angular range of the measured diffraction profile. R_e is the effective outer cut-off radius of dislocations, $A(L)$ the real part of the Fourier coefficients, A^S is the size Fourier coefficients. Due to the equipment used in this study, the data collected are not of a quality high enough to perform a correct Fourier analysis. Therefore all the peaks have been fitted by Voigt functions and the properties of these functions have been used to determine the Fourier coefficients [15]. The modified Warren-Averbach plot is shown in Fig. 6. The values of ρ (Tab. 1) are obtained by plotting $X(L)/L^2$ versus $\ln(L)$ and by using linear regression for the small values of L . The term $X(L) = \rho B L^2 \ln(R_e/L)$ is determined from Equation (19) for each L value.

4. Summary

Anisotropic line broadening of copper specimen after compression with oscillatory torsion is explained in terms of dislocation induced strain broadening. The dislocation density is of the order of 10^{15} m^{-2} . The application of compression and oscillatory torsion without previous compression resulted in decrease of crystallite size and increase of lattice distortion caused by dislocation if compared to combined method of deformation. Moreover, in specimen with smaller crystallite size higher fraction of edge dislocations was observed. The classical Williamson-Hall plot allows to determine a correct crystallite size, but is valid only if the data follows a straight line. In case of deformed copper a strong size anisotropy is present and the modified Williamson-Hall method is more useful to estimate the apparent size corresponding to the volume weighted mean columns-lengths of the crystallites. More detailed analysis of the results obtained with consideration of residual parameters (strain, R_e , μ etc), which can be determined from X-ray profile analysis, will be published later.

Acknowledgements

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References

- [1] M. Greger, R. Kocich, L. Cizek, L.A. Dobrzański, M. Widomska, B. Burek, A. Silbernagel, The structure and properties of chosen metals after ECAP, *Journal of Achievements in Materials and Manufacturing Engineering* 18 (2006) 103-106.
- [2] Z. Gronostajski, K. Jaśkiewicz, Influence of monotonic and cyclic deformation sequence on behavior of CuSi3.5 silicon bronze, *Proceedings of the 11th Scientific International Conference „Achievements in Mechanical and Materials Engineering” AMME’2002, Gliwice-Zakopane, 2002*, 219-222.
- [3] N. Hansen, X. Huang, R. Ueji, N. Tsuji, Structure and strength after large strain deformation, *Materials Science and Engineering A387-389* (2004) 191-194.
- [4] G. Niewielski, D. Kuc, K. Rodak, F. Grosman, J. Pawlicki, Influence of strain on the copper structure under controlled deformation path conditions, *Journal of Achievements in Materials and Manufacturing Engineering* 17 (2006) 109-112.
- [5] K. Rodak, Ultrafine grained Cu processed by compression with oscillatory torsion, *Journal of Achievements in Materials and Manufacturing Engineering* 20 (2007) 491-494.
- [6] K. Rodak, Microstructure of severely deformed Cu by using oscillatory compression method, *Journal of Achievements in Materials and Manufacturing Engineering* 20 (2007) 179-182.
- [7] R.Z. Valiev, R.K. Islamgaliev, I.V. Alexandrov, Bulk nanostructured materials from severe plastic deformation, *Progress in Materials Science* 45 (2000) 103-189.
- [8] G. Sakai, Z. Horita, T.G. Langdon, Grain refinement and superplasticity in aluminum alloy processed by high-pressure torsion, *Materials Science and Engineering A393* (2005) 344-351.
- [9] N. Krasilnikov, W. Lojkowski, Z. Pakiel, R. Valiev, tensile strength and ductility of ultra-fine-grained Nickel processed by severe plastic deformation, *Materials Science and Engineering A* 397 (2005) 330-337.
- [10] B. Mingler, H. P. Karnthaler, M. Zehetbauer, R. Z. Valiev, TEM investigation of multidirectionally deformed copper *Materials Science and Engineering A* 319-321 (2001) 242-245.
- [11] A. P. Zhilyaev, B.K. Kim, J.A. Szpunar, M.D. Baro, T.G. Langdon, The microstructural characteristics of ultrafine-grained nickel, *Materials Science and Engineering A* 391 (2005) 377-389.
- [12] T. Ungar, I. Dragomir, A. Revesz, A. Borbely, The contrast factor of dislocations in cubic crystals: the dislocation model of strain anisotropy in practice, *Journal Applied Crystallography* 32 (1999) 992-1002.
- [13] J. Gubicza, G. Ribarik, G.R. Goren-Muginstein, A.R. Rosen, T. Ungar, The density and the character of dislocation in cubic and hexagonal polycrystals determined by X-ray diffraction, *Materials Science and Engineering A309-310* (2001) 60-63.
- [14] T. Ungar, J. Gubicza, G. Ribarik, A. Borbely, Crystallite size distribution and dislocation structure determined by diffraction profile analysis: principles and practical application to cubic and hexagonal crystals, *Journal Applied Crystallography* 34 (2001) 298-310.
- [15] D. Balzar, in R.L. Snyder, H.J. Bunge, J. Fiala (Eds.), *Defect and Microstructure Analysis from Diffraction*, International Union of Crystallography, vol. 10, Oxford University Press, New York, 1999.