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
Volume 50 2005 Issue 1

K. WIERZBANOWSKI*, A. BACZMAŃSKI*, R. WAWSZCZAK*, J. TARASIUK*, P.H. GERBER**, B. BACROIX***, A. LODINI****

INTERNAL STRESS AND STORED ENERGY IN RECRYSTALLIZED COPPER

NAPRĘŻENIA WŁASNE I ENERGIA ZGROMADZONA W PROCESIE REKRYSTALIZACJI MIEDZI

The multi-reflection X-ray method was used to determine the stress level in deformed and recrystallized polycrystalline copper samples. The anisotropic diffraction elastic constants were calculated using the self- consistent model and crystallographic texture. A significant decrease of the first order residual stresses was observed during recrystallization, but they start to decrease already during recovery. Diffraction peak widths and intensities were also examined. A complementary study, using synchrotron radiation and electron back scattering diffraction techniques, was done in order to determine the stored energy in the examined material. This latter is strongly orientation dependent and has the lowest value in the cubic orientation, which is dominating one in recrystalization texture.
Keywords: residual stress, multireflection method, recrystallization, crystallographic texture, peak width, stored energy, modelling

Użyto rentgenowskiej metody wielo-odbiciowej w celu wyznaczenia naprężeń wewnętrznych w odkształconej i wyżarzonej rekrystalizująco polikrystalicznej miedzi. Anizotropowe dyfrakcyjne stałe sprężyste wyliczono stosując model samo-uzgodniony oraz funkcję tekstury. Zaobserwowano wyraźne zmniejszenie naprężeń wewnętrznych podczas rekrystalizacji, chociaż efekt ten rozpoczyna się już podczas zdrowienia. Zbadano również szerokość oraz intensywność pików dyfrakcyjnych. Przeprowadzono dodatkowe badania przy użyciu dyfrakcji promieniowania synchrotronowego oraz dyfrakcji elektronów rozproszonych wstecznie w celu wyznaczenia energii zgromadzonej w odkształconym materiale. Zależy ona silnie od orientacji krystalograficznej i ma najmniejszą wartość dla orientacji sześciennej, która jest dominującą składową tekstury rekrystalizacji.

* WYDZIAŁ FIZYKI I INFORMATYKI STOSOWANEJ, AKADEMIA GÓRNICZO-HUTNICZA, 30-059 KRAKÓW, AL. MICKIEWICZA 30
** LLB CEA SACLAY, BAT.563, 91 191 GIF-SUR-YVETTE CEDEX, FRANCE
*** LPMTM-CNRS, UNIVERSITE PARIS XIII, 99, AV. J.B.CLEMENT, 93 430 VILLETANEUSE, FRANCE
**** LACM, UNIVERSITE DE REIMS CHAMPAGNE ARDENNE, 9, BD. DE LA PAIX,51100 REIMS, FRANCE

1. Introduction

The residual stress and stored energy distributions characterize the recrystallization process. Diffraction techniques offer appropriate tools for the study of these quantities. Residual stress in rolled and recrystallized polycrystalline copper samples was studied using X-ray multi- reflection method. The advantage of this method is that various reflections *hkl* are simultaneously used in the fitting procedure, consequently the results are statistically more representative in comparison with the single reflection method. Independently of residual stress, the dislocations are accumulated in the sample during plastic deformation; it represents the stored energy (SE). The dislocation density can be examined using X-ray or synchrotron radiation diffraction (SRD) via peak profile study as well as using electron back scattered diffraction (EBSD). Residual stress and SE are strongly orientation dependent; consequently the crystallographic texture can exhibit some correlations with the above quantities. On the other hand, one expects that deformation models are able to predict most of the above characteristics.

2. Residual stress determination

The standard X-ray $sin^2\psi$ diffraction method of stress analysis is based on the measurement of peak positions for a given *hkl* reflection and for various directions of the scattering vector (defined by ϕ and ψ angles) — Fig. 1. Interplanar spacings are

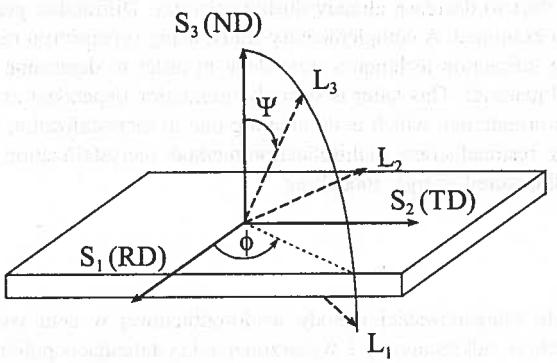

Fig. 1. Two coordinates systems used in the residual stress measurement by diffraction method: the sample coordinates system (**S**) is defined by rolling direction ($\mathbf{S_1}$ = **RD**), transverse direction ($\mathbf{S_2}$ = **TD**) and normal direction ($\mathbf{S_3}$ = **ND**), while the laboratory coordinates system (**L**) is defined by scattering vector **Q**($\mathbf{L_3}$ = **Q**), $\mathbf{L_2}$ axis lays in S_1S_2 plane and $\mathbf{L_3}$ is perpendicular to $\mathbf{L_1}$ and $\mathbf{L_2}$

calculated using Bragg's law and the macro-stress (the first order one) is determined applying a linear or elliptical regression procedure [1-3]. In *multi- reflection* method a few *hkl* reflections are measured and used in the fitting procedure [4-6]. This method, generalized for *anisotropic materials*, was used in the present work. The average strain determined by the diffraction method (i.e., measured along the scattering vector) is:

$$< \varepsilon'(\psi,\phi) >_{\{hkl\}} = F_{ij}(hkl,\psi,\phi,f(g))\,\sigma^{I}_{ij} \qquad (1)$$

where σij^{I} is the first order residual stress (expressed in the sample reference system — Fig. 1) and F_{ij} are diffraction elastic constants (DECs); repeated index summation convention is assumed in this work. The $< \ldots >$ symbol means the average over crystallites taking part in diffraction (i.e., for the grains with $\{hkl\}$ planes perpendicular to the scattering vector). DECs (F_{ij}) depend in general on hkl reflection, orientation of the scattering vector — ψ, ϕ and on texture function $f(g)$. Taking into account that:

$$< \varepsilon'(\psi,\phi) >_{\{hkl\}} = \frac{< d(\psi,\phi) >_{\{hkl\}} - d^{o}_{\{hkl\}}}{d^{o}_{\{hkl\}}} = \frac{< a(\psi,\phi) >_{\{hkl\}} - a^{o}}{a^{o}},$$

Eq. 1 can be rewritten as:

$$< a(\psi,\phi) >_{\{hkl\}} = F_{ij}(hkl,\psi,\phi,f(g))\,\sigma^{I}_{ij}\,a^{o} + a^{o} \qquad (2)$$

where $<\mathrm{d}(\phi,\psi)>_{\{\mathrm{hkl}\}}$ is the inter-planar distance and is $<\mathrm{a}(\phi,\psi)>_{\{\mathrm{hkl}\}}$ is the *equivalent* inter-planar distance. For the cubic structure:

$$< a(\psi,\phi) >_{\{hkl\}} =< d(\psi,\phi) >_{\{khl\}} \sqrt{h^2 + k^2 + l^2}$$

and

$$< a^{o}(\psi,\phi) >_{\{hkl\}} =< d^{o}(\psi,\phi) >_{\{khl\}} \sqrt{h^2 + k^2 + l^2}.$$

In the above equations $d_o(ora^{\circ})$ is a lattice parameter of a reference stress free sample. Having previously calculated $\mathrm{F_{ij}}(hkl,\psi,\varphi,\ \mathrm{f(g)})$, the values of a^o and the macro-stress σ^{I}_{ij} can be found, using the least squares procedure. Nonlinear DECs [7-9] were calculated using the self-consistent model and taking crystallographic texture into account [10, 11]. Cobalt radiation was used and sample surfaces were prepared by chemical etching before measurement. The polycrystalline copper sheets were of 3.8 mm thick and the surface layer about 10 μm was removed by etching prior to X-ray measurements. Rolled polycrystalline samples were examined after cold rolling (70% reduction) and after annealing during 15 min. at different temperatures from the range: 100°C — 250°C. The equivalent lattice parameters $<a>_{200}$, $<a>_{220}$ and $<a>_{311}$ vs $\sin^2\psi$ were determined for $\varphi = 0°, 45°$ and 90°. Based on the complete diffraction data set, the residual stress components σ^{I}_{11}, σ^{I}_{12} and σ^{I}_{22} were evaluated. Their variation as a function of annealing temperature is shown in Fig.2. A considerable decrease of main residual stress components (i.e., σ^{I}_{11} and σ^{I}_{22}) takes place in the temperature range between 150°C and 230°C; at the latter temperature the whole sample is completely recrystallized. It should be underlined that the decrease of main components (σ^{I}_{11} and σ^{I}_{22}) starts already before the beginning of recrystallization, especially σ^{I}_{11} component.

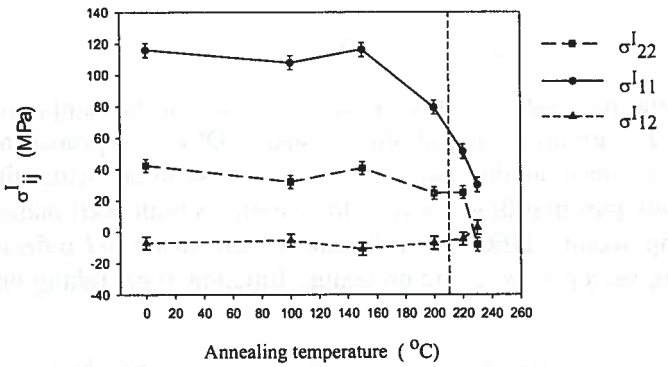

Fig. 2. First order residual stress components σ^I_{11}, σ^I_{12} and σ^I_{22} vs annealing temperature for rolled polycrystalline copper (vertical line shows the beginning of recrystallization)

The final decrease of residual stresses occurs during the recrystallization (temperatures above 210°C).

It is well known that a strong cubic texture component ($\varphi_1 = 0°, \phi = 0°, \varphi_2 = 0°$; see e.g., [12]) forms during recrystallization of the rolled polycrystalline copper (Fig. 3). In general, some amount of cubic orientation is already present after deformation and

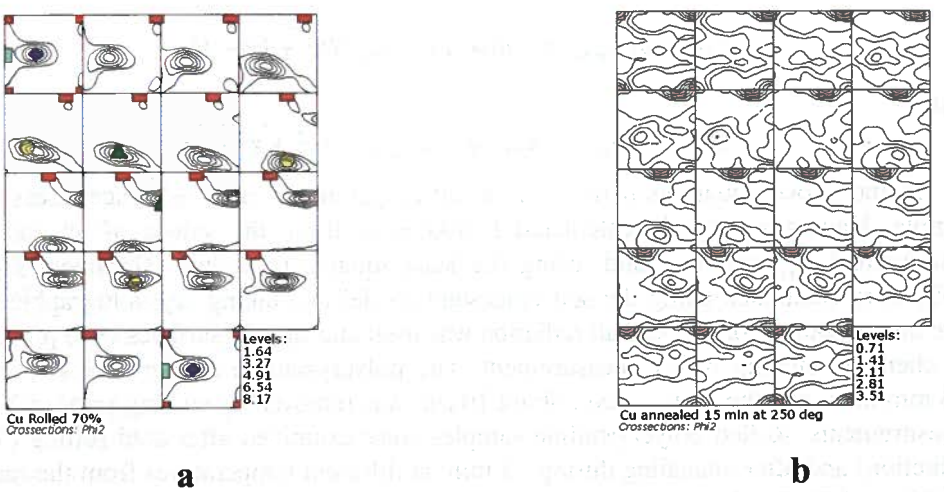

Fig. 3. Texture change in polycrystalline copper (X-rays). Orientation distribution functions are shown for: a. 70% cold rolled texture and b. recrystallized texture (15 min annealing at 250°C)
Texture components are also marked: ■ — cubic, ▲ — copper, ○ — S, □ — Goss, ♦ — brass

intensity of this orientation strongly increases during recrystallization. The intensities of 200 diffraction peaks for the cubic, copper ($\varphi_1 = 90°, \phi = 35°, \varphi_2 = 45°$) and brass ($\varphi_1 = 35°, \phi = 45°, \varphi_2 = 0°$) orientations are plotted versus annealing temperature

in Fig. 4a. It is visible that after the recrystallization process the cubic component dominates the final texture.

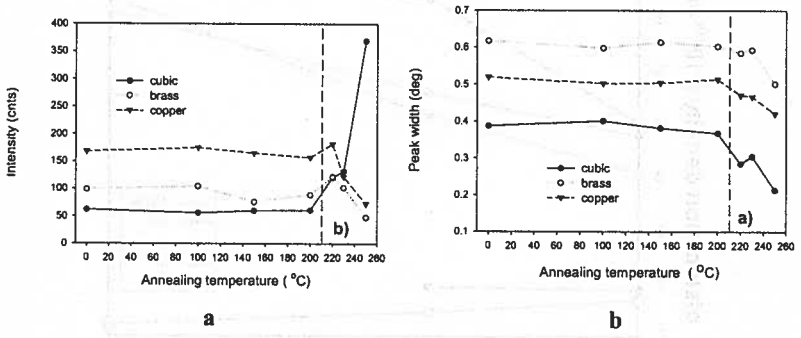

Fig. 4. (200) peak behaviour for three main texture components (cubic, copper and brass) vs annealing temperature: a. peak intensities, b. peak widths (FWHM)

The peak width was also studied (Fig. 4b). The as-rolled material has elongated grains (with the length of the order of 100 μm). It is known that the peak width depends on the square root of the dislocation density, ρ (i.e.: FWHM = a + b $\sqrt{\rho}$, where FWHM means: full width at half maximum and a are b constants). According to Fig. 4b, the peak widths (and consequently dislocation densities) of main texture components do not change much during recovery. A sharp decrease of the peak width during primary recrystallization (above temperature of 210°) is observed for three examined peaks. This is mostly caused by the reduction of dislocation density. The dislocation density (i.e., SE) is a crucial parameter for the recrystallization process. It was examined by two complementary techniques: SRD and EBSD.

3. Stored energy evaluation

Generally, SE is associated with the dislocation density. It can be evaluated from the peak width analysis, using the modified version of Warren-Averbach method [13, 14]. SRD measurements were performed for cold rolled samples with reductions of 70%, 90% and 98%. Estimated dislocation density vs equivalent strain is shown in Fig.5 [14]. It has clearly different levels in examined texture components. The lowest dislocation density was found for the cubic orientation grains. This result was confirmed by EBSD technique. This technique enables direct determination of crystal orientations and it furnishes also a very important parameter - image quality factor (IQF), which can be correlated with dislocation density. Grains with high IQF have low SE, and inversely. The orientation distribution of IQF is shown in Fig. 6. It is clearly seen that IQF has the highest value and consequently the lowest SE in the cubic orientation.

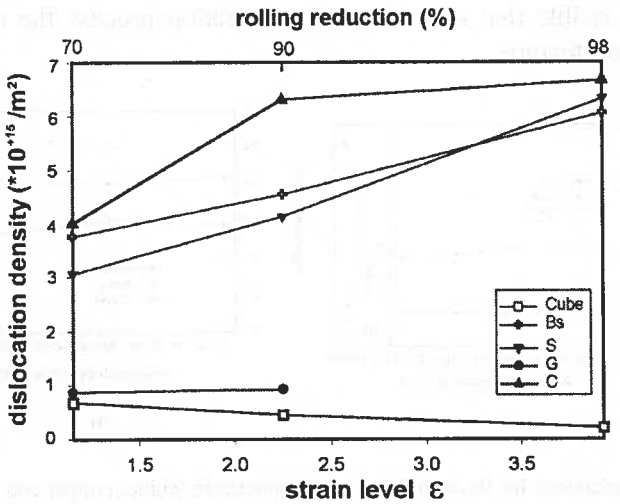

Fig. 5. Dislocation density vs deformation in polycrystalline copper for cubic (Cube), brass (Bs), S, Goss (G) and copper (C) orientations. (Warren — Averbach method, synchrotron radiation diffraction)[14]

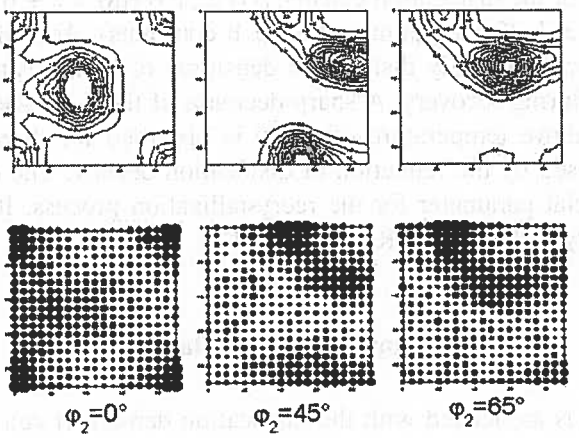

Fig. 6. Crystallographic texture and image quality factor (IQF) distribution vs crystal orientation in rolled polycrystalline copper (70% reduction) determined by EBSD technique (high IQF corresponds to low stored energy). The $\varphi_2 = 0°$, $\varphi_2 = 45°$ and $\varphi_2 = 65°$ section are shown

The SE was also calculated using the elasto-plastic deformation model (Leffers-Wierzbanowski model [15]). The dislocation density $\rho \sim <\tau^2>$ (τ is critical shear stress for slip) is shown in Fig. 7a. Independently, also the elastic energy connected with the second order residual stresses was calculated — Fig. 7b. Both predicted energies have low values in the cubic orientation.

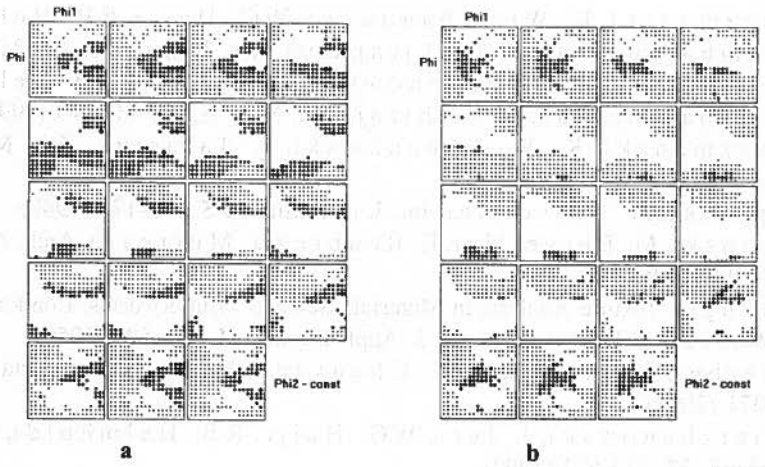

Fig. 7. Predicted distributions (using Leffers-Wierzbanowski model) of: (a) dislocation density, (b) elastic energy connected with the second order residual stresses

4. Conclusions

Residual stress state and stored energy in rolled polycrystalline copper were examined during consecutive annealing processes. A multi-reflection X-ray method was successfully used for measurement of the residual stresses in deformed and annealed polycrystalline copper. The main stress components are strongly reduced in the annealing temperature range 150°C – 230°C. They start to decrease already before the recrystallization process. Our study confirms exceptional role of the cubic texture component. Grains of this orientation have the lowest stored energy and this component dominates the final recrystallization texture.

Acknowledgements

The present work was supported by the Polish Commitee of Scientific Research (KBN).

REFERENCES

[1] I.C. Noyan, J.B. Cohen, Residual Streess, Springer-Verlag, New York (1987).
[2] H. Dölle, J. Appl. Cryst. **12**, 489-501 (1979).
[3] C.M. Brakma, Cryst. Res. Technol. **20**, 593-618 (1985).
[4] J.-D. Kamminga, Th.H. de Keijser, E.J. Mittemeijer, R. Delhez, J. Appl. Cryst. **33**, 1059-1066 (2000).
[5] A. Baczmański, C. Braham , W. Seiler, Phil. Mag. A. **83**, 3225-3246 (2003).
[6] S.J. Skrzypek, A. Baczmański, W. Ratuszek, E. Kusior, J. Appl. Cryst. **34**, 427-435 (2001).

[7] A. Baczmański, K. Wierzbanowski, W.G. Haije, R.B. Helmholdt, G. Ekambaranathan, B. Pathiraj, Cryst. Res. Technol. **28**, 229-243 (1993).
[8] A. Baczmański, K. Wierzbanowski, P. Lipinski, R.B. Helmholdt, G. Ekambaranathan, B. Pathiraj, Phil. Mag. A, **69**, 437-449 (1994).
[9] A. Baczmański, K. Wierzbanowski, J. Tarasiuk, Zeit. Metallk. **86**, 507-511 (1995).
[10] P. Lipinski, M. Berveiller, Int. J. of Plasticity, **5**, 149-172 (1989).
[11] P. Lipinski, M. Berveiller, E. Reubrez, J. Morreale, Arch. Appl. Mech. **65**, 291-311 (1995).
[12] H.J. Bunge, 'Texture Analysis in Materials Science': Butterworths, London (1982).
[13] B.E. Warren, B.L. Averbach, J. Appl. Physics **21**, 595-599 (1950).
[14] Ph. Gerber, J. Tarasiuk, Th. Chauveau, B. Bacroix, Acta Mater. **51**, 6359-6371 (2003).
[15] K. Wierzbanowski, J. Jura, W.G. Haije, R.B. Helmholdt, Cryst. Res. and Technol. **27**, 513-522 (1992).

Received: 24 January 2005.