DISLOCATION DENSITIES AND CHARACTER EVOLUTION IN COPPER DEFORMED BY ROLLING UNDER LIQUID NITROGEN FROM X-RAY PEAK PROFILE ANALYSIS

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ABSTRACT

The microstructure evolution in pure copper deformed by rolling at liquid nitrogen temperature was determined by using X-ray diffraction peak profile analysis. The crystallite size distribution and defects evolution were determined as a function of different reduction levels (e.g. 67%, 74%, 87%, and 97%). By using the Multiple Whole-Profile (MWP) fitting procedure the Fourier transforms of the experimental X-ray peak profiles were fitted all at once by theoretical calculated functions. Here it is assumed that the crystallites are spherical shape and have a log-normal size distribution. It is also supposed that the strain broadening of the profiles is caused by <110>{111}-type dislocations. The results show that the median and the variance of the crystallite size distribution decreases as the deformation reduction increases. The dislocation density has a minimum value at 74% reduction. The increase of the dislocation density at higher deformation levels is due to the nucleation of new generation of dislocations from the crystallite grain boundaries. It was found that the edge dislocation type is predominating the dislocation network formed during the deformation process.

INTRODUCTION

In recent years, research has shown that nano-structured metals and alloys have superior properties compared with their coarse-grained counterparts, such as high strength, superplasticity, less strain-hardening effect [1-3]. Those outstanding properties are believed to be owed by the reduction of crystallite size to the nano-domains [4, 5]. The deformation mechanisms and microstructure development in nano-crystalline materials is still not well understood. The microstructural parameters are extremely important for the fundamental understanding of the microstructure-properties relationship of the nano-structured materials.

The microstructural details can be obtained by using direct methods, such as transmission electron microscopy (TEM) techniques or by indirect methods, such as diffraction of X-ray (XRD) or neutrons techniques. TEM methods reveal microstructural information over very small areas of a sample, which in turn raise issues of how representative are the TEM microstructure parameters to the microstructure of the entire sample. On the other hand, XRD and neutron diffraction methods sample a large volume of the sample, which in turn provides an average representation of the studied material’s microstructure [6].

It has been shown in numerous works that X-ray diffraction peak profile analysis is a very valuable technique for the characterization of microstructures of crystalline materials in terms of crystallite size-distribution and dislocation structures [7]. In metals and alloys the effect of X-ray
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peak broadening caused by non-uniform strains can be parameterized in terms of dislocations densities and types by using the dislocation model of anisotropic broadening of X-ray diffraction profiles. In the present study pure copper specimens were deformed at different reduction rates at liquid nitrogen temperature. This deformation process led to the reduction of the crystallite size from few microns to 20 nm. XRD line profile analysis technique was used to determine the crystallite size distribution, dislocation densities, and dislocation type evolution as a function of deformation. The results presented here are in good correlation with the molecular dynamic simulations in [8], which have shown that inter-granular slip, involving edge dislocation emission and absorption at crystallites boundaries dominate the deformation at crystallite sizes of few tens of nm, while grain boundaries movement play an important roll in the deformation mechanism when the crystallite size is smaller than 10 nm.

EXPERIMENTAL PROCEDURES

Copper specimens were rolled at liquid nitrogen temperature to a reduction of 67%, 74%, 87%, and 97%. In order to obtain a homogeneous deformation a step size of 15% of reduction was applied. After each pass of 15% the sample was immediately returned in liquid nitrogen and again deformed to an additional reduction of 15%. The procedure was repeated until the desired reduction level was achieved. After the rolling deformation for each sample, a multi-step polishing procedure involving 240 grit, 320 grit, 400 grit, 600 grit, and 1000 grit was employed to minimize mechanical damage, and the samples were ultrasonically cleaned between each polishing step to minimize the retention of polishing media. Finally the samples were polished using the Struers ultra precision polishing machine involving the following steps: 9 micron at 30 Newton’s for 3 minutes, 3 micron at 25 Newton’s for 3 minutes, 1 micron at 30 Newton’s for 3 minutes and colloidal silica at 25 Newton’s for 4 minutes.

The diffraction profiles for the Peak Profile Analysis were measured using an Alpha-1 PANalytical Diffractometer set up in Bragg-Brentano geometry. With the use of a symmetrical incident beam Johansson monochromator, only the $K_{\alpha 1}$ component of copper radiation was used. The profile data acquisition was done using a solid-state position-sensitive ultra-fast linear detector (X'Celerator, PANalytical). For each sample the following reflections were measured: 111, 200, 220, 311, 222, 400 and 331. The instrumental broadening was measured using a NIST RSM (660a). As the measured profile is a convolution of the physical with the instrumental profile the Stokes-correction [9] based on the Fourier transforms of the profiles was used to determine the physical line profiles. Background and instrumental profile corrections were done using the MKDAT program described elsewhere [10].

EVALUATION OF THE X-RAY PEAK PROFILES

The X-ray Peak Profile Analysis was carried out using the Multiple Whole-Profile (MWP) fitting procedure. In this method the Fourier coefficients of the measured physical profiles are fitted all at once by the product of the theoretical functions for size ($A^5$) and strain caused by distortion in the crystal ($A^B$) broadening [10, 11]. In this evaluation process it is assumed that the peak broadening is caused by the crystallites size and by strain due to the dislocations. In the case of cubic crystals the MWP fitting procedure enables the evaluation of five microstructure parameters: (a) the dislocation density and arrangement parameter, $\rho$ and $M$. $M$ is defined by
Wilkens as the dislocation arrangement parameter in the Wilkens function [12, 13], the value of $M$ gives the strength of the dipole character of dislocations: the higher the value of $M$, the weaker the dipole character and the screening of the displacement fields of dislocations; (b) the median and the variance, $m$ and $\sigma$, of the size distribution; (c) $q$ the parameters of the dislocation contrast factors. Details of the MWP method can be found in Reference [10]. In the present work the crystallites are assumed to be spherical and have a lognormal size distribution. From $m$ and $\sigma$ the crystallite size distributions were calculated as shown in [10].

Due to the rolling deformation process the copper samples studied here show a strong texture. The presence of crystallographic texture complicates the evaluation of peak broadening observations [11]. In an attempt to emulate a random polycrystalline specimen for each sample the strongest diffraction peaks from the three different faces of the orthogonal sample were mixed to form a full diffraction pattern and used in the MWP evaluation.

THE DISLOCATION CHARACTER IN CUBIC CRYSTALS FROM X-RAY PEAK PROFILE ANALYSIS

It has been shown in References [15-17] that in the cubic crystal systems two parameters are sufficient to fully characterize the average contrast factors of dislocations in the selected slip system: $C_{h00}$ is the average contrast factor corresponding to the $h00$ reflections and $q$ is a parameter depending on the dislocation type and the elastic constants of the material.

$$C_{hkl} = C_{h00}(1-qH^2),$$

where $H^2=(h^2l^2+k^2l^2+h^2k^2)/(h^2+k^2+l^2)^2$ and $h, k, l$ are the Miller indices. The values of $q$ parameters contain the dependence of average contrast factor on dislocation types and geometry. Due to this by comparing the numerically evaluated $q$ factors with the ones obtained experimentally the dominant dislocation character can be determined. The $1/2 \langle 110 \rangle \{111\}$ is the most common slip system observed in the face-centred cubic materials [18, 19]. By assuming that the $1/2 \langle 110 \rangle \{111\}$ is the only dislocation slip system type activated in the deformed crystal, the measured average contrast factor of dislocation can be written in the terms of theoretical average contrast factors for pure edge and pure screw dislocations as:

$$C^m = f \bar{C}^e + (1-f) \bar{C}^s,$$

where $f$ is the fraction of edge dislocations and, $\bar{C}^e$ and $\bar{C}^s$ are the numerically calculated average contrast factors for pure edge and pure screw character, respectively. The numerical values of $\bar{C}^e$ and $\bar{C}^s$ have been evaluated as a function of the Zener constant, $A_z$, and the ratio of the elastic constants, $c_{12}/c_{44}$, in the case of the most common slip system in the cubic materials and published in [16]. In case of copper the elastic anisotropy parameter are $A_z=3.21$ and $c_{12}/c_{44}=1.61$. Using these values and the parametrical functions published in [16, 17] that the theoretical value of $q$ in the case of copper is 1.6 for pure edge dislocations and 2.38 for pure screw dislocations. This is illustrated in Figure 1. The theoretical values of $C_{h00}$ for the case of copper are obtained as 0.29 for screw and 0.31 for edge dislocations by using Figure 1 from [16].
In the next step, by inserting using Equation (1) in Equation (2) the fraction of edge and screw dislocation are evaluated.

![Graph showing the parameter q for screw and edge dislocations as a function of the Zener constant A_{Z} and the ratio of the elastic constants c_{12}/c_{44} in the case of the most common slip system in the face centered cubic materials: a/2 <110> {111}. In the case on copper A_{Z}=3.21, the theoretical value of q for pure edge dislocations is 1.6 and for pure screw dislocations is 2.38.]

**RESULTS AND DISCUSSION**

The X-ray peak profile analysis results for crystallite size distributions; dislocation density and types are illustrated in Figures 2 and 3. Figure 2 shows the crystallite size distribution for four different deformation levels studied here. It can be observed that the median, m, and the variance, \( \sigma \), of the size distributions decrease with increasing deformation level. This shows that by introducing extremely high strains at low temperatures, the dislocations movement is constrained leading to formation of dislocation walls determining the reduction of the X-ray diffraction coherent domain from 80 nm at 67% deformation reduction to about 20 nm at 97% rolling reduction. This result is in good correlation with the results of dislocation density shown in Figure 3, where a major decrease in the dislocation density as the material is rolled from 67% to 74% reduction. It can also be observed that from 74% to 97% reduction level the dislocation density increases slightly. This effect can be explained by the nucleation of new generation of dislocations.

The evolution of the dislocations type during cold rolling of the nanocrystalline copper is shown in Figure 3. It can be observed that the population of screw dislocations decreases at higher reduction levels, while the edge dislocation fraction increases. The results are in good agreement with molecular dynamics simulations of the deformation of nano-structured materials in [20].
The simulations predict that when the crystallite size decreases to few tens of nanometers the plastic deformation process in nano-structured materials is dominated by emission of edge dislocations from the grain boundaries.

![Figure 2](image2.png)

Figure 2. The crystallites size distribution function determined by X-ray Peak Profile Analysis corresponding to copper samples deformed under liquid nitrogen at different reduction levels.

![Figure 3](image3.png)

Figure 3. Dislocations character and dislocations densities evolution as a function of rolling reduction.
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