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Temperature dependence and size effects on strain hardening mechanisms in copper polycrystals

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ABSTRACT

The thermomechanical behavior of polycrystalline sheets of high purity copper with various numbers of grains (d) across the thickness (t) is experimentally analyzed. As for other face centered cubic materials, the t/d ratio strongly affects the work hardening of copper, especially for t/d values lower than a critical one around five. These results are in agreement with previous ones concerning nickel and can be correlated with surface effects which progressively take place in the material when few grains are present across the thickness. The t/d ratio mainly affects the second work hardening stage and an increase in temperature tends to reduce this effect due to the early beginning of cross slip which leads to a generalization of the surface properties on the overall volume of the material. This result is supported by the analysis of the dislocation structures during the second and third hardening stages for polycrystalline and multicrystalline specimens. As a consequence, the forming of thin metallic products should be ideally performed at moderate temperatures in order to avoid size effects which can deteriorate the reliability of the final product.

1. Introduction

The important efforts made recently by the scientific community to characterize the role played by the size of the samples on the mechanical behavior of metallic materials greatly improve our knowledge of the size effect mechanisms [1–4]. For polycrystalline specimens with sizes in the micrometer range (*i.e.* between a few micrometers and a few millimeters), one of the most important size effect seems to be linked to the decrease in the number of grains across the thickness t [5–8]. With a decrease of the latter, the flow stress, the hardening behavior and the fracture strain are modified [9–11]. This feature is due to the apparition of surface effects for specimens with less than a critical number of grains across the thickness (*i.e.* t/d ratio) which depends on the grain size d and on the stacking fault energy. These surface effects involve a stress gradient of around 30% between core and surface grains which had been previously characterized by Transmission Electron Microscopy (TEM) analysis for nickel [12] and then confirmed by finite element simulations with crystal plasticity models [13].

When we are dealing with forming industrial processes, it is necessary to take into account this size effect linked to the t/d ratio to correctly predict the force and geometry conditions and the appearance of fracture or necking during the forming process of microsized parts. Moreover, the effect of temperature generally associated with plastic straining during forming processes on this kind of size effect is of crude importance.

The objective of this work is to provide new scientific results regarding the influence of size effects coupled with temperature for copper polycrystals of constant thickness and various grain sizes. Copper was chosen because of its wide use in the microdevice industry which is strongly concerned by size effects from a technological point of view. Furthermore, copper has a lower stacking fault energy than nickel which was intensively studied by our team over recent years (typically 45 mJ/m^2 for Cu against 125 mJ/m^2 for Ni). Comparisons with the latter can therefore enhance conclusions on the influence of the t/d ratio on the mechanical behavior at room temperature. The second objective of this work is to carry out a detailed study devoted to the influence of temperature on the size effects previously highlighted. With this aim, the mechanical behavior of two kinds of samples, one with a t/d higher than the critical value (polycrystalline regime) and a second with a t/d lower than this value (multicrystalline regime [14]), was intensively studied at temperatures ranging between 203 K and 773 K. The interpretation of the results is supported by TEM observations on samples thermoplastically strained both in second and third stages of the work hardening. Conclusions are finally given on the effect of temperature on the formability of copper sheets with few grains across the thickness.

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2. Material properties and experimental procedure

2.1. Experimental details

Monotonous tensile tests were performed at various temperatures between 203 K and 773 K on dog-bone-shaped samples with a gauge section of 20 mm in length and 10 mm in width. The experiments were strain rate controlled at a strain rate of $2\times10^{-4}\ s^{-1}$. Lower temperatures were applied to the samples using a climatic room cooled down with liquid nitrogen. Axial deformation was measured by a traditional contact extensometer. High temperature tensile tests were carried out in a conventional furnace and the deformation was measured by temperature contact extensometer with two ceramic extremities.

Microstructural observations were performed thanks to conventional metallurgical setups. The grain size distribution of both surface and cross section of the samples was analyzed by light microscopy and Scanning Electron Microscopy (SEM). Crystallographic texture analysis was performed by Electron Backscatter Diffraction Analysis (EBSD) attached to the SEM. The dislocation structure analysis of the thermomechanically work hardened samples with various grain sizes was carried out by TEM. Thin foils were taken from the middle of the thickness and were prepared using a twin-jet electrolytical thinning apparatus. The dislocation substructures were studied by transmission using an electron microscope working at 200 kV.

2.2. Material properties

High purity polycrystalline (> 99.99 wt%) copper samples consisting of rolled sheets of t=0.5 mm thickness were annealed in a secondary vacuum ($PO_2 < 10^{-5}$ mbar) and then air cooled in order to obtain various grain sizes without oxide layers. Examples of various heat treatment conditions and corresponding microstructural features are reported in Table 1. Details of this experimental procedure and microstructural characterization were given previously for experiments concerning nickel studies [3,9]. Typical grain structures are shown in Fig. 1 using light microscopy observations (observations through the thickness) and EBSD (observations on the surface). In this case, the surface of the samples was electropolished and grains were revealed by an adequate etchant (H_3PO_4 in an alcoholic solution). Annealing twin boundaries were considered as grain boundaries for the estimation of the mean grain diameter. As reported in the literature, the proportion of the twin boundaries affects the mechanical behavior of metallic materials both for bulk [15] and small sized [2] metallic alloys. In this work, the fraction of the corresponding Σ_3 boundaries strongly fluctuates with the grain size of the samples and no correlation was observed between annealing twin boundaries and grain size. As a consequence, the modification of the mechanical behavior highlighted in this study cannot be due to a variation of twin boundaries proportion between samples. For samples with less than one grain across the thickness, the grain structure can be considered as columnar because of the low proportion of horizontal grain boundaries. The distribution of the mean grain diameter d follows a unimodal Gaussian statistic centered on 0.7 times the mean value. The mean grain size varies between 20 μm and 571 μm leading to t/d ratios ranging from 0.9 to 25. Crystallographic texture analysis shows a similar texture for all the samples. The corresponding maximal density I expressed in multiples of a random distribution (m.r.d. unit) takes values lower than five. Consequently, the behavior of copper is expected to be mechanically isotropic with no preferential deformation of the samples, especially along the thickness.

Table 1
Example of heat treatment conditions (temperature T and annealing time t_a, secondary vacuum). Corresponding values of grain size d, t/d values, maximal texture density I and Σ_3 boundary proportions.

T (K)	t_a (min)	d (μm)	t/d	I (m.r.d.)	Σ_3 proportion (%)
573	220	23	21.6	2.39	19.5
673	220	29	17	1.94	34.3
773	220	42	12	2.25	32.96
873	360	65	7.7	3.93	23
923	220	109	4.6	3.50	55.4
873	1440	120	4.2	4.39	9.4
1073	360	194	2.6	2.51	37.4
1073	1440	571	0.9	3.52	35.9

3. Thermomechanical strengthening of polycrystalline copper with various t/d ratios

3.1. Mechanical properties at room temperature

The grain size dependence of the stress σ for different strain levels ε is well represented by the conventional Hall and Petch (HP) relationship [16]:

$$\sigma(\varepsilon)=\sigma_0(\varepsilon)+\frac{k_{hp}(\varepsilon)}{\sqrt{d}} \quad (1)$$

$\sigma_0(\varepsilon)$ and $k_{hp}(\varepsilon)$ are material empirical constants which depend on strain. Fig. 2 shows HP plots for plastic strain values of 0.028 and 0.052. Two distinct regimes are clearly evidenced and a critical t/d ratio labeled $(t/d)_c$ can therefore be defined. The evolution of this parameter as a function of the plastic strain is given in the inset of Fig. 2. $(t/d)_c$ linearly increases with strain taking values ranging between 5 and 8. Regime I is representative of the polycrystalline behavior of copper and HP parameters are in agreement with literature [17–19]. In the second regime, the dependence on the grain size increases more significantly which leads to a sensitive decrease in stress. This effect strongly modifies the HP coefficients and characterizes the multicrystalline behavior of copper [4,14] located between the polycrystal and the single crystal ones.

As already observed for nickel, the strengthening mechanisms of copper are also affected by the t/d ratio. The work hardening rate $\theta=d\sigma/d\varepsilon_p$ of polycrystalline f.c.c. metals generally exhibits three different stages related to the activated gliding systems. Following a widely used analysis proposed by Mecking and Kocks [20,21], the different hardening stages can be studied using a plot of $\sigma\theta$ *vs* σ. An example of such a curve for copper polycrystals is presented in Fig. 3 with the corresponding monotonic tensile curve. Detailed descriptions of each stage can be found in traditional reviews [22–25]. Our results focus on the second stage of work hardening, related to the activation of multiple gliding systems and the existence of a few cross slips [26]. This stage occurs on a strain range $\Delta\varepsilon_{II}$ (called hereafter the length of the stage II) and ends for a critical stress value $\sigma_{II/III}$ corresponding to the generalization of cross slip in the material [26,27]. As described elsewhere [4,9], during the second work hardening stage, the evolution of the product $\sigma\theta$ with σ is linear and verifies the following relationship [24]:

$$\sigma\theta=\Delta_{II}\sigma+(\sigma\theta)_0 \quad (2)$$

where Δ_{II} is the latent hardening rate and corresponds to the interactions between forest dislocations [20]. The second term in the right side of Eq. (2) depends on the grain size and on the initial dislocation structure [4]. For pure recovered single crystals, this term is generally close to zero. Second phases or precipitation increases the value of this parameter [28] while the predominance