VERIFICATION OF HALL-PETCH EQUATION OF NANOCRYSTALLINE COPPER

Katarína Sülleiová
Michal Besterči
Tibor Kvačkaj

\(^{a}\) Institute of Materials Research of SAS, Watsonova 47, 040 01 Košice, SR, E-mail address: mbesterci@imr.saske.sk, ksulleiova@imr.saske.sk
\(^{b}\) Technical University Košice, Faculty of Metallurgy, Department of Metal Forming, Letná 9, 040 02 Košice, E-mail address: Tibor.Kvackaj@imr.saske.sk

Abstract

Hall-Petch relation for the yield stress as well as evolution of Cu microstructure after severe plastic deformation (SPD) realized by the method of the equal channel angular pressing (ECAP) are analyzed. On the basis of the experimental results and analyses the different mechanisms of plastic deformation generation for coarse grain and nanograin structures were identified. A final grain size decreased from the initial 50 \(\mu\)m to 100-300 nm after ten ECAP passes. Increase and deflection of the yield stress point were explained by the dislocation and diffusion theory and grain boundary sliding.

1. INTRODUCTION

Recently, large attention is devoted to the research and development of nanostructured materials. These materials with unique microstructure and grain size less than 100 nanometres (in one direction of 3D) have good physical and mechanical properties. The outstanding mechanical properties (superstrength, superhardness, enhanced tribological characteristics, enhanced tensile ductility and superplasticity) are of special importance. Several different methods for preparing of nanomaterials are elaborated, some of them based on powder metallurgy technique [1-5]. Residual porosity, pollution of the material system and grain coarsening are the problems that are unresolved satisfactorily up to now. These negatives can be eliminated by severe plastic deformations (SPD) using the method of the equal channel angular pressing (ECAP) where the experimental material is pressed in a special matrix through a special die consisting of two equal channels at the angle of 90°. The formation of high angle nanograins with the specific substructure of lattice and grain boundary dislocations is studied intensively and analyzed on models and real material systems, too [6-15].

The analysis of the yield stress point in relation to the evolution of nanostructure by SPD of copper using the ECAP method is the aim of this study.

2. EXPERIMENTAL MATERIAL AND TESTING METHODS

A pure conventional copper (99.9%) of the grain size \(~ 50 \mu\)m was used like the initial material. Cu bars of \(\Phi 10 \times 70\) mm were pressed in the ECAP hydraulic equipment at room temperature with the maximum power of cca 1 MN (Fig.1). Then static tensile test, hardness measurement, metallographic and TEM analyses have been carried out on the starting material as well as on ECAP-ed specimens.
3. RESULTS AND DISCUSSION

Mechanical properties as well as grain size of the initial Cu material are listed in Table 1. Cu is coarse grained with the middle grain size of ~ 50 µm and low yield stress and strength point but excellent reduction in area (65%).

Tab.1. Mechanical and plastic properties of initial Cu material

<table>
<thead>
<tr>
<th>$R_p$ 0.2 [MPa]</th>
<th>$R_m$ [MPa]</th>
<th>$A_5$ [%]</th>
<th>$Z$ [%]</th>
<th>HV</th>
<th>$d_z$ [µm]</th>
</tr>
</thead>
<tbody>
<tr>
<td>270</td>
<td>275</td>
<td>13</td>
<td>65</td>
<td>112</td>
<td>50</td>
</tr>
</tbody>
</table>

Fig.1. Scheme of ECAP

Fig.2. Microstructure of the initial Cu material

Fig.3. Dependence of the grain size on the ECAP passes
The grain microstructure of the initial material observed by light microscopy is shown in Fig. 2. Grains are polygonal and homogeneous. The grain size decreases with number of passes, Fig. 3. Despite of a specimen rotation at plastic deformation process in ECAP, the microstructure is heterogeneous, and the grains are partially elongated in the pressing direction. Grains after the maximum plastic deformation (10 passes) are already at the limit of the optical metallographic resolution; the mean grain size is < 1 µm. In order to estimate the grain size more accurately and to explain the grain formation during subsequent passes, thin foils from the neck area of the fractured specimens were prepared. The mean grain size after 10 passes has been estimated cca 100 – 300 nm, Fig. 4, [6]. The mechanism of forming grains with high angle boundaries is supposed to be by the forming of a cellular structure, and subgrains forming, which are transformed with increasing deformation into nanograins with high angle random orientation.

![Fig. 4 Nanograins with high angle random orientation](image)

It is known, that grain boundaries in metal systems play role of obstacles to the movement of dislocations and influence the yield point stress – the limit of the conventional plastic deformation. It is described by Hall-Petch equation, i. e. the relation between the yield stress point and the grain size:

\[ \sigma_y = \sigma_0 + k d^{-1/2}, \]  

where \( \sigma_y \) is the yield stress for a polycrystal, \( \sigma_0 \) is the yield stress for a single crystal or a polycrystal with an infinitely large grain size, \( d \) is the average grain size, and \( k \) is the Hall-Petch parameter.

Fig. 5 shows three different areas depending on the orientation of curves, [16]:
- relevance of Hall-Petch equation is in the range from monocrystal to the grain size of 1 µm, an exponent \( n \) has the value from -0.5 to 1.
- Hall-Petch equation is relevant, e. i. with the decreasing grain size the yield stress increases, but not so intensive as in the previous case, whereby \( n < -0.5 \); the equation is relevant in the grain size range from 1 to 100 nm,
- Hall-Petch equation loses its relevance, e. i. with the grain size decreasing the decrease of the yield stress occurs; a slope of straight line change occurs and the exponent can change its value, whereby the grain size is ≤ 10 nm and has extremely high volume fraction of grain boundaries over 50 %.
It has been established in [17] that for some pure metals the $k$ coefficient decreases with the grain refinement, and for copper it passes through zero and even takes on a negative value. Kozlov [18] analyzed behaviour of the $k$ parameter as a function of the grain size for Fe, Cu, Al, Ni and Ti and he stated that the $k$ parameter has the smallest value for fcc metals. The coefficient $k$ decreases rapidly as $d$ approaches 10 nm. The behaviour of the $k$ governed largely by the grain boundary sliding i.e. the properties of metals with the different crystal lattices match closely at high grain boundary densities. According to [19] diffusion mechanism, shear mechanism and combined mechanism are the mechanisms contributing to the grain boundary sliding. The different kinds of grain boundary shear both of the diffusive and dislocation types dominate for the grain size in the range of 10 nm. For a grain size of $> 1 \mu m$, the dislocation mechanism prevails. The processes involved in deformation are correlated.

Fig. 5 Microstructure classification

Fig. 6 Schematic representation of grain boundary sliding (GBS) accommodated by intergranular dislocation processes: GD is the gliding of the dislocations, MGB is the migrating grain boundary, DS is the dislocation sink, SD is the source of dislocations, and DC is the dislocation climb, [19].
Fig. 6 is a schematic representation of a polycrystal where the several intergranular processes are at work. Grain boundary sliding activates intergranular dislocation slip. The triple grain junctions are the sources of dislocations. The dislocation glide in grains initiates absorption of dislocations by the opposite boundary, sliding along the boundary and migration of the boundary. It is necessary to note that in fine grains ~10 nm, generation of dislocations is difficult and could be observed only in larger grains ~100 nm on thin foils.

According to Valiev et al. [8] at the grain size of 100 μm dislocations inside the grains were not observed, at grain size of 200 nm dislocations generated the cell substructure. It can be followed from these considerations that deformation mechanisms at the yield stress changed markedly e. i. shear dislocation mechanisms are replaced by combination of grain boundary mechanisms what causes a deflection of the k parameter in the Hall-Petch equation.

The experimental dependence of $R_p0.2$ on $d^{-1/2}$ for the ECAP-ed copper is shown in Fig. 7.

![Graph](image)

**Fig. 7** Dependence of hardening expressed by the yield point $R_p0.2$ on the grain size using the Hall-Petch equation

It is evident, that the $R_p0.2$ has the increasing tendency in the range of the grain sizes what is consistent with the theoretical considerations. Related to the deformation mechanisms, gliding of dislocations and grain boundary sliding are applied.

### 4. CONCLUSIONS

The following conclusions have been stated from the obtained results:

- Decrease of the average grain size from cca 50 μm to 100-300 nm with the increasing deformation by the method of ECAP (after 10 passes of ECAP).
- The experimental dependence $R_p0.2$ versus $d^{-1/2}$ for Cu after 10 passes of ECAP is in agreement with the theoretical considerations for the different
grain sizes. Gliding of dislocations and grain boundary sliding are feasible deformation mechanisms.

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