TEM analysis and determination of dislocation densities in nanostructured copper tube produced via parallel tubular channel angular pressing process

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A B S T R A C T

Parallel tubular channel angular pressing (PTCAP) is a recently developed novel intense plastic deformation method appropriate for fabrication ultrafine-grained and nanostructured cylindrical tubes. In the present work a commercially pure copper was processed via multi-pass PTCAP and the effects of number of passes on grain refinement and the dislocation density were studied. TEM analysis showed that after first pass elongated subgrains with interior tangled dislocations were formed. After pass number two the density of interior dislocations through the elongated grains was decreased. In the next stages of deformation, at pass number three, elongated grains almost disappeared and equiaxed grains with grain size about 150 nm are formed as a result of dynamic recovery. The dislocation densities were measured by hardness indentation size effect using the Nix–Gao model. The results showed that increase in the number of PTCAP passes leads to decrease in the dislocation densities. The dislocation density is decreased to $2.48 \times 10^{9}$ cm$^{-2}$ after fourth passes from $18.1 \times 10^{9}$ cm$^{-2}$ after first pass. TEM results verified calculated values from the Nix–Gao model. Microhardness of the PTCAP processed tube through four passes increased to ~142 HV from initial value of about 62 HV. Also, significant increase takes place after single pass and in the next passes the hardness value is saturated.

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1. Introduction

It is well-founded that there are superior mechanical and physical properties in ultrafine grain (UFG) and nanostructured (NS) materials [1]. Improving of the material properties using grain refinements via severe plastic deformation (SPD) has been interested in past two decades [2]. In recent decades different SPD methods were developed for producing UFG and NS bulks [3–7]. The ultra-high plastic strain imposed during SPD subdivides coarse grains into UFGs, and simultaneously creates a high density of dislocations on the order of $10^{14}$–$10^{16}$ m$^{-2}$ [8–10]. Tubular components have more application in different industries but few efforts have been done for producing UFG and NS tubes employing SPD techniques. An especially attractive method, tubular channel angular pressing (TCAP) based on equal channel angular pressing (ECAP), developed by Faraji et al. [11–13]. They applied this technique to AZ91 magnesium alloy and reported more grain refinement and significant improvement in the mechanical properties [11]. They also investigated the effects of process parameters such as channel geometry [12] and channel angle [13] on plastic deformation and process load using finite element modeling. Also, very recently they proposed a new SPD process named PTCAP method based on TCAP suitable for producing high strength tubes [14]. They showed that obtaining excellent strain and hardness homogeneity and also needing ~60% lower process loads which are two major advantages of PTCAP compared to TCAP. As is well-known, process load and hardness homogeneity are two important challenges in SPD of metallic materials, hence PTCAP is an excellent SPD method for producing UFG and NS tubular components [14].

During PTCAP process the material is severely deformed and UFG materials are achieved. To develop UFG structure the presence of excess dislocations are needed [15]. As reported in previous researches, the dislocation densities act as a determinant factor in controlling the mechanical properties of the metals [2,5,8,16,17]. Whereas existence of high density of dislocations plays a main role in producing UFG materials, estimation of the dislocation density is necessary. The dislocation density can be measured using TEM micrographs and also using the Nix–Gao model [18]. Graca et al. reported that it is possible to estimate the dislocation densities by the hardness indentation size using the Nix–Gao model [18]. In the current study a commercially pure copper tube was processed via multi-pass PTCAP and the effects of number of passes on grain refinement, mechanical properties and the dislocation density were studied.
In the PTCAP process there are two half cycles shown schematically in Fig. 1. The first punch presses the tube material into the gap between mandrel and die including two shear zones shown in Fig. 1(a) during the first half cycle. In this stage, the diameter of the tube increases to gain its maximum value. Then, the tube is pressed back using second punch in the second half cycle (Fig. 1(b)). The tubes thickness in initial and processed conditions is same. The process including two half cycles can be repeated to apply a distinct strain while no variation in the cross section area of the tube is occurred. After $N$ passes of PTCAP process, the total equivalent plastic strain can be estimated via the following equation [14]:

$$\varepsilon_{TN} = 2N \left\{ \sum_{i=1}^{2} \left[ \frac{2z \cot \left( \varphi_i / 2 + \psi_i / 2 \right)}{\sqrt{3}} + \psi_i \csc \left( \varphi_i / 2 + \psi_i / 2 \right) \right] + \frac{2}{\sqrt{3}} \ln \frac{R_2}{R_1} \right\}$$

These formulas are different from conventional ECAP because there are some peripheral tensile and compression strains [11,12] in PTCAP process. From Eq. (1), the equivalent plastic strain through 1, 2, 3 and 4 passes of PTCAP are 3, 6, 9 and 12, respectively.

2. Experimental procedures

The samples of commercially pure copper were taken as initial tubes of 2.5 mm in thickness, 20 mm in diameter, and 50 mm in length. Prior to PTCAP processing, tubes were heated to the temperature of 600 °C and annealed for 2 h to get a recrystallized homogeneous microstructure shown in Fig. 2(a). A PTCAP die including mandrel, die and punches from H13 tool steel was prepared and hardened to ~55 HRC. Before pressing
the tube, the die parts were lubricated with MoS$_2$ [19]. Die parameters including the channel angle $\phi_1 = \phi_2 = 120^\circ$, the angle of curvature $\psi_1 = \psi_2 = 0^\circ$ and $K = R_2 - R_1$ was equal to the tube thickness (2.5 mm) are shown in Fig. 1(d). PTCAP experiments were done in punch speed of 5 mm/min at RT. Microhardness measurements were performed in loads ranged from 0.1 to 5 N. The microstructure in the PTCAP processed tube was investigated by transmission electron microscopy (TEM). Lamella sample preparation was performed using focused ion beam (FIB). Two trenches are milled on each side of the area of interest until it is about ~1 μm thick. The upper and lower side of the TEM sample are then milled together with a U-cut under the sample leaving only a small uncut portion to hold the lamella to the main sample. The lamella is finally attached to a TEM Cu grid using platinum deposition and the needle is milled away. Final thinning is performed on the lamella sample. The grain sizes were measured using the linear interpret method.

3. Results and discussion

Fig. 2(a) shows initial recrystallized microstructure of the unprocessed sample in which a mean grain size of ~59 μm could be seen. The microstructure evolution of pure copper tube after PTCAP is shown in Fig. 2(b)–(g). Fig. 2(b) and (e) shows the TEM micrograph of PTCAP processed tube in perpendicular-with-axis direction through one pass, at equivalent strain ~3, in two different magnifications. As expected, the microstructures are more refined, and are significantly different from the initial state (Fig. 2(a)). TEM micrograph of first pass PTCAP processed specimen contains an elongated and subgrain structure with tangled dislocations in which distinct regular-shaped walls was observed. This event was obviously seen in pure copper after second pass in the ECAP process [20]. It is also found that the subgrain size in the microstructure of the initial tube is more coarser than that in the PTCAP processed one. After undergoing the first pass, original coarse grains are divided into subgrains. TEM micrographs of the
tube performed by PTCAP process for second pass which corresponds to equivalent strain of ~6 are shown in Fig. 2(c) and (f). In this figure, even though some of the local tangled dislocations still exist, the elongated grains with ~150 nm size can be distinguished by sharp boundaries. At this stage the density of interior dislocations is decreased but the grains is not still free of dislocations. This indicates that dynamic recovery (annihilation or reorganization) occurs near these boundaries [20]. The grain refinement process implies the arrangement and rearrangement of dislocation cells in original grains of the pure copper [21–23]. When the number of PTCAP passes increase to three, which is at equivalent strain ~9, elongated grains are almost disappeared, and equiaxed grains with grain size about 150 nm are formed (Fig. 2(d) and (g)). Tendency for formation of recrystallized equiaxed grains with sharp boundaries from elongated one when increasing in the accumulated strains to about 10 could also be seen in ECAP of high-purity Al [20]. Ferrasse et al. reported that the ultra-fine grains were developed by dynamic recrystallization through grain fragment rotation during ECAP deformation at room temperature at higher strains [24]. Small recrystallized grains were also observed in 99.95% copper deformed equivalent to a strain of 10 during ECAP [25].

The Vickers microhardness changes along the tube thickness after one to three PTCAP passes are shown in Fig. 3. Hardness increases notably after first pass of PTCAP and then increases gradually. Good hardness distribution was observed throughout the thickness of the PTCAP processed tube in all PTCAP passes. The figure shows that after third pass the microhardness is increased to ~142 HV from the primary value of 62 HV. It is also detected that the microhardness is saturated at equivalent plastic strain of about 6. This saturation could also be seen in ECAP process and it was mentioned to be related with the lower bound of the grain size which is in the range of about ~200 nm [21]. It is well known from Hall–Petch relationship that the hardness is related to the grain size. Microstructural investigations establish that the grain size is saturated at the value of 150 nm after one pass PTCAP. The changes in hardness, from the first to third pass of PTCAP, are in good agreement with the microstructural evolutions discussed in the previous sections. This is also verified by the works done by other researchers shown in the chart [26–28].

The dislocation densities of the samples could be estimated from indentation hardness measurement. Graca et al. [16,18] mentioned that the hardness values increase as the indentation depth decreases. The relation between the hardness value and indentation depth is indicated as following equation [16]:

\[
\left(\frac{H}{H_0}\right)^2 = 1 + \frac{h^*}{h} \left(\frac{1}{h}\right)
\]

where \(H_0\) is the hardness in the limitation of infinite depth (bulk hardness), \(h^*\) is a characteristic length and \(H\) is the hardness value corresponding to indentation depth \(h\). Fig. 4 shows the amount of \(h^*\) in the PTCAP processed specimens through 1, 2, 3 and 4 passes which are measured to be 788.53 nm, 2958.4 nm, 4268.3 nm and 5766.4 nm respectively determined by fitting Eq. (2) to experimental data.

The correlation between the dislocation density \(\rho_s\) statically stored in the lattice and the characteristic length \((h^*)\) is as following [8,17]:

\[
\rho_s = \frac{3}{2f^2 \pi^2} \frac{\tan^2 \theta}{bh^*}
\]

where \(b\) is the Burgers vector of the dislocations, \(\rho_s\) is the density of dislocations statistically stored in the lattice, \(\theta\) is the angle between the surfaces of the material and the indenter, and \(f\) is a correction factor for the size of the plastic zone. In this research \(\theta = 22^\circ, f = 1.9\) [8], and \(b = 0.25\) nm [8,17].

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The density of dislocations statistically stored in the lattice of processed samples versus PTCAP passes calculated from Eq. (3) is shown in Fig. 5. It is clear that an increase in the number of PTCAP passes and accumulated strain leads to decrease in the density of dislocations. As mentioned above, decrease in the density of dislocations with increase in the number of passes indicates that the dislocation annihilation by dynamic recovery occurs during additional straining [20]. The density of dislocations in PTCAP processed sample after one pass is about 18.1 \(\times 10^8\) cm\(^{-2}\) which is 3.7 times higher than that in processed sample after two passes. This can be verified by TEM micrographs of processed samples through one and two passes shown in Fig. 2(b) and (c), respectively. Increase in the number of PTCAP passes from two to three and four passes almost has not more effect on the density of dislocations. This can also verify the hardness saturation which was seen in Fig. 3.

Miyajima et al. estimated the density of dislocations values to be around \(10^{10}\) cm\(^{-2}\) in commercial pure aluminum samples with equivalent strain up to 10 processed by accumulative roll bonding process using the scanning transmission electron microscopy (STEM) [29]. The present values of dislocation densities estimated using hardness indentation have almost a good agreement with the previously reported values by other techniques such as STEM [29] and X-ray [30]. Barmouz et al. [8] estimated the dislocation densities of friction stir processed (FSPed) copper sample using the similar method used in this work to be in the range of \(10^9\) cm\(^{-2}\) which is lower than that of the values calculated in this research in the lower number of passes. This may be attributed to the heat input in the FSP process as a result of intense friction between tool and copper sample compared to PTCAP process done at room temperature [31].

4. Conclusions

A commercially pure copper was processed via multi-pass PTCAP and the effects of number of passes on grain refinement and accumulation of the dislocations were studied. TEM analysis showed that after first pass elongated subgrains with interior tangled dislocations were formed. After second pass, the density of interior dislocations through the elongated grains was decreased. In the next stages of deformation (third pass), elongated grains almost disappeared and equiaxed grains with grain size about 150 nm were formed as a result of dynamic recovery.
The dislocation densities were measured by hardness indentation size effect using the Nix–Gao model. The results showed that increase in the number of PTCAP passes leads to decrease in the dislocation densities. The dislocation density is decreased to $2.48 \times 10^9 \text{cm}^{-2}$ after fourth pass from $18.1 \times 10^9 \text{cm}^{-2}$ after first pass. TEM results verified calculated values from the Nix–Gao model. Microhardness of the four passes PTCAP processed tube increased to 142 HV from initial value of about 62 HV in which a significant increase had taken place after single pass and it is saturated in the next passes. There is also superior microstructure and hardness homogeneity through the processed tube.

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