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Production of Ultrafine Grain In Commercially Pure Copper By Ecap

[1]B.Praveen*, [2]V. Balusamy, [2]J.Krishnamoorthi, [2]P.C. Angelo
[1]Department of Materials Science and Engineering, CARE Group of Institutions, Tiruchirappalli – 620009
[2]Department of Metallurgical Engineering, PSG College of Technology Peelamedu, Coimbatore – 641 004
** Corresponding Author : nathanpraveen@gmail.com*

Abstract

Equal channel angular pressing, is a severe plastic deformation process to develop grains below 1□ m in diameter. The paper reports the formation of ultra fine grained structure in electrolytic copper by equal – channel angular pressing (ECAP). Electrolytic copper was successfully processed using a die having channel intersection angle Φ=90° and angle of curvature $\psi=20^0$ in route B_C. ECAP was carried out upto 10 passes at room temperature. The imposed strain resulted in a large reduction in the grain size to submicron level. Microstructure and mechanical properties of commercially pure copper rods processed by equal channel angular pressing were investigated. Special attention was paid to the features of the processed microstructures and micro hardness properties of the ECAP samples. The grain size and micro strain were calculated from XRD data using Williamson Hall Method. The optical morphology of each angle pressed copper was studied and correlated with grain size measured by William's Hall Method. Micro hardness and micro tensile values of ECA pressed copper varied with respect to each angle press.

Keywords: Severe plastic deformation, ECAP, Micro hardness, Microstructure, Mechanical properties, Grain size

Introduction

Recently several methods have been developed to obtain ultra fine grained metals. The ECAP technique allows the repetitive pressing of a billet through a die that has two intersecting channels without changing its cross sectional dimensions. Very high shear strains can therefore be produced in the material. Copper represents an ideal model material for studying the processes of deformation and microstructure evolution, due to its simple FCC structure, medium stacking-fault energy. (Nahed El

Mahallawy et.al., 2009) According to the fast developments in electronic industries, there is a great demand for high-strength and high-conductivity materials. The alloyed copper is the most widely used material in this respect. The electrical conductivity of these materials is inherently lower than unalloyed counterpart. To resolve this problem ultra-fine grains (UFG) or nanostructure commercial pure copper, can be used. This goal can be attended by severe plastic deformation (SPD) processes (Hosseini and Daneshmanesh, 2009). The application of ECAP to bulk solid produces grain refinement to the sub-micrometer or nanometer level (Han and Langdon, 2005; Han et al., 2002); fabricated ultra-fine grained pure coppers of 200nm in size by the equal channel angular extrusion (ECAE) and showed that the mechanical properties (ultimate tensile strength and yield strength) and hardness reach a maximum after four passes and then decrease with additional number of passes. Han et al. (2008) Most of the studies on ECAP process have mainly focused on Al, Mg alloys and steels; however, there is almost no report on the effect of ECAP process on pure copper properties. The present study mainly focused on the effect of ECAP pass on the microstructure, mechanical and electrical properties of commercial pure copper rod.

Experimental Procedures

ECAP Device

The device used in this study for the ECAP process with two split type die sets were made with channel angle $\phi = 90^0$ and $\psi = 20^0$ with 12 mm channel diameter (Fig 1).

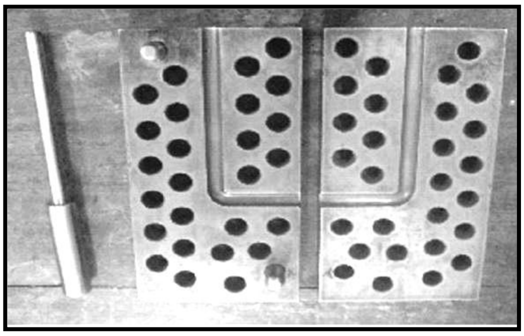

Figure 1. Die set-up with punch 20^0 ψ angle

The material used for the die and punch was H11 tool steel having average hardness of about 55 HRc. A 200 ton hydraulic press was used to carry out the pressures experiment. To accommodate the dies on the press suitable clamps were assembled on the work platform (Fig 2). To evaluate the variations of microstructure and mechanical properties, ECAP process was applied on pure copper rod upto10 times. The samples were rotated in the clockwise direction by 90◦ after each consecutive pressing. This kind of rotation, designated as route Bc in ECAP, led most expeditiously to homogeneous equiaxed-grained structure (Rusz et al., 2005).

Figure 2. 200 ton Hydraulic press with clam assembly

Procedures

Commercial pure copper rod was used in this study. The chemical composition of this material is given in Table 1. It was annealed at 600◦C for 1h, cut into rods of 60mm×12mm (length×diameter) and subjected to the ECAP process. To evaluate the changes in microstructure and mechanical properties the ECAP process was applied on pure copper rod upto 10 times. The samples were rotated around the longitudinal axis by 90◦ in the same direction after each passes.

Table 1: Chemical composition of base metal

Element	**Amount Present in %**
Cu	99.90
Fe	0.045
Zn	0.0295
Si	0.0067
Al	0.0045
Mn	0.012
Be	0.0008

Room temperature micro tensile test was conducted on round-type specimens that have been machined along the ECAP direction (Fig. 3) at a strain rate of $2.1\times10^{-3}s^{-1}$ by an Instron 8502 testing machine (capacity:30ton). The gauge length and width of the tensile specimens were 19.35 and 3.20 mm, respectively.

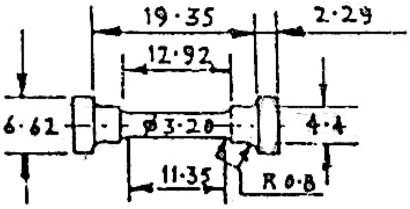

Figure 3. Micro tensile test specimen preparation

Microhardness was measured along the transverse direction at equal distances on the ECAPed samples. Hardness variations were measured by Vickers hardness tester with a pyramidal diamond indenter at 100 g load. For each sample, three measurements were recorded along the upper middle, and lower layer of the specimens. Every recorded measurement was the average of six hardness values. The optical microscope model ‘Card Zeiss Gotingen’ was used for the microstructure examination before and after the ECAP using dilute Nitric acid as etchant.

Results and Discussions

Microstructures

Fig. 4a–f shows the optical micrographs of the as-annealed and 2, 4, 6, 8 and 10th passes of ECAP’ed specimens. Grain sizes of specimens were measured by Image Tool software. The average grain size of as annealed specimen was about 30μm with a standard deviation of 22μm. It was heterogeneous and showed a typical recrystallized structure with angular grains (Fig. 4a). Grains of the 2-pass ECAPed specimen refined to approximately 14±9μm exciting increasing inhomogeneity of structure (Fig. 4b), while those of the 4, 6, 8 and 10th pass specimens were more refined that the grain size was hard to measure (Fig. 4c, 4d, 4e, and 4f).

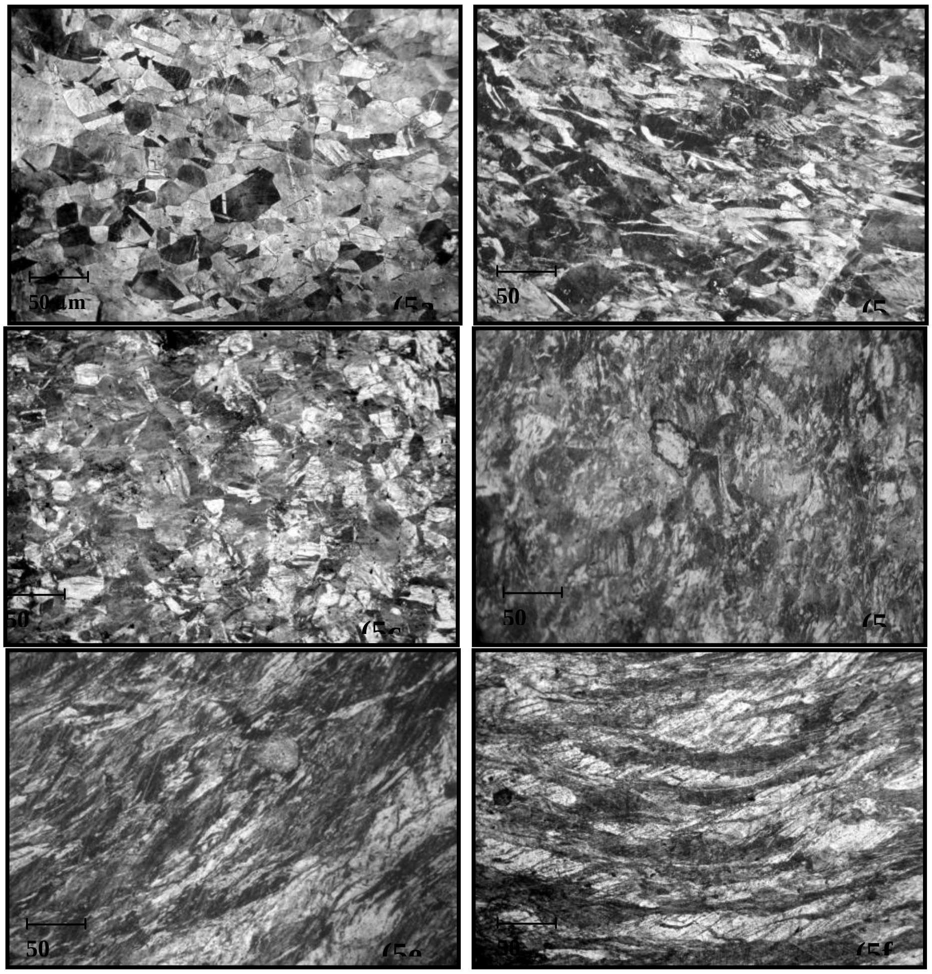

Fig. 4a–f is the optical micrographs of 2, 4, 6, 8 and 10th passes

Sub-grains sized 70–200nm were formed. Fig. 5, 6 and 7 are the TEM results confirming the formation of sub grains. Also some twin bands formed in the surface revealing the formation of dislocation cells responsible for the refinement of grains. The grains near the contact surfaces were smaller and more non-uniform than at the center of the specimen. Grain size increases with increasing distance from the contact surface. It is possible that further passes may expand this zone and reduce the differences between these two zones. Induction of severe shear deformation to materials by passing them through a die with two intersected channels produces ultra-fine grains in pure copper rod and with additional passes, new nano-sized sub-grains are formed inside the ultra-fine grains. For copper with a medium stacking fault energy (SFE:78mJ/m2), formation of dislocation cells is responsible for the refinement. The grain refinement in copper involves the formation of dislocation cells in original grains. With additional passes cell walls become thinner and the cell size reduces. Most boundaries are found to be high-angle ones while sub-boundaries with low-angle misorientations exist inside some refined grains. The refined grains are further subdivided by the sub-boundaries (Wang et al., 2006).

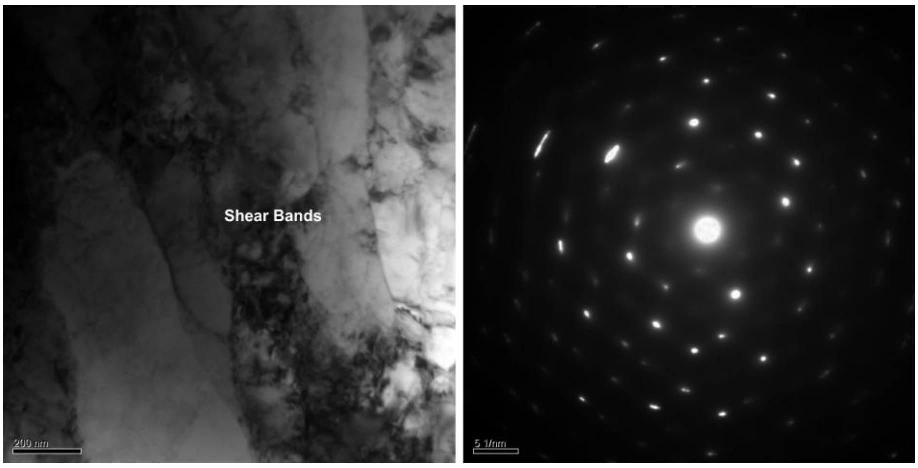

Fig. 5 TEM and their corresponding pattern for 2nd pass ECAPed Sample with formation of shear bands

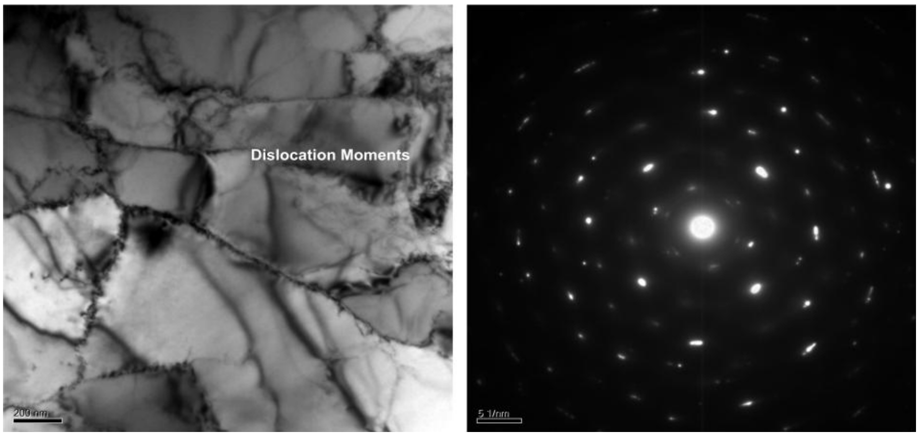

Fig. 6 TEM and their corresponding pattern for 4th pass ECAPed Sample with more no. of dislocation moments

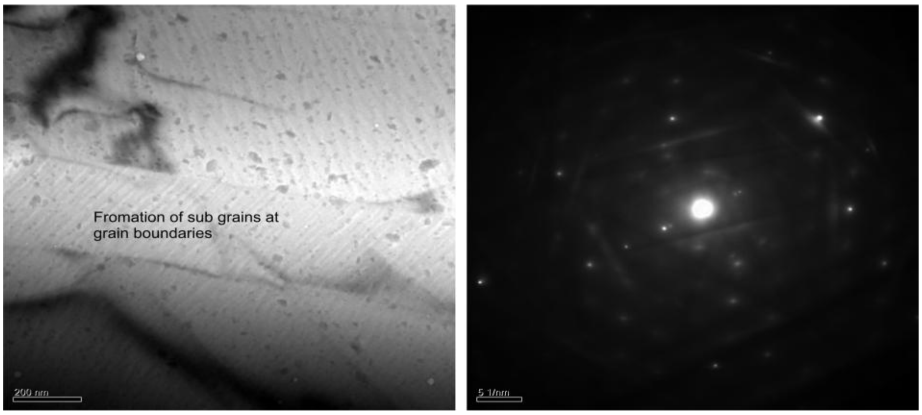

Fig. 7 TEM and their corresponding pattern for 6th pass ECAPed Sample with formation of sub grains at their grain bourdaries

X-ray diffraction of parent and ECA pressed copper

The ultra fine grain commercially pure copper was produced by equal channel angular pressing with different passes (such as 2nd, 4th, 6th, 8th and 10th) at the curvature angle (ψ) of 20°. The X-ray diffraction patterns of parent, and after 2nd, 4th, 6th, 8th and 10th pass are shown in Figure 8. It is found that the peaks observed are slightly shifted to higher angles after 2, 4 6, 8 and 10th passes at 2θ angles of 42.78°, 43.37°, 43.62°, 43.85°, 43.93° and 43.32° respectively. The peak shifts at higher angles generally cause an intrinsic fault or twin faults in the bulk copper. reveals that the observed line broadening to be asymmetric and negligible peak shift responsible for twin faults formed on the copper during ECAP process [Ter Verkrijging Van de Graad Van, thesis,1962].

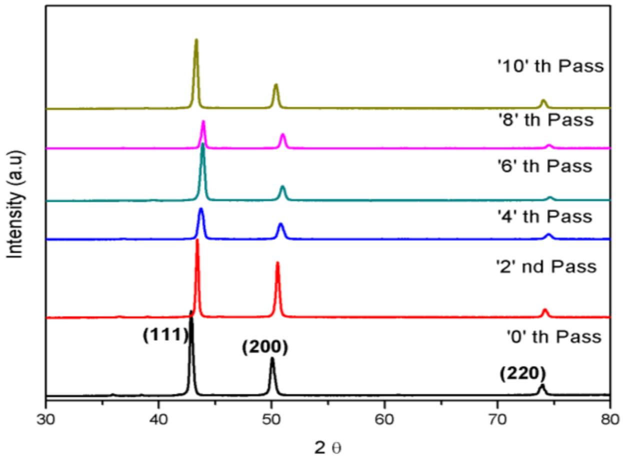

Fig. 8 : X-Ray diffraction pattern for parent (0th Passs), 2nd, 4th, 6th, 8th, and 10th passes

Effect of equal channel angular pressing passes on grain size

Equal Channel Angular Pressing (ECAP) on copper was carried out through route Bc. It means that the sample is pressed and rotated by 90^o angle in the clockwise direction after each pass. Generally, grains may be elongated when applying the load on the metals. ECAP is a continuous process so that there is a possibility of continuous elongation of the grains in respect of each pass. At the same time, the applied load creates a small strain on the materials, causing the breakage of elongated grains with the subsequent passes. This is confirmed by optical micrograph and grain size measurement via X-ray diffraction method (Hall Williamson method). The grain sizes after each pass were calculated using Hall-Williamson method using the equation given below :

$$\frac{\beta cos\theta}{\lambda} = \frac{K}{d} + 4\varepsilon\left(\frac{Sin\theta}{\lambda}\right)$$

The plot of $\frac{\beta cos\theta}{\lambda}$ and $\frac{Sin\theta}{\lambda}$ gives the microstrain from the slope and grain size from its intercept. The calculated grain size and micro strain is given in the Table 2. It shows that the grain size slightly increases for 2nd and 4th pass which is due to the elongation of the grain as seen in micrograph whereas subsequent passes such as 6th, 8th and 10th, the grain sizes are reasonably reduced which is attributed to the breaking of the elongated grains producing sub-grains. Figure 9 shows the HW-plot for 0th Pass and 10th Pass from this slope we had find the grain size.

Table 2: Calculated Grain Size and Microstrain from Hall Williamson Plot

No. of Passes	Microstrain	Grain size by HW-Plot (A)
0th Pass	0.0239	46.278
2nd Pass	0.0187	43.386
4th Pass	0.018	32.287
6th Pass	1.1519	27.843
8th Pass	1.149	27.645
10th Pass	0.9957	13.883

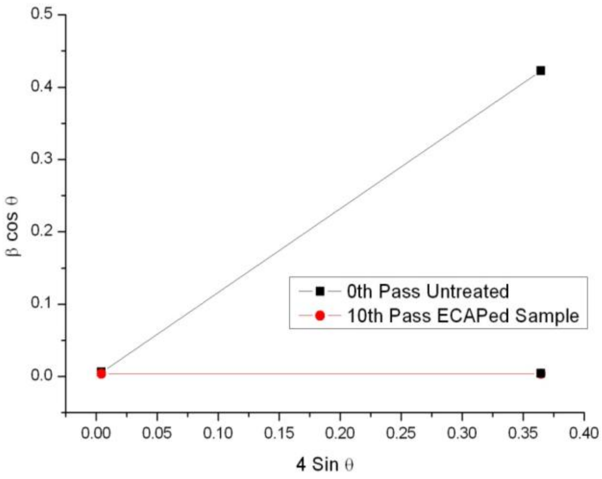

Fig 9 : The HW-Plot for Untreated and 4th Pass ECAP Samples

Micro Hardness

The Vickers microhardness values of pure copper processed for different passes of the ECAP processing are presented in Figure.10 It showed an increase (30%) in microhardness near the contact surfaces of die for the first two passes. It was followed by almost constant value up to sixth pass, then after a slight increase in seventh pass, the microhardness slightly decreased, for subsequent passing. The rapid increase of microhardness at relatively low passes can be attributed to strain hardening as a result of sub- grain boundaries formation rather than grain refinement (Shaarbaf and Toroghinejad, 2008). The grain refinement for additional passes did not cause much hardening. The hardening behavior showed saturation at additional upper passes as was previously reported in UFG materials fabricated by ECAP (Jianqiang et al., 2008). This phenomenon occurs because the materials reach the steady-state density of dislocation. The steady state density of dislocations is determined by a dynamic balance between dislocation generation during plastic deformation, and annihilation in the dynamic recovery processes reducing microhardness slightly. The hardness distribution along the side surface of ECAP'ed specimens shows higher values in the contact surfaces (Fig. 7). Jin et al. (2004) has proposed changes of the shape of the FEM elements. He indicated that the shear deformation, especially at the sheet surface, is not homogeneous throughout the strip thickness. However, close to the center layer of the strip, uniform shear deformation prevails. These are caused by strain large redundant shear strain induced by high friction between roller surface and specimen during the ECAP process. Such an uneven deformation observed from the surfaces is due to the presence of the dead zone located at outer corner of the die.

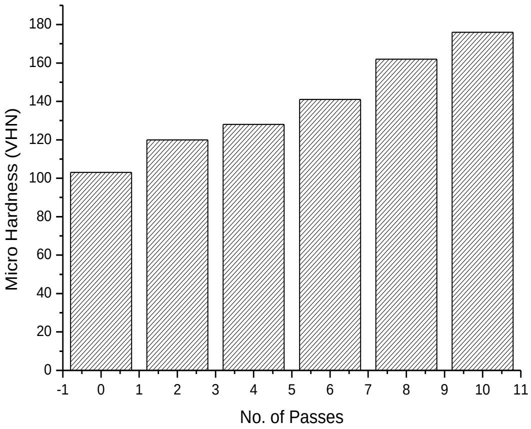

Fig 10 : Variation of Micro Hardness with Number of Passes

Tensile properties

Stress–strain curves in Figure 11 present the results of the room temperature tensile test. As can be seen, after ECAP process, stress–strain curves show higher strength and lower elongation. Fig. 12 shows the variation of yield point, tensile strength and elongation of pure copper in various passes of ECAP process. After the first pass, the yield strength significantly increases about 85%; while tensile strength did not experience much variation, as it increased only by about 6% Figure. 12 Though, the increase in strengths persisted until the 7th pass the increasing rate was lower than that in the first pass. ECAP process causes significant reduction of elongation, about 67%, until 3rd pass. Then this sharp trend changes to slight reduction of elongation until 10th pass. As a whole, Mechanical properties did not change significantly by increasing the number of passes by more than three passes. Table 3 shows the % of elongation and % of reduction after each passes and their corresponding strengths in MPa. Strain hardening decreased with increasing the number of passes and yield and tensile strengths have almost same values in higher number of passes. It is well known that yield strength of metallic materials is proportional to minus square root of grain size (Hall–Petch relation). Hence any variation of grain size leads to a significant change in yield strength of bulk materials. Increase in yield strength and ultimate tensile strength can be justified by strain hardening at initial stages. Large value of plastic deformation can occur in samples by ECAP process. Up to three passes, strain hardening or dislocation strengthening plays a main role in the strength increase. Also, the formation of sub-micron sub-grains or dislocation cells contribute to the strengthening. The dislocation density increases by the ECAP process. Especially in the first pass, the strain hardening plays a pivotal role in increasing the yield strength due to fine deformation bands formed by severe plastic deformation. From fourth pass, the strength increase is due to the evolution of the grain structure and formation of UFGs while strain hardening has less and less effect. The number of UFGs with high-angle grain boundaries increases with increasing ECAP passes up to seven passes so the boundaries with low-angles convert to boundaries with high- angles. Increase in misorientation angles in grain boundaries is the other reason for improved tensile properties. These high-angle boundaries and formation of sub-grains can prevent dislocation movements and cause higher yield and tensile properties. At higher ECAP passes, yield and tensile strengths decrease. These phenomena may be due to two reasons: the first one is the saturation in high-level accumulated strain in bulk material; at this stage, grains contain a large number of dislocations and it is impossible to produce new dislocation in these grains (Asiyeh Habibi et al., 2011).

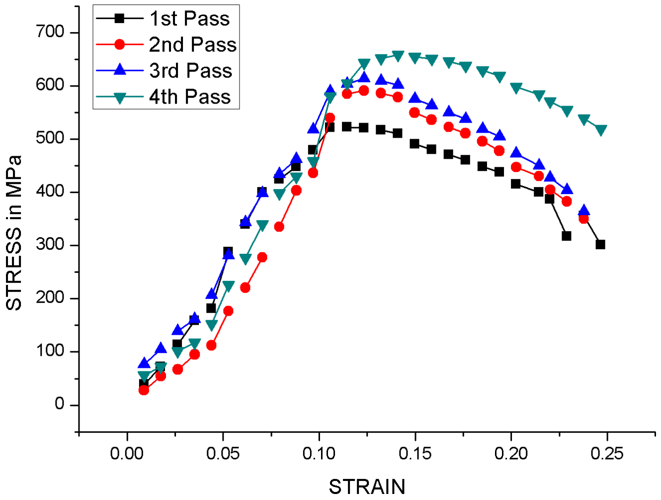

Fig. 11. True Stress-True Strain response of ECAP processed Pure Copper

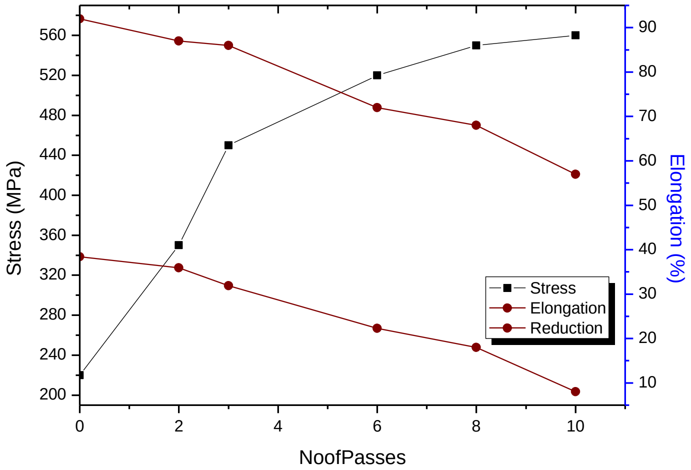

Fig. 12 Commercially Pure Cu Tensile Results after 4, 6, 8 and 10th Passes pressed using 90^O Channel angle and 20^O corner angle

Table 3. Commercially Pure Cu Tensile Results after 2, 4, 6, 8 and 10th Passes using 90^O Channel angle and 20^O corner angle

No. of Passes	**Initial Length**	**Final Length**	**Initial ϕ**	**Final ϕ**	**% elongation**	**% reduction in area**	**Strength in MPa**
Parent	16mm	22.15	4.53	1.28	38.43	92	220
2nd Pass	16 mm	21.75	4.53	1.60	35.93	87	350
4th Pass	13mm	17.15	3.2	1.2	31.9	86	450
6th Pass	13mm	15.9	3.2	1.7	22.3	72	520
8th Pass	13mm	15.3	3.2	1.8	18	68	550
10th Pass	13mm	14	3.2	2.1	8	57	560

Conclusion

1. Commercial purity grade copper was successfully processed upto 10th passes of ECAP at room temperature using a die with a channel angle of 90° and corner angle 20° at a pressing speed of 0.5 mm s-1.
2. The grain size was reduced from 30μm to 14μm in the 2nd pass and in further passes the grain size reduced and sub grains formed. Sub grain size (70 – 200 nm) was confirmed by TEM results. The microstructures of CP-Cu are composed of a mixture of grains with high angle grain boundaries (HAGBs), and low angle grain boundaries (LAGBs) This is suggested by the SAD patterns, which show rings with diffraction spots pointing out the

presence of HAGBS. These also show inclined dislocation walls that are formed by the accumulation of many glissile dislocations giving rise to LAGBs

3. From the XRD peak shift is observed and the grain size and micro strain have been calculated using Hall Williamson's method.
4. The strengthening of the material was homogeneous on both the cross-sectional and longitudinal planes. Increases of ~30% in hardness and ~67% in the ultimate tensile strength after 4 passes of ECAP at the slower speed were obtained. The elongation decreased by ~11% and percentage of reduction in area decreased by about 8% after 10 passes for commercially pure copper.

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