Equal Channel Angular Pressing to Produce Ultrafine Pure Copper with Excellent Electrical and Mechanical Properties

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Abstract:
In this article, commercially pure copper samples were severely deformed by equal channel angular pressing (ECAP) up to eight passes at room temperature. The effects of severe plastic deformation on the microstructure, mechanical properties, and electrical conductivity of the copper were investigated. The microstructure evolution was followed by optical microscope and field emission scanning electron microscope (FE-SEM). FE-SEM shows the extreme evolution of the microstructure after four to eight ECAP passes, in which a large amount of nanoscale and ultrafine grains are observable. The mechanical properties of the pure copper in each pass were studied by compression testing and Brinell hardness method at room temperature. In this respect, hardness and yield stress increased by ~390 MPa and 75HB, respectively, after five-pass ECAP because of finer boundary spacing. The electrical conductivity measurement at room temperature showed that there was no significant change in the conductivity of the processed samples compared with the initial specimen. Hence, by applying ECAP, one can obtain the ultrafine pure copper with sub-micro-meter grain sizes that can improve mechanical properties without impairing the electrical conductivity.
Keywords: *Equal channel angular pressing, Electrical conductivity, Grain refinement, Mechanical properties, Ultrafine grains.*

1. INTRODUCTION

Because of its excellent electrical and thermal conductivities, copper is used for electrical equipment such as electrical connector, vacuum contact interrupter, spot welding electrodes, rotating source neutron targets, and so on. With the rapid growth of both electrical and electronics industries, ultrahigh mechanical strength, as well as high electrical conductivity, of copper is to be improved further to achieve higher efficiency of these devices. Most of the studies have attempted to enhance the essential properties of copper by adding alloy elements such as beryllium, silver, titanium, and niobium [1–4]. However, their electrical conductivities are typically less than 20% International Annealed Copper Standard (IACS) [2–4]. The electrical conductivity of copper alloy is inherently lower than that of the unalloyed counterpart because of: (a) the impurities that increase electron scattering via thermal vibrations of the crystal lattice and (b) presence of structural imperfections, such as grain boundaries and dislocations[5, 6]. Therefore, a new material with a better combination of strength and electrical conductivity should be developed.

To alleviate the problem, ultrafine grains (UFGs) or nanostructure copper must be used without any change in chemical composition. UFG materials with grain sizes of 100nm to 1μmare preferred dowing to their unique physical and mechanical properties with respect to conventional coarse-grained materials [7–10].

Severe plastic deformation (SPD) techniques are considered the most promising route to produce UFG materials [7–10]. SPD techniques provide the capability of achieving a remarkable grain refinement in polycrystalline materials for industrial application, with advantages as porosity-free or contaminants-free and cost-effective [10, 11]. Equal channel angular pressing (ECAP) is one of the most important SPD techniques that can produce ultrafine to nanoscale grains in bulk materials [9–11]. In ECAP, a large amount of simple shear deformation can be imposed on the material via single or multiple passing steps without changing cross-sectional dimensions, resulting in fine-grained structures and enhanced mechanical and physical properties [9–13]. During ECAP, grain refinement occurs by the formation and gradual growth of cell structure [14–17]. As the number of passes increases, the cell size becomes smaller. Most of the grain boundaries are high-angle boundaries, whereas low-angle boundaries are formed inside the grains [14–17]. In ECAP, two mechanisms concurrently increase the strength of the material. The first is dislocation strengthening or strain hardening due to the presence of incidental dislocation boundaries of small misorientation. These boundaries result from some sort of statistical snaring of dislocations [10, 17]. The second is grain boundary strengthening via Hall–Petch relationship due to the formation of geometrically necessary boundaries owing to the differences in the slip system operating in neighboring grains or the local strain difference within each grain, which results in high-angle boundary misorientation at large strain [10, 17].

The aim of this article is to study the effect of grain refinement through each pass of ECAP with variations in the mechanical property and electrical conductivity of pure copper. The most efficient equivalent strain to obtain the best combination of strength and electrical conductivity was determined. Accordingly, commercially pure copper was processed through ECAP up to eight passes and the microstructure, mechanical properties, and electrical responses as a function of ECAP pass number were investigated.

2. EXPERIMENTAL PROCEDURES

2.1. Material

Acommercially pure copper rod was used and the same was processed by cutting and turning operation to achieve accurate size to obtain suitable samples for ECAP, with the dimensions of 16-mm diameter and 65-mm length. The chemical composition of this copper sample is given in Table 1.

2.2. Equal channel angular pressing procedures

The simplest die, a block with two channels that are connected by a 90° angle, was used to perform ECAP. The pure copper samples were pressed through the die with two circular intersecting channels and equal cross sections where the arc of curvature at the outer point was 37°. For these dimensions, the equivalent strain of ~1 was subjected to each sample in each pass. For manufacturing the die, AISI 1.2510 tool steel with a hardness of 40 HRC was used. AISI 1.2080 with a hardness of 50 HRC and AISI 1.1191 with a hardness of 30 HRC were used for manufacturing punch and subpress equipment such as punch holder and clamps, respectively. Samples were subjected to repetitive pressings in ECAP at room temperature according to route B_C [17], which rotates the samples by 90° in the same direction after each pass. Samples were sprayed with soap foam for lubrication and pressed into the ECAP die at a speed of 1 mm/s.

Table 1. *Chemical composition of the studied copper samples*

Elements	Sb	Sn	S	Ca	Cl	Mg	Cu
Weight %	0.007	0.006	0.02	0.02	0.026	0.47	99.44

2.3. Microstructure

Optical microscope and field emission scanning electron microscope (FE-SEM) microstructure the samples sectioned along their axial direction after ECAP. The metallographic samples were polished using 0.05-µm Al_2O_3 and then etched using a solution of 2g $K_2Cr_2O_7$, 4gNaCl, and 8m LH_2SO_4 at room temperature. The average grain size was calculated using linear intercept method.

2.4. Hardness test

The Brinell hardness method was used for hardness testing. Accordingly, samples were cut off usinga wire cutting machine from the top tobottom of the extruded materials. The hardness measurements were taken by Portable Hardness Tester HLN-11A. For each sample, the average of measurements at twelve points was taken as the hardness value. Figure 1 shows the points on which the hardness measurements were carried out.

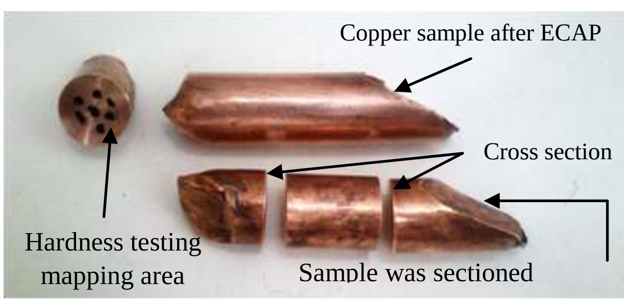

Figure 1. *Transverse section of the ECAP sample and hardness testing mapping areas*

2.5. Axial compression test

The compression test is a convenient method in determining the stress–strain response of materials at large strains ($\varepsilon > 0.5$), because the test is not influenced by the instability of necking that occurs in a tension test [18]. The compression test samples were machined from the materials obtained after ECAP, according to ASTM E-9 [19]. Compression tests were carried out at room temperature with a Zwick 155944 machine (capacity: 60 ton) operating at a constant crosshead displacement of 0.2 mm/s (corresponding to an initial strain rate of $0.02s^{-1}$). A layer of Teflon tape was put on the top and bottom of the sample and a film of lubricating grease was applied on the tape. The machine software aids for a constant displacement rate while loading the sample and, as a result, the true strain rate would increase in compression.

2.6. Electrical conductivity

Electrical conductivity was measured using the portable electrical conductivity tester SMP10 that measures electrical conductivity by using the eddy current method, according to ASTM E1004. Each sample was measured twice and the average was taken as the response (at 20°C temperature and parallel to the extrusion axis).

3. EXPERIMENTAL RESULT AND DISCUSSION

3.1. Microstructure

The optical micrographs of raw material (NON-ECAP) and ECAP samples of three and four passes are shown in Figure 2(a) to (c).

The microstructure of the raw material consists of conventional coarse grains with a mean grain size of 24µm. A large reduction in grain size was obtained in the first four passes through ECAP, as the original grains breakup into bands of sub grains which is due to the high cold work and grain refinements created by SPD of the copper after ECAP. After four passes, the crystalline structure of samples was much refined and the grain size was smaller to measure. Figure 3(a) and (b) shows the FE-SEM micrographs of the copper after four and eight passes, respectively. After four-pass ECAP, the average grain size became smaller, and the UFG obtained was about 360nm. Grains of the copper after eight-pass ECAP were much refined to nanograins of ~50–200 nm in size and show an increase in homogeneity of structure.

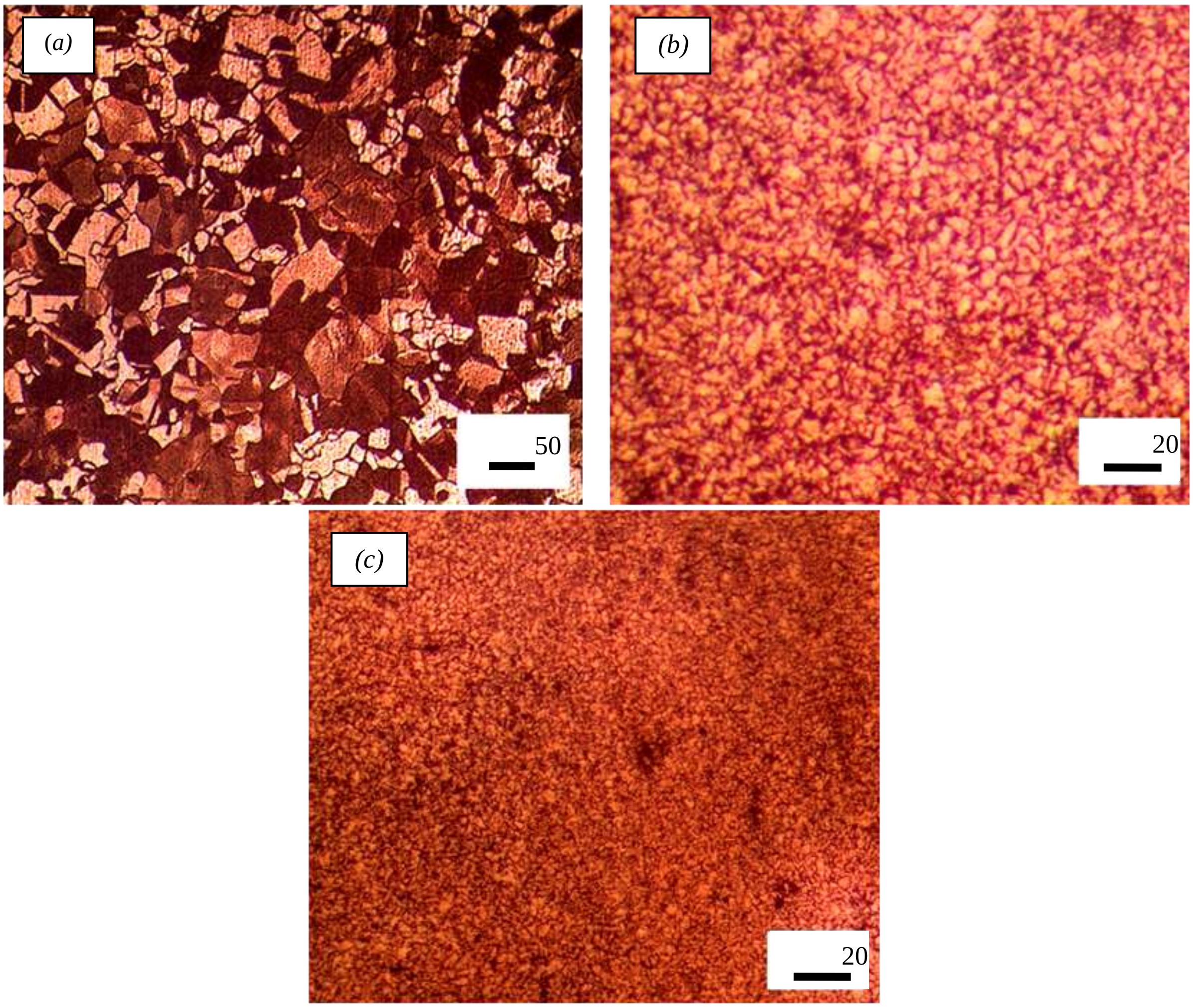

Figure 2. *Optical micrographs of copper samples. (a) NON-ECAP, (b) three-passECAP, and (d) four-pass ECAP*

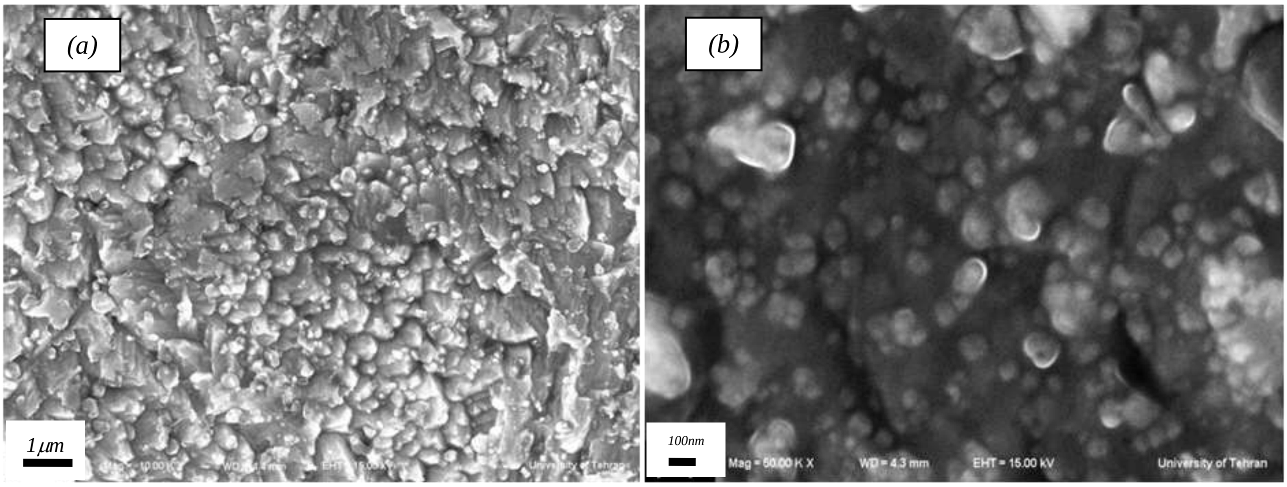

Figure 3. *FE-SEM micrographs of the copper samples produced by (a) four-pass (b) eight-pass ECAP*

3.2. Hardness

The average values of hardness of each sample as a function of ECAP passes are shown in Figure 4 with initial hardness (NON-ECAP) for comparison. As shown, an increased value of hardness will be achieved through ECAP. After five passes, the hardness reaches the maximum value, which is estimated to be approximately 75 HB, and then a gradual decrease in hardness is observed.

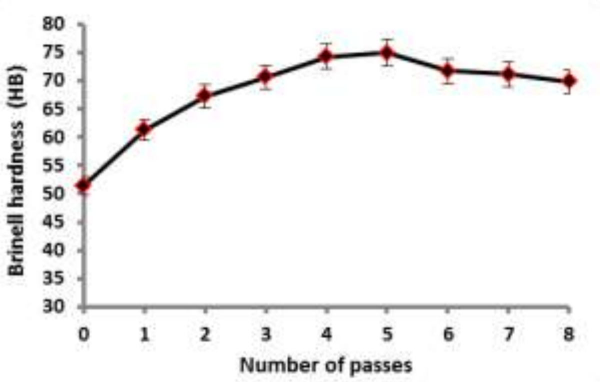

Figure 4. *Variation of hardness as a function of ECAP pass number*

3.3. Compression test

Figure 5 shows the true stress–strain curves obtained from the compression tests of samples after one to four ECAP passes. For comparison, the flow stress–strain curve of NON-ECAP is also plotted. The true stress–strain curves of the samples subjected to four, five, six, and eight passes are shown in Figure 6. Changes of yield stress are also shown in Figure 7 as a function of ECAP pass number. Results show that yield stress of copper until ECAP is 260MPa and after the first pass yield stress increases up to 300 MPa. On increasing number of ECAP passes, the yield stress increased gradually until the fifth pass, and thereafter the yield stress gradually reduced until the eighth pass. Although the increase in strength persisted until the fifth pass, the increasing rate was lower in the first and second passes comparatively. Thus, the increase in material strength up to four equivalent strains was considerable, beyond which their strength became insensitive to further deformation. On the other hand, as shown in stress–strain curves (Figures 5 and 6), after ECAP the work hardening of the material tremendously decreased.

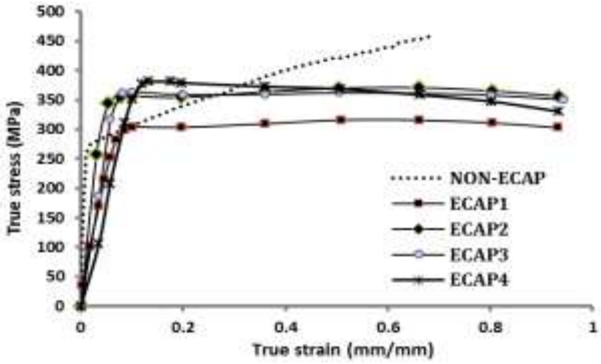

Figure 5. *Compression stress–strain curves of samples subjected toone to four passes and NON-ECAP*

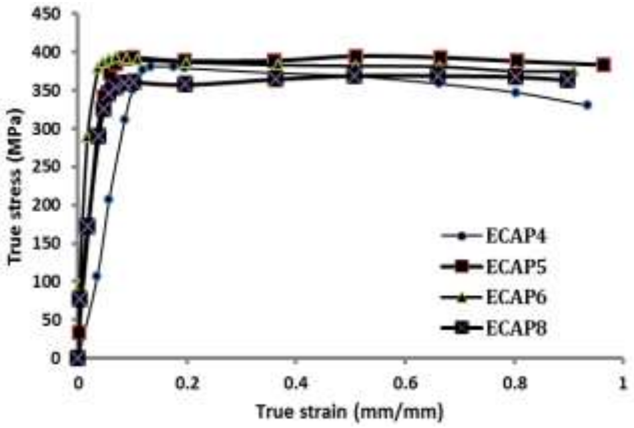

Figure 6. *Compression stress–strain curves of samples subjected to four, five, six, and eightpasses*

For coarse-grained metals, dislocation movement and twining are the primary deformation mechanisms. The strengthening of ECAP samples is generally due to the refinement of grains, formation of twins, and accumulation of dislocations. Using transmission electron microscopy (TEM), Dalla Torro et al. [20]measured the dislocation density, that was 1.6×10^{14} m^{-2} within cells and 1.5×10^{15} m^{-2}within the cell walls of coarse-grained copper after one pass of ECAP. Furthermore, according to Zhu et al.[9], the dislocation density of the pure copper after eight-pass ECAP with a strength of 368MPa was approximately 5×10^{15} m^{-2} (estimated from the high-resolution TEM images).

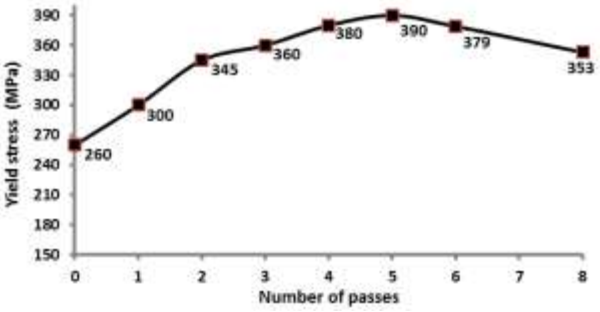

Figure 7*. Variation of yield stress as a function of ECAP pass number*

Increased yield strength and hardness can be justified by strain hardening at initial stages. Specifically from the first pass to the fourth passes, the strain hardening plays a pivotal role in yield strength, increasing as fine deformation bands formed by SPD. The number of UFGs increases with increasing ECAP passes up to four passes. Thereupon, at higher ECAP passes the strength increased mostly owing to the altered microstructure and grains that were refined to ultrafine or nanoscale range. On the other hand, with the formation of ultrafine and nanoscale grains, the strain hardening has a little effect. Ultrafine grains with high-angle boundaries impede the motion of dislocations[21]. Yield strength and hardness values decrease gradually from fifth to eighth passes because of softening phenomena. After four passes, the contribution of dislocation density to strengthening decreases, whereas the contribution of grain boundaries almost remains constant. Therefore, a decrease in yield strength and hardness was observed.

It has been observed that when metals are subjected to SPD at ambient temperature softening behavior, which is due to dynamic recovery [22, 23], dynamic recrystallization [14, 24, 25], and high-angle grain boundaries' formation [26],will occur.

3.4. Electrical conductivity

The relative electrical conductivity (relative to IACS) of commercially pure copper is shown in Figure 8 as a function of ECAP pass numbers. As shown, the conductivity of samples decreased concurrently with an increase in the number of ECAP passes. The conductivity of the material is only reduced by approximately 8% IACS after five

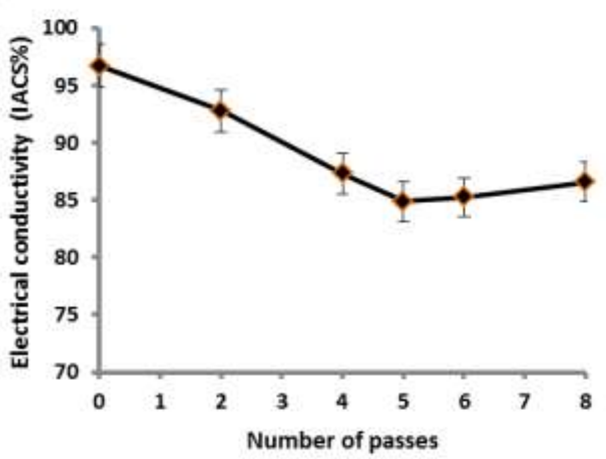

Figure 8. *Variation of relative electrical conductivity as a function of ECAP pass number*

passes and then slightly increased.

Decreasing the values of electrical conductivity can be justified by the creation of more grain boundaries owing to breakage of the initial coarse grains. Creation of numerous grain boundaries and increased dislocation density in the UFG structure shorten the free movement path of electrons. Thus, the electrical conductivity of samples had been decreasing up to five passes. After which, the electrical conductivity of samples slightly increased because of softening phenomena and reduction in point defects. The increase could be accelerated by the higher values of temperature rise in the final passes compared with the initial passes.

Similar results were observed by Hosseini and Daneshmanesh [27] in UFG copper produced by accumulative roll bonding (ARB). They found that the electrical conductivity of commercially pure copper decreased concurrently with increasing ARB cycles up to six cycles and then up to eight cycles. This rise in electrical conductivity is due to the elimination of point defects because of the dynamic recovery that happened.

Although ECAP significantly improves mechanical properties, the electrical conductivity decreased with respect to increasing ECAP passes. However, their values remain quite high and enough for many electrical applications. Figure 9 shows the relationship between yield strength and electrical conductivity of copper produced by ECAP and NON-ECAP.

The relationships between yield strength and electrical conductivity of different Cu–Zn alloys with different chemical compositions [28] are also shown for comparison. As shown in Figure 9, with increasing Zn metal in Cu–Zn alloy, the mechanical strength increases concurrently but the electrical conductivity reduces drastically. It can be concluded that the grain refinement of copper by ECAP improves its mechanical properties without impairing the electrical conductivity.

Thus, ECAP is an effective method for producing pure copper with high strength and high electrical conductivity.

4. CONCLUSIONS

In this article, commercially pure copper samples were severely deformed by ECAP for up to eight passes in room temperature. The effects of SPD on the microstructure, mechanical properties, electrical conductivity, and electrical wear resistance of copper were investigated. The main results are summarized as follows.

The grain size of the ECAP-passed copper is greatly reduced to an ultrafine and nano-grain-sized structure.

UFG microstructures were formed in commercially pure copper after four-pass ECAP and the number of nanoscale grains increased concurrently with increasing the number of passes up to the eighth pass.

High hardness and yield stress was achieved in pure copper by ECAP. The hardness and yield stress of copper reached their maximum approximately 75 HB and 390 MPa after five passes and decreased from the fifth to the eighth pass. Thus, the relevant mechanical properties can be achieved using ECAP only up to four passes. On the other hand, the work hardening of copper greatly reduced after ECAP. Although the strength of the samples are increased using ECAP, their electrical conductivity is decreased up to five passes, and it was estimated to be 84.87% IACS. After these five passes, electrical conductivity had been increasing slightly until the eighth pass. In addition, the electrical conductivity of the ECAP-passed copper is higher than that of its alloyed counterpart with equal strength. Thus, high-strength, high-conductivity commercially pure copper can be obtained. According to the results, the four-pass ECAP is an effective method to produce commercially pure copper with high strength and high conductivity.

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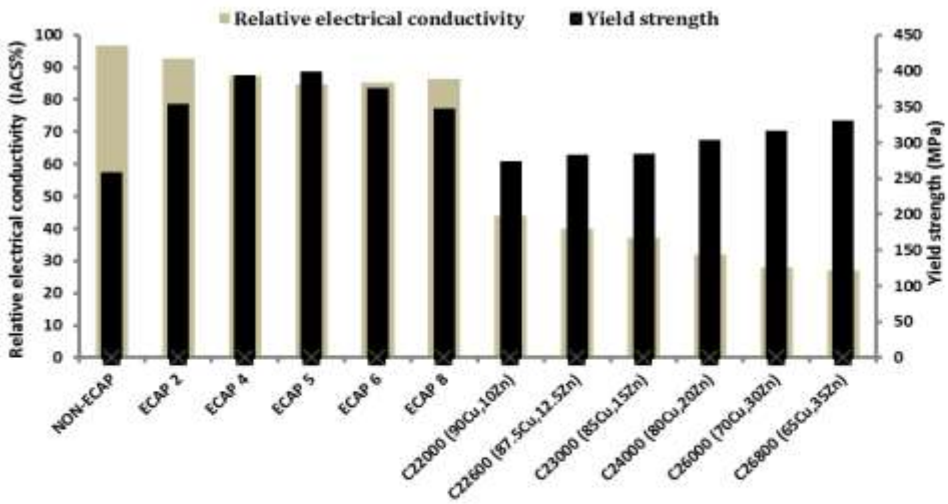

Figure 9. *Comparison of electrical conductivity and yield stress of pure copper samples subjected to two-, four-, five-, six-, and eight-pass ECAP, Non-ECAP, and Cu–Zn alloys [28] with different chemical compositions*

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