Communication

Enhancing the Electrical Conductivity of Electrolytic Tough Pitch Copper Rods Processed by Incremental Equal Channel Angular Pressing

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Electrolytic tough pitch copper rods were processed by Incremental Equal Channel Angular Pressing and subjected to short-term annealing. Conductivity of 94 pct IACS without significant changes in the material's microstructure and mechanical properties, up to 130 HV0.2, was achieved, which gives a very good ratio of electrical conductivity to strength in comparison with other processing methods. The proposed method offers a solution for manufacturing rods of significant sizes with good mechanical strength and electrical conductivity.

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The use of Electrolytic Tough Pitch (ETP) copper as a structural component is limited because, although it has advantageous electrical and thermal conductivity, its mechanical strength is poor. The strengthening mechanisms available (that do not involve a change in chemical composition) include strain hardening and grain size refinement. Both of these can be achieved by means of Severe Plastic Deformation (SPD), which, for low applied strains, generates a high density of dislocations that for higher strains transform into an array of high-angle grain boundaries. It has been demonstrated for pure copper that such processing improves mechanical strength without excessively reducing electrical conductivity.[1–4] Typically, SPD-processed copper exhibit a microhardness (strength) of 130 HV and an electrical conductivity of 88 pct IACS. To further enhance the combination of high mechanical strength and high electrical conductivity, various strategies have been proposed, including deep cryogenic treatment,[5] post-processing annealing,[3] aging,[6] and the introduction of nano-scale twins.[7] The best combination of strength (YS = 610 MPa) and electrical conductivity (95 pct IACS) obtained by means of SPD processing was reported for copper acquired by dynamic plastic deformation (DPD),[8] whereas the highest values ever reported in the literature, achieved in a copper film produced by electrodeposition, were 900 MPa and 97 pct IACS.[7]

However, most SPD methods, including both DPD and Equal Channel Angular Pressing (ECAP) in its basic form, are capable of producing only limited quantities of ultrafine-grained (UFG) material, limiting the applicability of SPD-processed copper in structural applications. In this paper, we propose a continuous method, namely Incremental ECAP (I-ECAP),[9] for processing virtually infinite copper rods. In this process, friction is substantially reduced by developing the plastic shear in small increments. Therefore, products can be manufactured that have a shape suitable for structural components. The effectiveness of the method in terms of grain refinement has been demonstrated for aluminum,[10] magnesium,[11] and titanium[12] bars, as well as for pure aluminum sheets.[13, 14]

Generally, copper is easily strengthened by plastic deformation as imposing a strain of only 1 results in an improvement in microhardness by as much as 200 pct. Further processing does not significantly strengthen the material, but results in a steady state[3] and introduces changes in the microstructure. Depending on the strain imposed, different types of microstructure can be created that govern the material's mechanical and electrical properties. These microstructures can be further modified by short-term annealing. The aim of this work is to propose the most suitable combination of thermal and mechanical processing to tailor the microstructure of ETP copper and achieve a good balance between its electrical conductivity and mechanical strength.

The material studied was ETP copper in the form of rods, which were machined to a square cross section with sides 10 and 200 mm long and processed by I-ECAP at room temperature up to 8 passes with a rotation of 90 deg in one direction after each pass.

The rods after 1 and 8 passes, further referred to as 1p and 8p, respectively, were subjected to short-term annealing at 50, 100, 150, and 200 °C for 10 minutes and evaluated in terms of their electrical conductivity and microhardness. A microstructural evaluation was performed for selected samples using the EBSD technique on a Hitachi SU70 and a TEM Jeol 1200 with acceleration voltage of 120 kV. The mechanical strength

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was evaluated by a standard Vickers microhardness test under a load of 200 g, which was carried out in the Y plane, *i.e.*, parallel to the pressing direction. The electrical conductivity was investigated using the standard four-point method.

The initial coarse grains of an average size $d = 28 \pm 16.70$ μm (Figure 1(a)) were reduced to 1.41 ± 1.33 μm after 1 I-ECAP pass, and a lamellar microstructure is created with a fraction of high-angle grain boundaries (HAGBs) equal to 27 pct. After annealing at 100 °C and 150 °C, the grain size and HAGB fraction changed, reaching 1.64 ± 2.24 μm and 15 pct and 1.73 ± 4.42 μm and 33 pct, respectively. Sample 8p is characterized by equiaxed grains of $d = 0.57 \pm 0.20$ μm with 67 pct HAGBs, as listed in Table I, which changed to 0.90 ± 0.20 μm and 61 pct, and 0.92 ± 0.23 μm and 54 pct, after annealing at 100 °C and 150 °C, respectively. Samples 1p and 8p also differ in their dislocation substructure, as illustrated in Figures 1(b) and (c). In the former, dislocations are present inside the grains and near grain boundaries (GBs), whereas in the latter, there are well-developed grains with few dislocations inside.

Table 1. Change in Grain Size and HAGB Fraction in Samples 1p and 8p in As-Processed Condition and After Annealing at 100 °C and 150 °C for 10 min

Y Plane	$d \pm \sigma$ [μm]	Pct HAGBs
Initial	27.63 ± 16.70	91
1 Pass	1.41 ± 1.33	27
1 Pass + A100 °C	1.64 ± 2.24	15
1 Pass + A150 °C	1.73 ± 4.42	33
8 Passes	0.57 ± 0.20	67
8 Passes + A100 °C	0.90 ± 0.20	61
8 Passes + A150 °C	0.92 ± 0.23	54

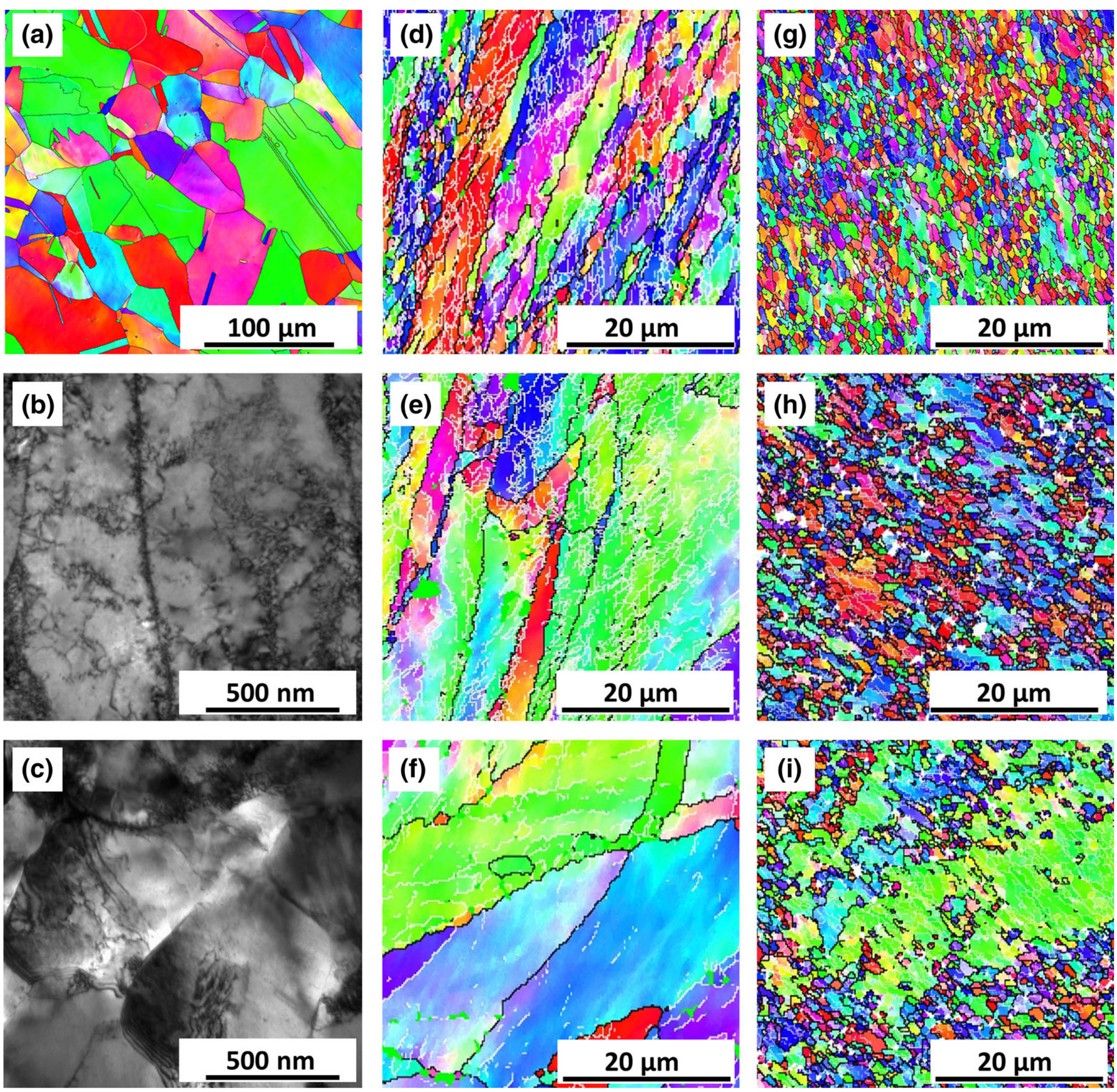

Fig. 1—EBSD map of the initial material (*a*); TEM photograph of sample 1p (*b*) and 8p (*c*); EBSD map of a sample after 1 pass (*d*) and annealed at 100 °C (*e*) and 150 °C (*f*); after 8 passes (*g*) and annealed at 100 °C (*h*) and 150 °C (*i*).

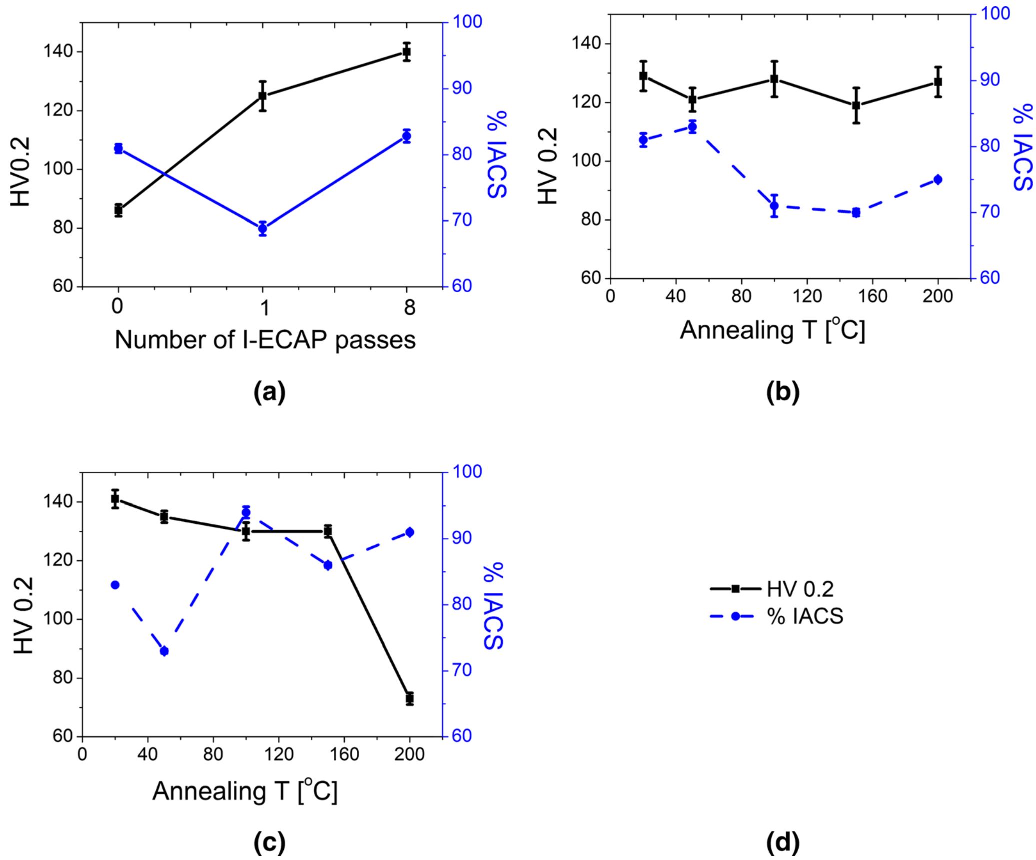

Fig. 2—Change in microhardness and conductivity with the number of I-ECAP passes (*a*); rod after 1 pass (*b*) and 8 I-ECAP passes (*c*) with annealing; (*d*) legend.

Figure 2(a) shows electrical conductivity and microhardness as a function of the number of I-ECAP passes, and Figures 2(b) and (c) as a function of annealing temperature. The initial microhardness value of 87 HV0.2 increased to 125 and 140 HV0.2 for samples 1p and 8p, respectively. Thermal processing had a stronger effect on the microhardness of sample 8p, whose value decreased by around 10 HV0.2 for annealing up to 150 °C and was further reduced down to 73 HV0.2 after annealing at 200 °C. The microhardness of sample 1p fluctuated around 125 HV0.2 regardless of the annealing temperature.

One can observe that 1 pass of I-ECAP decreased electrical conductivity from 81 to 69 pct IACS, while after 8 passes of I-ECAP that value recovered to 83 pct IACS. The annealing of sample 1p resulted in a reduction in electrical conductivity, whereas the annealing of sample 8p increased it to a value above that achieved directly after SPD processing.

Our studies showed that the mechanical properties can be enhanced after even just 1 I-ECAP pass, and that short-term annealing did not cause a serious deterioration in the microhardness in the investigated samples. In sample 8p, a significant impact of heat treatment is noticeable only after the recrystallization temperature is reached, manifest as a sharp drop in the microhardness value observed between the temperatures of 150 °C and 200 °C. This indicates a faster onset of recrystallization than in the 1p sample, where the microhardness did not change after annealing at 200 °C. This phenomenon is attributed to the presence of many recrystallization sites due to the strain path, *i.e.*, places where the shearing planes intersect.[15] Nevertheless, there is a substantial difference in electrical conductivity in samples 1p and 8p, which can be explained based on their microstructural features. After the first pass, a lamellar microstructure was created, with subgrains (marked as white lines in Figure 1(d)) inside the elongated grains and with a dominant fraction of LAGBs. Due to the imposed strain, a substantial amount of dislocations inside the grains were created, as visible in the TEM micrograph in Figure 1(b); the strain deformed the lattice and suppressed the flow of electrons. The thermal treatment of sample 1p caused an even further deterioration of conductivity. After annealing at 100 °C, the fraction of HABGs was reduced to 15 pct, as even more LAGBs were created (see Figure 1(e)); the grain size increased slightly. The annealing at 150 °C resulted in grain growth up to a size of 1.73 ± 4.42 μm and an increase in the fraction of HAGBs to 33 pct. The microstructure obtained, depicted in Figure 1(f), is also characterized by an array of LAGBs inside the grains. Similar observations were reported in References 16 and 17, where it was shown that imposing a limited strain suppresses electrical conductivity, which can, however, be restored by introducing a higher strain. During the

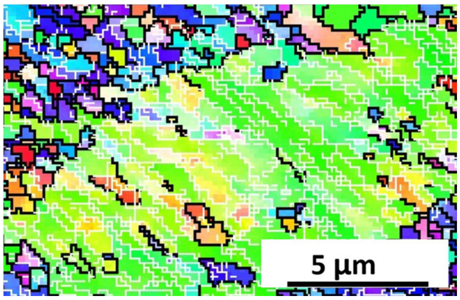

Fig. 3—LAGBs in the sample 8p annealed at 150 °C.

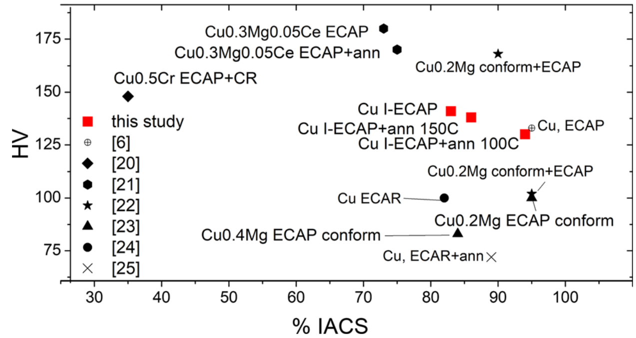

Fig. 4—Comparison of the results obtained in this study and others.

processing up to 8 passes, the dislocations rearrange and annihilate creating new grains.[18] In sample 8p, well-developed grains with a majority of HAGBs were created (Figures 1(c) and (g)) where, due to the reduced amount of dislocations, electrons were able to flow more freely. As indicated in Reference 19, annealing changes the state of the HAGBs from non-equilibrium to more equilibrium as the dislocations present in the GBs are annihilated, which enhances electrical conductivity. Annealing conditions at 100 °C and 10 minutes cause a decrease in the density of the grain boundary dislocations without excessive grain growth (Figure 1(h)), which is confirmed by the change in grain size from 0.57 to 0.90 μm and the change in pct HAGBs from 67 to 61. The high fraction of HAGBs is preserved, and so high mechanical strength remains, but as the GB state changes to more equilibrium, the electron flow is not restricted. Whereas at 150 °C, the fraction of HAGBs drops to 54 pct and conductivity decreases because, during this heat treatment, relatively large grains are created having LABGs inside, as can be observed in Figure 1(i) on the grains colored in green and in Figure 3, which is a close-up of Figure 1(i). This causes a reduction in electrical conductivity.

In Figure 4, microhardness as a function of electrical conductivity is shown for pure and alloyed copper processed by either continuous ECAP-based processes or conventional ECAP. It is not possible to achieve both high microhardness and high electrical conductivity, and therefore the right balance between them must be found. The values achieved in the present study seem to provide a reasonable compromise. Higher microhardness than that achieved in this study is featured by copper alloys that have lower electrical conductivity,[20–22] whereas larger electrical conductivity is achieved in samples having lower mechanical properties.[2, 23–25] The only other study that reported results similar to the present one is,[6] which, however, concerned pure copper processed by conventional ECAP, implying a limited amount of processed material.

The results indicate that I-ECAP processing followed by subsequent annealing result in a good balance between electrical conductivity and mechanical strength in rods of considerable size, which offers a possible solution for manufacture structural conductive copper rods. A significant enhancement of mechanical strength is possible even after just 1 pass, although acquiring both good electrical conductivity and mechanical strength requires further processing. By implementing 8 passes of I-ECAP followed by short-term annealing at 100 °C for 10 minutes, it is possible to achieve an electrical conductivity of 94 pct IACS and a microhardness of 130 HV0.2.

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