Enhanced tensile ductility and toughness in nanostructured Cu

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Pure copper with ultrafine grain sizes and nanoscale subgrain (dislocation) structures was prepared by using severe plastic deformation through cold rolling at subambient temperatures, with or without subsequent recovery annealing. We report coexisting high strength and tensile ductility (large elongation to failure and ductile fracture). Factors leading to the simultaneous strengthening and toughening with increasing cold deformation and microstructural refinement are discussed. © *2002 American Institute of Physics.* [DOI: 10.1063/1.1465528]

An enhancement in both the strength and ductility has been predicted for nanocrystalline (nc) materials (grain sizes $<\sim 100$ nm) based on extrapolations of the grain size dependence of mechanical properties of conventional materials.[1] While the promised gain in strength has indeed been realized in nc and ultrafine-grained (UFG) (grain sizes $<\sim 300$ nm) metals, which are typically 5–10 times stronger than their coarse-grained (CG) counterparts,[1–3] it is disappointing that almost all of the nc metals studied to date exhibit a RT tensile elongation to failure of no more than a few percent.[3] This is true even for nc metals such as Cu that are very ductile in the CG form (tensile elongation to failure $>60\%$) or under compression.

One factor reducing the uniform tensile elongation, as discussed in our previous letter,[4] is the propensity for plastic instability (necking) in the early stage of plastic deformation due to the diminishing strain hardening capacity and minimal strain rate sensitivity frequently observed for nc and UFG metals. On the other hand, the refined grain size could potentially also be helpful in enhancing the ductility, as pointed out by Koch *et al.* in a discussion of the fracture criteria as applied to the analysis of ductility in nc and UFG metals.[3] This possibility is based on our experience with some conventional materials where the fracture stress may increase faster than the yield stress with decreasing grain size such that ductile/brittle transitions can occur. One may expect an enhancement in ductility if the small grain sizes lead to reduced flaw sizes and increased difficulties for the imposed stress concentration at the flaw to exceed the critical toughness of the material, thus suppressing the crack nucleation/propagation instability.

Whether and in which metal this actually happens upon the extreme grain size reduction to the nc and UFG regime is unknown. Given the strong increase in flow stress, the competition between plastic flow and fracture is difficult to predict.[3] The low tensile ductility observed in experiments so far may not be an intrinsic property of nc and UFG metals, but is most definitely related to premature catastrophic failure caused by excessive flaws.[5,6] Such artifacts as porosity, poor interparticle bonding, impurities, and high internal stresses are often impossible to eliminate in processing nc metals.

Therefore, for a meaningful evaluation of the ductility in nc and UFG metals and its dependence on grain size, sufficiently large and porosity-free samples are required. We hence resorted to the bulk processing route of severe plastic deformation,[7,8] plus suitable low-temperature recovery annealing, for sample preparation. Uniaxial tensile tests are necessary for assessing ductility, because in compression tests ductile failure instabilities and brittle crack propagation may be inhibited or much more difficult.[3] Our results demonstrate that both the tensile strength and ductility can be enhanced to high levels simultaneously in Cu by modifying the microstructure in the nc and UFG regime.

A commercial high purity Cu (99.99%) 10 mm in diameter was processed using the well known equal channel angular pressing (ECAP) method.[7–9] Figure 1(a) displays the transmission electron microscopy (TEM) micrograph of the transverse cross section of the Cu sample after eight passes (route B_c). Our ECAP Cu reached an average grain size of the order of 300 nm.[9] Some grains appear elongated and the microstructure is somewhat inhomogeneous.[9] The selected area diffraction (SAD) pattern shows nonuniform and discontinuous rings, suggesting that the majority of the grain boundaries are of fairly low angles,[8,9] which is also confirmed by TEM tilting experiments. Samples from the ECAP Cu were cut and polished to a 1 mm$\times$1.6 mm cross section, and a gauge length of 7.5 mm, for tensile tests at an initial quasi-static strain rate of 1×10^{-4} s^{-1} (the same parameters were used for all other tensile test samples in this work). The engineering stress-strain curve is shown in Fig. 2. The ECAP Cu has a strength (σ) and an elongation to failure ($\varepsilon\%$) larger than, or at least comparable with, those of Cu heavily cold rolled at RT (typically σ is in the range of 300–400 MPa and $\varepsilon\%$ is a few percent or less).[10] Compared with ECAP Cu, RT rolled Cu is known to be even more dominated by elongated grains with low-angle subgrain structures (dislocation cell blocks).[11] Figure 2 also shows that necking sets in rather early as expected from the small strain hardening capability (due to fast dynamic recovery in the small grains) at RT and

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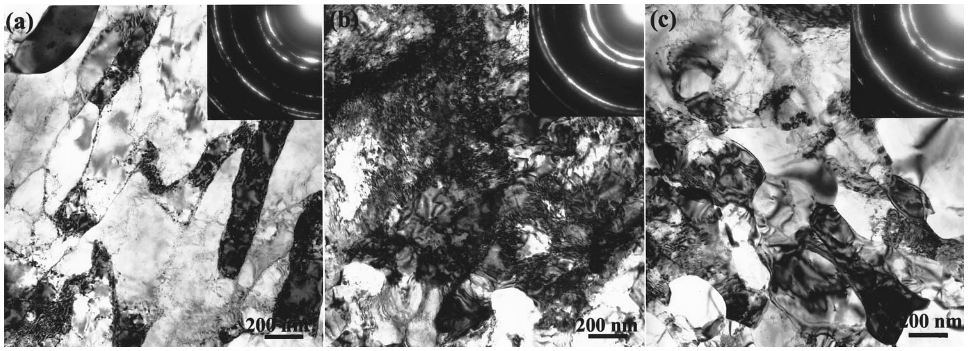

FIG. 1. TEM micrographs and corresponding SAD patterns of (a) as-received ECAP Cu, (b) after additional LNT rolling to 1340%, and (c) the sample in (b) after annealing at 100 °C for 2 h.

minimal strain rate sensitivity known for Cu of such grain sizes.[8,12]

To further refine the microstructure, we subjected the ECAP Cu to additional plastic deformation by cold rolling at the liquid nitrogen temperature (LNT). The use of the low temperature is necessary to increase the effectiveness of the process: due to the dynamic recovery that opposes the accumulation of dislocations and grain boundaries, the steady-state grain size achievable by RT deformation is very limited. As a result, rolling at RT, which is not a deformation route that can efficiently introduce very large strains, was unable to refine the grain size to levels beyond what was achieved by ECAP. We therefore cooled the sample to LNT between consecutive rolling passes. Figure 1 (b) shows the microstructure after rolling to an accumulative cross sectional area reduction of 1340%. The high densities of dislocations in nanoscale networks are typical of such heavily deformed microstructure.[13] The grain sizes is refined to $\sim$200 nm (see the grains resolved in the bottom portion of the micrograph). After a low-temperature anneal at 100 °C for 2 h, the dislocation density is reduced and the microstructure consists of much better defined grains with an estimated average size of 190 nm, as shown in Fig. 2(c). Such a brief anneal was shown before for heavily cold-rolled or ECAP Cu to lead to the rearrangement of dislocations, more regular grain structures, and an associated decrease in internal stresses, without causing grain growth.[9,10] A comparison with the micrograph in Fig. 1(a) shows that the grain size is obviously reduced. The grains are more equiaxed and uniform in size. Also, the more distinct SAD ring pattern is suggestive of a higher population of boundaries separated by large angles of misorientations. Numerous grain boundaries and subgrain boundaries remain small angle in nature, which can be easily revealed upon tilting in TEM.

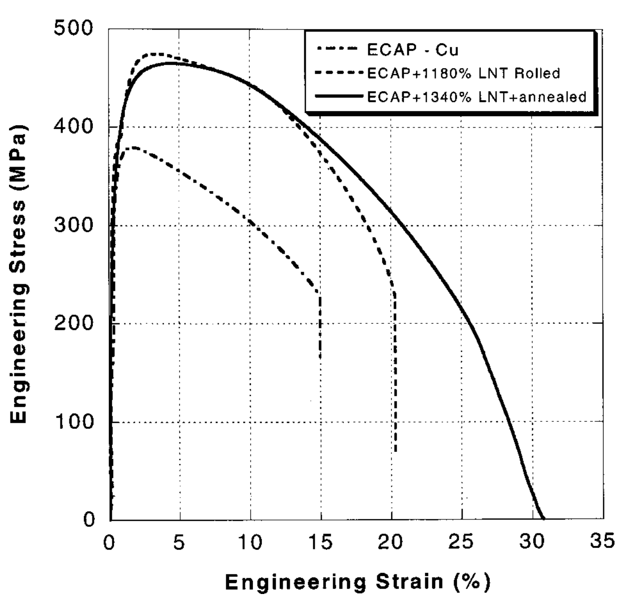

FIG. 2. Tensile engineering stress-strain curves for three Cu samples: as-received ECAP Cu, after additional LNT rolling to 1180%, and after LNT rolling to 1340% plus annealing at 100 °C for 2 h.

After LNT rolling to 1180%, a significant increase in tensile strength over the ECAP Cu is observed, Fig. 2. Interestingly, the tensile ε% also increased significantly rather than decreased. The result after increasing the rolling to 1340% is similar. After the additional recovery annealing that led to a relief of internal stresses, ε% increased further. A part of this increase is due to the increase in uniform deformation (compare peak positions in Fig. 2) because some strain hardening capacity is regained after annealing, delaying necking. The strength decreased only slightly. Figure 2 demonstrates an obvious concurrent strengthening and toughening (enhanced ε% and area under the curve) upon modifying the grain size/dislocation structures. To establish the reproducibility of this observation, four additional samples from the same ECAP Cu were LNT rolled to similar strain levels. In all cases, strength and ductility simultaneously increased, consistent with those seen in Fig. 2.

Figure 3 shows scanning electron microscopy (SEM) micrographs of the fracture surfaces after the tensile tests. The as-received ECAP Cu fractured with some ductile features similar to that known for Cu heavily cold-rolled at RT.[10] The Cu with additional LNT processing, in comparison, appears to be more ductile, showing fracture features dominated by microvoid formation on a much finer scale.

Several factors contribute to the improved ductility and toughness (area under the stress-strain curve). With increasing plastic deformation we observe increasingly more dislocations, subgrain boundaries, and small- and high-angle grain boundaries. The refined grain/dislocation structures reduce the size of the nucleating flaws and increase the resistance to crack propagation, leading to a higher fracture stress. They do so by offering a higher resistance to shear localization and shear fracture and stabilizing the hydrostatic triaxial stress state that promotes ductile fracture through mi-

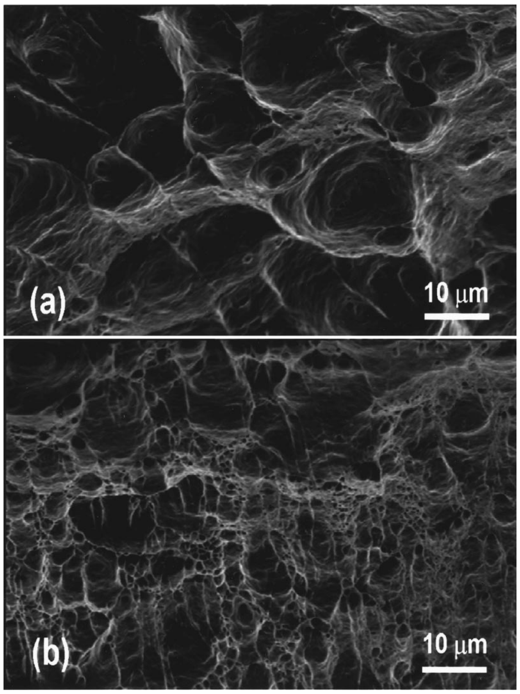

FIG. 3. SEM micrographs of the fracture surfaces of (a) as-received ECAP Cu and (b) after additional LNT rolling to 1340% plus annealing at 100° C for 2 h (the features before annealing are similar).

crovoid nucleation and coalescence. The as-received ECAP Cu fractured after a cross-sectional area reduction of 70%. The LNT rolled sample, on the other hand, fractured when only 10% of the original cross-sectional area was left, clearly indicating a higher fracture stress. In addition, the dislocations observed to accumulate at the nonequilibrium boundaries (high magnification TEM micrographs not shown), some of which are not geometrically necessary, may move to contribute to plastic deformation. The boundaries may actually act as sources of dislocations.[14] It has in fact been proposed that different stress-strain curves are possible even for a given nominal grain size, so long as the nonequilibrium grain boundaries are of different nature.[16] As some grain boundaries gradually increase their angles of misorientations,[15,16] they may even facilitate deformation through grain boundary mechanisms such as grain rotation or grain boundary sliding.[8,17] Consistent with this notion and with the slowing down of necking, we observed an increase in the strain rate sensitivity of the flow stress, m, from 0.015 for the ECAP Cu to 0.03 after LNT processing, in compression tests (not shown). Finally, LNT rolling led to {110} texture, similar to the cases reported for several other processing routes.[9,13,18] However, the rolling texture alone cannot explain the enhanced ductility, as the conventional Cu heavily cold rolled at RT only shows less ε% than the ECAP Cu.[10] Our tensile tests using different in-plane orientations yielded similar results. A detailed study of all the earlier parameters is beyond the scope of this letter.

In summary, we report that by refining the grains and modifying the boundary/dislocation structures in the UFG and nc range, a combination of a high strength of ~500 MPa and a large nominal tensile elongation of ~30% can be achieved in pure Cu. The Hall–Petch-type strengthening is accompanied by increased tensile ε% and enhanced ductile fracture features. Such a concurrent enhancement in both the strength and toughness with decreasing grain size is consistent with an extrapolation of the trend for many materials with conventional grain sizes.[22] As the majority of the elongation is post necking and thus inhomogeneous deformation, our samples would have exhibited an even larger ε% if we used a smaller gauge length such as those typically used for UFG and nc metals (1–5 mm).[19,20] The only published report of an impressive tensile ductility of similar magnitude (30%) was for an electrodeposited Cu.[21] However, this ''nanostructured Cu,''[17,20] different from all typical nc metals, had nanoscale subgrain mosaics with very low angle boundaries but otherwise micrometer-sized grains and consequently a low yield strength of only $\lesssim$100 MPA. Progress in our additional efforts to increase strain hardening so as to further improve the more useful uniform elongation will be reported in a forthcoming publication.

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