Grain size dependence of tensile properties in ultrafine-grained Cu with nanoscale twins

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Received 17 June 2010; revised 6 September 2010; accepted 12 October 2010
Available online 18 October 2010

We investigate the grain size dependence of tensile behaviors of nanotwinned Cu with fixed twin thickness. With an increased grain size, ductility and work hardening of nanotwinned Cu are effectively promoted, but strength is not sacrificed to any notable degree. This may be attributed to the highly anisotropic plastic deformation of nanoscale twins.

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Keywords: Coherent twin boundary; Grain size dependence; Strengthening; Ductility; Work hardening

The strength, ductility, work hardening and other mechanical characteristics of metals and alloys are strongly dependent on their grain sizes [1,2]. It has been recognized that the strength of a polycrystalline material can be effectively elevated by refining its grain size following the empirical Hall–Petch (H–P) relationship [3,4]. Extremely high strength and hardness have been observed in nanocrystalline (nc) metals with average grain size below 100 nm [5,6]. However, nc metals are very brittle, and have usually failed permanently when elongated only a few percent in tensile tests, even those that are very ductile coarse-grained (CG) metals [5–7].

Recently, we synthesized an ultrafine-grained copper with a high density of nanoscale growth twins embedded in individual grains via a pulsed electrodeposition technique [8–11]. Systematic studies show that twin lamellar thickness (λ) has a significant effect on the strength, ductility and work hardening of nanotwinned Cu (nt-Cu). For example, the yield strength of such nt-Cu first increases as λ decreases [9,11], reaching a maximal strength of 900 MPa at λ ≈ 15 nm [12]. Interestingly, a pronounced increment in tensile uniform ductility and work hardening is observed in nt-Cu with monotonically decreasing λ [12]. The twin refinement-induced increases in ductility and work hardening are the inverse of the general trend in ultrafine-grained and nc materials, where ductility and work hardening continuously decrease with decreasing grain size, d [5]. The trend of increasing strength in nt-Cu with decreasing λ can be explained by the H–P relationship because the twin boundaries (TBs) serve as effective barriers to dislocation gliding on an inclined slip plane, like grain boundaries (GBs) [8,11,12]. The superior uniform elongation of nt-Cu is due to the fact that TBs are much more hardenable and afford a huge room for dislocation storage when they gradually lose coherency during deformation.

Since the twin lamellae are embedded within equiaxed ultrafine grains in the nt-Cu samples, the twin structure inherently has two microstructural dimensions: the dimension in the direction perpendicular to the TBs and the dimension parallel to them. The former dimension is generally known as the twin/matrix lamellar thickness, and the latter one corresponds to the grain size. Although there are many studies about the effect of the twin lamellar thickness on the mechanical properties, the dependence of the mechanical properties and the deformation mechanism of nanotwinned metals on grain size have rarely been reported. Understanding the dependence of the deformation of nt-metals and its mechanism on the grain size, with the aim of improving the metal’s strength/plasticity, are of central interest with regard to the microstructure design of engineered alloys wherein twinning is purposely orchestrated at
that the average grain sizes, Cu specimens consisted of roughly equiaxed grains with morphologies of the two as-deposited nt-Cu specimens. Both of as-deposited and as-deformed twinning structures. Therefore, obtaining a high density of nanotwins inside grains with a wider size distribution, while keeping the average twin thickness roughly unchanged, is still a big challenge in practice. In the present study, a high density of nanoscale twins can only be obtained experimentally with a narrow grain size distribution: nt-Cu with \( d = 500 \) nm is prepared with a peak current density of \( 7 \) A cm\(^{-2} \) \([9]\) and nt-Cu with \( d = 1500 \) nm has a lower peak current density of \( 3 \) A cm\(^{-2} \).

Uniaxial tensile tests were performed on a Tytron 250 Microforce Testing System (MTS System Corporation, Eden Prairie, MN, USA) at a constant strain rate of \( 6 \times 10^{-3} \) s\(^{-1} \) at room temperature. Dog-bone-shaped tensile specimens with a gauge length of \( 4 \) mm and a width of \( 2 \) mm were prepared by means of electrodischarging from the as-deposited Cu foils. The final sample thickness for tension tests after electropolishing is about \( 20 \pm 5 \) μm, as measured by a LEICA MPS 30 optical microscope. The tensile test was carried out at least five times for each nt-Cu sample.

Microstructural characterizations of both as-deposited and deformed samples were performed in a scanning electron microscope (SEM: Cambridge S-360, UK) with an electron channeling contrast (ECC) technique. Finer microstructures were characterized in a transmission electron microscope (TEM; JEOL 2010, UK) with an accelerating voltage of 200 kV. For the tensile nt-Cu samples, the TEM foils were cut from the localized necking area. The plastic strains, which are estimated by measuring the average cross-section reduction, are 3–7% and 15–32% for 460-nt-Cu and 1500-nt-Cu, respectively. Select area electron diffraction (SAED) patterns for nt-Cu were taken with aperture diameters of 0.2 ~ 1.25 μm for a detailed observation of as-deposited and as-deformed twinning structures.

Figure 1a and d shows the typical SEM planar morphologies of the two as-deposited nt-Cu specimens. Both Cu specimens consisted of roughly equiaxed grains with random orientations. Statistical measurements showed that the average grain sizes, \( d \), are 460 ± 50 and 1500 ± 130 nm, respectively \([5,6]\). For simplicity, the two nt-Cu samples are referred to as 460-nt-Cu and 1500-nt-Cu. TEM observations of two as-deposited nt-Cu samples are shown in Figure 1b and e. The SAED pattern indicated that the internal interfaces inside the crystals are typical TBs. The average \( \lambda \) between two adjacent TBs is \( 57 \pm 10 \) nm for 460-nt-Cu and \( 62 \pm 10 \) nm for 1500-nt-Cu. It is clear that the two nt-Cu samples with different grain sizes but possessing comparable twin thickness offer a good opportunity to explore the grain size effect on the mechanical properties of nanotwinned metals.

Typical tension true stress–strain curves of nt-Cu samples are displayed in Figure 2. It is interesting to see that the grain size does not have a clear influence on yield strength. The yield strength (0.2% offset, \( \sigma_y \)) of 1500-nt-Cu is about 510 MPa, which is very close to that of 460-nt-Cu (525 MPa). However, the plastic deformation stages of nt-Cu samples are affected significantly by \( d \). When \( d \) increases from 460 to 1500 nm, the ultimate tensile strength is enhanced from 570 to 710 MPa. A negligible work hardening is observed in 460-nt-Cu with an elongation-to-failure of 3.1%. More significantly, a distinct work hardening and a uniform strain of 12% are detected in 1500-nt-Cu, which are rarely observed in other nanostructured materials \([6,13,14]\). The work hardening rate of nt-Cu samples is defined as \([15,16]\).

\[
\Theta = \frac{d\sigma}{d\varepsilon}
\]  

where \( \sigma \) and \( \varepsilon \) are the macroscopic true stress and true plastic strain, respectively. Based on true stress–strain curves, \( \Theta \) is derived and plotted vs. the true stress and
strain for the two nt-Cu specimens (Fig. 3a and b, respectively). 1500-nt-Cu offers a distinctly improved hardening rate at both high stress and high strain. Comparing the hardening rate values of two nt-Cu samples, it is clear that the larger grain size enhances H appreciably. For example, at 2% plastic strain, H of 460-nt-Cu is 500 MPa, whereas for 1500-nt-Cu it is fivefold that value. The hardening rate of 1500-nt-Cu is beyond that of 460-nt-Cu over a wide range of stress and strain, which is in agreement with the postoned onset necking of 1500-nt-Cu in Figure 2. According to the instability criterion for geometrical softening via localized necking for sheet samples [17]:

\[ \Theta < \sigma/2 \]  

(2)

the estimated uniform strain for 1500-nt-Cu is 10.5%, which is about three times higher than that for 460-nt-Cu (3.2%), which is consistent with the tension results in Figure 2.

The plastic strain is not uniform in 460-nt-Cu (Fig. 4a). Some TBs are stepped or curved, or have even disappeared, as indicated by the black arrow, while others remain as they were. The morphology of the GB is still clear and sharp, but stress-concentrated GBs are prevalent even at a strain of more than 3%. The SAED pattern (inset in Fig. 4a) reveals that the diffraction spots are arc shaped. It is estimated that TBs deviate from the standard 70.5° (<1 1 2>), with a disorientation of 0–3° from SAED calculation [18].

Most TBs and GBs are not distinct in 1500-nt-Cu with a strain level of 15–32% (Fig. 4b). Abundant dislocations and some elongated sub-grains are generally presented. However, the SAED patterns (the inset in Fig. 4b) of two adjacent grains (outlined by the white dashed circle) show that the sub-boundary still keeps the twin-matrix symmetrical relationship with a disorientation of TBs of roughly 12°. Compared with the TBs of 460-nt-Cu in Figure 4a, only little TB debris is detected in 1500-nt-Cu. According to the microstrain from the XRD analysis [19], the density of dislocations in 1500-nt-Cu is estimated to be about 3.74 × 10^{14} m^{-2}, which is five times higher than that stored in 460-nt-Cu (6.90 × 10^{13} m^{-2}). The above finding indicates that a larger grain size is not only propitious to TBs gradually losing coherency during deformation by interacting with dislocations, but also facilitates the storage of more dislocations at twin planes, both processes sustaining a pronounced work hardening.

Kocks [15] and Mecking [16] considered that work hardening of material is determined by the evolutionary rate of dislocation density, which is usually governed by two competing processes: one strengthening and the other softening. In coarse-grained polycrystalline materials, the former is generally related to lattice dislocation storage and the latter is mainly from the dynamic recovery of lattice dislocations. The origin of hardening capacity of nt-Cu can be interpreted by the Mecking–Kocks theory as:

\[ \Theta = \Theta_0 - K \sigma \]  

(3)

where \( \Theta_0 \), which is independent of strain rate but dependent upon temperature, is the extreme hardening rate when \( \sigma = 0 \), and \( K \) is a constant, which is proportional to the softening process during deformation. Fitting the \( \Theta-\sigma \) curves in Figure 3a using Eq. (4), we obtained a \( K \) value of 126 in 460-nt-Cu, which is five times higher than that of 1500-nt-Cu. This suggests that 1500-nt-Cu has a lower softening rate at the uniform plastic strain stage than 460-nt-Cu.

For the nt-Cu samples, both GBs and TBs could be barriers for the motion of dislocations and both could contribute to the strengthening. Here the Hall–Petch-type relationship of yield strength as a function of GBs and TBs is generalized as follows:

\[ \sigma = \sigma_0 + k_{GB} d^{-1/2} + k_{TB} \lambda^{-1/2} \]  

(4)

where \( \sigma_0 \) is the initial friction stress, \( k_{GB} \) and \( k_{TB} \) are constants, \( d \) is the average grain size and \( \lambda \) is the average twin thickness. We suppose that \( k_{GB} = k_{TB} = 3478 \) MPa nm^{-1/2}, approximately. Based on Eq. (4), the yield strengths for nt-Cu with a twin thickness of 60 nm and different grain sizes were predicted to be about 675 and 621 MPa for 460-nt-Cu and 1500-nt-Cu, respectively, which are slightly higher than the experimental data. Since the scale of \( \lambda \) (nanometer scale) is one order of magnitude smaller than that of \( d \) (submicrometer scale), it is reasonable to consider the strengthening mechanism of the nt-Cu to be primarily determined by the twin thickness, rather than the twin length (grain size).
Experimental results found that a maximum strength is achieved for the nanotwinned Cu ($d = 500$ nm) when $\lambda = 15$ nm. When $\lambda$ is decreased further, a strength softening (rather than strengthening) will be seen [12]. Large-scale molecular dynamic simulation also showed that such a critical twin thickness for the maximum strength depends on the grain size: the smaller the grain size, the smaller the critical twin thickness and the higher the maximum strength of the materials [20]. For Cu samples with $d = 1500$ nm, the estimated critical $\lambda$ of maximum strength is about 30 nm from Eq. (1) in Ref. [20], which is much smaller than the average value (62 nm) from the experiments. That is consistent with the present conclusion that the grain size does not substantially alter the strength of both nt-Cu samples.

Because of the anisotropic of the twin lamellar structure, the distribution of dislocations is not spatially uniform. Most dislocations accumulate mainly in the vicinity of TBs and GBs, which may contribute to its softening process. For TBs, dislocation activities were confined within the two-dimensional twin plane. The grain size, i.e. the length of the TB, plays important roles in the dislocation accumulation, rearrangement and dynamic recovery. The nanotwinned samples with a larger grain size should have more room for dislocation storage and work hardening, similar to that in conventional polycrystalline materials. On the other hand, since the GB/TB intersections are potential dislocation sources and sinks, the localized deformation regions with respect to GBs possibly develop into the sites of void nucleation, growth, coalescence and ultimately failure, as usually observed experimentally, and dominate the softening process. As a consequence, such a GB-related failure/softening mechanism is directly correlated to the GB region and is roughly inverse to its grain size. The smaller the grain, the more GBs and the earlier necking and failure, which confirms the experimental results.

As discussed previously, most attention had been focused on the effect of the twin thickness or twin density on the mechanical properties [8–11,21]. However, the grain size also plays an important role in ntm-nt-metals. It is interesting to note that increasing the grain size results in an impressive increase in tensile ductility and ultimate tensile strength but does not sacrifice the yield strength obviously. The enhanced ductility and work hardening are believed to be associated with a higher density of stored dislocations and suppressed GB softening.

In conclusion, we found that grain size plays an important role in determining the mechanical properties of nanotwinned materials. While keeping the twin thickness fixed at the nanometer scale, increasing the grain size results in an impressive increase in tensile ductility and ultimate tensile strength but does not sacrifice the yield strength obviously. The enhanced ductility and work hardening are believed to be associated with a higher density of stored dislocations and suppressed GB softening.

The authors acknowledge the financial support from the National Science Foundation of China (Grant Nos. 50725103, 50621091, 50890171 & 51071153) and the MOST project of China (Grant No. 2005CB623604). L.L. and K.L. gratefully acknowledge support from the Danish National Research Foundation and the National Natural Science Foundation of China (Grant No. 5091130230) for the Danish–Chinese Center for Nanometals, within which part of this work was performed. The authors are grateful to Drs. X. Huang and M. Dao for stimulating discussions, and Mr. X. Si for assistance with sample preparation.