Temperature effect on rolling behavior of nano-twinned copper

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The effect of temperature on the rolling behavior of an ultrafine-grained Cu with nano-scale twins is reported. Rolling the nano-twinned Cu at liquid nitrogen temperature (LNT) led to a slight drop in hardness with an obvious thickening of twins, while rolling at room temperature (RT) resulted in obvious hardening without change in twin thickness. The strengthening mechanism via dislocation–twin boundary interactions dominates the rolling-induced hardening at RT, and the twinning partial activities are responsible for the de-twinning and softening in rolling at LNT.

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The novel mechanical properties and deformation mechanism of nanostructured materials have attracted a broad range of attention [1]. Fascinating strengthening has been achieved in nanocrystalline materials, but at the expense of ductility and toughness [2]. Recently, Lu et al. showed that the introduction of a high concentration of coherent nano-scale twins in Cu, generated through electro-deposition or dynamic plastic deformation, imparts significantly high strength while preserving considerable ductility [3–7]. Tensile tests at room temperature (RT) indicated that both the strength and ductility of the nano-twinned Cu (nt-Cu) increased gradually with decreasing average twin thickness [4], and maximum strength was achieved with an average twin thickness of 15 nm [8]. The strengthening mechanism is associated with the interaction between dislocations and twin boundaries (TBs), i.e., TBs serve as effective barriers to dislocation motion. Molecular dynamic (MD) simulations [9,10] also supported the fact that interactions of dislocations and TBs provide one of the principal mechanistic reasons for the strengthening of nt-Cu.

The temperature and strain rate dependence of deformation behaviors may shed light on the operating deformation mechanism. The strain rate sensitivity and reduced activation volume were found with decreasing twin thickness. This trend is similar to the grain size effect in face-centered cubic (fcc) nanocrystalline metals. Zhu et al. [9] also suggested that the TB-mediated slip transfer is a rate-controlling mechanism for the observed enhanced rate sensitivity with decreasing twin thickness from MD simulations. However, the temperature dependence of deformation behaviors has not been pursued so far for nano-twinned materials. The effect of temperature on deformation behavior and the deformation mechanism in the nt-Cu samples is studied in this paper.

High-purity ultrafine-grained Cu with nano-scale twins was synthesized using the pulse electro-deposition technique from an electrolyte of CuSO$_4$ [3]. Chemical analysis indicated that the purity of as-deposited Cu is better than 99.998 at.%. The density of each as-deposited sample is 8.93 ± 0.03 g cm$^{-3}$.

In order to investigate the temperature effect on the mechanical properties and deformation mechanism of nt-Cu samples, cold rolling at RT (293 K) and at liquid nitrogen temperature (LNT, 77 K) were performed, respectively. As-deposited Cu sheet samples (size $8 \times 4$ mm$^2$ and initial thickness $\sim 100$ μm) were cut and polished using SiC papers. Then they were chemically polished to a final thickness of $\sim 40$ μm with mirror surfaces. Two pieces of stainless steel sheet with $\sim 200$ μm thick were used to sandwich the nt-Cu samples during rolling. Cold rolling was carried out on a twin-roller apparatus with roller diameter 50 mm. The rolling strain ($\varepsilon$) is defined as $\varepsilon = (\delta_0 - \delta)/\delta$, where $\delta_0$ is the initial thickness.
initial sample thickness, and $\delta$ is the final thickness after rolling. The strain rate is estimated to be $\approx 10^{-1} - 10^{-2} \text{s}^{-1}$. In this study, three final rolling strains were chosen: $\approx 15\%$, 30\% and 40\%. For LNT rolling, the samples were immersed in liquid nitrogen for $\approx 2$ min before each rolling pass to cool the samples completely.

Microstructures of the as-deposited and as-rolled samples were characterized by transmission electron microscopy (TEM; JEOL 2010) operating with an accelerating voltage of 200 kV. The microhardness of the nt-Cu samples was measured on a MVK H3 hardness tester with load 5 g and duration time 10 s.

Microhardness measurements showed a clear increase in hardness in the RT-rolled nt-Cu from 2.5 ± 0.07 GPa to 3.0 ± 0.07 GPa with the rolling strain increasing from 0\% to 40\%, as shown in Figure 1. This suggests clear strain-hardening during rolling at RT, which is consistent with the previous tensile test results [14]. However, for the LNT-rolled nt-Cu, microhardness shows a slight decrease from 2.33 ± 0.05 to 2.21 ± 0.06 GPa with increasing rolling strain (see Fig. 1). The different hardness variation tendencies indicate a distinct temperature effect of the plastic deformation and the different underlying deformation mechanisms.

The planar-view TEM observations indicate that the as-deposited Cu sample consists of roughly equiaxed irregular grains with an average grain size of 400–500 nm (as shown in Fig. 2a). Inside each grain, there is a high concentration of growth twins which subdivide the ultrafine-grained crystals into a nano-scale twin/matrix lamellar structure. Most of TBs are coherent $\Sigma3$ and edged-on. They are perpendicular to the deposition surface and parallel to the (1 1 0) beam direction. From the TEM statistics, the average twin thickness for the as-deposited nt-Cu is $\approx 30 \pm 5$ nm. The microstrain of the as-deposited sample determined from X-ray diffraction analysis is negligible, suggesting a relatively low dislocation density.

After cold rolling, there is no visible change in the average grain size and morphology of nt-Cu with different strains at RT and LNT, as shown in Figure 2b and c. Initially, most of the dislocations pile up against TBs and grain boundaries (GBs). With increasing rolling strain, the dislocation density of nt-Cu increases obviously, and the majority of TBs are much strained, stepped and even curved owing to the high density of the stored dislocations. The difference in microstructure between the RT- and LNT-rolled Cu samples is not obvious at small rolling strains. Upon rolling to 40\%, it is interesting to note that more and more thick twin lamellae appeared in the LNT-rolled nt-Cu samples (see Fig. 2c). Twin lamellae with thickness $>200$ nm are seen more frequently, but thick twins are rarely seen in the RT-rolled sample with the same strain (Fig. 2b). In addition, the interior of thick twin lamellae in the LNT-rolled nt-Cu have a clear contrast, which is distinct from the highly concentrated tangled dislocations observed in the interior of the twins in the RT-rolled Cu.

Statistical distribution of twin thickness shows that the peak position of twin thickness of the RT-rolled nt-Cu exhibits a negligible change with rolling strain increasing to $\approx 40\%$, as shown in Figure 3a. In contrast, an obvious rightward shift of twin thickness distributions is seen in the LNT-rolled nt-Cu (Fig. 3b). The statistic volume fraction of twin lamellae with thickness $>100$ nm is only 12\% in the as-deposited state, while it reaches $\approx 28\%$ in the LNT-rolled sample with a strain of 40\%. The variation in average twin thickness (defined as the Gaussian peak value of the twin thickness distribution, similar to the definition in Ref. [4]) in Figure 3c shows that the average twin thickness increases from 31 to 53 nm in the case of LNT, while it remains constant at $\approx 34$ nm in RT rolling with a rolling strain.
up to 40%. The different variation trends in microhardness, twin thickness distribution and average twin thickness with rolling strain suggest that the deformation processes at RT and LNT are dominated by different mechanisms.

Dislocation nucleation and slip (full dislocation activity) and deformation twinning (controlled by partial dislocation activity) are two of the most common and competitive plastic deformation modes in fcc metals. The stress necessary to form twins is generally greater, but less sensitive to temperature [15,16], than that for full dislocation slip. However, the stress necessary for full dislocation activities strongly depends on the deformation temperature, i.e., a much higher stress is required for full dislocation activities at lower temperatures [15,17]. As such, a critical temperature exists, below which the critical stress for activation of twinning partial dislocations is lower than that for full dislocations; hence the activities of full dislocations would be suppressed, and partial twins would take over. Cu and other fcc metals usually deform by dislocation slip at RT, while twinning partial activities are more prevalent at low temperatures or at high strain rates [16].

Previous studies indicated that deformation of nt-Cu at RT is dominated by full dislocation activities, whereas the dislocations nucleate at grain boundaries, most likely at the TB–GB intersections where significant stress concentrations may develop. These full dislocations may slip along any (1 1 1) planes and then interact with the abundant growth coherent TBs via slip transfer [9], leading to apparent work hardening at RT, as shown in Figure 1.

With a decrease in the deformation temperature to LNT, the critical stress necessarily required for full dislocation activities will be much higher than that for twinning partial activities. As such, the twinning (Shockley) partial activity will be favorable at low temperatures in comparison with full dislocation activities. Specifically, the twinning partials on the (1 1 1) plane parallel to the pre-existing twin interfaces will be easily emitted, because such partial dislocation nucleation only results in twin boundary migration, without creating new twin interfaces. TB would migrate one atomic layer towards the matrix (twin thickening) or towards the twin (detwinning or twin shrinking), as illustrated in Figure 4a, depending on the local stress and orientation of the activated slip system [18,19]. The twinning partial activity associated with growth of existing twins is an energetically favorable process. Li and Ghoniem [20] revealed the twinning partials nucleation mechanism in nt-Cu in MD simulations. They suggested that the emission of Shockley partial dislocations on (1 1 1) planes adjacent to the TB could be dominant when the external ultrahigh shear stress was applied parallel to the associated TB (i.e., with smaller twin thickness or at high strain rates or low temperatures). This result is consistent with experimental observations that the deformation mechanism of nt-Cu is dominated by partial dislocation activities at LNT, rather than full dislocation activities as at RT.

Continuous nucleation and motion of the associated twinning partial dislocations lead to TB migration, formation of TB ledges/steps [21] and partial elimination of some thin twins (as illustrated in Fig. 4a), which eventually result in thickening of twin/matrix lamellae (i.e., detwinning). This is consistent with the microstructure observations that numerous TB ledges and steps are observed in LNT-rolled nt-Cu with a strain of 40%.
In as-deposited nt-Cu, the ledges are from deposition-induced twinning only, while in the as-rolled ones, the ledges are either from deposition-induced twinning or rolling-induced de-twinning (or twinning). In Figure 4c, the TB frequency and ledge density among the as-deposited, the RT-rolled and the LNT-Rolled nt-Cu samples with a strain of 40% are compared. Obviously, the TB frequency and ledge density of the RT-rolled nt-Cu are comparable with those of as-deposited Cu. However, the ledge density of the LNT-rolled nt-Cu is much larger, while the TB frequency is decreased, which can be reasonably attributed to the dominant deformation mechanism of the twinning partial activities at LNT.

Both the twin lamella thickening and TB ledge forming would result in softening. When the average twin thickness increases from 31 nm in the as-deposited state to 51 nm with a rolling strain of 40% at LNT, a substantial drop in hardness is anticipated according to the literature [4]. The high density of ledges consisting of mobile Shockley partial arrays may also be softening factors [22]. Such analyses above imply phenomenological responses for significant softening behavior of nt-Cu deformed at low temperature. However, only a slight softening phenomenon was seen experimentally in the LNT-rolled nt-Cu. The possible reason for this may be related to the concomitant strain-hardening due to dislocation accumulation during cold rolling at LNT, which would balance the softening to some extent.

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