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Viewpoint Paper

Repeated frictional sliding properties of copper containing nanoscale twins

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Abstract—Bulk dynamic plastic deformation (DPD) materials comprise a composite structure of nanoscale twin bundles and nanoscale grains. The tribological properties of DPD-processed pure nano-Cu have been investigated in this study and compared with conventional coarse-grained (CG) Cu under both monotonic and repeated frictional sliding. We demonstrate that DPD nano-Cu and CG Cu exhibit steady-state mechanical characteristics after repeated frictional sliding that are similar to those seen in nanotwinned (NT) Cu produced by pulsed electrodeposition.

Keywords: Contact sliding; Wear; Nanotwin; Nanocrystalline; Fatigue

1. Introduction

Grain refinement has been a traditional way to attain marked improvements in such properties as strength, wear and corrosion resistance, and diffusivity in metals [1–8]. Recent studies have also revealed that nanoscale twins introduced in the microstructure lead to improved mechanical properties not only in terms of higher strength, but also in retention of reasonable ductility, higher rate-sensitivity of deformation, and greater resistance to both fatigue crack initiation and growth [9–11]. These effects can be further enhanced through refining nanotwin (NT) spacing. However, high-quality pure nanotwinned specimens have thus far been produced successfully only in thin-film form [9–13]. This can impose severe constraints on potential applications of nanotwinned materials for bulk structural components.

Dynamic plastic deformation (DPD) offers a means to manufacture large specimens of nanostructured materials that comprise a mixture of nanotwins and nanograins [14,15]. While the basic mechanical properties of DPD Cu have been studied previously [14,15], the tribological response and the attendant microstructural stability of DPD nano-Cu under repeated frictional sliding have thus far not been investigated. Design against wear damage is a topic of considerable technological interest as it is one of the most common ways of material loss in most engineering applications [16]. Excessive material removal due to repeated frictional sliding and rubbing between contacting surfaces can lead to device dysfunction in micro electromechanical systems, failure of engineering components such as ball bearings and adverse immune response in the human body to metal-based biological implants. There have been only limited studies done on the wear response of nanograined (NG) and NT materials in general. In NG Ni, wear damage has been shown to decrease with decreasing grain size, but beyond the Hall–Petch breakdown point increased material loss was observed with grain refinement [6]. However, degraded wear response with strengthening has been observed in NG Ni–B alloy film produced by electrodeposition [17] and NG iron produced by rolling [18]. Grain coarsening under repeated sliding has been documented for NG Ni [8] as well as for NG Ni–W [19]. Moreover, a recent experimental study on repeated frictional sliding of NT Cu within ultrafine grains averaging 450 nm in size [11] confirmed that higher-density NT Cu exhibited greater resistance to both microstructure changes and surface damage after a frictional sliding pass; however, after many repeated sliding passes, copper samples with different NT densities were found to exhibit similar surface hardness and microstructure. This behavior bears a

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striking resemblance to the uniaxial strain-controlled fatigue behavior of medium-to-high stacking fault energy (SFE) metals, in which a uniform steady-state structure evolves after repeated mechanical loading as a result of rearrangements in defect structure facilitated by cyclic deformation.

Given the complexity of its microstructure, it is a challenging task to predict the tribological response of DPD nano-Cu only from its elasto-plastic properties. Moreover, the heterogeneous microstructure of DPD nano-Cu that comprises mixtures of nanotwin bundles and nano grains undergoes complex structural changes during frictional sliding because NG and NT materials influence structural evolution in different ways. It is therefore desirable to undertake experimental studies to ascertain the repeated frictional sliding response of DPD and contrast it with coarse-grained (CG) Cu under the same sliding test conditions, and to compare these results with NT Cu. Repeated frictional sliding tests provide controlled and quantitative measures that enable a fundamental understanding of the mechanisms underlying structural evolution during cyclic contact and the attendant wear damage processes.

The present work examines the tribological response of DPD nano-Cu and CG Cu by means of instrumented scratch tests under monotonic and repeated frictional sliding conditions. Friction coefficient and pile-up height are documented as functions of the sliding pass number. We have also studied the deformation-induced hardness evolution by indenting within the scratch tracks. These measurements enable the establishment of possible connections between tribological properties that develop in response to damage accumulation under repeated sliding and hardness/microstructure evolution. Repeated contact sliding (contact fatigue) is also seen to have an apparent similarity to strain-controlled uniaxial fatigue loading in that both types of loading produce plastic properties and the final corresponding microstructure that depend on the specific stress state, temperature and strain rate applied, but do not depend on the loading history for high SFE metals [20,21]. It is interesting to find that the eventual flow strength attained for CG Cu and DPD nano-Cu under similar experimental conditions was slightly lower than that obtained for NT Cu produced by pulsed electrodeposition (PED) [11]. This suggests that the processing method and the nature of twin boundaries (coherent twin boundaries in the case of PED vs. nanotwins generated from deformation twinning for DPD) may also be important factors that influence the final hardness and the corresponding microstructure attained in the deformation-affected zone.

2. Materials and experimental method

2.1. Materials

A copper cylinder with a diameter of 16 mm and a height of 26 mm was mechanically polished with SiC paper and subsequently annealed for 120 min in a vacuum (10^{-3} torr) at 700 °C to achieve a final average grain size value of approximately 250 μm. DPD treatment involved placing the copper cylinder on a lower anvil and compressing with an upper impact anvil at a strain rate of 10^2–10^3 s^{-1}. The specimen was totally immersed in a liquid nitrogen bath during each impact to prevent dynamic recovery and recrystallization. The deformation strain during the DPD process, defined as $\varepsilon = \ln(L_0/L_f)$, where L_0 and L_f are the initial and final thickness of the specimen, respectively, was 2.1. The resulting DPD nano-Cu specimen was essentially without any flaws or pores at the level of structural dimensions, and the final density and purity were found to be comparable to those of the original CG sample. The microstructure comprised ~67 vol.% of nano grains and ~33 vol.% of deformation twin bundles. The yield strength was measured to be 600 MPa and the ductility was 11% [14]. Essentially no strain hardening was observed. Further details of specimen preparation and microstructural and mechanical characterization can be found in [14]. In order to develop a basis for comparison for the foregoing DPD nano-Cu, a copper bar with an initial size of 8 mm × 8 mm × 12 mm was also mechanically polished with SiC paper and subsequently annealed for 120 min in a vacuum (10^{-3} torr) at 700 °C to achieve a final average grain size of approximately 250 μm. The bar, hereafter referred to as CG Cu, was directly used for companion experiments.

2.2. Experimental procedure

The frictional sliding experiments on DPD nano-Cu and CG Cu were performed using a NanoTest™ (Micromaterials, Wrexham, UK) instrumented indenter. A conical diamond tip (with a half tip angle of 70.3° and a tip radius of 2 μm) was used to make an array of scratches on the sample surface. This was accomplished by first making initial contact with the polished specimens (thoroughly cleaned in an ultrasonic ethanol bath), subsequent to which a normal load was applied on the specimen as the sample stage was moved laterally at a steady rate of 5 μm s^{-1}. The normal load was ramped up to its maximum value of 500 mN over the initial 50 μm of the sample-stage motion and allowed to stay constant for the remaining 450 μm length of the scratch. Thus, a 500 μm long scratch was introduced at the termination of one sliding pass. After that, the sample stage was first retracted by 15 μm away from the tip, and then returned to its original position

before the subsequent, new sliding pass. Multiple sliding over the same scratch path can be accomplished by repeating the process described above. The indenter was programmed to make 1, 17, 34, 50, 66, 82 and 98 sliding cycles, which left seven parallel tracks on DPD nano-Cu as well as CG Cu specimens. In order to avoid the interaction of strain-induced deformation zones between adjacent scratches, the grooved tracks were spaced amply apart from each other. Tangential loads were also acquired for the entire length of the scratch by force transducers mounted on either side of the indenter tip.

The same instrumented indenter was used to conduct all the indentations. First, 25 indentations were made on each specimen in order to ascertain the hardness. The same conical diamond indenter tip (with half angle of 70.3° and a tip radius of 2 μm) was used. The indentations were depth-controlled and the load required to reach a depth of 3 μm was recorded. Hardness was evaluated for both DPD and CG specimens. All these indentations were performed at a loading rate of 10 mN s^{-1}.

In order to investigate deformation-induced hardness change under repeated sliding, depth controlled indentations with a maximum depth of 1 μm were performed within the seven grooved tracks. Again, the same 70.3° half-angle diamond tip was used. The indentation loading rate was 10 mN s^{-1}. The method outlined in [11] was applied in order to extract the flow strength of the deformation-affected zone immediately beneath the scratch surface from the *P–h* (i.e. load vs. depth) response. These repeated sliding passes were made in the same direction.

The specimens were subsequently removed from the steel sample holders and a Tencor P10 profilometer (KLA-Tencor, San Jose, California) was used to scan the surface profile of the scratches. The probing tip of the profilometer was conical and made of diamond, with an apex angel of 45° and a tip radius of 2 μm. About 35 cross-sectional profiles were obtained from the steady-state region of the sliding track. These scanned profiles were in turn used for extracting the pile-up height for both specimens as a function of the number of sliding passes. Note that the tip of the profilometer has an apex angle of 45°, which is much smaller than the scratch indenter tip. In this case, there will be some errors for capturing the cross-sectional profile due to the tip radius effect, especially near the bottom of the groove. However, the primary aim of the profilometry study was to see the maximum depth of the groove and the maximum amount of the pile-up. These maximum values will not be affected by the tip radius effect.

3. Results

3.1. Hardness measurements

DPD nano-Cu processed at high strain rates and cryogenic temperatures has an enhanced hardness compared to CG Cu. The latter has a hardness of 700 MPa, while DPD nano-Cu exhibits a hardness value as high as 1.90 GPa. An abundance of nano grains and deformation twin bundles, formed as a result of low-temperature, high-strain-rate deformation, are considered responsible for the much higher hardness of DPD nano-Cu than CG Cu.

3.2. Friction coefficient

Figure 1 shows how the total friction coefficient between the indenter and the specimens evolves as a function of the number of sliding passes. The friction coefficient for both DPD and CG Cu decreases monotonically with increasing number of sliding cycles; however, the rate of decline is higher initially, and after about 60 passes the friction coefficient values does not decrease significantly any further, as a steady-state microstructure is being developed. Friction coefficient values for DPD nano-Cu are comparable to those observed for NT Cu [11]. CG Cu exhibits similar values of friction coefficient as DPD nano-Cu, in spite of different initial microstructures and consequently different initial hardness values.

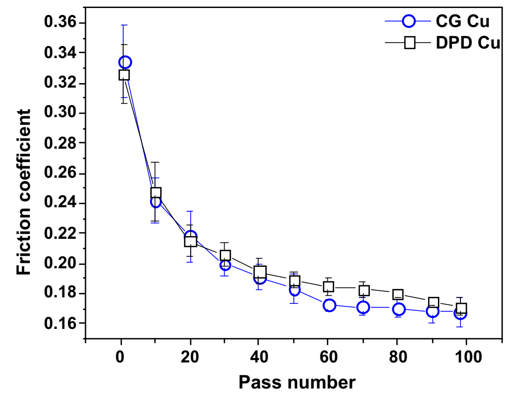

Figure 1. Friction coefficient as a function of pass number for both DPD nano-Cu and CG Cu.

3.3. Pile-up height

Owing to the indenter tip penetration and subsequent repeated scratch motion, the specimen surface is worn and the material removed from the specimen is piled up along both sides of the scratch. The pile-up height gives a measure of the wear damage and material removal produced by the frictional sliding process. Figure 2 shows that the pile-up height increases with increasing number of sliding cycles; however, there is a progressive decrease in the rate at which the pile-up height increases. CG Cu showed a much higher initial pile-up height owing to its lower initial hardness than DPD nano-Cu. Compared with the results of ultrafine-grained (UFG) Cu and NT Cu [11] under the same testing conditions, CG Cu presented the highest pile-up height among all the samples considered (CG, DPD, UFG, low-density NT (LDNT) and high-density NT Cu (HDNT)), while DPD nano-Cu showed a similar level of pile-up to LDNT Cu.

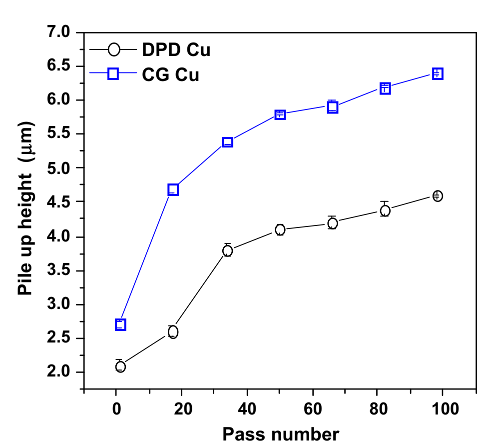

Figure 2. Pile-up height as a function of pass number for DPD and CG Cu.

3.4. Hardness evolution

The flow strength near the bottom of the scratch-groove center line for both DPD and CG Cu is plotted as a function of the number of sliding cycles in Figure 3; the results of LDNT Cu and HDNT Cu [11] are also included for comparison. High strains beneath the indenter lead to strengthening of the deformation-affected zone in CG Cu and the relatively softer LDNT Cu, and the hardness monotonically increases with the number of sliding passes. However, repeated frictional sliding has a softening effect on DPD nano-Cu in that the flow strength decreases from its peak value after the first pass with subsequent sliding cycles, and after 82 sliding cycles both CG and DPD nano-Cu exhibit similar flow strength in the deformation-affected zone. Similar converging surface material flow strength vs. the number of sliding passes was also observed for LDNT and HDNT Cu [11], although the final flow strength appeared to be a little higher for LDNT/HDNT Cu than that for CG and DPD nano-Cu.

4. Discussion

The DPD process leads to significant strengthening, as the hardness of DPD nano-Cu is much higher than that of CG Cu. This can be attributed to the high number of deformation twin boundaries that contribute to strength by blocking dislocation motion [22], and to grain refinement, as nano grains comprise about 67 vol.% of DPD nano-Cu.

DPD nano-Cu shows a lower pile-up height than CG Cu because of its much higher strength. The pile-up heights of both DPD nano-Cu and CG Cu increase with the sliding pass number, but the rate of increase progressively diminishes as structure evolution begins to stabilize after about 50 passes. Pile-up height, which is an indirect measure of the material removed by the motion of the sharp indenter, provides an estimate of the material loss incurred by the repeated frictional sliding process. Higher pile-up increases the contact adhesion between the indenter tip and the material surface being scratched, which leads to increased tangential loads required to perform frictional sliding tests. Comparing the results obtained under the same testing conditions for UFG, LDNT and HDNT Cu, we find that CG Cu shows the highest pile-up while DPD nano-Cu exhibits a similar level of pile-up to LDNT Cu. The introduction of nano grains and nanotwins in DPD nano-Cu ostensibly reduces the material removal under frictional sliding. The attainment of similar rates of pile-up height increase for DPD nano-Cu and CG Cu is consistent with the converging flow strength observed near the bottom of the scratch surface after 50 passes.

The value of friction coefficient developed during relative motion between the indenter tip and the specimen is a complex function of elastic modulus, yield strength, strain hardening exponent, elastic modulus and local fracture processes. Figure 1 shows that DPD nano-Cu and CG Cu exhibit similar values of friction coefficient at all pass numbers in spite of having very different initial hardness values. We believe these interesting results are accidental rather than universal. It has been shown that, although the friction coefficient decreases with increasing yield strength, a lower strain hardening exponent leads to an increase in the friction coefficient [23,24]. It is shown in [14] that DPD nano-Cu exhibits negligible strain hardening whereas CG Cu shows considerable work hardening. CG Cu, with its significantly lower strength but much higher strain-hardening exponent, has similar values of friction coefficient to DPD nano-Cu.

Frictional sliding by a sharp conical indenter imposes high strains on the material, thereby promoting significant changes in the local microstructure and hardness. Figure 3 shows that the flow strength near the bottom of the scratch surface changes progressively with the number of sliding passes. Measured right after the first sliding pass, DPD nano-Cu exhibits a high flow strength of 700 MPa. After the first pass, the hardness of DPD nano-Cu decreases monotonically with the number of sliding passes, with pronounced changes occurring in the first 50 passes. After 82 sliding passes the flow strength for DPD nano-Cu is around 500 MPa. On the other hand, CG Cu exhibits a low strength of 150 MPa after the first sliding pass. The CG Cu hardens with increasing number of sliding passes, so that after 82 cycles it has attained a flow strength of about 450 MPa – close to that of DPD nano-Cu. This is consistent with our earlier study conducted on NT materials in which high- and low-density NT Cu specimens acquire similar microstructure and hardness after repeated sliding [11]. Based on the flow strength values extracted from indentation inside wear tracks, the average grain size in the deformation-affected zone after 82 passes is expected to be close to 200–300 nm from the Hall–Petch relationship for Cu, with 450–500 MPa flow strength. A grain size close to 200–300 nm is also consistent with what has been found for Cu subjected to various severe plastic deformation processes [25]. However, following the Hall–Petch relationship to estimate the average grain size assumes that there is a negligible fraction of deformation twin boundaries remaining in the multiple

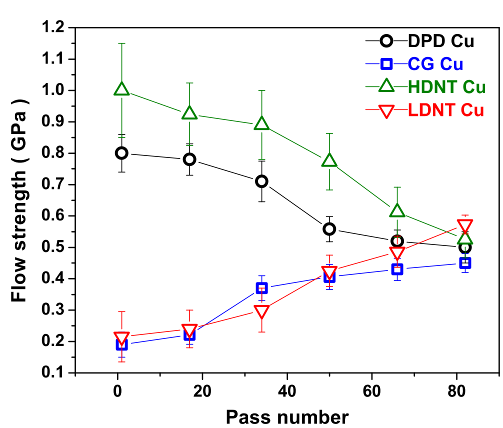

Figure 3. Flow strength evolution of the deformation-affected region just below the scratch in DPD and CG Cu with increasing number of sliding cycles. With repetitions in sliding, the region below the scratch gets harder for CG Cu whereas it gets softer for DPD nano-Cu. The results of LDNT Cu and HDNT Cu [11] are also included for comparison.

scratched deformation zones. This should be a reasonable assumption, considering the fact that detwinning has also been observed in Cu thin film layers with growth twins under uniaxial cyclic loading as well as indentation [26,27].

Strain-controlled uniaxial fatigue loading [20,21] has been shown to alter the microstructure and strength of medium-to-high SFE metals. Under repeated uniaxial loading, cold-worked high-strength metals soften and annealed softer metals harden until their hardness values converge and their microstructures reach a common saturated state, provided that they have a sufficiently high SFE such that cross-slip of dislocations is not hindered. In our current study, copper, which has a moderate SFE (4.0×10^{-2} Jm^{-2}) [28], exhibited a similar behavior under repeated contact sliding to that observed under uniaxial fatigue loading. The convergence of flow strength near the bottom of the scratch surface for both DPD nano- and CG Cu specimens after multiple sliding cycles is ascribed to the evolution of similar microstructures. The final average grain size (200–300 nm) estimated from hardness values within the 1 μm region close to the scratch surface, however, is different from the average grain size (∼50 nm) observed for nanotwinned Cu with initially coherent twin boundaries after repeated frictional sliding under the same experimental conditions [11]. This implies that the initial processing conditions for specimen processing/preparation may significantly influence the subsequent microstructure and hardness attained after repeated sliding.

5. Conclusion

The wear response of DPD and CG Cu was studied through monotonic and repeated frictional sliding, and the results were compared with those obtained from similar experiments performed previously [11] on LDNT and HDNT Cu. The following conclusions are drawn from this study:

- The DPD process improves hardness while providing improved tribological properties compared to CG Cu. The introduction of nano grains and nanotwins in DPD nano-Cu appears to aid in reducing the material removal under repeated frictional sliding.
- The friction coefficients of DPD nano-Cu and CG Cu during repeated sliding were found to be similar in spite of significant differences in their initial hardness values. This may be attributed to the significantly higher strain hardening capability of CG Cu than that of DPD nano-Cu.
- Under repeated frictional sliding, DPD nano-Cu softens and CG Cu hardens, and eventually both specimens exhibit similar values of hardness within the deformation-affected zone. The corresponding flow strength attained was found lower than that observed for LDNT and HDNT Cu [11].

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