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Enhanced fatigue resistance of Cu with a gradient nanograined surface layer

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Cyclic deformation was studied in Cu samples with a gradient nanograined (GNG) surface layer. Compared with the coarse-grained sample, the Cu samples with a GNG surface layer exhibit a greatly enhanced fatigue limit under stress-controlled cyclic deformation. The cyclic deformation induced an abnormal grain coarsening that initiated from the subsurface layer and grew along 45° to the stress axis toward the top surface layer, where the fatigue cracks were formed.

Keywords: Gradient nanograined surface layer; Fatigue limit; Grain coarsening; Crack nucleation; Copper

Refining grains to the nanometer scale results in materials that exhibit greatly enhanced strength/hardness [1,2], wear resistance [3] and other properties in comparison with their coarse-grained (CG) counterparts. However, nanosized grains do not guarantee better ductility [1] and fatigue performance. Mughrabi et al. [4] studied the cyclic deformation and fatigue behavior of ultrafine-grained (UFG) metals produced by equal-channel angular pressing (ECAP) and showed that the fatigue behavior of UFG Cu was even worse than that of CG Cu in terms of the low-cycle fatigue Coffin–Manson data for strain controlled fatigue tests, while the high-cycle fatigue behavior showed a remarkable improvement over the CG one in stress-controlled tests. Hanlon et al. [5] showed that grain refinement in the fully dense nanocrystalline (NC) and UFG Ni samples generally led to an increase in resistance to failure under stress-controlled fatigue, whereas a generally deleterious effect was seen on resistance to fatigue crack growth.

In fact, most fatigue failures occur on material surfaces and then propagate into the interior. Thus, a structural architecture comprising a nanostructured surface layer and a CG interior has been considered optimal for enhanced fatigue behaviors. This is because fatigue crack initiation would be suppressed by the strong nanostructured surface layer. Once the crack nucleates, crack propagation can be arrested by the CG interior. Roland et al. [6] demonstrated this idea with elegant experiments proving that significant improved fatigue strength in both high-cycle and low-cycle fatigue regimes could be achieved in a 316L stainless steel with a nanocrystalline surface layer generated through surface mechanical attrition treatment (SMAT). An enhanced fatigue limit has also been achieved in Ni-based C2000 alloy by surface nanocrystallization; it was hypothesized that the nanocrystalline, work-hardened region as well as the residual compressive stress in the top surface layer played a significant role in the fatigue performance [7].

Most recently, a novel surface treatment technique, namely surface mechanical grinding treatment (SMGT) [8], was utilized to prepare a spatial gradient nanograined (GNG) structured surface layer on bulk CG Cu rod. Compared with other surface plastic deformation techniques, such as friction sliding [9], SMAT [10] and wire-brushing [11], the SMGT sample offers the advantages of a thicker nanostructured layer, better structural homogeneity in the surface layer and much smaller surface roughness. Moreover, significant uniform tensile plasticity has been achieved in the nanocrystalline surface layer where the strain localization is effectively suppressed [12]; this plasticity was believed to originate from the deformation mechanism with concomitant mechanically driven growth of nanosized grains.

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In this paper, fatigue behaviors are investigated on a CG Cu sample with a GNG surface layer processed by means of SMGT at cryogenic temperature. The possible fatigue mechanism and the microstructure stability of the GNG Cu during cyclic deformation will be discussed.

A commercial-purity copper rod (99.7 wt.%, prepared in the shape of a cylindrical dog-bone with a gauge length of 12 mm and a diameter of 6 mm) was chosen for the SMGT experiment. The rod was annealed at 723 K for 1 h to obtain a uniform CG polycrystalline structure. The detailed procedure for sample preparation was described in Ref. [12]. The SMGT processing parameters are as follows: rod rotation speed 600 rpm; amount of feed 40 μm; feed rate (Z-axis) 20 mm min^{-1}. The Cu rod was cooled to cryogenic temperature (~173 K) with liquid nitrogen. In order to effectively refine grains in the surface layer and increase the thickness of the nanostructured layer, the SMGT processes were repeated 11 times with the same processing parameters.

SMGT Cu samples were fatigued with tension–compression in a MTS 810 servohydraulic testing machine. The fatigue tests, at constant stress amplitudes of 84, 98, 112 and 140 MPa, were carried out with a frequency of 30 Hz. All fatigue tests were conducted with a stress ratio ($\sigma_{min}/\sigma_{max}$) of −1. The microstructure of specimens was studied by transmission electron microscopy (TEM; Tecnai F20G^2) and scanning electron microscopy (SEM; Nova Nano 430). Laser confocal microscopy (LCM; LEXT OLS4000) was also used to characterize the three-dimensional fracture surface topography after the fatigue tests.

A typical cross-sectional microstructural characterization of an 11-pass SMGT Cu sample is shown in Figure 1. Homogeneous plastic deformation is seen in the top layer with a very small surface roughness ($R_a \approx 0.3$ μm). No crack can be identified in the deformed top surface layer. A spatial gradient structure is formed with continuously increasing grain sizes from nanoscale to submicro/microscale on the CG substrate after SMGT. TEM observations showed a gradually increasing grain size with increasing depth. The top surface layer of specimens is composed of nanosized elongated grains with random crystallographic orientations, as indicated by the selected-area electron diffraction patterns shown in Figure 1b. The average transverse axis size and longitudinal axis grain size are 56 and 106 nm, respectively, in the top 15 μm thick layer (Fig. 1b). The grain size gradually increases to about 300 nm in the depth range of 15–75 μm (Fig. 1b and c), where the grains can be approximately considered to be equiaxial. Typical deformation structures at depths of >75 μm are coarse grains with sizes ranging from submicrometers to micrometers and characterized by dislocation tangles or dislocation cells. The total thickness of the deformed CG layer is ~500–700 μm.

Tensile tests of GNG Cu samples showed a yield strength (0.2% offset) of 126 MPa, almost twice that of the CG sample, but there is no obvious improvement in the fracture strength. This increment of yield strength can be reasonably attributed to the deformed CG layer and the strong GNG surface layer [12]. The uniform elongation of GNG Cu is about 27%, similar to that of the CG Cu tensile sample. The dependence of the fatigue life N_f on the cyclic maximum stress for GNG Cu and annealed CG Cu are compared in Figure 2. The fatigue strengths of GNG Cu and CG Cu are 98 and 56 MPa, respectively, based on a fatigue life of 10^7 cycles. The fatigue behavior of CG Cu in this study is consistent with the literature data [10]. This means that the SMGT process has improved the fatigue strength of the Cu sample by almost 75%. Moreover, the fatigue life of GNG Cu is remarkably longer than that of the CG Cu at low-cycle fatigue tests. For example, the fatigue life of the GNG Cu is more than 14 times that of CG Cu at a stress amplitude of 140 MPa. It is interesting to find that the enhancement of fatigue life in the stress-controlled fatigue is comparable to what has been reported for fatigued UFG materials, compared to CG metals. Such an unexpected similarity has also been observed for both the fatigued nanotwinned Cu and fatigued UFG Cu [13]. This similarity may be a coincidence or be caused by other unknown factors, and needs further study.

The fractograph of the fatigued GNG Cu displayed in Figure 3a is similar to that of CG Cu. Three obvious regions can be distinguished from the fracture surface of the GNG Cu: a crack initiation region (region 1), a crack propagation region (region 2) and an instant rupture region (region 3). From the low-magnification SEM observation in region 1 (Fig. 3a), it can be seen that all the cracks initiate from the surface region of samples. Many fatigue striations can be identified in region 2; these are

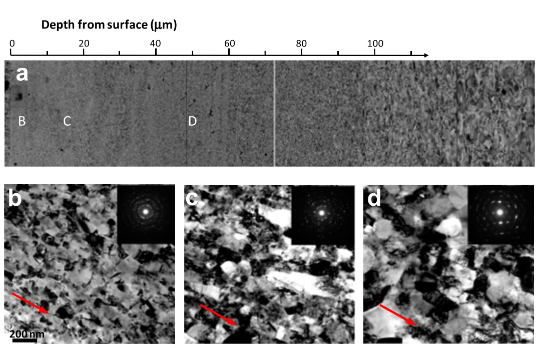

Figure 1. (a) A typical cross-sectional SEM image of the GNG Cu sample. The bright-field TEM images at positions B, C and D indicated in (a) are shown in (b–d), respectively. The arrows indicate fatigue loading directions.

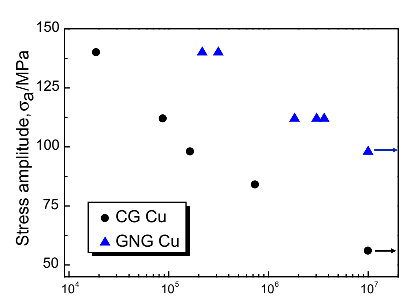

Figure 2. Dependence of the fatigue life (N_f) on the cyclic maximum stress for CG Cu and GNG Cu.

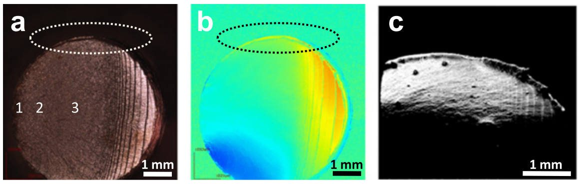

Figure 3. Fracture surface of the fatigued GNG Cu observed by (a) SEM and (b) LCM and (c) a high-magnification image of the area in (a).

caused by the repetitive crack blunting/sharpening under cyclic loading according to Laird's proposal [14]. Compared with the flat fractograph of the CG Cu sample, the obvious difference is that a shell surface is easily detected in the top surface of GNG Cu at a high magnification, as shown in Figure 3c. LCM measurement indicated that the height of the surface shell, defined as the difference in height between the top surface and the center of the fractograph, is <100 μm (Fig. 3b).

The fatigue properties of the GNG Cu samples are increased considerably compared with the CG Cu for both high- and low-cycle regimes; this is analogous to the results reported for other surface nanocrystallization samples [6]. According to the Basquin equation [14], higher true fracture strength will result in a longer fatigue life at the same stress amplitude for most metals. Because the fracture strength of SMGT Cu in this study shows no obvious improvement compared with CG Cu, the enhanced fatigue behaviors of the GNG Cu samples can be mainly attributed to the increased resistance for fatigue crack initiation by the gradient nanostructure (including nanograined surface layer and UFG subsurface layer), compressive residual stress and small surface roughness. These factors will be discussed in the following paragraphs.

The surface roughness of the SMGT sample is less than that of samples processed by other surface nanocrystallization techniques, such as surface nanocrystallization and hardening which give a surface roughness of >40 μm [15]. Large surface roughness may induce stress concentration and facilitate crack initiation under fatigue conditions. The decreased surface roughness in GNG Cu rod may suppress fatigue crack initiation because of the reduced notch effects. In the high-cycle regime, the fatigue endurance limit is typically controlled by the number of fatigue cycles required to initiate a dominant crack, which could account for about 90% of the total life [14]. Therefore, effective suppression of crack nucleation and/or facilitation of crack closure should be valid paths for improving the fatigue properties. The surface compressive residual stresses, generally induced during SMGT, can act as the closure stress for the short fatigue cracks and also constrain the plastic deformation developing at the crack tip [16]. Similar to the compressive residual stress, a nanostructured surface and UFG subsurface also can postpone crack nucleation by suppressing plastic deformation at the surface region.

In order to explore the intrinsic fatigue fracture mechanisms of the GNG Cu, the transverse microstructure of GNG Cu after fatigue test at a constant stress amplitude of 140 MPa was investigated. Positions A1–A4 in Figure 4 indicate continuous SEM observations along the long axis from the fracture surface to the grip ends of the GNG Cu. The larger the fatigue plastic strain sustained, the closer the site is to the fracture surface (A1). Therefore, the typical microstructure evolution during cyclic deformation can be roughly envisaged from positions A4 to A1. Microstructure instability, i.e. abnormal grain coarsening, is prevalently seen in the subsurface layer of GNG Cu. This is different from the homogeneous grain coarsening of the GNG layer during uniaxial tensile testing which is interpreted as a mechanically driven grain boundary migration process [12].

In the early stage (Fig. 4A4, A3, far away from the fracture surface), few coarsened grains are seen in the subsurface layer with a depth of about 50 μm from the top surface. The initial coarse grains have typical abnormal grain growth characteristics with approximately equiaxed shape. However, the coarse grains become elongated with further cyclic deformation. As shown in Figure 4A2, grain coarsening develops along the maximum shear stress direction to a rather large extent, i.e. along ~45° to the stress axis (Fig. 4A2). In the later stage, universal grain coarsening close to the main crack is shown in the A1 position. More and more abnormal grain coarsening continuously occurs and grows towards the top surface. Careful observations indicate that some small cracks initiate in the coarse grains at the top surface. The cracks propagate along 45° with respect to the loading direction, parallel to the maximum shear stress direction, as shown in Figure 4A4. Thus, the main crack of GNG Cu is believed to be nucleated from the top surface and occurred from the CG regime. Such a grain-coarsening phenomenon accompanied by strain localization, leading to nucleation sites of cracks which dominate the final failure, should be similar to the failure process of fcc CG metals. However, no shear band was detected along the direction 45° to the stress axis, which is different from the fatigue-induced microstructure coarsening observed in UFG metals prepared by severe plastic deformation [17].

Although the underlying mechanism for the cyclic deformation-induced grain coarsening and failure observed in the GNG Cu samples is still unclear at the moment, the phenomenon of grain coarsening from

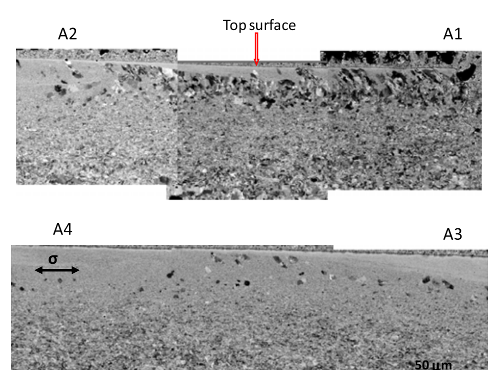

Figure 4. A typical cross-sectional SEM image of the GNG Cu fatigued to failure at a stress amplitude of 140 MPa.

the subsurface layer can be clearly seen (as shown in Fig. 4). In brief, the microstructure evolution of GNG Cu during cyclic deformation can be sketched out as follows: at the beginning stage of cyclic deformation, localized grain coarsening occurs in the subsurface layer and then grows toward to the surface along 45°, which sustains the main cyclic plastic deformation. The final fatigue failure is postponed by the spatial gradient nanostructure and governed by the surface crack nucleation when the grain coarsening reaches the top surface. Clearly, the improved fatigue properties of the GNG Cu sample can be attributed to the GNG surface layer, which not only contributes to the high fatigue strength but also suppresses crack nucleation. However, fracture failure occurs mainly in the coarsening regime, more like what happens in CG fcc metals. If the microstructure instability or grain coarsening during the fatigue process could be effectively suppressed or postponed, better fatigue properties will be expected.

A comparison of the microstructures before and after fatigue tests between Figures 1 and 4 indicates that grain coarsening initially occurs in the subsurface layer with an average grain size of ~200–300 nm, which is the typical stable grain size achieved by plastic deformation processes such as ECAP, accumulative roll bonding, etc. [18–20]. Such an abnormal grain-coarsening phenomenon is also frequently observed in UFG metals upon cyclic deformation [21]. The average grain size and the relatively high-energy state of UFG microstructure should act as the major, though not necessarily the only, driving forces for the initiation of the coarsening process in GNG Cu.

Based on the experimental results, a dynamic recrystallization mechanism, related to the migration of grain and subgrain boundaries, is suggested to be responsible for the cyclic grain coarsening of UFG metals [22]. The grain boundary motion may be affected strongly by the high-energy state of the plastically deformed sample, which may arise either from the internal energy associated with the residual strains or from the grain boundary energy associated with the small grain size. Decreasing the internal energy and/or grain boundary energy may result in grain boundary migration and cause grain coarsening. The closer to the surface, the higher the energies of the grains and the higher the driving force for grain boundary migration. Hence, grain coarsening would tend to develop in the top nanocrystalline layer along the plane of maximum shear stress. The fatigue fracture mechanism can also explain the experimental observations that a small shell surface was left after fatigue tests, as shown in Figure 3c.

In summary, a GNG surface layer was developed in Cu samples by means of SMGT. The improved fatigue strength and fatigue lifetime of the GNG Cu sample are attributed to the GNG surface layer, which not only contributes to the high fatigue strength but also suppresses crack nucleation. Microstructural observations on the fatigued GNG Cu show that abnormal grain coarsening was initiated in the subsurface layer and then propagated to the top surface, where the fatigue cracks nucleate and propagate.

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