SCIENCE CHINA Materials

LETTER

mater.scichina.com link.springer.com

Published online 20 July 2016 | doi: 10.1007/s40843-016-5068-6
Sci China Mater 2016, (7): 531–537

Intrinsic high cycle fatigue behavior of ultrafine grained pure Cu with stable structure

Peng Xue, Zhiye Huang, Beibei Wang, Yanzhong Tian, Wenguang Wang, Bolv Xiao and Zongyi Ma*

ABSTRACT Ultrafine grained (UFG) materials have attracted considerable attention owing to their unique microstructure and mechanical properties. However, the easy formation of large-scale shear bands and severe grain coarsening during cyclic deformation gives rise to enormous difficulties when investigating the intrinsic fatigue behavior of UFG materials. Herein, we discuss the fabrication of an ideal model material, based on pure Cu, by friction stir processing (FSP), which exhibits equiaxed ultrafine grains, low dislocation density, and a high ratio of high-angle grain boundaries. This model material was used to investigate the intrinsic high cycle fatigue behavior of UFG material. It was found that an enhanced fatigue limit and fatigue ratio can be achieved by FSP Cu due to its uniform and stable UFG structure. Instead of traditional large-scale shear bands, protrusion was found to be the main surface damage morphology for FSP Cu during high cycle fatigue deformation, and no obvious grain coarsening was observed. Dislocation related activity also dominated, but was limited to the ultrafine grains without the formation of regular dislocation structures.

Keywords: friction stir processing, ultrafine grained microstructure, high cycle fatigue behavior, dislocation structure, grain growth

In recent decades, ultrafine grained (UFG) materials have attracted considerable interest due to their unique microstructure and mechanical properties [1,2]. Many previous studies on UFG materials focused on microstructure evolution, strength enhancement and the strength-ductility relationship. Regarding the fatigue of UFG materials, most studies have concentrated on cyclic deformation, fatigue life, surface damage formation and underlying microstructural mechanisms [3]. However, despite the amount of research in this field, there is as yet no unequivocal knowledge of the fatigue deformation mechanism of UFG materials, which is of primary importance for practical engineering applications [3–10].

At present, various severe plastic deformation (SPD) methods, such as equal channel angular pressing (ECAP), high pressure torsion (HPT), and dynamic plastic deformation (DPD), provide practical approaches to prepare bulk metal and alloy samples with UFG structures [1,2,11–16]. The fabricated UFG materials exist mostly in a metastable state, the microstructures of which are easy to manipulate by dynamic recovery and recrystallization under applied loading, even at room temperature [1–3]. It has been commonly recognized that room temperature fatigue damage in UFG materials can be attributed to grain coarsening and the formation of large-scale shear bands (SBs), which can extend over a greater scale than the initial grain size [3–10].

As a typical face center cubic (fcc) metal, pure Cu has been widely used as a model material in investigating the fatigue behavior of UFG materials [3]. The results show that although the fatigue strength of Cu can be improved to some extent in the high stress amplitude range, its high cycle fatigue limit remains essentially unchanged (~80–100 MPa) and a relatively low fatigue ratio (fatigue limit/ultimate tensile strength) of ~0.20–0.25 is achieved [3–8]. Coarse grains, together with large-scale SBs of several tens of micrometers, can be observed due to the excessive grain growth caused by strain localization and dynamic recrystallization. Furthermore, various typical dislocation configurations of fatigued coarse-grained (CG) Cu, such as dislocation walls/cells, can be observed in fatigued UFG Cu, and the intrinsic fatigue behavior of UFG Cu may be concealed in this case.

Based on the basic principle of friction stir welding (FSW), a new processing technique–friction stir processing (FSP), has been developed for structural modification [17].

Shenyang National Laboratory for Materials Science, Institute of Metal Research, Chinese Academy of Sciences, Shenyang 110016, China
* Corresponding author (email: zyma@imr.ac.cn)

Recently, FSP has been proven to be an effective method to prepare bulk UFG materials, such as Al, Mg, Cu, Ni and their alloys [18–23]. Compared to the SPD UFG materials, a more stable structure should be achieved in FSP UFG materials, which exhibits an equiaxed recrystallized microstructure, with low dislocation density and a high ratio of high-angle grain boundaries (HAGBs). Therefore, enhanced strain hardening can be achieved in FSP UFG Cu due to its unique microstructure [23].

FSP UFG Cu acts as an ideal model for the investigation of mechanical properties, especially when examining fatigue behavior, which is strongly associated with the microstructure. As a preliminary investigation, stress-controlled high cycle fatigue tests were conducted on FSP UFG Cu. The aim of this study was to investigate the intrinsic high cycle fatigue behavior of UFG materials.

Commercially pure Cu (99.98 wt.%) plates of 3 mm thickness were used in this study, and they were annealed for 2 h at 700°C to provide the initial CG state. To prepare the UFG Cu, the plates were first fixed in water and then subjected to FSP at a rotation rate of 600 rpm using a tool with a shoulder 12 mm in diameter. During FSP, additional rapid cooling by flowing water was utilized. Detailed parameters regarding water cooling were described in the previous study [24].

Microstructural examination of samples was completed using scanning electron microscopy (SEM), transmission electron microscopy (TEM) and electron backscatter diffraction (EBSD). The specimens for microstructural observation were machined from the cross-sectional plane perpendicular to the processing direction. Data collection by EBSD was performed using a program developed by TSL (OIM analysis system) on a field emission (FE) SEM with step size of 70 nm. Flat dog-bone-shaped fatigue specimens with a gauge dimension of 4×3×2 mm^3 were cut from the processed zone along the FSP direction. Symmetrical cyclic pull-push (R=−1) fatigue tests under fully reversed constant stress were carried out with the use of a Shimadzu 4830 fatigue testing machine with a frequency of 30 Hz.

Fig. 1 shows the typical microstructure of the FSP UFG Cu observed from EBSD and TEM. Similar to other FSP UFG materials [18–22], uniform equiaxed grains were obtained in FSP Cu, as shown in Fig. 1a. From the detailed TEM microstructure in Fig. 1b, a low dislocation density is observed in these equiaxed ultrafine grains. Moreover,

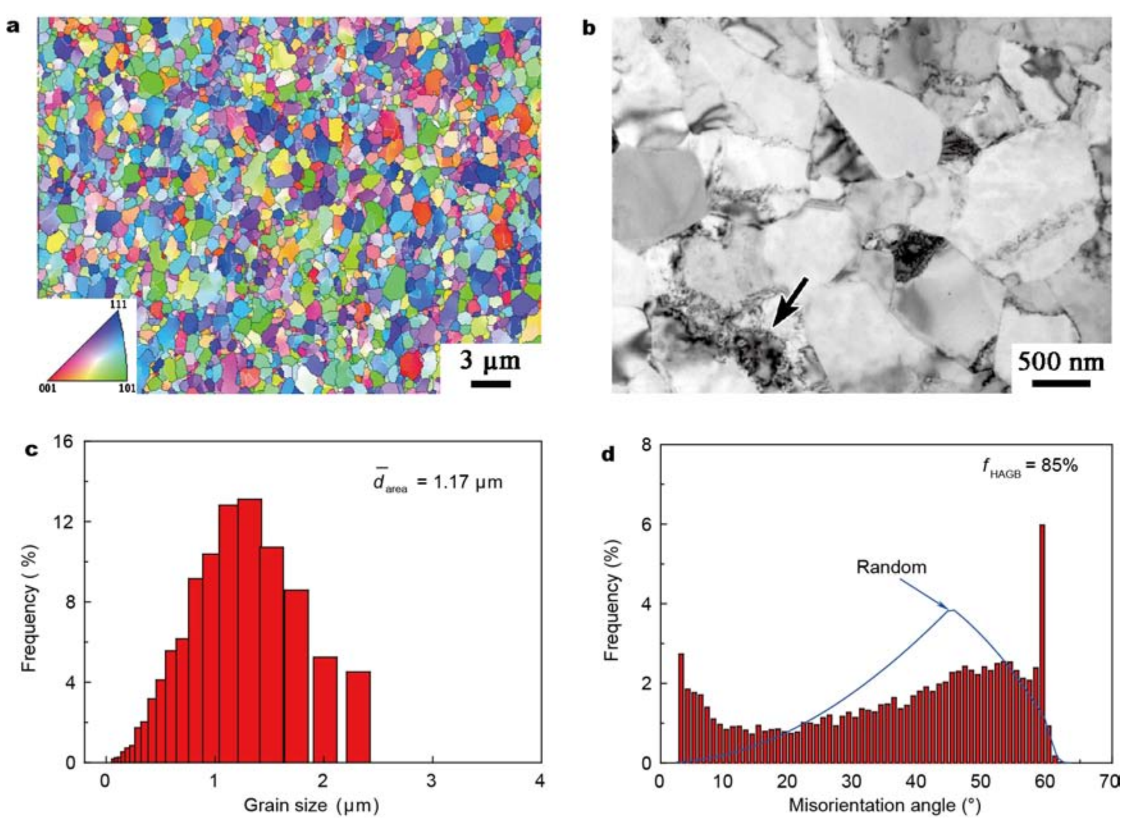

Figure 1 (a) EBSD and (b) TEM microstructures, and (c) grain size and (d) grain boundary misorientation angle distributions of FSP Cu.

most grain boundaries are sharp, clear, and relatively straight, which are characterized as HAGBs. Wavy, diffuse and ill-defined low angle GBs (LAGBs), as indicated in Fig. 1b, are also observed, which are similar to those in SPD UFG Cu [1,2,25,26]. An average grain (sub-grain) size of ~800 nm could be calculated from the TEM microstructure; however, an average grain size of 1.17 μm was calculated automatically by the grain area determination method of the TSL software, and the grain size distribution is shown in Fig. 1c. Differences from the SPD Cu, which has a low ratio of HAGBs (~60%) [26,27], a high ratio of HAGBs (85%), can be obtained in FSP Cu, which is in accordance with other FSP UFG materials [18–22]. Furthermore, the grain boundary distribution of FSP Cu is close to the random distribution of a cubic metal, as shown in Fig. 1d. From these microstructural characteristics, it is clear that FSP Cu provides an ideal model to investigate the high cycle fatigue behavior of UFG materials.

Fig. 2 shows the stress amplitude-number of cycles to failure (*S*-*N*) curves of FSP Cu and other SPD Cu as well as the CG Cu by log-log form. The ultimate tensile strength (UTS), fatigue limit (10^7 cycles), together with the calculated fatigue ratios are shown in Table 1. It can be seen that CG Cu exhibited a low fatigue limit of 60 MPa, and enhanced fatigue limits were obtained for various UFG Cu. Though a relatively low UTS of 330 MPa was achieved for FSP Cu compared to that of ECAP and HPT Cu, FSP Cu exhibited a higher fatigue limit of 120 MPa; i.e., a higher fatigue ratio (~0.36) can be obtained for FSP Cu, which was even larger than that of CG Cu.

The relationship between stress amplitude $\Delta\sigma/2$ and fatigue life $2N_f$ can be expressed by the following Basquin equation [26]:

$$\Delta\sigma / 2 = \sigma'_{\mathrm{f}}(2N_{\mathrm{f}})^{b}, \qquad (1)$$

where σ'_f is the fatigue strength coefficient and b is the fatigue strength exponent (Basquin exponent). Obviously, strong linear relationships can be observed between $\log\Delta\sigma/2$ and $\log 2N_f$ for the CG Cu and various UFG Cu

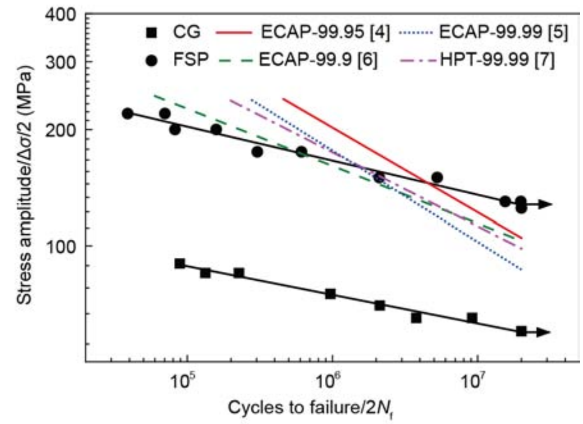

Figure 2 log-log form of stress-fatigue life (*S*-*N*) curves for CG Cu and various UFG Cu specimens [4–7].

specimens (Fig. 2), and the σ'_f and b values can be calculated from the intercept with the amplitude axis and the linear slope, respectively, as shown in Table 1.

From Equation (1), it is clear that in order to increase the high cycle fatigue strength, increased σ'_f and b values are needed. Usually, σ'_f is nearly equal to the true tensile strength after modification with necking [28]. Clearly, FSP Cu exhibited a much lower σ'_f compared to SPD Cu, due to its relatively low tensile strength (Table 1). However, a larger b value was achieved for FSP Cu, resulting in a higher fatigue limit (Fig. 2 and Table 1). In an ideal circumstance (b=0), the fatigue strength equals to σ'_f, indicating a damage immunizing microstructure during cyclic deformation. Actually, fatigue damage accumulates during cyclic deformation and the fatigue strength is always lower than the tensile strength, so b<0. To a certain extent, b represents the degree of the fatigue damage, which is associated with the initial microstructure characteristics.

Grain refinement is a double-edged sword for the improvement of fatigue strength. In the UFG materials, GBs are the dominate obstacles to impede dislocations, in which case, b decreases sharply for unstable microstructure and highly localized strain. It is found that the well-developed

Table 1 Tensile and fatigue properties of CG Cu and various UFG Cu specimens

Materials	σ_{UTS} (MPa)	Fatigue limit (MPa)	Fatigue ratio	σ'_y (MPa)	b
CG-99.98	200	60	0.30	207	−0.074
FSP-99.98	330	120	0.36	559	−0.088
ECAP-99.95 [4]	419	105	0.25	4089	−0.218
ECAP-99.99 [5]	402	85	0.21	4666	−0.237
ECAP-99.9 [6]	402	100	0.25	1329	−0.153
HPT-99.99 [7]	481	100	0.21	2238	−0.186

equilibrium HAGBs are more stable than the non-equilibrium boundaries or the low angle/sub boundaries after SPD procedures, which may contribute relatively high *b* and high fatigue strength [29,30]. Obviously, *b* value would decrease clearly in extremely unstable microstructure with high density of defects and non-equilibrium structures in the SPD materials. By contrast, stable structure with uniform equiaxed fine grains and well-developed equilibrium HAGBs was obtained in FSP Cu, resulting in the enhanced *b* value compared to that of the SPD Cu.

Fig. 3 shows the typical surface damage morphologies after fatigue for the CG Cu and FSP Cu. It is clear that very large slip bands, can be observed on the damaged surface of CG Cu, and the cracks propagated along the slip bands or the grain boundaries (Fig. 3a). Moreover, many extrusions can be observed protruding from the slip bands, as shown in Fig. 3b, which is similar to that of the single crystal Cu [28,31]. For FSP Cu, no large-scale slip bands could be observed, and no obvious crack initiating sites were found near the fracture edge. Under low stress amplitudes, i.e., for high cycle fatigue circumstances, only protrusions were observed and no slip bands were found on the fatigued surfaces (Fig. 3c). Therefore, protrusion was observed to be the main damage morphology for FSP Cu during the high cycle fatigue deformation process. Under a high fatigue stress of 200 MPa, i.e., near the low cycle fatigue conditions, few small-scale slip bands could be found in the grains with a relatively large size, although protrusion is still the main damage morphology, as shown in Fig. 3d. Although no obvious crack initiation sites were observed in FSP Cu, cracks should initiate preferentially at the GBs and protrusions which are the main surface damages.

The damage morphology of SPD Cu is very different from that in FSP Cu observed in this study [3–10]. It is well known that large-scale slip bands, several tens of micrometers in size have been observed on the damaged surfaces of SPD Cu, even at very low stress amplitudes (less than 0.5 UTS) [4,5,7,10,31]. Furthermore, Goto *et al.* [32] indicated that the slip bands formed in ECAP Cu were longer and had an inferior uniformity of distribution density at a lower stress amplitude of 120 MPa, when compared to the slip bands at a high stress amplitude of 240 MPa. Similarly, large-scale slip bands can also be observed on the damaged surfaces of HPT Cu specimens after fatigue tests [7,33]. It is well accepted that fatigue cracks easily initiate and propagate along large-scale slip bands in SPD Cu, resulting in earlier fractures with a low fatigue limit.

There is still no general consensus on the formation mechanism of large-scale slip bands in SPD UFG materials during cyclic deformation. Grain coarsening has been

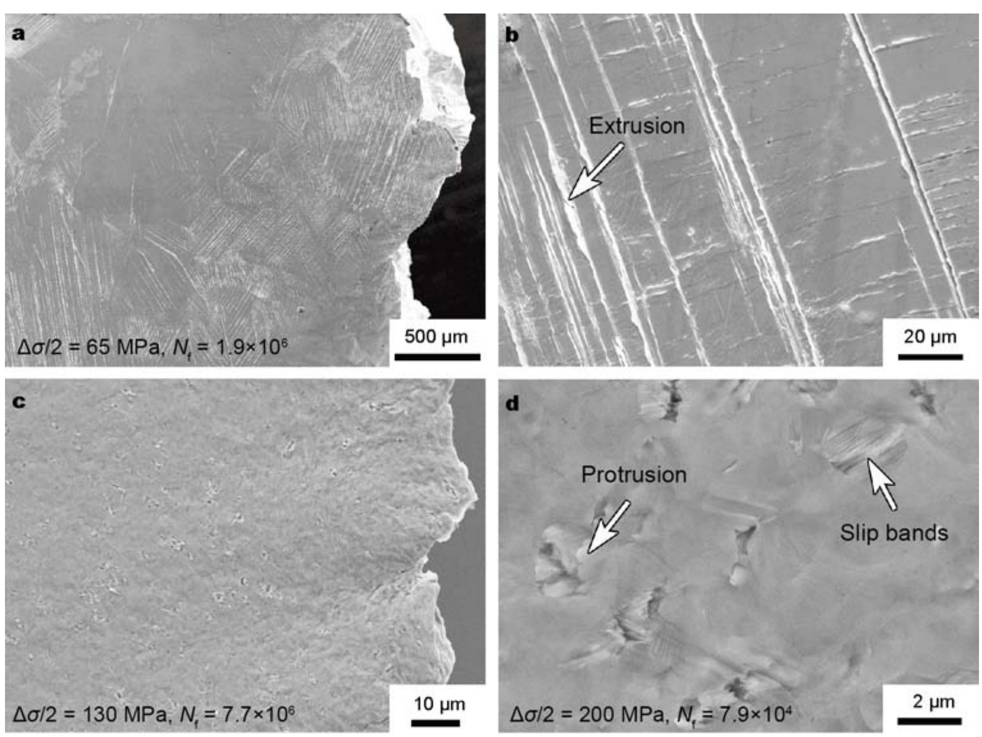

Figure 3 SEM micrographs of the damaged surfaces of (a and b) CG Cu, and (c and d) FSP Cu specimens.

taken as an explanation of slip band formation. However, this explanation carries with it the problem of a dimensional mismatch between SBs and coarse grains [3–10]. Therefore, some researchers suggest that the formation of slip bands in ECAP UFG materials may be related to the oriented distributions of defects along the shear plane of the last pressing [3,5,32].

An unstable microstructure was purported to be the intrinsic origin for the formation of large-scale slip bands. A high degree of non-equilibrium at grain boundaries with high energy, excessive volume, and long-range stress fields was proven to exist in SPD UFG materials. Grain boundary sliding assisted dynamic recrystallization easily occurred during cyclic deformation, resulting in the formation of coarse grains [3–5,8]. A strong shear texture was present in the SPD Cu due to severe shear deformation, and pre-existing slip bands are believed to exist in SPD UFG materials [1–3,5,7]. Therefore, large-scale slip bands are easy to form locally in SPD UFG materials with an unstable microstructure.

For the present FSP Cu, the increased b value significantly reduced the fatigue damage due to its stable microstructure. Usually, FSP Cu contains low dislocation density, and dislocation slip can occur during cyclic deformation, facilitating the formation of dislocation structures in the grains. More important, a uniform microstructure with high density of equilibrium HAGBs was obtained in FSP Cu. So, plastic deformation was very homogeneous during high cycle fatigue process, and no unstable area deformed preferentially. That is to say, the dislocation activity originated from plastic deformation should be restricted in the ultrafine grains of FSP Cu during high cycle fatigue process, therefore inhibiting the formation of large-scale damage morphologies.

Fig. 4 shows the microstructures after fatigue tests on FSP Cu specimens under a low stress of 130 MPa, which is near the fatigue limit of 120 MPa. From the EBSD microstructure, it is clear that a microstructure similar to the original FSP Cu (Fig. 1a) could be observed, as shown in Fig. 4a. Further, no obvious grain growth was detected, with the average grain size only slightly increasing to ~1.21 μm (Fig. 4b). TEM microstructure further proves that the grain structure retains a morphology similar to that of the original, without obvious grain growth. However, a high density of dislocations could be observed, as shown in Fig. 4c.

Grain coarsening was frequently observed in SPD Cu after fatigue tests, and regular dislocation structures (subgrain boundaries) were found in most of the grown grains, which are similar to those in post-fatigue CG Cu [3,6–8,10].

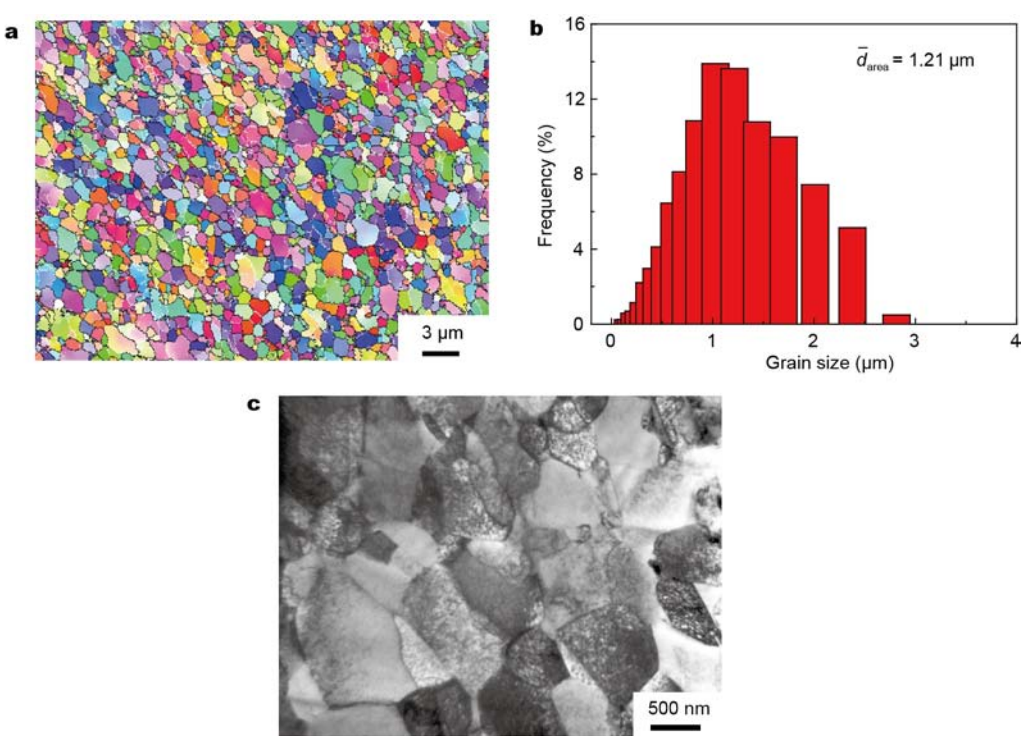

Figure 4 (a) EBSD microstructure, (b) grain size distribution, and (c) TEM microstructure of the cyclic deformed FSP Cu under a stress of 130 MPa ($N_f = 7.7\times10^6$).

However, FSP Cu exhibited very different dislocation structures after fatigue test, with the HAGBs still keeping their original characteristics and a high density of dislocation was observed in the ultrafine grains (Fig. 4c). This implies that the dislocation related activities also dominate during the high cycle fatigue deformation of the FSP UFG materials, and the ultrafine grains have a strong capability to initiate and store dislocations in their interior. In CG Cu, regular dislocation patterns in the forms of walls and PSBs can be readily observed after cyclic deformation [28]. Similarly, dislocation walls and cells can be observed in the SPD UFG Cu due to the grain coarsening [3,6–8,10]. In FSP Cu, high density of dislocations were observed in the ultrafine grains, however, well-defined patterns (high-density dislocation in the walls and extremely low-density dislocations in the interior) cannot be formed.

In summary, the present results demonstrate that enhanced high cycle fatigue properties can be achieved in FSP UFG Cu compared to that of SPD Cu, and the fatigue limit and fatigue ratio of FSP Cu were 120 MPa and 0.36, respectively. FSP Cu still exhibited a uniform equiaxed UFG structure without grain coarsening after high cycle fatigue tests, and no large-scale slip bands formed on the damaged surfaces. Instead, protrusion is the main damage morphology for FSP Cu during high cycle fatigue deformation. Further, a high density of dislocations could be observed in the ultrafine grains of FSP Cu after high cycle fatigue tests.

Received 10 May 2016; accepted 8 July 2016; published online 20 July 2016

1 Meyers MA, Mishra A, Benson DJ. Mechanical properties of nanocrystalline materials. Prog Mater Sci, 2006, 51: 427–556
2 Valiev RZ, Islamgaliev RK, Alexandrov IV. Bulk nanostructured materials from severe plastic deformation. Prog Mater Sci, 2000, 45: 103–189
3 Mughrabi H, Höppel HW. Cyclic deformation and fatigue properties of very fine-grained metals and alloys. Int J Fatigue, 2010, 32: 1413–1427
4 Goto M, Han S, Yakushiji T, *et al.* Fatigue strength and formation behavior of surface damage in ultrafine grained copper with different non-equilibrium microstructures. Int J Fatigue, 2008, 30: 1333–1344
5 Sheikh-ali AD. Coupling of grain boundary sliding and migration within the range of boundary specialness. Acta Mater, 2010, 58: 6249–6255
6 Zhang ZJ, An XH, Zhang P, *et al.* Effects of dislocation slip mode on high-cycle fatigue behaviors of ultrafine-grained Cu–Zn alloy processed by equal-channel angular pressing. Scripta Mater, 2013, 68: 389–392
7 An X, Lin Q, Wu S, *et al.* Improved fatigue strengths of nanocrystalline Cu and Cu–Al alloys. Mater Res Lett, 2015, 3: 135–141
8 An XH, Wu SD, Wang ZG, *et al.* Enhanced cyclic deformation responses of ultrafine-grained Cu and nanocrystalline Cu–Al alloys. Acta Mater, 2014, 74: 200–214
9 Pan Q, Lu L. Dislocation characterization in fatigued Cu with nanoscale twins. Sci China Mater, 2015, 58: 915–920
10 Li XW, Jiang QW, Wu Y, *et al.* Stress-amplitude-dependent deformation characteristics and microstructures of cyclically stressed ultrafine-grained copper. Adv Eng Mater, 2008, 10: 720–726
11 Malekjani S, Hodgson PD, Stanford NE, *et al.* Shear bands evolution in ultrafine-grained aluminium under cyclic loading. Scripta Mater, 2013, 68: 821–824
12 Cheng S, Xie J, Stoica AD, *et al.* Cyclic deformation of nanocrystalline and ultrafine-grained nickel. Acta Mater, 2009, 57: 1272–1280
13 Zha M, Li Y, Mathiesen RH, *et al.* Microstructure evolution and mechanical behavior of a binary Al–7Mg alloy processed by equal-channel angular pressing. Acta Mater, 2015, 84: 42–54
14 Reglitz G, Oberdorfer B, Fleischmann N, *et al.* Combined volumetric, energetic and microstructural defect analysis of ECAP-processed nickel. Acta Mater, 2016, 103: 396–406
15 Gu CF, Toth LS, Zhang YD, *et al.* Unexpected brass-type texture in rolling of ultrafine-grained copper. Scripta Mater, 2014, 92: 51–54
16 Sun LX, Tao NR, Lu K. A high strength and high electrical conductivity bulk CuCrZr alloy with nanotwins. Scripta Mater, 2015, 99: 73–76
17 Mishra RS, Ma ZY. Friction stir welding and processing. Mater Sci Eng-R-Rep, 2005, 50: 1–78
18 Xue P, Xiao BL, Ma ZY. Achieving ultrafine-grained structure in a pure nickel by friction stir processing with additional cooling. Mater Design, 2014, 56: 848–851
19 Su JQ, Nelson TW, Sterling CJ. Friction stir processing of large-area bulk UFG aluminum alloys. Scripta Mater, 2005, 52: 135–140
20 Xue P, Ma ZY, Komizo Y, *et al.* Achieving ultrafine-grained ferrite structure in friction stir processed weld metal. Mater Lett, 2016, 162: 161–164
21 Xue P, Xiao BL, Ma ZY. Enhanced strength and ductility of friction stir processed Cu–Al alloys with abundant twin boundaries. Scripta Mater, 2013, 68: 751–754
22 Chang CI, Du XH, Huang JC. Achieving ultrafine grain size in Mg–Al–Zn alloy by friction stir processing. Scripta Mater, 2007, 57: 209–212
23 Xue P, Xiao BL, Ma ZY. High tensile ductility via enhanced strain hardening in ultrafine-grained Cu. Mater Sci Eng-A, 2012, 532: 106–110
24 Xue P, Xiao BL, Zhang Q, *et al.* Achieving friction stir welded pure copper joints with nearly equal strength to the parent metal via additional rapid cooling. Scripta Mater, 2011, 64: 1051–1054
25 Dalla torre F, Lapovok R, Sandlin J, *et al.* Microstructures and properties of copper processed by equal channel angular extrusion for 1–16 passes. Acta Mater, 2004, 52: 4819–4832
26 Zhao YH, Bingert JF, Zhu YT, *et al.* Tougher ultrafine grain Cu via high-angle grain boundaries and low dislocation density. Appl Phys Lett, 2008, 92: 081903
27 Huang CX, Yang HJ, Wu SD, *et al.* Microstructural characterizations of Cu processed by ECAP from 4 to 24 passes. MSF, 2008, 584-586: 333–337
28 Suresh S. Fatigue of Materials. 2nd Edition. Cambrige: Cambrige University Press. 1998
29 Vinogradov A, Hashimoto S. Multiscale phenomena in fatigue of ultra-fine grain materials-an overview.. Mater Trans, 2001, 42: 74–84
30 Zhang ZJ, Pang JC, Zhang ZF. Optimizing the fatigue strength of ultrafine-grained Cu–Zn alloys. Mater Sci Eng-A, 2016, 666: 305–313

31 Li P, Li S, Wang Z, *et al.* Basic criterion for the formation of self-organized dislocation patterns in fatigued FCC pure metals. Sci China Mater, 2015, 58: 921–928
32 Goto M, Han SZ, Yakushiji T, *et al.* Formation process of shear bands and protrusions in ultrafine grained copper under cyclic stresses. Scripta Mater, 2006, 54: 2101–2106
33 Khatibi G, Horky J, Weiss B, *et al.* High cycle fatigue behaviour of copper deformed by high pressure torsion. Int J Fatigue, 2010, 32: 269–278

Acknowledgments This work was supported by the National Natural Science Foundation of China (51301178 and 51331008).

Author contributions Ma Z and Xiao B supervised the project. Xue P performed the experiments with help from Huang Z, Wang B, Wang W and Tian Y; Xue P wrote the paper with support from Ma Z, Xiao B and Tian Y.

Conflict of interest The authors declare that they have no conflict of interest.

Peng Xue is an associate professor at Shenyang National Laboratory for Materials Science at the Institute of Metal Research, Chinese Academy of Sciences. His research focuses on the friction stir welding and processing, and the mechanical properties of ultrafine grained materials.

Zongyi Ma is a professor at Shenyang National Laboratory for Materials Science at the Institute of Metal Research, Chinese Academy of Sciences. His research focuses on the advanced metal matrix composite, friction stir welding and processing, and superplastic deformation behavior of fine grained materials. He authored and co-authored more than 240 international journal publications with citations more than 6,300.

超细晶纯铜的本征高周疲劳行为研究

薛鹏, 黄治治, 王贝贝, 田艳中, 王文广, 肖伯律, 马宗义*

摘要 超细晶材料由于独特的组织和性能引起了广泛的关注, 然而由于在循环变形过程中极易发生应变局部化和动态再结晶, 导致产生大尺度剪切带和严重的晶粒粗化, 使得人们对超细晶材料的本征疲劳行为一直缺乏深入认识. 本研究利用搅拌摩擦加工(FSP)方法在纯铜中制备出理想的等轴超细晶组织, 对其高周疲劳行为的研究表明, FSP纯铜的疲劳极限和疲劳比与其他超细晶纯铜相比明显提高, 而且疲劳后没有出现大尺度的剪切带和严重的晶粒粗化, 疲劳损伤主要以挤出机制为主. 在超细晶尺度内, 位错相关的活动仍然占主导, 但仅局限于超细晶内部, 没有形成粗晶中常见的规则位错结构.