Microstructures and mechanical properties of a Cu–Zn alloy subjected to cryogenic dynamic plastic deformation

G.H. Xiao, N.R. Tao *, K. Lu
Shenyang National Laboratory for Materials Science, Institute of Metal Research, Chinese Academy of Sciences, Shenyang 110016, China

ARTICLE INFO

Article history:
Received 5 November 2008
Received in revised form 10 January 2009
Accepted 12 January 2009

Keywords:
Cu–Zn alloy
Dynamic plastic deformation
Nano-scale twins
Shear band
Nanograin

ABSTRACT

The microstructures and mechanical properties of a Cu–Zn alloy subjected to dynamic plastic deformation (DPD) at liquid nitrogen temperature (77 K) were investigated. The planar dislocation activities, deformation twinning and shear banding dominate the plastic deformation of samples with different strains, respectively. Twin boundary spacing decreases with strain and nearly saturates when the strain exceeds 1.2. Shear bands occur within high-density twin region at higher strains, inside which nano-sized grains are formed. Tensile tests show that the DPD brass exhibits a high strength and a limited tensile ductility. Deformation twins play an important role in the strengthening while the appearance of shear bands obviously reduces the ductility of materials.

© 2009 Elsevier B.V. All rights reserved.

1. Introduction

Plastic deformation is one of the most promising approaches to synthesize dense, contamination-free nanostructured materials. During the past decades, various plastic deformation techniques have been developed, such as cold rolling [1,2], equal channel angular pressing (ECAP) [3], high pressure torsion (HPT) [4], ball milling (BM) [5]. Previous experimental results showed that high strain rate or low deformation temperature could enhance the efficiency of plastic deformation and facilitate grain refinement [6,7]. Although numerous investigations on these deformation approaches have been carried out, considerably less attention has been paid to high-strain-rate deformation at cryogenic temperatures. Recently, a new deformation approach, dynamic plastic deformation (DPD), was developed [8,9] in order to realize high-strain-rate deformation of bulk materials at liquid nitrogen temperature (LNT). The microstructures and mechanical properties of bulk pure copper subjected to LNT-DPD have been investigated systematically [10–12]. Mixed microstructures consisting of high-density mechanical twins and nano-sized grains were induced into DPD Cu, which exhibits a high tensile strength of 630 MPa. However, little has been reported so far on the microstructures and mechanical properties of low stacking fault energy (SFE) metals and alloys subjected to high-strain-rate and cryogenic deformation.

In addition, previous work showed that annealed twin boundary (TB) was equivalent to grain boundary in strengthening when TB spacing is in a few tens of micrometers range [13]. As TB spacing is down to nanometer scale, the significant strengthening is also observed [14,15]. Recently, the strengthening effect of nano-scale twins has been investigated extensively. Nevertheless, the investigations mainly focus on nano-scale growth-in twins with coherent boundaries in thin foil samples due to the limitation of preparation techniques [14,15]. Actually, high-density deformation twins could be induced into bulk low SFE fcc materials via cryogenic and high-strain-rate deformation [16], which could provide an ideal sample to investigate the effects of deformation twins on mechanical properties. Furthermore, it has been reported that twinning plays an important role in the high strength and high ductility in low SFE fcc metals and alloys [17,18]. Hence, it is of great interest to investigate the microstructures and mechanical properties of low SFE fcc materials subjected to high-strain-rate and cryogenic deformation.

2. Experimental

The material used in this study is a commercial 68/32 brass alloy with 68 wt.% Cu and 32 wt.% Zn. Its SFE is approximated as that of 70/30 brass, being 14 mJ/m² [19]. The received samples were annealed at 700 °C in argon for 1 h and the obtained average grain size was 110 μm with a few annealing twins. Plastic deformation was conducted by means of LNT-DPD, for which the detailed set-up and procedure can be found in [9,10]. The strain rate during LNT-DPD was estimated to be 10²−³/s. The deformation strain...
was defined as $\varepsilon_t = \ln (L_0/L_f)$, where $L_0$ and $L_f$ are the initial and final thicknesses of the deformed sample, respectively. Multiple impacts were applied to obtain a pancake-like sample eventually. The treated sample was immersed into liquid nitrogen during DPD.

Microstructure characterization was performed on a Leica DMRX optical microscope (OM), Quanta 600 scanning electron microscopy (SEM) and JEOL 2010 transmission electron microscope (TEM), respectively. The cross-sectional thin foils for TEM were prepared by means of mechanical grinding and electrochemical polishing at $-10^\circ$ C. Mechanical properties were evaluated using uniaxial tensile tests at ambient temperatures. The pancake-like LNT-DPD samples were machined into flat dog-bone tensile specimens along the radial direction, with a gauge length of 5 mm and width of 1 mm. The gauge sections of the tensile specimens were polished to reduce the effect of surface defects. Tensile tests were performed on an Instron 5848 Microtester at a strain rate of $6 \times 10^{-3}$/s. The displacement of the tensile gauge section was measured using a laser extensometer with an accuracy of 1 $\mu$m.

3. Results

3.1. OM observations

Fig. 1 shows the optical micrographs of LNT-DPD samples observed in longitudinal cross-section with different strain levels. At $\varepsilon_t = 0.2$, the original equiaxed grains were deformed slightly and no obvious deformation marking was observed inside grains (Fig. 1a). TEM observations testify that the deformation is accommodated by the planar dislocations when $\varepsilon_t < 0.2$. At $\varepsilon_t = 0.6$, numerous parallel strips were observed in deformed grains (Fig. 1b). These strips usually initiate at grain boundaries and are confined inside one grain. They are proved to be clusters of deformation twins through the following TEM investigations, which is consistent with the previous result that deformation twinning only occurs after a certain strain [20]. At $\varepsilon_t = 0.8$, shear bands were developed to cut through high-density mechanical twins (Fig. 1c). They appeared as dark-etching, narrow and wavy bands aligned at about 55° to the loading direction. At $\varepsilon_t = 1.6$, the density of shear bands obviously increased and divided the material into numer-
ous rhomboidal prisms (Fig. 1d). Obviously, the whole deformation process can be classified into three stages. At stage I ($\varepsilon_t = 0–0.2$), the deformation is mainly accommodated by dislocation slip. At stage II ($\varepsilon_t = 0.2–0.8$), deformation twinning is activated and becomes the dominant microstructure. At stage III ($\varepsilon_t = 0.8–1.6$), the profuse shear bandings start to carry the plastic deformation. The various microstructures are induced into DPD sample at three different stages, which will be described in following sections based on the detailed TEM observations.

3.2. TEM observations

3.2.1. Dislocations and deformation twins

Fig. 2a shows the planar dislocations at $\varepsilon_t = 0.2$ and the corresponding selected area electron diffraction (SAED) pattern. Because of the low SFE in 68/32 brass, the large separation between partial dislocations inhibits the cross-slip and causes dislocations to organize themselves into planar arrays or planar slip bands.

In addition to dislocation substructures, SFE can also affect the propensity of twinning. In low SFE metals, twinning is more favorable than slip [21]. When $\varepsilon_t > 0.2$, it is found that deformation twins are activated and twinning is the major deformation mode at stage II. The twins divide the grains into twin/matrix lamellae. As deformation proceeds, new twins are activated to accommodate deformation so that the twin density increases gradually until it reaches saturation state [16], as shown in Fig. 2b–d.

3.2.2. Shear bands

Fig. 3 presents typical TEM images of shear bands in DPD samples with different strain levels. The initial stage of shear band (Fig. 3a) shows the severe twist of TBs, which indicated that this narrow band was subjected to intense localized shearing. At the initial stage, the width of narrow bands is about 100 nm. As deformation continues, shear bands broaden and twin structures within bands are disrupted, as indicated in Fig. 3b–d. The continuous ring patterns in SAED indicate the occurrence of grain refinement within shear bands (Fig. 3c–d). The statistical results of the width of shear bands are shown in Fig. 4. A total of 200 shear bands were measured to obtain each average value. The average width of shear bands increases from 223 nm at $\varepsilon_t = 0.8$ to 960 nm at $\varepsilon_t = 1.6$. Meanwhile, the distribution range of band width broadens from 100 to 925 nm at $\varepsilon_t = 0.8$, 100–1450 nm at $\varepsilon_t = 1.2$ to 100–2000 nm at $\varepsilon_t = 1.6$. It is
suggested that new shear bands form continuously accompanied with broadening of pre-existing bands with the increasing strain. Hence, the density of shear bands gradually increases and the interval between them obviously decreases with strain.

Another important feature is the grain refinement in shear bands, as seen in Fig. 3c and d. Most of the grains in shear bands are elongated along shear direction and have a plate shape. Therefore, the sizes of grains in both longitudinal and transverse direction were measured as grain length and width, respectively. About 250 grains obtained from TEM dark-field (DF) images were measured. As presented in Fig. 5, with the shear band thickness increasing, the grain length reduces steeply from 190 nm to 65 nm, while the grain width changes a little from 39 nm to 47 nm. The aspect ratio of the grains also decreases from 4 to 1.7. It is well known that the shear strain within shear bands is proportional to the thickness of shear bands. The higher shear strain in wider shear band leads to the decrease of grain length. Meanwhile, the little change in the width of grain finally results in the grains with a smaller aspect ratio, indicating that the microstructure within shear bands develops from elongated grains into roughly equiaxed ones.

Additionally, some isolated nanograin (NG) regions were also observed at stage III. Fig. 6 presents the typical TEM micrographs of these NG regions and the corresponding statistical results of grain sizes. The average grain width and length are 38 nm and 63 nm, respectively. Interestingly, both average grain sizes in these isolated NG regions are close to those in the widest shear bands. Meanwhile, it is noted that most of the grains are aligned along shear direction (Fig. 6b). All above results indicate that these isolated NG regions may derive from the thicker shear bands whose borders are out of sight in TEM observations.

Fig. 7 summarizes the TB spacing ($\lambda$), grain length ($D_L$), grain width ($D_T$), the volume fraction of twinned regions ($f_{\text{twin}}$) and the volume fraction of NG regions ($f_{\text{NG}}$) in LNT-DPD samples with different strains. With the increasing strain, the average TB spacing decreases from 48 nm at $\varepsilon_t = 0.2$ to 14 nm at $\varepsilon_t = 1.6$. Additionally, it is noted that TB spacing almost saturates when strain exceeds
1.2. The grain size also decreases with strain and the grain width $D_T$ gradually approaches a saturated value of 39 nm. Both $f_{\text{tw}}$ and $f_{\text{NG}}$ are obtained from the statistical results through TEM observations. The $f_{\text{tw}}$ increases with strain at stage II and is up to peak at $\varepsilon_t = 0.6$. When the deformation proceeds into stage III, the $f_{\text{tw}}$ starts to decrease with strain due to the occurrence of shear banding within twinned regions while $f_{\text{NG}}$ starts to increase rapidly at this stage. The rapid increase of $f_{\text{NG}}$ at stage III can be attributed to grain refinement in shear bands.

3.2.3. Final mixed structures
At $\varepsilon_t = 1.6$, the microstructure is composed of NGs within shear bands and dispersed nano-scale twin bundles. Fig. 8a presents a typical micrograph of mixed structures and its corresponding schematic graph is also given in Fig. 8e. A series of SAED patterns from the diffraction pattern of twin to continuous ring pattern (Fig. 8b–d) indicated the twin structures were gradually replaced by the NGs inside shear bands. Also, the SAED pattern (Fig. 8d) of continuous ring indicated that larger misorientations were induced between the NGs. Obviously, the mixed microstructures of NGs ($D_L = 65$ nm, $D_T = 39$ nm) and nano-scale deformation twins ($\Delta = 14$ nm) can be obtained in brass by means of LNT-DPD.

3.3. Mechanical properties
Engineering stress–strain curves of LNT-DPD brass with different strains are presented compared with coarse-grained (CG) brass in Fig. 9a. It is obvious that the LNT-DPD sample exhibits much
higher strength than that of CG counterpart. With the increasing strain, the yield strength \(\sigma_y\) and ultimate tensile strength \(\sigma_b\) increase from 248 MPa, 370 MPa at \(\varepsilon_t = 0.2\) to 717 MPa, 783 MPa at \(\varepsilon_t = 1.6\), respectively. However, for the CG sample, the \(\sigma_y\) is 80 MPa and the \(\sigma_b\) is 256 MPa. Fig. 9b summarizes the tensile results of LNT-DPD samples with different strains. The significant reduction in the uniform elongation is clearly seen. The uniform elongation decreases from 50% in CG sample to less than 5% when \(\varepsilon_t > 0.8\). It is believed that the lack of strain hardening leads to the poor uniform elongation, making the tensile curves peak rapidly after yielding.

Fig. 10a–b compares the SEM micrographs of the fracture surface of LNT-DPD and CG brass. It can be seen that the fracture patterns of both of them are characterized by dimples, suggesting that the LNT-DPD sample also deforms in a ductile manner. However, the dimples in LNT-DPD sample are smaller and smoother than those in CG sample, corresponding to the worse ductility of LNT-DPD sample. A view of the side flat face of the fractured LNT-DPD brass is shown in Fig. 10c. The necking, as indicated by the horizontal arrows, is observed obviously. The cross-sectional area of LNT-DPD brass was reduced to about 60% compared with DPD Cu [8]. It is supposed that the necking-controlled failure would lead to the fracture surface perpendicular to the applied stress [22]. However, the fracture surface at the edge of the flat specimen aligns at about 55° to the tensile axis, indicating that shear bands may play an important role in the fracture of LNT-DPD brass [23].

4. Discussion

4.1. Shear banding within high-density twins

In LNT-DPD brass samples, shear bands always form in the twinned regions where the TB spacing is very thin (Fig. 3a). When deformation strain is larger than 0.8, shear banding becomes the dominant deformation mode instead of deformation twinning. As mentioned in [16], the propagation of deformation twins is achieved by continuous insertion of twin layers into the pre-existing TB spacing. When the twin density is saturated, it is quite difficult for deformation twins to nucleate and grow. In order to accommodate the following strain, shear banding is activated in some local areas with great stress concentration and thermal softening.

According to the Considère criterion, inhomogeneous deformation such as necking and shear banding sets in when, 
\[
\left(\frac{d\sigma}{d\varepsilon}\right)_\sigma \geq \sigma
\]
where \(\sigma\) and \(\varepsilon\) are true stress and strain, respectively. This equation indicates that sufficiently large strain hardening need to be present to sustain the uniform straining before the onset of localized deformation [24]. For LNT-DPD samples, the strain hardening capacity reduces significantly at stage III where the average TB spacing decreases below 20 nm (see Figs. 7 and 9). Hence, the lack of strain hardening may lead to the poor ductility and fracture behavior of LNT-DPD brass.
of strain hardening promotes the enhanced tendency for plastic instabilities, i.e. shear banding.

At the initial stage of shear band development, intense plastic deformation takes place within a narrow band and TBs are strongly twisted (Fig. 3a). As twin structure is disrupted, elongated dislocation cells can form along shear direction. As deformation continues, they would break up into roughly equiaxed subgrains accompanied by increasing misorientations to accommodate the strain and to obtain equilibrium with the increasing flow stress (Fig. 3c and d). Equiaxed ultrafine subgrains can further increase misorientations by means of rotation, resulting in formation of random oriented NGs. This grain refinement process is called rotational dynamic recrystallization in [25]. Dependent upon the strain level, the structures within shear band vary from elongated dislocation cells to equiaxed NGs, as seen in Figs. 3 and 7.

From Fig. 3, it is found that the shear bands grow in thickness as strain increases, which is consistent with the observations in [26]. Since shear band is soft area compared with twin matrix, the following deformation will be accommodated preferably by the “soft” shear bands. As grain refinement leads to hardening of shear band, the adjacent twin matrix is sheared to accommodate intense plastic deformation, resulting in the growth of shear band thickness. The shear strain is proportional to the thickness of shear bands and increases with strain [26], responsible for the grain refinement within shear bands [25]. This grain refinement is mainly accomplished by an obvious decrease in the size of long axis while the size of short axis changes a little. The sizes of both long axis and short axis of refined grains gradually tend to their stable values (about 65 nm and 39 nm), much smaller than those of ECAP Cu (100–200 nm) [27], which might be attributed to high shear strain, high strain rate and cryogenic deformation temperature in LNT-DPD brass.

It should be noted that the material nature also plays an important role in the structural evolution within shear bands. The grain coarsening was observed in shear bands of DPD Cu while grain refinement in the present sample. This contradiction may be attributed to the difference in recrystallization temperature between Cu–Zn alloy and Cu. The formation and thickening process of shear bands are always accompanied by severe plastic deformation and adiabatic heating. The severe plastic deformation can lead
to grain refinement while the adiabatic heating causes grain coarsening. It was found that the transient temperature within shear bands was close to 500 K [25]. Such a high thermal pulse may induce dynamic recovery or recrystallization in DPD Cu whose recrystallization temperature is about 423 K [28]. However, for DPD Cu–Zn alloy, the size of grain in shear band is less susceptible to the thermal pulse due to higher recrystallization temperature 580 K which will be shown in our other results.

4.2. Twin boundary strengthening

The yield strength of material as a function of grain size can be described by a Hall-Petch (H-P) relationship [29]:

$$\sigma_y = \sigma_0 + kd^{-1/2}$$  

(1)

where $\sigma_y$ is the yield strength of material, $\sigma_0$ and $k$ are constants, and $d$ is the grain size. It was found that H-P type relationship was also applicable to the materials with micro-sized twins [30]. For instance, deformation twins in shock-loaded metals contributed to the residual yield stress through a H-P relationship in the form of $\sigma \sim \Delta^{-1/2}$, where $\Delta$ was the TB spacing [30]. When TB spacing decreases into nanometer regime, TBs can also offer significant strengthening in [31]. By means of LNT-DPD, bulk brass samples with high-density nano-scale deformation twins were produced. The LNT-DPD brass exhibited high yield strength of 717 MPa, obviously higher than that of cold-rolled brass [32]. This significant strengthening is believed to be attributed to the nano-scale deformation twins whose average TB spacing is about 14 nm. However, it is noted that the LNT-DPD brass exhibits smaller strength than the electrodeposited Cu with growth-in twins [34], although they have similar TB spacing. This may be due to the following factors: (1) unlike growth-in twin boundaries, the boundaries of deformation twins are not perfectly coherent, which may facilitate dislocation slip and decrease the strengthening; (2) as mentioned in Section 3.2.3, the final microstructure of LNT-DPD brass is a mixed structure. Deformation twins account for only about 30% of the total volume. Shear banding inside deformation twins decreases the twin strengthening, which will be discussed in the following part.

4.3. Effect of shear bands on mechanical properties

Shear banding is a much complex phenomenon that involves localized adiabatic heating with the associated thermal softening, leading to extreme localization of strain along certain narrow bands. In LNT-DPD brass, profuse shear bands were observed in the high-density twins after significant homogeneous deformation. These shear bands have an effect of softening on the matrix, which includes geometrical softening and thermal softening. First, the geometrical softening occurs as a result of lattice reorientation. For metallic polycrystalline, the transmission of shear bands across the grain boundaries is often favored by lattice rotations which reduce the grain boundary misalignment and provide easy paths for the transmission of shear bands [33]. Second, the shear banding process is essentially adiabatic and the thermal excision associated with this process can be significant. The temperature rise can be estimated by assuming that 90% of deformation work is converted into heat [25],

$$dT = \frac{9}{10c_0} \varepsilon \sigma$$

(2)

where $c$ is the density of sample, $\varepsilon$ is the heat capacity of sample, $\sigma$ is the flow stress and $\varepsilon$ is the flow strain. According to (2), the temperature in brass samples after LNT-DPD is in the range of 200–300 K. However, it should be noted that the calculated temperature rise is for the overall specimen. Since the localized shear strain within shear bands is much larger than that in the overall sample, the local temperature within shear bands is high enough to cause thermal softening [25]. On the other hand, when shear banding happens, the pre-existing twin lamellae are replaced by the elongated NGs inside shear bands. The average NG width is about 39 nm, much larger than the pre-existing TB spacing 14 nm. The grain coarsening may also play an important role in the softening.

The uniform elongation of LNT-DPD samples reduces significantly at stage III ($\varepsilon > 0.8$) where shear bands become dominant microstructures (Fig. 9b). It is suggested that these profuse shear bands were deleterious to the overall ductility of LNT-DPD samples. The occurrence of shear bands confines the plastic deformation in local regions and correspondingly suppresses the uniform deformation of the whole sample, which is deleterious to the plasticity. Shear bands accommodate strain preferentially compared with the adjoining harder twin lamellae, which leads to broadening of shear bands and accelerates the evolution from microscopic shear bands into macroscopic ones. It is well known that macroscopic shear bands are preferential crack nucleation sites and fragmentation of materials always occurs along the shear band propagation path [33,34], which is also indicated in our results (Fig. 10c).

5. Summary

LNT-DPD induces high-density deformation twins and nanograins into bulk Cu–Zn alloy. Planar dislocation activities, deformation twinning and shear banding dominate the plastic deformation with increasing strain, respectively. TB spacing decreases from 48 to 14 nm with increasing strain and steps into saturated state when $\varepsilon \sim 1.2$. Shear bands occur within high-density twins at higher strains, inside which intense shear deformation causes transformation of twin/matrix lamellae into NGs. As strain increases, the thickness of shear bands broadens accompanied with a reduction of NG sizes. The mixed microstructures of nano-scale twins and NGs exhibit yield strength of 717 MPa and a limited uniform elongation less than 5%. It is suggested that deformation twins play an important role in the strengthening while the occurrence of shear bands obviously reduces the ductility of materials.

Acknowledgement

Financial support from the National Natural Science Foundation of China (Grants No (Grants Nos. 50671106, 50431010 and 50071061), Shenyang Science & Technology Project (Grant No. 1071107-1-00) and the Ministry of Science and Technology of China is acknowledged.

References