Materials Science & Engineering A 703 (2017) 470–476

Contents lists available at ScienceDirect

Materials Science & Engineering A

journal homepage: www.elsevier.com/locate/msea

Grain refinement and nanostructure formation in pure copper during cryogenic friction stir processing

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ARTICLE INFO

Keywords:
Copper
Grain refinement
Microstructure evolution
Nanocrystalline materials
Cryogenic friction stir processing

ABSTRACT

Two different mechanisms in term of dislocation manipulation were identified for the microstructure evolutions in pure copper treated by cryogenic friction stir processing (CFSP) at different parameters. The final average grain size in the nuggets increased first and then decreased with increasing rotational speed. In the conditions of low rotational speed, the refined grains evolved from dislocation cells. In the conditions of high rotational speed, the refined grains transformed from the subdivision of nanoscale lamellar structures combined with the effect of dynamic recovery. The high proportion of nano grains in high rotational speed conditions should be attributed to the large strain from the strong constrain effect of "hard shell" structure around the nugget during CFSP.

1. Introduction

Numerous investigations have demonstrated the considerable advantages of the mechanical properties of ultrafine-grained (UFG) or nanostructured materials compared with conventional coarse-grained structures. Of the many techniques used for processing fine-grained materials directly in bulk form, severe plastic deformation (SPD), such as equal channel angular pressing (ECAP) [1], accumulative roll bonding (ARB) [2] and high-pressure torsion (HPT) [3], has been considered to be a promising route. It was also found that twinning or dynamic recrystallization (DRX) were responsible for the grain refinement during these processes [4,5].

In addition to the conventional SPD technique, friction stir processing (FSP), which is derived from friction stir welding (FSW) in principle, have been widely used in some metallic materials as a microstructure modification technique in the past decades [6–8]. Usually, the grains can only be refined down to 1–5 μm after traditional FSP because the grain growth is inevitable under higher temperature state in the FSPed region (referred as "nugget" here). Meanwhile, continuous or discontinuous DRX is responsible for the formation of the fine grains because of the inherently involve severe plastic deformation as well as the transients and gradients in strain, strain rate and temperature [9–11]. Such transients and gradients strongly depend on the processing conditions and governing factors include tool design, processing parameters and imposed cooling rates [12].

Successful microstructural refinement below the ultrafine scale rang (< 1 μm) via "cold" FSP combined with rapid heat sink **have** been achieved [9,13–15]. The grains smaller than 200 nm (even nanoscale crystallites) without dislocations and separated by high-angle grain boundaries (HAGBs) were detected in the nuggets. For example, Su et al. found small crystallites with a few tens of nanometers initially formed around the pin tool during FSP of copper conducted with continuous dry ice quenching [12]. These small crystallites rapidly finally transform to randomly oriented equiaxed grains by shape adjustment rather than DRX. Fonda et al. employed a stop-action technology combined with water quenching in FSP to "freeze in" the dynamic microstructures around the tool for Al–Li 2195 alloy [14]. They found that the subdivision of original grains companied with dynamic recovery accounted for the grain refinement without the need to invoke DRX mechanism. It is necessary to investigate the refinement mechanism during such "cold" FSP under a broader range of processing parameter. Nevertheless, the smaller fraction of the nanostructures formed in the "cold" FSP limits in-depth investigation on the evolutions of nanoscale structures. Fortunately, by using our special shoulderless stir tool combining with liquid nitrogen cooling during FSP, defined as cryogenic friction stir processing (CFSP), a mixture microstructure of nano grain (NG) and UFG has been successfully prepared in pure copper [16]. The average grain size is 109 nm with 45% of NG. The nanostructures were detected without dislocations and separated by HAGBs in the nugget. The mechanical properties of the UFG/NG mixture structured Cu exhibited an enhanced ultimate tensile strength and high tensile elongation.

The objective of the present work focuses on the microstructure evolution and nanostructure development in pure copper subjected to

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http://dx.doi.org/10.1016/j.msea.2017.07.090
Received 12 June 2017; Accepted 27 July 2017

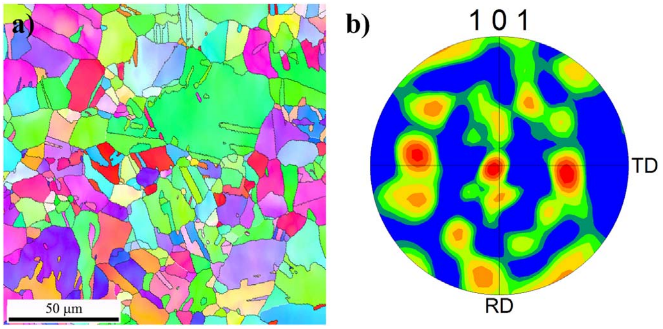

Fig. 1. (a) Orientation map and (b) (1 0 1) pole figure constructed by EBSD data of the starting base metal. High angle boundaries (> 15°) and low angle boundaries (< 15°) are sketched by black and white lines, respectively. Pole figure implies {1 1 1} ⟨1 1 2⟩ texture.

CFSP. It is of significance for both the fundamental understanding and technological applications of the CFSP process.

2. Experimental

A polycrystalline copper plate (120 mm × 40 mm ×3 mm) with a purity of 99.95% was chosen as experimental material. The plate was annealed at 623 K for 1 h before processed. The starting base metal sample for Electron backscattered diffraction (EBSD) examination was mechanical polishing, followed by electrolytic polishing in electrolyte (H_3PO_4: H_2O = 3:7) at ambient temperature. The characterization was conducted by the EBSD technique using a TSL OIM Analysis 7 software. The orientation map and pole figure of the annealed microstructure of base metal are shown in Fig. 1. The grains exhibit {1 1 1} ⟨1 1 2⟩ texture. The average grain size is about 15 μm and the fraction of HAGBs (> 15°) is of ~ 95%.

The details of the CFSP technique can be found in Ref. [16]. In the present investigation, four different rotational speeds (i.e., 400, 600, 800 rpm and 1200 rpm) were used. And the tool's travel speed was 20 mm/min for all samples. The as-processed plates were sectioned parallel to the top surface about in the middle of the plates and the sampling positions were illustrated in Fig. 2.

EBSD technology has been used to reveal the microstructure evolution by investigating the structure surrounding the tool used in FSW or FSP [14,17,18]. The subgrains transformation with greater misorientations close to the tool were usually observed. However, it should be pointed out that the boundaries with the misorientation angle < 2° are excluded for statistics due to the uncertainty in EBSD [19].

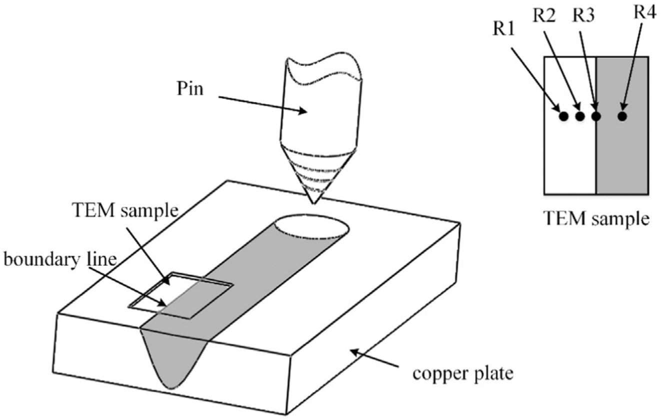

Fig. 2. Schematic illustration of the sampling position and the positions scanned by TEM in the TEM sample defined as region R1, R2, R3 and R4, respectively. R1, R2 and R3 are in the edge of the boundary line and the distance between the regions is about 50 μm, and R4 is in the interior of nugget.

Moreover, the grain structures very close to the tool were hard to characterize by the usual EBSD technology. If the observed structures is far away from the tool, the grain structures is not the real case undergone the severe plastic deformation in the nugget. It has been reported that for the metals with medium or high stacking fault energies, such as Fe, Cu and Al, the coarse grains are refined upon continued straining by various dislocation activities [20]. It is believed that the microstructure evolution can begin with dislocations motion usually in small misorientation. Hence, we use transmission electron microscope (TEM) to examine the dislocation motions in deformed Cu very close to the pin. It has been found that there is a clear boundary line between the nugget and base metal [16]. Therefore, different observed regions close to the boundary line, defined as R1, R2, R3 and R4 (as labeled in Fig. 2), were determined to reveal the microstructure evolution of the nugget. Disks with 3 mm in diameter corresponding to the above four positions were cut from the CFSP sample and thinned at 273 K with a twin-jet polisher under the condition of 12 V using a solution of 30% HNO_3 in ethanol. TEM observations were carried out on JEOL-2010 transmission electron microscope at a voltage of 200 kV. The grain size, grain boundary character and orientation data of the nuggets of the different samples were determined by an ASTAR™ system installed in the NanoMEGAS Precession Electron Diffraction (PED) platform. The details of how the NanoMEGAS platform works can be found in Ref. [21]. The PED scanning was operated with a 0.3° procession angle and a scanning step size of 3 nm, in the same size regions of interest, which were 1.2 μm × 1.2 μm. After scanning, the data was converted for analysis using TSL OIM Analysis 7 software.

3. Results

3.1. Crystallographic textures

The evolutions of crystallographic textures under different processing parameters can provide insight into the development of grain structure during CFSP of pure copper. Fig. 3 shows the orientation data arranged as (1 1 1) and (1 0 1) pole figures derived from the PED at R3 region of 1200-20 sample and the nuggets of 400-20 and 1200-20 samples, respectively. Similar to the case in FSW, the predominant deformation of the CFSP is expected to be simple shear, which is typically displayed in pole figures with a vertical shear plane-normal (SPN) and a horizontal shear direction (SD). The SD is tangential to the tool surface and the SPN is perpendicular to that surface, which aligns the rotation direction with the inclined vertical surface of the tool [22].

From the Fig. 3a, it can be found that the textures of R3 region of 1200-20 sample can be described in terms of $A/\bar{A}$ {1 1 1} ⟨1 1 0⟩ simple shear texture by contrasting with the exact pole figures in Fig. 3d, distinct from that of {1 1 1} ⟨1 1 2⟩ texture of the base metal (see Fig. 1b). The similar textures is also detested in the R3 region of 400-20 sample. This kind of shear texture was frequently observed in the nuggets near the pin of the FSW by a stop action technique or with a rapid heat sink as reported by Mironov et al. for S31254 super austenitic stainless steel [23] and by Xu et al. [18] and Mironov et al. [17] for pure copper. The formation of shear texture in R3 region is attributed to the large shear strain and high strain rate during CFSP. Moreover, this $A/\bar{A}$ shear texture is also detected in the nuggets of 400-20 and 1200-20 samples, respectively (see Fig. 3b and c). The experimental peaks are all deviated from the ideal $A/\bar{A}$ positions in all cases, but can be brought into very close correspondence by suitable adjusting rotations. These additional required rotations indicate the influence of rigid rotation of material and the complexity of the flow field close to the pin during CFSP [11].

As well known, the texture observed in the nuggets are typically weak and the grain orientation is of random distribution in some parameters of FSW/FSP [24,25]. In addition, the cube recrystallization texture component, which is different from the texture close to the pin, was also found in some nuggets [18,26]. In the present work, the