MICROSTRUCTURAL STABILITY OF ULTRAFINE GRAINED COPPER AT ELEVATED TEMPERATURE

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Abstract
Thermal stability of ultrafine grained (UFG) structure of 99.9% pure copper produced by eight equal channel angular pressing (ECAP) passes was studied. The annealing experiments were conducted at temperature range from 180 °C to 300 °C in a tube furnace under argon as a protecting gas. The dwell times were in the range from 10 min to 300 min. The electron backscattering diffraction (EBSD) analyses were performed before and after annealing at exactly the same area in order to quantify the changes of UFG structure. More advanced analysis of the EBSD results based on kernel average misorientation (KAM) parameter was performed.

Keywords: ultrafine grained microstructure, copper, microstructural stability, electron backscattering diffraction (EBSD)

1 Introduction

Microstructural stability of ultra-fine grained materials prepared by severe plastic deformation (SPD) is one of the most discussed problems. It was found that the grain boundary structure of different materials is sensitive to elevated temperatures [1]. However, information about the stability of UFG microstructures is still poor; there is a number of opened questions about the details of grain coarsening mechanisms.

At elevated temperature, the grain coarsening of UFG structures generally occurs [2]. Changes in grain size and formation of bimodal structures, as well as changes of amount of low angle boundary (LAB) and the high angle boundaries (HAB) were reported [3, 4].

Evolution of bimodal structure results in changes of mechanical properties like tensile characteristics, microhardness and fatigue properties [4-6]. Susceptibility to temperature activated processes leading to grain coarsening (normal or abnormal) depends on the level of strain introduced during SPD process.

Currently, there is no sufficient description of the effect of elevated temperature on the microstructure of UFG materials, especially those prepared by SPD techniques. One of the factors frequently deemed as influencing the high temperature behaviour of UFG materials is the materials' purity, which is based on a fact that alloys have higher thermal stability than pure metals. Other factors likely to affect the UFG materials' behaviour are the details of grain boundary arrangement.

The EBSD technique is a method used for the assessment of microstructural changes of UFG materials prevalently, together with the transmission electron microscopy (TEM). Most of the assessments are done conventionally, i.e. by comparison of different samples before and after exposure (the technique of TEM sample preparation does not even allow any other procedure, putting aside the "in-situ" observation). This approach can lead to misinterpretation of the level of microstructural changes that occur during thermal exposure due to the inherent inhomogeneity of the SPD-prepared materials. This bias can be to some extent reduced by employing the technique of observation of the same place before and after exposure.

The goal of this article is to assess the thermal stability of microstructure of UFG copper by means of the EBSD technique using the "site-specific" approach.

2 Material and Experimental Methods

UFG copper of commercial purity (99.9 %) was used in this study. Cylindrical billets of 20 mm in diameter and 120 mm in length were processed by eight passes of ECAP using route B_C. After ECAP process, the billets were turned to 16 mm diameter and 100 mm length. Specimens with cross section approximately 4×4 mm and length of 10 - 16 mm were machined from the billets. Specimen surface, parallel to the longitudinal axis of the billet was carefully mechanically and finally electrolytically polished to enable the EBSD application. Electropolishing was performed in phosphoric acid-ethanol-water-isopropyl alcohol solution at 10 V with holding time of 60 s. The temperature of the electrolyte varied between 9 and 13 °C.

Annealing experiments were conducted in the temperature range from 180 to 300 °C in a tube furnace under argon as a protecting gas. The dwell times were in the range of 10 to 30 min. Temperature of the sample was controlled by Pt 100 thermal probe immersed into a shallow bore inside a specimen. Specimens were inserted to the preheated furnace and then, upon reaching the required temperature of the specimen, the dwell time was measured. After thermal exposition, the specimens were cooled down in the cold zone of the tube by argon flow.

The EBSD analyses were made before and after annealing at specific areas in order to quantify the degree of decomposition of the UFG structure. EBSD analyses were carried out with a Philips XL 30 scanning electron microscope with TSL EBSD system. Area of the EBSD scans was 30×60 μm with 0.15 μm step. The grain sizes were calculated as diameter of a circle with the same area as the observed grain.

3 Results and Discussion

It follows from the comparison of the microstructure in the same area before and after annealing (180 °C/10 min./argon) that only slight redistribution in misorientation angles occurred by modest increase in occupancy of the LAB class (below 15°) from 69.42 % to 70.94 % on account of the HAB classes (over 15°) – table 1. The annealing affected also the local average misorientation (KAM parameter) manifested by detectable shift towards lower misorientation values and by slight increase in number fraction of the classes 0° – 0.1° and 0.6° –1.1°. The grain size distribution also changed a little by redistribution of grain size classes' occupancy while the average grain size remained almost the same. Annealing at 180 °C for 10 min caused no considerable effect in texture because the texture intensity value changed negligibly [7].

Based on the results obtained it can be stated that annealing at 180 °C – 225 °C lead neither to considerable changes in the grain size nor to notable misorientation angle redistribution for all investigated dwell times. The annealing led only to moderate changes in texture and local lattice distortions (measured by the KAM parameter). The bimodal structure generation started to occur at the temperature 250 °C after 30 min dwell, when the localised grain coarsening was detected – **Fig. 1** and **2** – which became more pronounced with increasing dwell – see **Fig. 3** and **4**. Remarkable misorientation angle redistribution took place at the same time (increase in HAGB from 29.02 % to 31.82 %). KAM parameter also underwent gradual redistribution in classes below 1° resulting in a drop in average KAM value from 1.081° (as pressed) to 0.931° (annealed for 250 °C/300 min.), which means decrease in local lattice distortion level. Increasing the dwell at 250 °C did not result in erasure of the ECAP-generated texture, only partial changes in distribution of the preferred orientations occurred.

Table 1 Low and high angle boundary fraction in as-pressed state and after annealing

180 °C/argon	dwell time [min.]			
	10	30	60	120
state	LAB/HAB [%]	LAB/HAB [%]	LAB/HAB [%]	LAB/HAB [%]
as received	69.42/30.58	68.55/31.45	76.95/23.05	74.79/25.21
annealed	70.94/29.06	72.95/27.05	81.28/18.72	75.39/24.61

Table 2 Low angle and high angle boundary fraction in as-pressed state and after annealing

300 °C/argon	dwell time [min.]	
	60	120
state	LAB/HAB [%]	LAB/HAB [%]
ECAP	72.36/27.64	76.25/23.75
annealed	37.78/62.22	25.79/74.21

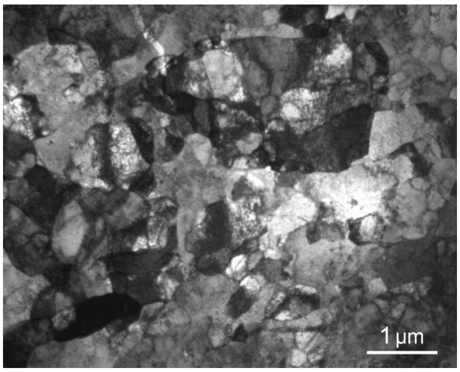

Fig.1 Microstructure after annealing – region of fine graines (250 °C/30 min), TEM

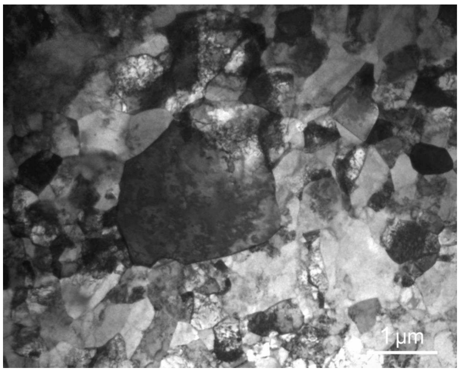

Fig.2 Region of bimodal structure after annealing (250 °C/30 min), TEM

Considerable changes took place just during annealing at 300 °C when substantial grain growth was observed, diminishing the LAB fraction and increasing the HAB content. Quantitatively, the 60 min. dwell resulted in HAB fraction increase from 27.63 % to 62.22 % (**Fig. 6**) and the 120 min. one in the increase from 23.74 % to 74.21 % (**Table 2**); this trend is in accordance with [3] and [8]. The local lattice distortion expressed in average KAM value also decreased from 1.1° in as-pressed state to 0.47° after annealing 300 °C/60 min. (**Fig. 7**) and from 1.2°to 0.39° for the case of 300 °C/120 min. annealing. Increase in annealing temperature from 250 °C to 300 °C led to deep changes in texture in all three investigated directions RD, TD, ND.

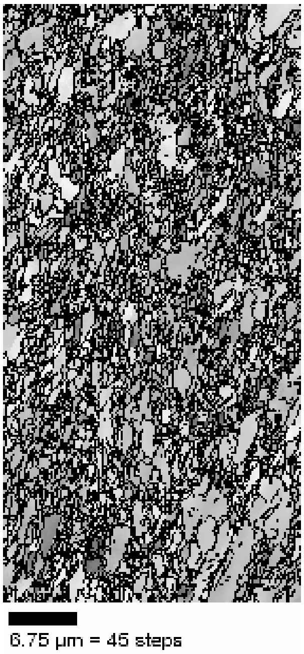

Fig.3 IPF map with grain boundary network – as received state

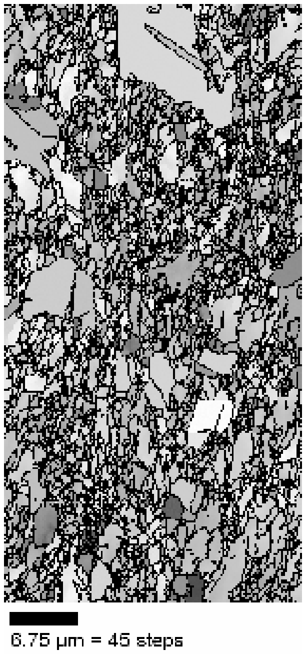

Fig.4 IPF map with grain boundary network, after annealing (250 °C/300 min, argon)

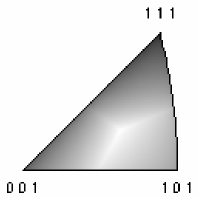

Fig.5 Colour code for inverse pole figure (IPF) map

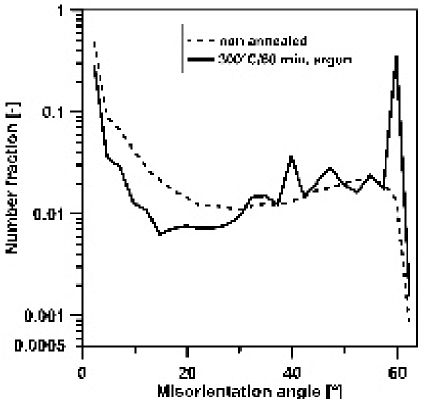

Fig.6 Redistribution of misorientation angles due to thermal exposure

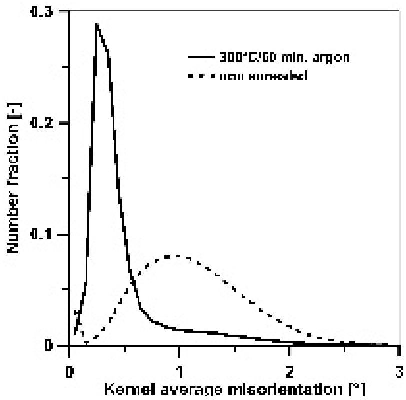

Fig.7 Redistribution of KAMs due to thermal exposure

The results obtained by extensive EBSD examinations within this work point towards higher microstructural stability in comparison with the literature [1], [6], [9-11]. Higher thermal stability of the UFG copper investigated is probably affected by purity of the material and also by the details of grain boundary dislocation arrangement. Impurities segregated along the grain boundaries could have hindered the boundary migration that normally takes place due to thermally activated processes [12-15].

4 Conclusions

Thermal stability experiments showed that the bimodal structure starts to create just after 30 min. dwell at 250 °C, while considerable grain coarsening takes place at 300 °C; extensive misorientation angle redistribution, texture changes and local lattice distortion decrease occur at the same temperature.

Acknowledgements

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