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Statistics of GB misorientations in 2D polycrystalline copper foil

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ABSTRACT

The two-dimensional grain structure in polycrystalline foil of pure copper has been studied with EBSD and compared with model for polycrystal with random grain orientations. The texture with ⟨1 1 0⟩ crystallographic direction preferentially oriented perpendicular to foil surface has been observed. The Σ3, Σ9 and Σ33 grain boundaries (GBs) were more frequent in experimental dataset than in simulated random one. The {1 1 1} GB planes are the most frequently observed. Similar enrichment in {1 1 1} planes was obtained for simulated set of GBs if texture of the foil and GB planes orientation is taken into account.

1. Introduction

It is well established that physico-chemical and mechanical properties of polycrystalline materials are tightly linked to the geometry of grain boundaries (GBs). An approach based on directed manipulation of GB structure and character (GB engineering) was developed during last decades and demonstrates significant progress [1]. It is based on the relation of material properties with GBs properties, which by-turn depends on GB geometry. The aim of this study was to reveal "special" GBs in copper polycrystalline foil from their appearance statistics assuming that they should not disappear during annealing and some of general GBs will tend to change their geometry toward special character. Both GB misorientation and plane orientation were analyzed.

2. Experimental

Electrolytically deposited 18 μm thick copper foil of 99.997% purity was held at 1000 °C (i.e. 0.95 of melting temperature T_m) for 6 h under flowing H_2 and then air quenched. The sample was investigated from both sides by means of the optical interferometer MII-4 in order to measure the average grain size and to determine GB plane inclination. Orientation of 480 individual grains was calculated on the basis of data, obtained with JSM-840A SEM equipped with electron-back-scattering (EBSD) analyzer.

3. Results and discussion

3.1. Grains orientation

In-plane average grain size about 30 μm was obtained. Inspection of the foil surface from both sides revealed mainly "monolayer" grain structure with GBs oriented perpendicular to the sample surface (Fig. 1a). Thus the use of thin foil allows to avoid serial sectioning [2] for GB plane orientation detection.

Orientation of the grains was found to be not random: the ⟨1 1 0⟩ orientation normal to foil plane dominates (Fig. 1b). Such texture is typical for electrodeposited copper foils [9]. In order to compare experimental distribution with random ones two large sets (40,000) of grain orientations were simulated. Set I was a set of random orientations uniformly distributed among all possible directions using algorithm proposed in [3]. Second set of grains orientations was generated in order to simulate foil texture (set II). Euler angles (φ_1, Φ, φ_2) which describes grains orientation were generated as follows: φ_1 was uniformly distributed in (0, 360°) interval, Φ and φ_1 values were distributed using (45°, 10°) and (0°, 10°) normal distributions correspondingly. These parameters were adjusted to reproduce experimental distribution (Fig. 1b),

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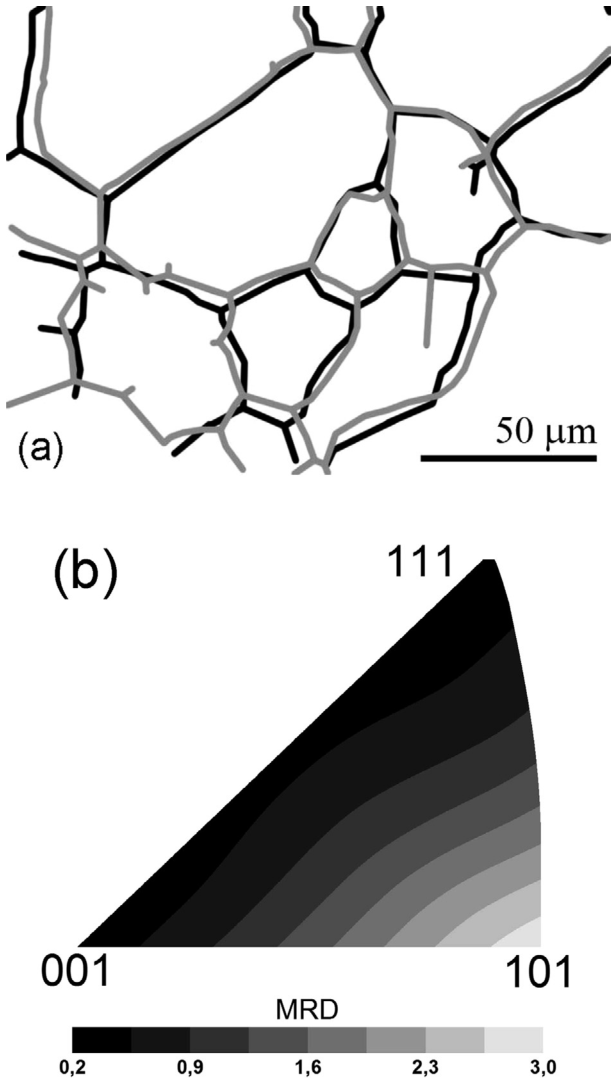

Fig. 1. Superposition of GB traces from both sides of copper foil (a) and experimental distribution of grains orientation relative to foil surface normalized by uniform distribution (b).

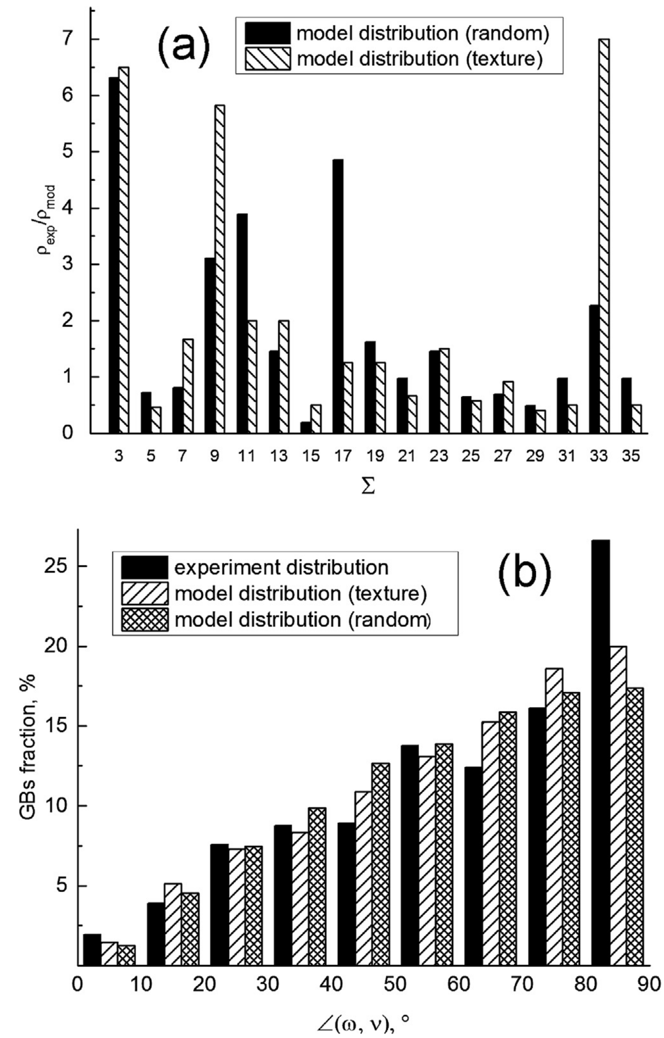

Fig. 2. (a) Probability of special CSL boundaries appearance in copper foil relative to uniform distribution (filled) and textured ⟨1 1 0⟩ distribution (dashed). (b) GB distribution vs. the misorientation axis / GB plane normal angle for experimental, random model and textured model distributions

namely to minimize disagreement in unitary probability line position on polar diagram.

3.1.1. GBs misorientation

Misorientation of 515 GBs was calculated from orientation of adjacent grains. The misorientations for 20,000 grain couples randomly selected from simulated sets I and II were calculated for comparison. The first task was to reveal whether GBs classified as a "special" in a framework of CSL model appear more frequently than other "general" misorientations. Brandon criterion [4] with $\theta_0 = 15°$ was used to classify GB as a "special". For both experimental and simulated datasets relative amount of special GB's with $\Sigma \leq 35$ was calculated. Relations of experimental probability to that simulated from set I (black bars) and set II (dashed bars) are presented in Fig. 2a.

It could be seen from Fig. 2a, that in the analyzed set of CSL boundaries only Σ3, Σ9 and Σ33 boundaries were found considerably more frequent in the foil, than in simulated set II. Σ11 and Σ13 boundaries appear in the foil only two times more frequently. This result is in a good agreement with the hypothesis, that special GBs exist in finite temperature interval and "special to general" GB transition can occur below melting temperature [5,6]. Thus, it was demonstrated [7] from misorientation analysis of single crystalline copper particles sintered to single crystalline copper plates at 0.99 T_m that Σ3, Σ11 are the most frequent GBs (strong diffraction peaks) and Σ9, Σ33 GBs were attributed to medium intensity peaks. Σ13 boundaries were attributed to low intensity peaks, but it could be linked to relatively higher temperature, used in [7]. Relative increase of Σ17 frequency is due to texture of copper foil (Fig. 2a). According to Ref. 8, the Σ3 and Σ9 are the most frequent CSL boundaries in copper, and the amount of Σ27 GBs is not much different from one for random distribution which coincides well with our data.

It could be noticed that probability of special GB appearance changes as a wave-like function with reciprocal volume density of coincidence site Σ (Fig. 2a). The same behavior is intrinsic for surface density of coincidence sites as a function of Σ [8], but probability of special GBs and surface density of CS are hardly could be linked directly. Maxima of CS densities for twin boundaries are located at Σ11, 19, 27, 35, etc [9]. We have not observed maxima at these values on probability curve (Fig. 2a). This is a reasonable result taking into account that most of special boundaries found in the foil are not symmetrical twins.

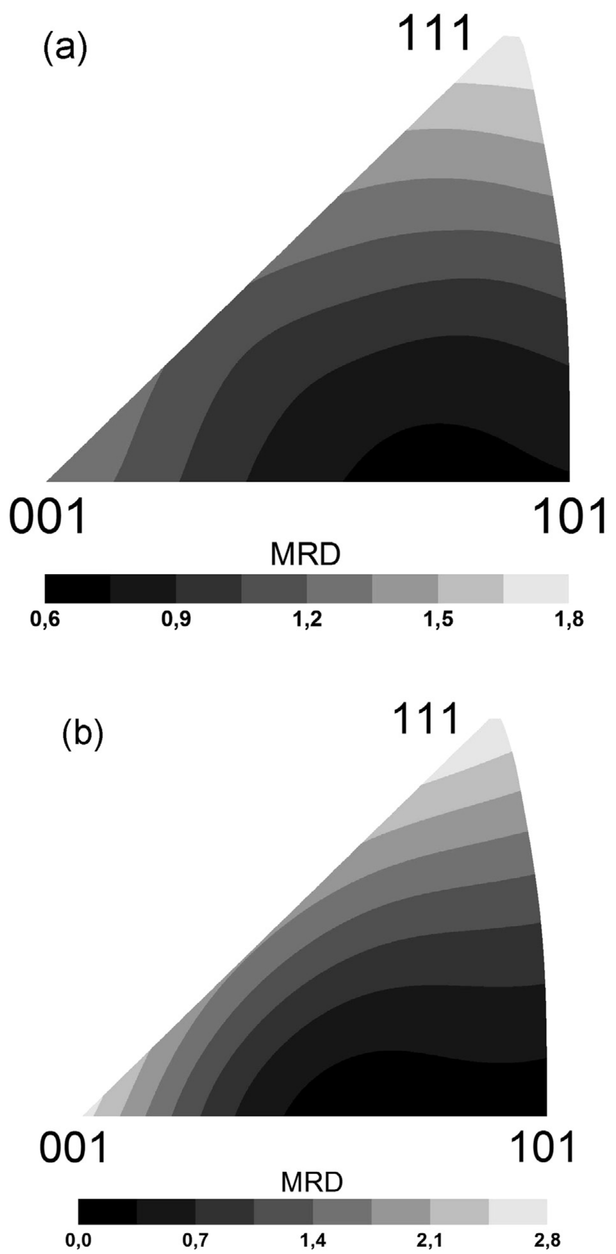

Fig. 3. Distribution of GB plane orientations normalized by the random plane orientation in copper foil (a) and in model polycrystal with ⟨1 1 0⟩ grain texture (Set II) and GB planes parallel to texture direction.

3.1.2. GB plane orientation

In further analysis GB planes were supposed to be planar and perpendicular to the foil surface (Fig. 1a), thus orientation of GB plane was calculated from orientation of GB trace on the foil surface and GB area was assumed to be proportional to the length of GB trace. Frequently used approach for analysis of GB plane orientation is the classification of GBs as a tilt or twist (or close to tilt or twist configuration). Fig. 2b represents experimental GB distribution as a function of the angle between GB plane normal and misorientation axis. Experimental distribution was compared with model distributions generated for set I with random plane orientation and set II with GB plane oriented perpendicular to the foil surface and rotated arbitrary around normal to the foil surface.

All distribution functions increase significantly with tilt component (Fig. 2b). Experimental distribution is considerably enriched by tilt boundaries comparing to totally random distribution. This could be explained by the presence of ⟨1 1 0⟩ texture. Together with simultaneous GB plane orientation perpendicular to foil surface it leads to the increase of tilt boundaries fraction (Fig. 2b, hatched bars). Significant amount (about 10%) of twin boundaries in the experimental set increases the tilt boundaries fraction as well. Thus, we could not detect any specific variation in tilt and twist boundaries fractions, experimental distribution is very close to model one, which takes into account foil texture and GB planes orientation.

The impact of foil texture on GB plane orientation statistics is presented in Fig. 3a. The number of GB planes close to {1 1 1} orientation exceeds significantly the amount specific for uniform distribution. We have also observed slight enrichment in {1 0 0} orientation. Boundary planes with {1 1 0} orientation are comparatively rare. Domination of {1 1 1} GB plane orientation was reported for polycrystalline copper samples of different type: so-called GB engineered copper [10], coarse grained copper and electroplated copper foil [9]. In the same time GB plane distribution obtained for model textured data Set II with GB plane normal perpendicular to {1 1 0} orientation yields almost the same distribution (Fig. 3b).

The effect of increased {1 1 1} probability is often attributed to the lowest energy of GBs with {1 1 1} GB planes and, in general, inverse dependence of GB frequency (or cumulative surface) on GB energy was reported for copper [10], nickel [11,12], magnesia [13]. The appearance of various GB planes is dependent also on the so-called faceting-roughening transitions [14]. However, an accurate determination of GB energy for large sets of GBs is not completely solved task. Recent studies have been demonstrated a good agreement between calculated GB energies and those extracted from GB triple junction geometry [15] in Ni only for $\Sigma3$ and $\Sigma9$ GBs [16].

Distribution of GB plane orientations in studied copper foil could be explained by texture, during annealing there was no reconstruction towards specific GB plane orientations. The same conclusion is probably valid for electroplated copper foil, studied in [10]. On the other hand, coarse grained copper have no ⟨1 1 0⟩ texture, but probability of {1 1 1} planes increases as well [10]. It could be speculated that for copper foils with columnar and almost "monolayer" grain structure the random plane orientation was preserved due to the absence of heat treatment in [10] and high temperature of heat treatment in our study while the coarse grained copper in [10] was annealed for 2 h at 300 °C.

4. Conclusions

On the basis of data for grain misorientation and GB plane position collected for electrodeposited copper foil we can draw several conclusions:

1. Texture with ⟨1 1 0⟩ crystallographic direction preferentially oriented perpendicular to the foil surface is revealed. It is probably formed during film growth and preserved after annealing.
2. Only $\Sigma3$, $\Sigma9$ and $\Sigma33$ GBs were more frequent in experimental dataset than in simulated one, when simulation was performed taking into account texture of polycrystalline foil. This result is in a good agreement with data [7], were the GBs with special structure were revealed by annealing of copper microspheres on the surface of copper single crystal.
3. Analysis of tilt-to-twist relation in experimental set of GBs did not reveal any specific features comparing to randomly generated GB set except of some enrichment by tilt boundaries due to the twinning. {1 1 1} GB planes are the most frequently observed, which is common for FCC polycrystals [10,11,14]. This is commonly explained by the low energy of GBs containing {1 1 1} planes, but in our case this is not evident. Similar enrich-

ment in {1 1 1} planes was obtained for simulated set of GBs if texture of the foil and GB planes orientation is taken into account.

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