Microstructure and crystallographic texture of Cu-Zn alloys subjected to ECAP and flat rolling

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Abstract. This paper presents the results of the microstructure and crystallographic texture investigations of the Cu-Zn alloys system with different stacking fault energies (SFE) subjected to severe plastic deformation (SPD) by equal channel angular pressing (ECAP) and subsequent flat rolling. It is shown that ECAP leads to the formation of an ultrafine-grained (UFG) structure. Further flat rolling is accompanied by a decrease in the size of structural elements and the formation of nanoscale twins, which are more likely to be detected in an alloy with a lower SFE. As the deformation degree increases, the main crystallographic textures components of the investigated alloys become Brass and Goss components.

1. Introduction
Severe plastic deformation (SPD) methods allow the formation of ultrafine-grained (UFG) and nanostructured (NS) states in various metallic materials. The SPD process is accompanied by active texture formation processes. Metals and alloys subjected to SPD are characterized by a combination of high physical and mechanical properties, including strength, ductility, electrical conductivity, magnetic and some others [1]. The main schemes for the implementation of SPD are high pressure torsion (HPT), equal channel angular pressing (ECAP) or equal channel angular extrusion (ECAE) [1].

The nature of the microstructure formed in the process of SPD is significantly influenced by the stacking fault energy (SFE) value. Alloys with a lower SFE are characterized by a smaller average grain size, increased dislocation density and deformation twins concentration and, accordingly, increased strength characteristics [2]. The character of the crystallographic texture formed in the process of SPD also depends on the SFE value [3].

The above regularities are also characteristic of the Cu-Zn alloys system, which are widely used as model alloys with different SFE. An increase in the Zn concentration in alloys of this system is accompanied by a significant decrease in the SFE. At some optimal SFE, a balance is observed between the activity of dislocations and twins, leading to the simultaneous manifestation of high strength and ductility [2,4,5]. A decrease in SFE leads to a texture transition from a copper-type texture to a brass-type texture during SPD, realized by joining sheets of Cu-Zn alloys by accumulative roll bonding [6]. As the number of passes during ECAE increases, the value of the main texture components changes [7]. A decrease in SFE helps to reduce the intensity of the texture. This effect of the SFE can be associated with the formation of shear bands and deformation twins.

Flat rolling is one of the industrial methods of deformation processing, the most widely used to give blanks of various metal materials the required geometric shapes and sizes. During cold flat rolling of the Cu-Zn alloys system, the grains are refined, a developed subgrain structure is formed, the density of
crystal structure defects increases, and a sharp crystallographic texture is formed. Depending on the SFE value, two types of crystallographic texture are distinguished. An increased SFE leads to the formation of the "copper" type $\{111\} <112>$ texture, while its decrease leads to a transition to the "brass" type $\{110\} <112>$ texture [6].

The consistent application of SPD and flat rolling methods in laboratory conditions leads to a further decrease in the crystallite size, an increase in the dislocation density and deformation twins concentration, which is important for achieving high strength and ductility of the Cu-Zn alloys system [8, 9]. It was found that the twins density increases with a decrease in the average grain size and reaches its maximum value at a certain critical grain size. The optimum grain size for the highest twins concentration is independent of the processing conditions. Below the critical grain size, the twins concentration decreases with a further decrease in the grain size. This can be caused by the transformation of twin boundaries into grain boundaries by detwinning, i.e. the inverse size effect on deformation twinning [9]. The indicated processes of microstructure evolution should be accompanied by noticeable changes in the nature of the crystallographic texture during flat rolling of UFG alloys of the Cu-Zn system.

The purpose of this paper is to establish the influence of the accumulated deformation degree on the features of the microstructure and crystallographic texture formation as a result of ECAP and subsequent flat rolling of the Cu-Zn alloys system with different SFE values.

2. Experimental procedure

Initial billets of Cu-10 wt.% Zn and Cu-30 wt.% Zn alloys with SFE values equal to 35 and 14 mJ∙m$^{-2}$, respectively [10], before deformation were annealed at 800 °C for 2 h, then slowly cooled to a temperature of 500 °C and quenched in water in order to avoid the possible formation of short-range order in the arrangement of atoms, which can lead to inhomogeneous plastic deformation during ECAP and further flat rolling. ECAP of blanks with dimensions of 8 mm × 8 mm × 60 mm was carried out at a speed of 37 mm/min at a temperature of $T = 150$ °C (for Cu-10 wt.% Zn alloy) and $T = 300$ °C (for Cu-30 wt.% Zn). The choice of ECAP temperatures was dictated by the task of obtaining integral billets without chips and cracks. The external angle of the channels intersection in the tooling was $\psi = 0°$, the internal angle was $\phi = 90°$. The number of ECAP passes for both alloys was two. ECAP was implemented using the $B_c$ route with the workpieces turning around the longitudinal axis at a 90° angle clockwise. Subsequent flat rolling of billets was carried out at temperatures equal to the ECAP temperatures for each alloy. The reduction rates varied from 30% to 95% with a gradual (no more than 10%) accumulation of deformation for each pass.

The initial coarse-grained (CG) billets (CG state) of the investigated alloys was subjected to flat rolling under similar conditions. The equivalent deformation degree during flat rolling of the CG billets to the reduction degree of 95% was 3.45 (FR - state). After two passes of ECAP, it was equal to 2.30 (UFG state), after ECAP and subsequent flat rolling with a reduction degree of 95% - 5.75 (UFG + FR state).

The microstructure was investigated using a JEOL 2100 TEM. Thin foils were investigated in dark and bright fields at an accelerating voltage of 200 kV. Thinning of foils was carried out by the jet electrolytic polishing method on a special installation Tenupol-5 using an electrolyte of the following composition: 25% phosphoric acid, 25% ethanol, 5% isopropyl alcohol, 5% urea, and 40% distilled water with a current of 50–80 mA.

X-ray diffraction (XRD) analysis was performed using a Rigaku Ultima IV X-ray diffractometer at an accelerating voltage of 40 kV, a current of 40 mA at room temperature. The X-ray tube anode was copper, and the corresponding characteristic radiation length was $K_{\alpha} = 0.15406$ nm. X-ray survey was carried out from the longitudinal horizontal section of the workpieces after ECAP and from the rolled plane of the samples. The MAUD software was used to estimate the coherent scattering domains (CSD) sizes and twins probability [11].

The analysis of the crystallographic texture formation processes was carried out using a DRON-3M diffractometer. The shooting of incomplete pole figures (PFs) for the crystallographic planes (111),
(200) and (220) was carried out using Cu Kα radiation (U = 40 kV, I = 40 mA) within the variation range of the radial angle γ from 0° to 75° and the azimuthal angle δ from 0° to 360° with a step of 5° from the longitudinal horizontal section of the blanks after ECAP, and from the rolled plane of the samples. To construct the complete PFs and the orientation distribution functions (ODFs), as well as to determine the type and volume fraction of ideal crystallographic orientations, the LaboTEX software was used [12].

3. Results and discussion

Figure 1 shows the TEM investigation results of the Cu-10 wt.% Zn and Cu-30 wt.% Zn alloys fine structure in the FR state. Bright-field images indicate the formation of a submicrocrystalline structure in the investigated alloys. The dependences of the structural elements (Dav) sizes on the deformation degree are shown in figure 2. As a result of CG alloys rolling, a subgrain structure with boundaries consisting of dislocation walls is formed. Subgrains have an irregular shape, elongated along the deformation direction. In the Cu-10 wt.% Zn alloy, flat rolling leads to the structural elements formation with an average size Dav = 200 ± 55 nm (figure 1a and 2). In the Cu-30 wt.% Zn alloy, rolling promotes the structural elements formation with an average size of 385 ± 23 nm (figure 1b and 2). The latter value is greater than the corresponding value for the Cu-10 wt.% Zn alloy, although the SFE of the Cu-30 wt.% Zn alloy has a lower value. Apparently, this is due to the higher rolling temperature of this alloy. Also, after rolling, in the defect structure of alloys, many grains contain deformation twins (figure 1a and b). The twins length and thickness are on the order of 200–300 nm and the thickness is 6–10 nm, respectively. The twins probability, determined by X-ray diffraction analysis, for the Cu-30 wt.% Zn alloy is higher than for the Cu-10 wt.% Zn alloy and is 0.98% and 0.39%, respectively (figure 2).

Rolling of the UFG Cu-10 wt.% Zn alloy (figure 1c and 2) leads to the microstructure of the UFG + FR state formation with an average size of structural elements of 220 ± 30 nm. Deformation twins are clearly visible in the fine structure. The twins probability according to X-ray diffraction data as a result of flat rolling after ECAP, i.e. as the applied deformation degree increases, it increases and amounts to 0.79%.

![Figure 1](image-url)
In the UFG state of the Cu-30 wt.% Zn alloy, the average size of the structural elements was 120 ± 9 nm (figure 1d and 2). Flat rolling also leads to the formation of deformation twins in the UFG + FR state of this alloy (figure 1d and 2). The twins probability increases to 1.30% (figure 2).

According to the X-ray structural analysis results, the CSD size (figure 2) of the alloys decreases with an increase in the deformation degree. In the UFG + FR state of the Cu-10 wt.% Zn alloy, the CSD size was 100 ± 6 nm; in the Cu-30 wt.% Zn alloy, it was 65 ± 2 nm (figure 2). As can be seen, the difference in the data obtained by the TEM and X-ray diffraction methods is very significant for the investigated materials. This can be explained by the fact that XRD measurements are carried out in a direction perpendicular to the sample surface. At the same time, the TEM method measures the total grain size, which includes highly distorted near-boundary regions, and the measurements are carried out parallel to the sample surface [13].

![Figure 2](image_url)

**Figure 2.** Dependences of $D_{av}$, $D_{XRD}$ and the twins probability on the equivalent deformation degree.

As the deformation degree increases, firstly, the size of structural elements and twins monotonically decreases, and secondly, in the Cu-30 wt.% Zn alloy, refinement to nanosizes occurs more strongly, which is associated with a lower SFE value. In the Cu-10 wt.% Zn alloy, the twin length becomes about 130–150 nm, the thickness is 4–6 nm, and in the Cu-30 wt.% Zn alloy, the length is 100–110 nm, the thickness is 3–4 nm at a similar deformation degree. In this case, the twins probability in the Cu-30 wt.% Zn alloy increases to 1.30%, which is higher than for the Cu-10 wt.% Zn alloy.

Figure 3 shows the ODFs of the Cu-10 wt.% Zn and Cu-30 wt.% Zn alloys subjected to flat rolling in the CG and UFG states. As known, rolling of materials leads to the formation of various texture components due to the presence of grains in the microstructure, each of which, due to a certain
orientation in the workpiece, responds with its specific deformation reactions to external applied deformation. The known ideal deformation texture orientations of FCC materials are shown schematically in the upper part of figure 3.

The main texture components observed during rolling of the investigated alloys are typical rolling texture orientations, namely: Copper \(\{112\}<111>\), Brass \(\{110\}<112>\), Goss \(\{011\}<100>\) and Copper twin \(\{552\}<115>\). The change in the volume fractions of the orientations depending on the deformation degree is shown in figure 4.

In the Cu-10 wt.% Zn alloy, which has a large SFE, with an increase in the applied deformation degree, the most intense texture components are Brass \(\{110\}<112>\) and Goss \(\{011\}<100>\) (figure 3 and 4), which are formed after due to the appearance of shear bands and deformation twins in the microstructure. The Copper \(\{112\}<111>\) component, which occurs during deformation due to dislocation slip, which was present at the initial deformation degrees, subsequently disappears completely, transforming into the Copper Twin \(\{552\}<115>\) texture component, the intensity of which increases with increasing deformation degree. The presence of this orientation reflects the activity of deformation twinning processes. This indicates that an additional deformation mode is activated during rolling. This additional deformation mode is deformation twinning.

A similar tendency to change the basic orientations during rolling is observed in the Cu-30 wt.% Zn alloy. The intensity of the main Brass \(\{110\}<112>\) and Goss \(\{011\}<100>\) orientations during CG rolling is higher than for the Cu-10 wt.% Zn alloy. However, later on, as the deformation degree changes, the Brass \(\{110\}<112>\) component is static, which may be due to the low SFE value of this alloy (figure 4).

**Figure 3.** Schematic representation of the ideal rolling texture orientations of FCC metals on ODFs (top) and experimental ODFs of alloys after flat rolling, ECAP and subsequent flat rolling (bottom). Sections \(\varphi_2 = 0^\circ\) и \(\varphi_2 = 45^\circ\).

It should be noted that the most intense orientation is Goss \(\{011\}<100>\), which intensifies as the deformation degree increases. The intensity of this orientation is higher in the Cu-30 wt.% Zn alloy. A low SFE promotes deformation twinning during deformation and, as a consequence, the formation of massive shear bands, which leads to texture evolution. The Copper \(\{112\}<111>\) texture component
completely disappears, and the intensity of the Copper Twin \(\{552\}<115>\) component is higher than for the Cu-10 wt.% Zn alloy.

\[\text{Figure 4.} \text{ Changes in the main orientations of alloys with an increase in the deformation degree.}\]

It is known that shear bands provide the main contribution at the late stages of texture development with the Brass \(\{110\}<112>\) component [14]. Shear bands consist of very small, elongated crystallites, which are separated by grain boundaries at a large angle and have a certain preference for the Goss \(\{011\}<100>\) component [15]. In this regard, the orientation of the crystallites can contribute to the rolling texture, therefore, as the deformation degree in these alloys increases, the intensity of the Goss \(\{011\}<100>\) component continuously increases.

Thus, it can be concluded that with an increase in the deformation degree, both Brass \(\{110\}<112>\) and Goss \(\{011\}<100>\) components increase. As the deformation increases, the Copper \(\{112\}<111>\) component is weakened and the Copper twin \(\{552\}<115>\) component is formed. These observations are confirmed by other methods of investigation the structure being formed.

### 4. Conclusions

The possibility of obtaining integral UFG billets of Cu-10 wt.% Zn alloys (with a higher SFE) and Cu-30 wt.% Zn (with a lower SFE) as a result of deformation-heat treatment, including ECAP and subsequent flat rolling with high reduction degrees up to 95%. A tendency for a decrease with saturation in the average size of structural elements (according to TEM data) and CSD (according to X-ray diffraction data) in both alloys was established as the accumulated deformation degree increased and somewhat lower corresponding values were reached in the alloy with a lower SFE. X-ray diffraction data indicate the activation of twinning processes with an increase in the accumulated deformation degree in both alloys. The twins probability increases linearly in both alloys with an increase in the accumulated deformation degree and is higher in the Cu-30 wt.% Zn alloy with a lower SFE than in the Cu-10% wt.Zn alloy with a higher SFE. An increase in the proportion of both Brass \(\{110\}<112>\) and Goss \(\{011\}<100>\) texture components with an increase in the accumulated deformation degree according to the data of texture analysis also indicates an increase in the activity of deformation twinning.

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