On the rolling and annealing texture in a Cu-15Ni-8Sn (wt.%) alloy

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Abstract

The results of an extensive neutron diffraction study of rolling and annealing texture evolution in a Cu-15%Ni-8%Sn (wt.%) alloy are presented. The alloy showed a markedly different texture consequent upon cold rolling to 50, 70 and 90 % thickness reduction. The recrystallized and aged at 400 °C and 600 °C alloys that have undergone a discontinuous precipitation reaction exhibit the same texture (similar to the α-Brass) as that of the cold worked one, but without any tendency towards a randomization. Some texture components are shown to emerge.

Key words: copper alloy, texture, recrystallization, precipitation, neutron diffraction

1. Introduction

Cu-15Ni-XSn (X = 3–15 wt.%) are commercial alloys widely used in the electronic industry. Their mechanical, electrical and corrosion resistance properties have been studied intensively. For the most studied alloy, *Pfinodal,* Cu-15Ni-8Sn (wt.%), Zhao & Notis [1] reviewed the literature data and presented their results of phases transformation kinetics obtained, by transmission electron microscopy and resistivity measurements, in a TTT diagram. It was established that this alloy undergoes at least 6 different kinds of precipitation reactions.

Among them are inter- and intragranular precipitation, spinodal decomposition, ordered microprecipitates, discontinuous (DP) and continuous precipitation. The kinetics and the temperature of these transformations are well summarized in Fig. 2 of [1]. The process of the phase transformations that can occur during the ageing of the deformed alloy becomes more complicated because of the interaction with recrystallization [2].

So far, the deformation, recrystallization and ageing textures of polycrystalline Cu-based alloys have been the subject of several studies [3]. Although, the development of annealing texture in the supersaturated Cu-Ni-Sn alloys has received little attention. In the literature, except the pioneer work of [4] there is a total lack of correlation studies between phase transformations such as recrystallization or precipitation and texture evolution in Cu-Ni-Sn alloys. Ray & Chandra Narayanan [4] presented a qualitative description of the texture evolution during the course of combined recrystallization and precipitation in Cu-Ni--Sn alloys with X-ray diffraction study. They followed the preferred orientation variations by the pole figures observation with a detailed attention for the Brass and Goss components. Such a procedure yielded only rough determination of texture. A more detailed and quantitative study associated with the occurrence of new minor components requires the use of the orientation distribution function (ODF) [5].

For this purpose, extensive neutron diffraction analysis using modern tools of texture characterization has been undertaken in order to assess quantitatively their findings. This work attempts also to search for

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eventual minor components and to confirm the tendency towards a texture randomization during discontinuous precipitation as evidenced in other systems [6].

2. Experimental

The original Cu-15Ni-8Sn (wt.%) alloy was provided by Tréfimetaux (France) in the form of strips with diameter of 1 mm. The alloy was re-melted in an induction furnace under high vacuum and cast into rods of 12 mm diameter. The ingots were then homogenized at 825 ℃ for 10 hours, quenched in water and cut to slabs of 3 mm diameter. The slabs were cold rolled with thickness reduction ranging from 50 to 90 %.

Texture analysis by neutron diffraction has been performed at Laboratoire Léon Brillouin (LLB, CEA, Saclay, France), on the four-circles 6T1 diffractometer. The full description of the experimental details and general layout of the 6T1 can be found in the LLB web site (www.llb.cea.fr). Sheet packets (1 cm^3) were prepared from the cold rolled as well as from cold rolled and annealed materials. A first set of the material was heat treated at 400 and 600 ℃ for 10 hours and 2 minutes respectively in order to ensure a certain amount of discontinuous precipitates (DP) concomitant to the onset of recrystallization. A second set underwent a prolonged ageing treatment at the same temperatures for 100 and 1 hour respectively in order to ensure a complete recrystallization and a considerable amount of DP.

The ageing conditions are based on those used in [4] and the phase transformations time-temperature-transformation kinetics (TTT diagram) presented by [2].

From the {111}, {200} and {220} complete pole figures measured using a 5° × 5° grid, the orientation distribution functions (ODF) were calculated using the LaboTex software that enables a full determination of the volume fraction (integration method taking into account the overlapping of orientations) of the texture components. The main references of the theoretical bases of the arbitrary defined cell method implemented in Labotex software can be found in Labosoft web site (www.labosoft.com.pl).

3. Results and discussion

3.1. Deformation texture

The {111} pole figures and the complete ODF of the cold rolled samples to 50 %, 70 % and 90 % reduction are shown in Figs. 1 and 2, respectively. The rolling texture of the material is typical transition type rolling texture, consisting essentially of Brass {011}⟨211⟩ and Goss orientation {011}⟨100⟩, also with reinforcements close to the S orientation {123}⟨634⟩ and to a minor Copper orientation {112}⟨100⟩, {525}⟨1-51⟩ (Rotated Copper) and {122}⟨2-21⟩ (Twin Cube).

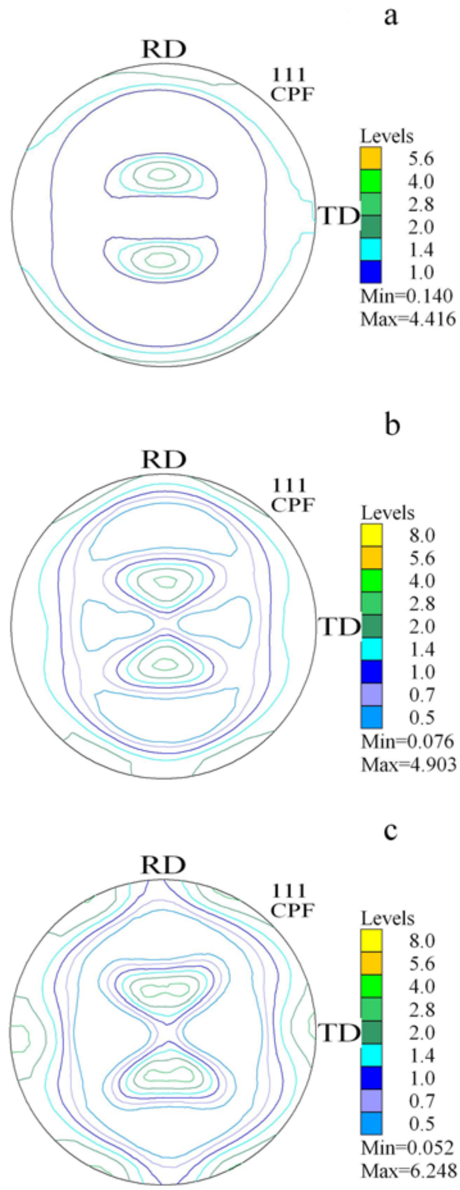

Fig. 1. {111} pole figures of Cu-15Ni-8Sn (wt.%) alloy after cold rolling, thickness reduction of: a) 50 %, b) 70 %, c) 90 %.

The quantitative ODF and volume fraction texture components results versus the degree of deformation are reported in Table 1. It is clearly seen that the textures are subdivided into two types, major and minor components, respectively. The major components are {110}⟨1-12⟩ (Brass), {110}⟨001⟩ (Goss) and {213}⟨-3-64⟩ (S). Whereas the Brass component volume frac-

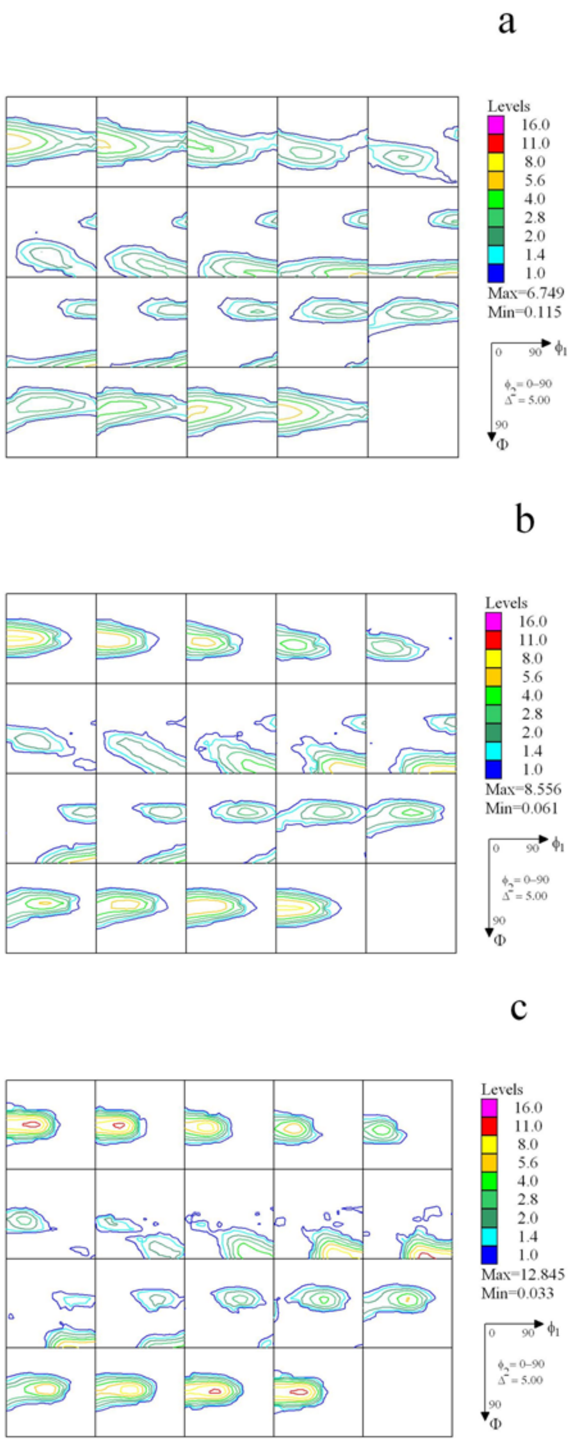

Fig. 2. ODF of Cu-15Ni-8Sn (wt.%) alloy after cold rolling, thickness reduction of: a) 50 %, b) 70 %, c) 90 % and ideal important orientations on φ_2 sections as follows d): {110}⟨1-12⟩ B, {110}⟨001⟩ G, {213}⟨-3-64⟩ S, {112}⟨11-1⟩ C, {525}⟨1-51⟩ Rot C, {100}⟨001⟩ Cube, {122}⟨2-21⟩ Twin cube.

tion increases continuously with the reduction ratio, the Goss component volume fraction is stable until 70 % and decreased for 90 % reduction.

While the minor components {525}⟨1-51⟩ (Rotated Copper) increase and become stable for 70 and 90 % reduction, the {112}⟨11-1⟩ (Copper) component is relatively stable for 50 and 70 % and decreased at 90 % thickness reduction.

The {122}⟨2-21⟩ (Twin cube) component increases from 50 to 90 % reduction. We found that the ODF and volume fraction values of the {001}⟨100⟩ (Cube) orientation were both far less that random ones.

The results of the ODF analysis performed after cold rolling indicate that the Cu-Ni-Sn texture can be described by the Brass type (or Silver type). Most of the major and minor texture components belonging to this texture type are present in Cu-Ni-Sn alloys. The occurrence of the major components has been widely discussed in the literature [7–10]. Our discussion will focus especially on the occurrence of the minor components, i.e. {525}⟨1-51⟩ (Rotated Copper) and {122}⟨2-21⟩ (Twin Cube). With increasing the thickness reduction ratio from 50 to 90 %, a net texture transition is depicted from Table 1 with a rather abrupt decrease of the Copper {112}⟨11-1⟩ orientation.

From Table 1 it is clear that the Rotated Copper orientation is present and it is relatively stable upon increasing the thickness reduction from 70 %. The occurrence of said orientation has already been reported in transition rolling texture [11].

The Rotated Copper orientation {525}⟨1-51⟩ ($\varphi_1 = 90°$, $\Phi = 74°$, $\varphi_2 = 45°$) which would be produced from the Copper {112}⟨11-1⟩ orientation by Wassermann's [12] mode of deformation twinning, reached higher volume fraction than both the {112}⟨11-1⟩ (Copper) and {122}⟨2-21⟩ (Twin Cube) orientations at all deformation amounts. It is evident from Table 1 that the decrease of the {112}⟨11-1⟩ (Copper) component volume fraction at 90 % deformation is linked to the decrease of the {525}⟨1-51⟩ (Rotated Copper) component volume fraction.

The Rotated Copper orientation {525}⟨1-51⟩ (Rotated Copper) ($\varphi_1 = 90°$, $\Phi = 74°$, $\varphi_2 = 45°$) belongs to the τ fibre which corresponds to orientations having a ⟨110⟩ parallel to the transverse direction (TD) and extends along the line $\varphi_1 = 90°$, in the $\varphi_2 = 45°$ section from the Copper {112}⟨11-1⟩, to the {110}⟨001⟩, Goss orientation at $\Phi = 35°$ and 90°, respectively [11]. We have roughly estimated the value of the stacking fault energy (γ_{SFE}) of CuNiSn alloys in the deformed state from the results of X-ray line profile analysis results of Sahu et al. [13]. In the literature γ_{SFE} is given by the following equation [14]:

$$\gamma_{\mathrm{SFE}} = \frac{\mu a_0^3 \rho}{24\sqrt{3}\pi\alpha}, \qquad (1)$$

where μ, a_0, ρ and α are the shear modulus, the cell parameter, the dislocation density and the net stack-

Table 1. ODF value and volume fraction of the texture components calculated for Cu-15Ni-8Sn (wt.%) alloy after cold rolling

Component \ Reduction	50 %		70 %		90 %	
	$F(g)$	F_{v} (%)	$F(g)$	F_{v} (%)	$F(g)$	F_{v} (%)
Brass {110}⟨1-12⟩	4.6	13.3	6.2	15.5	10.4	20.4
Goss {110}⟨001⟩	6.7	12.4	8.4	12.4	10.1	12.1
S {213}⟨-3-64⟩	2.7	13.3	3.3	13.7	2.9	14.5
Copper {112}⟨11-1⟩	2.6	5.8	2.2	5.7	0.6	3.9
Rotated Copper {525}⟨1-51⟩	1.2	7.3	2.0	9.9	1.8	9.6
Twin Cube {122}⟨2-21⟩	1.4	6.1	1.4	9.3	1.6	10.2

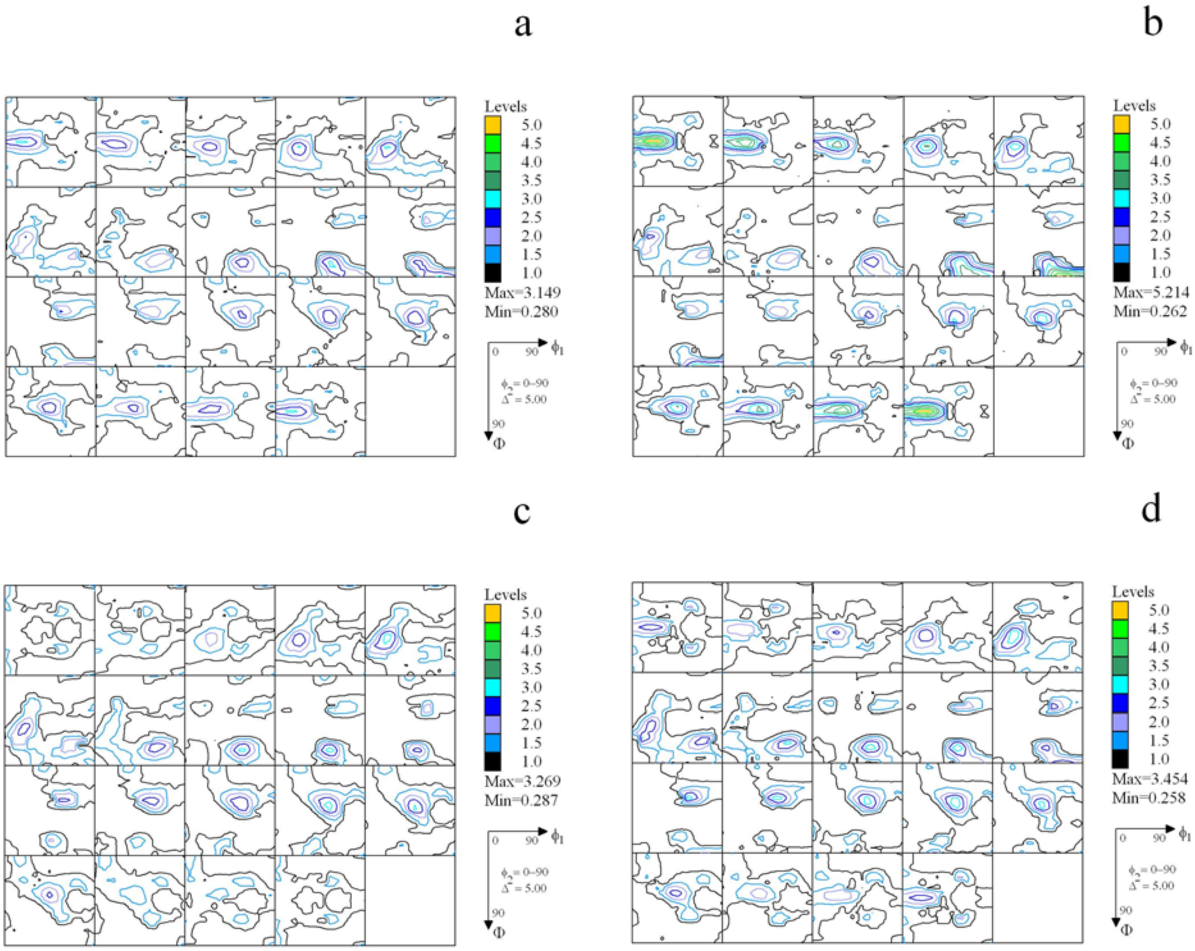

Fig. 3a,b,c,d. ODFs for a Cu-15Ni-8Sn (wt.%) alloy after cold rolling, thickness reduction of 90 % and recrystallization and ageing at 400 and 600 °C.

ing fault probability, respectively. The latter is given by the difference between α' and α, that are intrinsic and extrinsic deformation fault probabilities, respectively. Both are two kinds of planar defects in FCC structures, usually determined by X-ray Diffraction Line Profile Analysis. The estimated value is around $50\,\mathrm{mJ\,m^{-2}}$, which indicates that Cu-Ni-Sn alloys have an intermediate value of γ_{SFE}. The existence of the Rotated Copper orientation and probable other orientations belonging to the τ fibre is not surprising since the τ fibre is a special feature of the texture of materials with intermediate values of γ_{SFE}. It has been found that Cu-Ni has a higher stacking fault energy [10] which means that Sn has a lowering effect as Si, Ge and Pb alloyed to Cu, all these elements belonging to the IVA group of the periodic table.

The {122}⟨2-21⟩ (Twin Cube) texture component has already been observed in some FCC metals and

T a b l e 2. ODF and volume fraction of the texture components calculated for the Cu-15Ni-8Sn (wt.%) alloy after cold rolling, thickness reduction of 90 % and recrystallization and ageing at 400 and 600 ℃

Texture Component	400 ℃/10 h $F(g)$	400 ℃/10 h F_v(%)	600 ℃/2 min $F(g)$	600 ℃/2 min F_v (%)	400 ℃/100 h $F(g)$	400 ℃/100 h F_v (%)	600 ℃/1 h $F(g)$	600 ℃/1 h F_v (%)
B	2.2	7.1	3.7	8.8	0.9	5.3	1.9	6.1
Goss	2.6	6.4	4.2	7.3	1.0	5.2	2.4	5.9
S	2.4	10.8	2.4	11.0	2.7	10.8	2.7	11.2
Cu	1.2	4.3	1.1	4.0	1.0	4.2	1.0	4.4
Cube	1.8	3.3	1.4	3.0	1.8	3.3	1.5	3.3
Twin Cube	1.7	7.2	1.6	7.0	1.9	7.7	1.7	7.5

alloys such as Cu, Ag and Ni-Co [15, 16]. According to Verbraak [17], the {122}⟨2-21⟩ (Twin Cube) orientation is produced (during the last stage of the rolling process and the first stage of recrystallization process) by the inverse Rowland transformation together with {001}⟨100⟩ (Cube) texture in order to provide an extra accommodation between the cube nucleus and the surrounding matrix. This first generation twin cannot be stable and transforms into second generation twin having the orientation {841}⟨474⟩ (2$^{\mathrm{nd}}$ Twin Cube) as observed upon recrystallization in a Cu-Ag alloy. This is not the case in the Cu-Ni-Sn system, in which any trace of this latter orientation was present.

3.2. Recrystallization and annealing textures

Figures 3 a–d present the complete ODF of cold rolled (90 % thickness reduction) Cu-15Ni-8Sn alloy and annealed at 400 ℃ and 600 ℃ for various time intervals. The latter are representative of the tendencies existing for the two other deformation levels. Figures 3a and 3b correspond to the onset of the recrystallization concomitant to a fine discontinuous precipitation while Figs. 3c and d to annealing conditions beyond the complete recrystallization at stages where the discontinuous precipitation reaction is well developed. This latter reaction is known to initiate at grain boundaries and invades the adjacent grains [1, 4]. Manifestly, the maximum ODF intensity levels are smaller than those corresponding to the cold rolled alloys. The textures are not fairly flat and some components can be recognized from the ODF.

The quantitative results of the recrystallization and annealing texture at 400 and 600 ℃ are unambiguous and quite different from the deformation texture. Table 2 summarizes the results of the ODF values and the volume fraction of the components versus the annealing conditions at temperatures 400 and 600 ℃. We can notice a net decrease of all the values against those of the deformed state. A net decrease is depicted upon long time ageing versus small time. This decrease is more pronounced for the annealing at 400 ℃ than at 600 ℃. We can depict a relative tendency towards randomization when ageing at 400 ℃ for 100 h and at 600 ℃ for 1 h. A noticeable finding that contradicts the randomization is the presence of the {213}⟨-3-64⟩ (S) and {122}⟨2-21⟩ (Twin Cube) components whose volume fraction value keeps practically constant around 10.9 and 7.3, respectively. These two texture components have always been reported in recrystallization texture of deformed and recrystallized FCC alloys and metals [11] and seem to be stable end orientations.

Our findings are very similar to those of [18] in the Cu-Ag system where a discontinuous precipitation occurred at 600 ℃ concomitant to the recrystallization. The authors evidenced an emerging Cube and its twin orientation with noticeable volume fractions that contrast with the overall randomization. For samples aged, rolled and recrystallized at 550 ℃, the authors found that the (S) {213}⟨-3-64⟩ (S) component was dominant whereas it was absent for samples annealed at 600 ℃ after rolling.

Both our results and those of [18] are in contradiction with those of [6] in which many binary and ternary systems such as Al-Cu, Ni-Al, Ni-Al-Cr, Cu-Co and Ni-Be showed a strong tendency towards a randomization of their texture after having undergone a discontinuous precipitation reaction. This randomization is due to a loss of growth anisotropy owing to solute segregation. As assumed by [18], their work focussed on a system where adverse segregation effects were less severe. This is not the same in our case where tin is known to have a strong segregation tendency in the Cu-Ni-Sn system [19, 20].

As pointed out by [18], concomitant occurrence of precipitation and recrystallization complicate strongly the causes of the texture evolution. Furthermore, [1] has been established that at least Cu-15Ni-8Sn (wt.%) alloy undergoes 6 different kinds of precipitation reactions (inter- and intra-granular precipitation, spinodal decomposition, ordered micro-precipitates, discontinuous (DP) and continuous precipitation). It has also been shown that there is a critical ageing temperature ($T_{\mathrm{R}} \approx 530$ ℃) above which only pure discontinuous mode of decomposition takes place while below this temperature a defined sequence of reactions takes place. This sequence consists of the spinodal decomposition and the formation of a successive fine ordered DO_{22}, then $L1_2(Cu,Ni)_3Sn$ phases and a continuous growth/structural change of the ordered particles into equilibrium phase. The last stage of the sequence was evidenced by Ray et al. [4] as a combined discontinuous recrystallization and precipitation of equilibrium phase and on further ageing, the completion of these two reactions. Similar observations were made on a Pb-Ca-Sn alloy on which a heat treatment (1.5 h at 100 ℃) immediately after casting and rolling accelerates and induces in a short time a complete precipitation of the ordered L_{12}, which hinders the recovery and recrystallization [21]. When ageing at 600 ℃ (above the critical temperature), Zhao & Notis [1] evidenced a suppression of all the preliminary transitional decompositions and the phase transformations were combined recrystallization and discontinuous precipitation. The occurrence sequence of the two reactions and the influence of temperature, supersaturation and annealing time have been discussed qualitatively in [2]. The Cu-15Ni-8Sn (wt.%) system seems to obey the proposed schemes (see Fig. 1b in [2]) only at ageing temperature of 400 ℃ for which the precipitated particles influence the rearrangement of dislocations to form recrystallization front and its migration.

Ageing at 600 ℃ induces combined recrystallization and discontinuous precipitation reactions that are dissimilar to the second part of the scheme proposed by ([2], p. 159) in which the recrystallization is influenced only by segregation and the precipitation occurs after the completion of the recrystallization.

4. Conclusions

The detailed study of the evolution of the rolling and annealing texture of a supersaturated CuNiSn solid solution by means of extensive ODF analysis through a neutron diffraction characterization revealed that not only the main expected Brass type texture (Brass, Goss and S) component but also several minor reinforcements are quantitatively determined.

Beside a tendency towards randomization of the major preferred orientations upon annealing conditions, reinforced S and Twin Cube texture components are observed, which are strictly stable for different stages of precipitation-recrystallization. Ageing at 400 and 600 ℃ shows that the texture depends univoquely on the recrystallized-precipitated structure, which itself depends on the annealing duration.

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