Evolution of recrystallization texture for Cu-30%Zn alloy sheets with high Young’s modulus in the transverse direction

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Abstract. Miniaturization of electric connecters with higher stability in the coupling force is demanded as components of electric vehicles. Copper alloys are attractive materials from the perspective of electric conductivity. It is considered that Brass orientations {110}<112> has high Young’s modulus along the transverse direction (TD) since the Young’s moduli of copper single crystals show a maximum value at the <111> direction. The authors could successfully develop the near-Brass orientation in TD of the annealed sheets by performing additional warm rolling at a low reduction in thickness for cold rolled Cu-30% Zn alloy (brass) sheets with material cost lower than pure copper. The near-Brass recrystallization texture with a main component {110}<223> showed maximum orientation density after annealing at 723 K when warm rolling was performed at 623 K. In addition, we discussed the mechanism of the recrystallization texture evolution for Cu-30% Zn alloy sheets, which were subjected to the combined rolling consisting of cold and warm rolling using a conventional rolling mill and subsequently annealed at 723 K, in view of annealing twinning in the early stage of recrystallization.

1. Introduction

Miniaturization of electronic devices is proceeding in recent years. Materials with good properties of the strength, conductivity, bendability and stress relaxation resistance, and so on are demanded for electric connecters. Among the copper alloys for connecters, solid solution strengthening Cu-Zn alloys are low costs as raw materials and conventionally used for automobiles. Recently a Cu-Zn alloy for onboard connecters are developed and it is reported that it has good stress relaxation resistance [1]. A strong coupling force is required as copper alloys for connecters and therefore materials with high Young’s moduli are promising. However it is difficult to greatly increase the Young’s moduli by adding a small amount of alloy elements.

The Young’s modulus of copper single crystals shows great dependence on crystallographic orientation. It is known that the maximum value is the <111> direction (205.9 GPa) about three times higher than the minimum one of <100> (66.7 GPa) [2]. The authors have recently reported that for cold rolled and asymmetrically warm rolled Cu-30%Zn alloy sheets the recrystallization texture has the {011}<322> main components close to Brass orientations {011}<211> after annealing [3]. In the
case of the Brass orientations, the Young’s modulus in the transvers direction (TD) perpendicular to the rolling direction (RD) is a maximum, namely TD/⟨111⟩. Since in polycrystalline metals the properties are leveled by individual grains, they cannot reach those of copper single crystals. However there is a possibility that high Young’s modulus in TD of Cu-30%Zn alloy sheets is obtained. It is shown in the previous study [3] that asymmetric rolling is related to the asymmetry with respect to TD in rolling texture, and that the recrystallization texture of the annealed sheets is affected by warm rolling temperature. Since the Brass orientations ⟨011⟩⟨211⟩ are symmetrical with respect to TD, (symmetric) warm rolling is performed instead of asymmetric warm rolling, and if the ⟨111⟩ direction in TD can be strongly developed for the annealed sheets, a great increase in the Young’s modulus of TD is possible. In the present study, the Cu-30%Zn alloys were cold rolled, warm rolled and subsequently annealed. The Brass orientations symmetrical with respect to TD in the annealed sheets were developed by recrystallization annealing, and the influence of warm rolling temperature on the recrystallization texture and the Young’s modulus was investigated. The recrystallization textures were quantitatively analyzed by orientation distribution functions (ODFs). Using data of the Young’s modulus in copper single crystals, a change in the Young’s modulus of the 90° direction (TD) at warm rolling temperature for annealed sheets was predicted from ODFs and compared with the experimental result. In addition, a change in recrystallization texture at 623 K warm rolling, where was the most favorable for the Young’s modulus, was investigated in the early stage of recrystallization by EBSD, and then the evolution of the recrystallization texture through annealing twins has been discussed in this paper.

2. Experimental

The annealing condition we adopted the recrystallization-completed temperature and time which were measured in the micro-Vickers hardness test. Hereinafter, a cold rolled and annealed sheet, and cold and warm rolled and annealed sheets are called the CR sample, and the WR samples, respectively.

Four Pole figures of {111}, {200}, {220} and {311} are measured in the mid-thickness of the sheets at tilt angles of φ = 15°, 90° with a step of 5° by CuKα radiation using Schultz reflection method [4], and then the ODFs were calculated by the iterative series expansion method [5]. In preparation of the specimens for pole figure measurements, they were carefully polished to the #1500 water-proof paper, not to enter the strain by mechanical grinding.

By using the resonance method at room temperature, the Young’s moduli were measured specimens with a size of 50 mm in length and 8 mm in width cut out from the annealed sheets, and the in-plane anisotropy of the Young’s moduli were investigated along three directions of 0° (RD), 45° and 90° (TD) to RD. In addition, the Young’s moduli in 90° direction were predicted from the calculated ODFs using the Voigt and Reuss methods, and the Hill approximation which their arithmetic mean is [6,7]. The predicted results were compared with the measured values. To discuss the evolution of recrystallization texture, the annealing twins were observed by the EBSD analysis in the RD-ND section (the TD plane).

3. Results and Discussion

3.1 Texture. In the cold rolled sheet, the rolling texture showed the alloy type (α-fiber) with Brass orientations as main components. In the sheets cold rolled and warm rolled at temperatures higher than 623 K, pure-metal type (β-fiber) rolling texture was observed other than the α-fiber. This is a similar effect to cold rolled metals with a high stacking fault energy (SFE). When these rolled sheets were recrystallization-annealed, in the CR sample a conventional alloy type recrstallization texture consisting mainly of the {236}⟨385⟩ orientations close to {113}⟨121⟩ was formed as main components (Fig. 1(a)). On the other hand, as seen in the ODFs of the WR samples (Fig. 1(b)-1(c)), the recrystallization texture consisting mainly of the (110)[2-23] (about the 8° angle difference with the Brass orientation, φ1 = −43°, Φ = 90°, φ2 = 45°) close to the Brass orientation (110)[1-12] was observed with a spread around the ND/⟨110⟩ axis. The orientation components of the β-fiber were observed other than this orientation were observed. This corresponds to the transition of rolling texture
from the α-fiber to the β-fiber. Although a reason that the \{011\} \langle 322 \rangle orientations are developed is not clear, by performing warm rolling after cold rolling, the orientations close to Brass orientations present in both the α-fiber and the β-fiber are main components of the recrystallization texture.

3.2 Young’s modulus. Figure 2 shows rolling temperature dependence of the Young’s modulus in annealed sheets. The Young’s modulus in the 90° direction (TD) for the WR samples increased greatly. It was the highest at 623 K warm rolling and decreased as the rolling temperature was raised further. In the 0° (RD) and 45° directions, it is not so much changed from the CR sample compared to that in the 90° direction. Since this will be related to a change in recrystallization texture by warm rolling which is conducted after cold rolling. The maximum orientation density in the \{011\} \langle 322 \rangle orientation ($\phi_1 = 40^\circ$, $\Phi = 45^\circ$, $\phi_2 = 0^\circ$) was determined. As shown in Fig. 3, there is a clear correlation between the \{011\} \langle 322 \rangle orientation density and the Young’s modulus in TD. It is concluded that 623 K warm rolling is the most effective in increasing the Young’s modulus in TD. It is considered that the transition from alloy type (α-fiber) to pure-metal type (β-fiber) occurred since deformation by cross slips are possible on heating in the furnace during warm rolling. This corresponds to raising stacking fault energy (SFE). It is unclear whether the SFE was really raised at rolling temperatures not less than 573 K. In austenitic stainless steel sheets, it is known that the transition from alloy type to pure-metal type occurs as rolling temperature rises [8]. It is considered that a similar phenomenon occurred even in Cu-30%Zn alloy.

3.3 Prediction of the Young’s modulus from ODF. Recrystallized polycrystalline metals are composed of many grains with various orientations. Since there is a possibility that texture components other than the \{011\} \langle 322 \rangle orientations affect the Young’s moduli, they was predicted from ODFs.
Figure 4 shows the calculated and measured results. The measured values are closer to the Reuss method than the Hill method. We used copper single crystals’ data instead of Cu-30%Zn alloy. Since zinc (48 GPa [9]) with a lower value than copper (129.8 GPa [10]) is contained 30%, the Young’s modulus prediction of Cu-Zn alloys (brass, 103 GPa [10]) with the same face-centered cubic structure is necessary to quantitatively consider the Zn content. However, a qualitative tendency of the predicted value, on the whole, corresponds to the measured value. It is suggested that the prediction of Young’s modulus as a function of rolling temperature is possible. A large number of future problems such that the reduction of the Young’s modulus at 723 K and the distribution condition of the Zn content in the thickness direction are present. Fig. 5 Rolling and recrystallization textures for cold-rolled and additionally warm-rolled Cu-30%Zn sheets and only cold-rolled ones.

3.4 Effect of warm rolling on recrystallization texture. Rolling and recrystallization textures of the sample 85% cold rolled and 40% warm rolled at 623 K (the WR sample), and 91% cold rolled sample (the CR sample) are shown in Fig. 5. The rolling textures are nearly the same, but their recrystallization textures are quite different.

It is possible that the recrystallization texture is changed by adding warm rolling with a low reduction. Texture control is a very effective means to change material properties with large anisotropy in single crystals such as the Young’s modulus. A reason for preferred orientations of \{011\}<322> close to Brass orientations as main components of the recrystallization texture is unclear at present. As this reason, since two equivalent Brass orientation variants have a twin relationship each other, it is considered that annealing twins of the Brass orientations are formed and grown when recrystallized nuclei of the Brass orientations grow to recrystallized grains. Although the rolling textures are similar to each other, this explains the different recrystallization textures well after recrystallization was completed. Adding warm rolling leads to slip deformation and recovery. Alloy type rolling texture (α-fiber) consisting mainly of the Brass orientations, which will form annealing twins in partial regions and will grow to recrystallized grains. The mechanism of the \{011\}<322> recrystallization texture evolution is investigated using EBSD analysis.
3.5 Discussion for the \{011\}<322> recrystallization texture evolution. A change in hardness with recrystallization annealing (softening curve) was measured. The results showed a large change in hardness when the samples were heated to 723 K. Therefore it is considered that softening occurs at 723 K heating. To observe a change in orientation in the early stage of recrystallization, microstructures of 723 K-12 s and 723 K-15 s annealed samples were investigated by EBSD measurements, as time condition is 0 s when samples were entered in the furnace of 723 K holding. The results were shown in Figure 6. The Brass orientations \{011\}<211> and the near-Brass \{011\}<322> orientations clearly increased with increasing annealing time. The Ishii et al. [3] mentioned that the near-Brass orientations were developed through asymmetric warm rolling after cold rolling. The effect of asymmetric rolling (additional shear deformation) contributed to only the slight asymmetry with respect to TD of pole figure. The (symmetric) warm rolling was expected to occur the transition of rolling texture, and it is considered to contribute to the evolution of the near-Brass orientations. As described in the reference [3], it is suggested that this is related to the twin relationship. An orientation change from the Brass orientations \{011\}<211> to the near-Brass \{011\}<322> orientations was observed, as shown in Figure 6. This is suggested that an orientation change which leads to grain boundaries migration in grain growth during primary recrystallization (a rotation around the \{011\> axis due to the twin relationship is considered.) As a result, the \{011\}<322> recrystallization texture with a wide spread would be formed by longer time (1.8 ks) annealing. The volume fractions of the Brass orientations and the near-Brass orientations \{011\}<322> obtained from EBSD analysis are shown in Figure 7. It is clear that these orientations increased with an increasing annealing time for any orientation. In other words, it is suggested that the nucleation of the Brass orientation \{011\}<211> and thereafter the evolution of \{011\}<322> orientations may be attributed to the formation of annealing twins with high order.

4. Conclusions

Cu-30%Zn alloys were cold rolled to a high reduction in thickness and warm rolled to a low

![Fig. 6 Changes in grain with an increase of annealing time. (a, b) \{011\}<211> and (c, d) \{011\}<322> orientations. (a, c) a 12 s annealed sample and (b, d) a 15 s annealed one.](image)

![Fig. 7 Change in volume fraction of each oriented grain in the early stages of recrystallization.](image)
reduction. The relationship between recrystallization texture and the Young’s modulus was investigated, and the mechanism of recrystallization texture evolution was discussed. The results obtained in this study are as follows:

1) By performing warm rolling at temperatures more than 573 K for a cold rolled Cu-30%Zn alloy sheet, the recrystallization texture consisting mainly of the \{011\}<322> orientations was obtained for the annealed sheet.

2) Through the evolution of the above recrystallization texture, the Young’s modulus in TD was greatly increased. The WR sample warm rolled to 40% reduction at 623 K showed a relatively high Young’s modulus of 122 GPa.

3) A change in the Young’s modulus with warm rolling temperature can be qualitatively predicted from the ODFs for cold rolled, warm rolled and annealed sheets.

4) An increase in volume fraction of annealing twins in each oriented grain was observed in the early stages of recrystallization.

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References
[1] Mori H., Maki K. and Yamasita D.: Materia 53 (2014), 69-71.
[2] Karashima S.: Strength of Metals and Alloys, Japan Inst. Met (1972) 6.
[3] Ishi Y. and Inoue H: Copper and copper alloys 56 (2017), 51-55.
[4] Schluz L G.: J. Appl. Phys. 20 (1949), 1030-1033.
[5] Inoue H. and Inakazu N.: J. Japan Inst. Met 58 (1994), 892-898.
[6] Bunge H. J.: Texture Analysis in Materials Science, Translated by Morris, Butterworths, (1982), 321-327.
[7] Inoue H. and Iwata J.: Copper and copper alloys 50 (2011), 204-209.
[8] Goodman S. R. and Hu H.: Trans. Metall. Soc. AIME 230 (1964), 1413-1419.
[9] Koide S.: Physics, Syokabou (2003), 89.
[10] Takano K.: How to Choose and Use Plastic Materials for prevent Trouble, Kogyo Chosakai Publishing Co., Ltd. (2005), 61.