Changes in Strength and Microstructure of Cu (100) [001] Single Crystals Caused by Accumulative Roll-Bonding

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Plate-like copper single crystals with (100) [001] orientation were subjected up to nine cycles of the ARB process. Ultrafine grains less than 1 μm were obtained in the single crystal ARB processed by six cycles. Changes in strength and microstructure of the ARB processed single crystals are compared with those of the ARB processed polycrystals. Origins of high strength of the ARB processed single crystals are discussed. [\[doi:10.2320/matertrans.MD201116\]](https://doi.org/10.2320/matertrans.MD201116)

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1. Introduction

Metals having ultrafine grains less than about 1 μm have been produced by severe plastic deformation (SPD) processes; these metals have higher strength than coarse-grained metals.^{1–4)} Changes in strength and microstructure during SPD processes have been studied in many previous papers.^{5–9)} Although there are a few studies using single crystals,^{10–13)} polycrystals have been used as starting materials for the SPD processes in most of the previous studies. Since plastic deformation is affected by grain boundaries, the changes in strength and microstructure during SPD processes may be also affected by the grain boundaries. Basic experimental results to reveal the changes of strength and microstructure are still needed to understand how grain refinement occurs and high strength is achieved during SPD processes. In the present paper, we study the changes in strength and microstructure of Cu single crystals caused by an accumulative roll-bonding (ARB) process which is one type of SPD processes.

2. Experimental Procedures

Using Cu with the purity of 99.99%, plate-like single crystals with 2 mm thickness, 50 mm width and 120 mm length were grown by the Bridgman method in vacuum with a seed. The Cu single crystals have the (100) [001] orientation as shown by the stereographic projection in Fig. 1, where RD and TD are the rolling and transverse directions of the subsequent ARB process, respectively. The (100) [001] orientation was selected as an idealized cube orientation which is a typical recrystallization texture of rolled FCC polycrystalline metals.

The ARB process^{1,9)} was carried out at room temperature without lubrication. The first cycle of the ARB process was made by stacking two (100) [001] single crystals so that the orientations of the two single crystals were the same. The total thickness of the stacked two single crystals before ARB was 4 mm. During the roll-bonding, about 50% thickness reduction was made for the stacked single crystals. This 50%

Fig. 1 A stereographic projection of the Cu single crystal used for the ARB process.

Table 1 A list of parameters for the ARB process made in the present study.

ARB cycles	Start	1c	2c	3c	4c	6c	8c	9c
Equivalent strain	0	1.0	2.1	2.8	3.6	5.1	6.9	7.8
Number of layers	1	2	2	4	8	16	128	254
Thickness [mm]	2.00	1.63	0.90	1.02	1.06	1.09	0.98	0.90

roll-bonding was repeated up to 9 cycles (9c), except that at 2c which was the normal 50% cold rolling without bonding. This rolling without bonding was made to reduce the specimen thickness from about 2 to 1 mm. Details of the present ARB process are shown in Table 1. If just 50% roll-bonding is repeated, the equivalent strain after the N th cycle is given by $0.8 \times N$.^{4,9)} The values of equivalent strain in Table 1 are slightly different from the ideal values due to deviations in the thickness reduction from 50%. For comparison, the ARB process up to 8c was also performed on polycrystalline Cu annealed at 873 K for 2 h before ARB. The initial grain size was 34 μm .

Tensile tests were carried out at room temperature in order to investigate mechanical properties of the ARB processed specimens. The gauge length and width were 10 and 3 mm, respectively. The tensile axis was parallel to the TD of the ARB process. After mechanical polishing, the specimens

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Fig. 2 Stress-strain curves of the Cu specimens after various cycles of ARB processing.

were tensile tested at room temperature under an initial strain rate of $8.3 \times 10^{-4} \text{ s}^{-1}$.

Microstructures of the ARB processed specimens were observed using a scanning electron microscope, JEM-7000F. Orientations of ultrafine grains of the ARB processed specimens were determined by analyzing electron backscatter diffraction pattern (EBSP) obtained with the JEM-7000F. After electrolytic polishing at -50°C in a mixture of nitric acid and water with the ratio of 1:4 in volume, EBSP measurements with a scan area of $20 \mu\text{m}$ (RD) and $30 \mu\text{m}$ (normal direction: ND) and a step size of $0.05 \mu\text{m}$ were made on the RD-ND plane at the center of the specimen thickness.

3. Results

Figure 2 shows stress-strain curves of the specimens ARB processed for various cycles. These stress-strain curves are drawn using the nominal tensile stress and the nominal plastic strain including that for the 0c specimen. The 0c specimen is a specimen before ARB, as cut from the original (100) [001] single crystal. The low strength and high ductility seen in Fig. 2 is consistent with the behavior expected for a single crystal of this composition. The stress-strain curve for the 0c specimen in Fig. 2 shows these characteristics. The tensile stress σ for the 0c specimen can be converted to the shear stress τ acting on the slip system of this single crystal using the Schmid factor ($S_F = 0.416$ in this study). If we evaluate the tensile flow stress as the product of the shear stress τ and the Taylor factor for FCC metals ($M = 3.06$), the evaluated tensile stress increases about 30% from σ . The ARB process increases strength and decreases elongation to fracture drastically. The stress-strain curve for the 1c specimen also shows much higher strength and much lower ductility compared with those of the 0c specimen.

Figure 3 shows the relationship between the 0.2% proof stress $\sigma_{0.2}$ and N . In Fig. 3, filled circles indicate the present results for the ARB processed Cu single crystals, while open triangles indicate the results for the ARB processed Cu polycrystals obtained by Takata *et al.*¹⁴⁾ The first cycle of the ARB process rapidly increases $\sigma_{0.2}$ for both single crystalline and polycrystalline Cu. Before the ARB process, $\sigma_{0.2}$ for the annealed polycrystal is higher than that for the (100) [001] single crystal as shown by the data points for $N = 0$. However, after the first cycle of the ARB process, the values

Fig. 3 The relationship between the 0.2% proof stress $\sigma_{0.2}$ and the number of ARB cycles N . Filled circles indicate the present results for the ARB processed Cu single crystals, while open triangles indicate the results for ARB processed Cu polycrystals.¹⁴⁾

of $\sigma_{0.2}$ for the single crystalline and polycrystalline Cu are almost the same.

Figure 4 shows boundary maps obtained by the EBSP analyses of the ARB processed single crystals. In the maps, grain boundaries are classified into two types depending on the misorientation angle θ between neighboring points in the EBSP measurements: Low-angle boundaries (LABs; $15^\circ > \theta \geq 2^\circ$) drawn with red lines and high-angle boundaries (HABs; $\theta \geq 15^\circ$) drawn with green lines. Here, the misorientation angle less than two degrees was cut off due to the limitation of the EBSP measurement. Neither LABs nor HABs were observed in single crystals before ARB. Only LABs were observed in the 1c specimen as shown in Fig. 4(a). HABs appeared clearly after 4c as shown in Fig. 4(c).

From Figs. 4(a) to 4(f), it seems that the total length of LABs and HABs in the samples generally increases with increasing N . In order to discuss this quantitatively, the length of LABs and HABs per unit area, X_{LAB} and X_{HAB} , respectively, were evaluated from Figs. 4(a) to 4(f). X_{LAB} and X_{HAB} are given by the total length of LABs and HABs and the area of scanned region.

Figure 5(a) shows the variations of X_{LAB} and X_{HAB} as a function of N . As shown by Fig. 5(a), X_{LAB} starts increasing from $N = 1$, while X_{HAB} starts to increase after $N = 3$. Then, X_{LAB} saturates after $N = 4$, while X_{HAB} saturates after $N = 6$.

In addition to X_{LAB} and X_{HAB} , the fraction of LABs f_L and HABs f_H are defined as the total length of LABs and HABs divided by the sum of the total length of LABs and HABs. Figure 5(b) shows the changes of f_L and f_H with N . Initially, f_H decreases from about 20% at $N = 1$ to 5% at $N = 2$, and then f_H increases with increasing N up to ARB 6c. Finally, f_H saturates at around 60% after $N = 6$. f_L shows the opposite trend which is easily understood from the definition.

Figure 6 shows the variations of the boundary spacings between LABs and HABs along the ND as a function of N . The results for both the ARB processed single crystal and polycrystal are shown in Fig. 6. In Fig. 6, four variables are used, of which superscript denotes the type of starting materials: (S) is a single crystal and (P) is a polycrystal, whereas their subscript indicates the spacing between boundaries: H is only HAB ($\theta \geq 15^\circ$) and A is all boundaries (both LAB and HAB, $\theta \geq 2^\circ$).

Fig. 4 Boundary maps obtained by EBSD analyses of the ARB processed single crystals; (a) 1c, (b) 2c, (c) 4c, (d) 6c, (e) 8c and (f) 9c.

Fig. 5 (a) The variations of the length of LABs and HABs per unit area (X_{LAB} and X_{HAB}), and (b) the changes of the fractions of LABs and HABs (f_L and f_H) as a function of the number of ARB cycles N .

All the spacings, $d_H^{(S)}$, $d_A^{(S)}$, $d_H^{(P)}$ and $d_A^{(P)}$ decrease with increasing N . When $N \geq 6$, these values become less than $1 \mu m$ and ultrafine grains are obtained in both ARB processed single crystal and polycrystal. It is noted that the evolution of the grain refinement of the single crystal is affected by the initial orientation of the single crystal. We have found that the grain refinement of the single crystal having the initial orientation of (122) [278] is faster than that having cube orientation.

4. Discussion

It has been reported that the macroscopic deformation band is formed when a single crystal having the initial orientation of cube is cold rolled.¹⁵⁾ The length scale along ND of the macroscopic deformation bands in the single crystal study of

Fig. 6 Variations of the boundary spacings between LABs and HABs as a function of the number of the ARB cycles N . The spacings were measured along ND. The results for both the ARB processed single crystal and polycrystal are indicated. See text for details of the variables and symbols in this figure. $d_H^{(S)}$ shown by $\bullet$ is the spacing between HABs in the ARB processed single crystal, $d_A^{(S)}$ shown by $\blacktriangle$ is the spacing between both LABs and HABs in the ARB processed single crystal, $d_H^{(P)}$ shown by $\circ$ is the spacing between HABs in the ARB processed polycrystal, and $d_A^{(P)}$ shown by $\triangle$ is the spacing between both LABs and HABs in the ARB processed polycrystal.

Liu and Hansen is about a few hundreds μm in the case of 50% rolling reduction,¹⁵⁾ whereas the length scale along ND of the EBSD measurements in this study is a few tens μm . The EBSD observations correspond to changes within a single band appeared in 50% cold rolled single crystal. The boundary maps of the early stage of the ARB processed samples show inclined LABs having an angle of 30–35° against RD as shown in Figs. 4(a) and 4(b). These LABs are microscopic components of the macroscopic deformation bands, which contribute the drastic increase of the strength of a single crystalline Cu after ARB 1c (Fig. 2). As shown in Fig. 6, when $N \geq 6$, the boundary spacings $d_H^{(S)}$ and $d_A^{(S)}$ for the ARB processed single crystals are the same as $d_H^{(P)}$ and $d_A^{(P)}$ for the ARB processed polycrystals, respectively. Moreover, ultrafine grains less than $1 \mu m$ are commonly obtained when $N \geq 6$. In accordance with the similar microstructure, the values of $\sigma_{0.2}$ when $N \geq 6$ are also the same for the ARB processed single crystals and polycrystals. Kunimine *et al.*^{16–18)} have made systematic studies to

understand the changes in mechanical properties of Cu throughout the ARB process. According to Kunimine *et al.*, $N=6$ is the critical ARB cycle where characteristic mechanical properties appear and deformation mechanisms of ARB processed polycrystalline Cu change qualitatively. Examples of the characteristic mechanical properties are the higher strain-rate sensitivity of flow stress and the decrease in activation volume for the ARB processed Cu when $N \geq 6$.^{16,17)} The changes of the mechanical properties occur when ultrafine-grained structure appears in the specimens and this suggests the difference in the deformation mechanisms between materials with deformation structure and ultrafine-grain structure. The similarities of the strength and microstructure between the ARB processed single crystals and polycrystals when $N \geq 6$ mean that grain boundaries in Cu before ARB do not affect the critical number of the ARB cycle $N=6$, required to form ultrafine-grain structure showing characteristic mechanical properties.

As shown in Fig. 6, the boundary spacings for the ARB processed single crystals are much longer than those for the ARB processed polycrystals when $N=1-4$. The boundary spacings when $N=5$ were not measured in the present study. For example, when $N=2$, $d_A^{(S)} \approx 1 \mu\text{m}$ and $d_H^{(S)} \approx 20 \mu\text{m}$ were observed for the ARB processed single crystal. On the other hand at the same $N=2$, $d_A^{(P)} \approx 0.4 \mu\text{m}$ and $d_H^{(P)} \approx 2 \mu\text{m}$ for the ARB processed polycrystal. However, as shown in Fig. 3, the values of $\sigma_{0.2}$ were similar for the ARB processed single crystal and polycrystal at about 330 MPa, when $N=1$. We find that the strength of the ARB processed specimens when $N=1-4$ should be explained by the deformation structure obstructing the motion of dislocations in grains. As shown in Fig. 6, the grain refinement continues until $N=6$ and then the grain size saturates. However, the proof stress $\sigma_{0.2}$ of the ARB processed single crystal remains relatively constant at about 400 MPa when $2 \leq N \leq 9$. To understand the relatively constant $\sigma_{0.2}$ of 400 MPa when $2 \leq N \leq 9$, quantitative consideration based on the theory of dislocations is needed to evaluate the upper limit of strength. This is the subject of our future work.

5. Summary and Conclusions

Plate-like copper single crystals with (100) [001] orientation grown by the Bridgman method were subjected to the ARB process. The ARB process was carried out without lubrication at room temperature up to nine cycles. Ultrafine grains less than $1 \mu\text{m}$ were obtained in the ARB processed single crystal when the number of ARB cycles, $N \geq 6$. The microstructure of the ARB processed single crystals when $N \geq 6$ are the same as that of ARB processed

polycrystals with the same N . When $N < 4$, the fraction of high-angle boundaries in the ARB processed single crystals is much less than that in the ARB processed polycrystals. The proof stress $\sigma_{0.2}$ of the ARB processed single crystals remains constant at a value around 400 MPa when $2 \leq N \leq 9$, although the origin of high strength changes from the in-grain deformation structure to the ultrafine-grain structure at $N \approx 6$.

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