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ARTICLE

Effects of Cold Rolling Reduction and Annealing Temperature on Microstructure and Texture Evolution of Cu-44%Ni Alloy

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Abstract: The effects of cold rolling reduction and annealing temperatures on the formation of cube texture and microstructural evolution have been investigated in Cu-44%Ni alloy. The results show that the recrystallized cube texture is strengthened by either increasing the rolling reduction or increasing the annealing temperature. And the strong cube texture can be observed for the severely cold-rolled (>90%) alloy after annealing at high temperature (>900 °C). Furthermore, high-angle grain boundaries (HAGBs) and annealing twin boundaries (Σ3 boundaries) decrease with the increase of rolling reduction. However, during isochronal annealing, the trend of Σ3 boundaries is consistent with that of HAGBs, which firstly increases during recrystallization process and then decreases with further increasing temperature. After annealing at 1100 °C for 1 h, the 99% cold-rolled Cu-44%Ni alloy obtains the fraction of the cube texture 99.8%, and the fractions of HAGBs and Σ3 boundaries are 2.5% and 1.3%, respectively.

Key words: cold rolling reduction; recrystallization; Cu-44%Ni alloy; texture; microstructure

Ni-5%W is one of the most widely employed Ni-based alloys as a textured substrate for $YBa_2Cu_3O_{7-x}$ (YBCO) coated conductors[1]. However, one main drawback of this material is its ferromagnetic property at the application temperature of 77 K, which leads to unwanted energy losses in superconductor[2,3]. Then, several studies were carried out on non-magnetic materials. Alternative Cu-Ni alloys have thus been proposed since they are non-magnetic at operating temperatures for Cu concentrations above 54 at%[4]. Furthermore, taking into account that well textured and lower cost for Cu-Ni alloys compared to the more commonly used Ni-W substrates[5], the Cu-Ni alloys are expected to be the promising substrate materials.

One of essential requirements for a substrate material is that it should contain as few high-angle grain boundaries (HAGBs) as possible to avoid a typical 'weak-link' behavior that drastically reduces the critical current carried in epitaxially grown superconducting layer[6]. Therefore, the substrate material should consist of grains with predominantly low-angle grain boundaries (LAGBs), in other words the material should be strongly textured[7]. As a consequence, it is evident that the use of a substrate with a sharp cube texture {001}<100> is of fundamental importance for the realization of YBCO-coated conductors.

Cube texture in face-centered cubic (fcc) metals such as Cu, Al and Ni can be formed after rolling and subsequent annealing as the primary recrystallization texture[8,9]. The strength and sharpness of cube texture have been found to depend on a lot of thermo-mechanical processing parameters. Among them are such factors as the total rolling reduction, deformation temperature, annealing time and annealing temperature[1,8-13]. It is worth emphasizing that the rolling reduction and annealing temperature are always considered as two common methods to optimize the cube textured substrates. For instance, Specht et al.[8] reported that a combination of high reduction and high-temperature annealing in a reducing atmosphere leads to >99% cube texture in high-purity Ni. Recently, Cu-Ni based alloys have also been studied by several authors for developing the biaxial texture. Tian et al.[14] reviewed that the high-temperature annealing after heavy rolling generates strong cube texture in Cu-Ni alloys. And Vannozzi et al.[15]

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indicated that high annealing temperatures enabled the fraction of cube orientation as high as 95% to be obtained in Cu-Ni-Co alloys, demonstrating the suitability of this alloy tape as substrate for YBCO-coated conductors. The encouraging results obtained with this alloy substrate have driven further research on Cu-Ni based alloy tape. However, previous studies of Cu-Ni alloys mostly focused on optimization of the thermo-mechanical processing to realize a strong cube texture, with very little effort to investigate the formation of deformation and recrystalliza- tion textures as well as the relationship between them.

In the present study, Cu-44%Ni alloys were subjected to 70%, 80%, 90%, 99% cold-rolling and subsequent isothermal annealing at a variety of temperatures. The focus of the research was to study the effects of rolling reduction and annealing temperature on the evolution of microstructure and texture in Cu-Ni alloys. We also aimed to determine the optimal conditions for both rolling reduction and annealing temperature for such a substrate in order to make sure it can be used in superconductor industry.

1 Experiment

In the present work, the Cu-44%Ni alloy was prepared from molten Cu and Ni metals both with a high purity of 99.99% in a high-frequency induction furnace. The obtained ingot was hot deformed and machined into 10 mm thick bars. The bars were further cold rolled using mirror-finished rolls in several passes with an average of ~5% cold reduction per rolling pass. In order to study the effects of rolling reduction, the bars were cold rolled to a final thickness of 3, 2, 1 and 0.1 mm corresponding to 70%, 80%, 90% and 99% total reduction in thickness, respectively. Then, the deformed materials were annealed at 900 °C for 1 h in a protective Ar-4% H_2 atmosphere. The effects of annealing temperature on the development of the cube texture were also investigated. The bars were rolled to 99% reduction in thickness at room temperature, and then annealed at various temperatures ranging from 500 °C to 1100 °C for 1 h in a protective Ar-4%H_2 atmosphere. The heat treated samples were quenched in cold water immediately.

Bulk textures of the samples were characterized on a Rigaku D2500 X-ray texture goniometer using Cu Kα radiation. {111}, {200} and {220} pole figures were measured. From three incomplete pole figures, the orientation distribution functions (ODF) were calculated using the Bunge method provided in the OIM analysis software. The fractions of different texture components were determined within a spread of 15° around their respective ideal locations in Euler space.

Microstructures and textures of the annealing samples were also characterized using a fully automated Channel 5 EBSD system attached to a TESCAN MIRA 3 field emission gun scanning electron microscope. The samples for texture investigation were first ground to SiC4000 and then electropolished in mixed solution $HClO_4$:C_2H_5COOH:C_2H_5OH=1:3:4 at −10 °C. Typically, for the rolled samples, a step size of 0.1 μm or 0.2 μm was used, while for the fully recrystallized samples, a step size of 1 or 2 μm was used. The micro-texture measurements for the samples were taken from the rolling plane. The scanned areas were randomly selected and at least three maps were taken for each condition. The EBSD data was analyzed by the HKL Channel 5 software. The area fraction of the relevant texture component was calculated from the orientation maps constructed from the EBSD data with maximum deviation angle of 15° from the ideal orientation. The texture components considered are {001}<100> (Cube), {112}<111> (Cu), {123}<634> (S), {110}<112> (B) and {011}<100> (Goss). The remaining texture are grouped together and referred to as "other". Low-angle grain boundaries (LAGBs) and high-angle grain boundaries (HAGBs) are defined as boundaries with misorientations 2°~15° and more than 15°, respectively. Annealing twin boundaries (Σ3 boundaries) were classified in the present experiment by satisfying the Brandom criterion $\Delta\theta_{max}=15°\Sigma^{-1/2}$, namely 8.66° [16]. The average grain size was calculated without considering twin boundaries.

2 Results

2.1 Effect of rolling reduction on deformation and recrystallization textures

Fig.1 shows the calculated ODF sections (φ_2=0°, 45°, 65°) of Cu-44%Ni alloys cold rolled 70%, 80%, 90% and 99%. After cold rolling, the typical pure metal or Cu-type deformation texture is formed, which is characterized by the development of preferred orientation along the β-fiber running from the Cu orientation {112}<111> over the S orientation {123}<634> to the B orientation {011}<211>[17]. And this texture becomes more pronounced with increasing rolling reduction.

The volume fractions of the main deformation texture components are calculated from the ODF and are summarized in Fig.2. With increasing rolling reduction from medium (70%) to high (99%) deformation degree, the tendency of a strongly increasing β-fiber texture can be observed, and S component tends to be the main rolling texture. Most importantly, the fraction of unwanted Goss component shows a slight decrease. Besides, the cube orientation fraction is about 5% and remains quasi constant for whatever reduction. The above results show that the larger rolling reduction contributes to the formation of the preferred β-fiber texture.

X-ray diffraction was performed on the annealed Cu-44%Ni alloys to understand the global recrystallization texture. From the {111} pole figures of the recrystallized samples(Fig.3), it

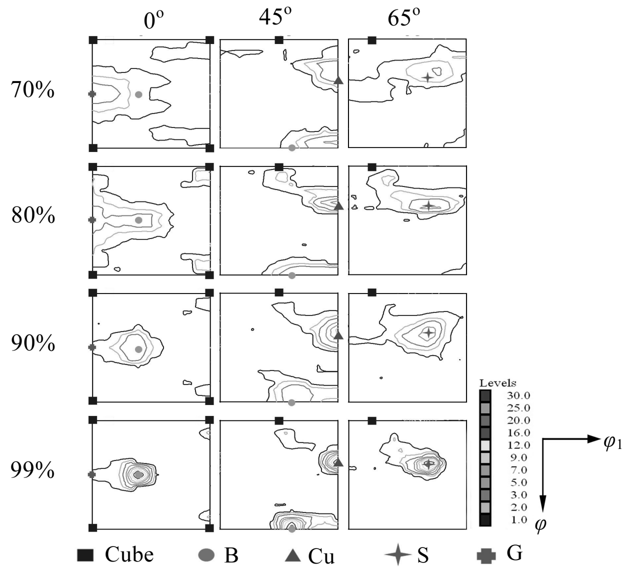

Fig.1 ODF sections (φ_2=0°, 45°, 65°) of Cu-44%Ni alloy cold rolled by different rolling reductions (70%, 80%, 90% 99%)

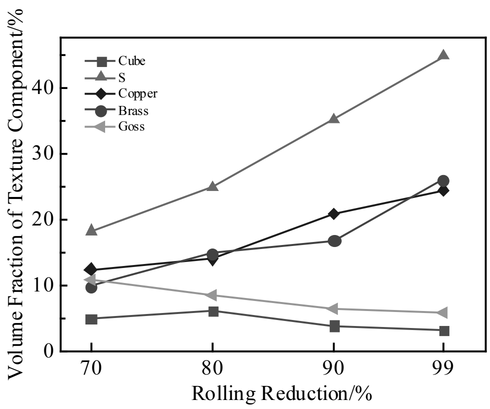

Fig.2 Volume fraction of deformation texture components in the cold-rolled Cu-44%Ni alloy affected by different rolling reductions

is found that there is a perceptible difference in the texture components of Cu-44%Ni alloy after annealing at 900 °C. The 70% cold-rolled Cu-44%Ni after high temperature annealing (Fig.3a) shows only a very weak cube texture together with a diffuse spread of orientations. But the cube texture gradually sharpens with increasing rolling reduction.

In order to understand the effect of rolling reduction on the formation of the texture and microstructures on a local scale, the annealed Cu-Ni alloys were also analyzed by EBSD. Fig.4 presents the EBSD maps of Cu-44%Ni alloys annealed at 900 °C for 1 h. Fig.5 shows the fractions of the main texture components which are measured by means of EBSD. From Fig.4 and Fig.5, it is clearly seen that the

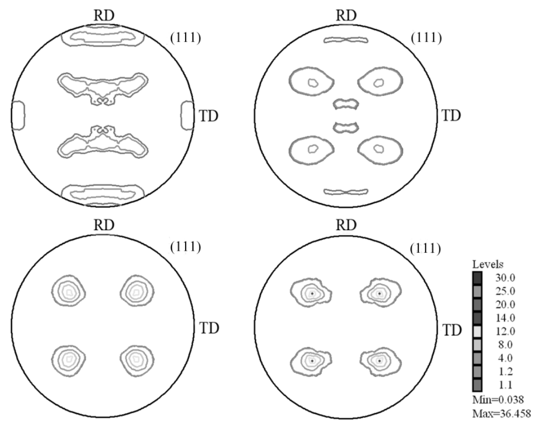

Fig. 3 {111} pole figures of cold-rolled Cu-44%Ni alloy annealed at 900 °C for 1 h after different cold-rolling reductions: (a) 70%, (b) 80%, (c) 90%, and (d) 99%

rolling reduction exerts a great influence on achieving a strong cube texture. After annealing at 900 °C, the 70% cold-rolled Cu-44%Ni shows the presence of only a few recrystallized cube grains with the majority of non-cube grains. With the rolling reduction up to 80%, the fraction of cube component in the annealed specimen increases slightly, but still has many non-cube grains. However, as rolling reduction increases from 80% to 90%, the recrystallization microstructure is dominated by the cube grains. When the rolling reduction reaches to 99%, the cube component is further strengthened after annealing at 900 °C resulting in 93.9%. Furthermore, the average recrystallized grain size decreases from 28.3 μm to 18.4 μm with increasing rolling reduction. And the sharpening of cube texture is also accompanied by the reduction of HAGBs and Σ3 boundaries. The results mentioned above are displayed in Table 1.

From above results, it is verified that increasing the rolling reduction can develop a stronger cube texture after high temperature annealed. Though the 99% cold-rolled Cu-44%Ni alloy annealed at 900 °C for 1 h makes it possible to form more than 90% of the cube grains, a small percentage of Σ3 and HAGBs (5.3% and 15.1%, respectively) are still discerned in such fully recrystallized substrates. Since Σ3 boundaries and HAGBs dramatically decrease the critical current density of the epitaxially grown YBCO layer[6], the fraction of such boundaries in the substrate must be as low as possible. Therefore, further study is needed to optimize the final texture and microstructure. The effect of annealing temperature on the cube texture and microstructure of the annealed substrate is considered in the following subsection.

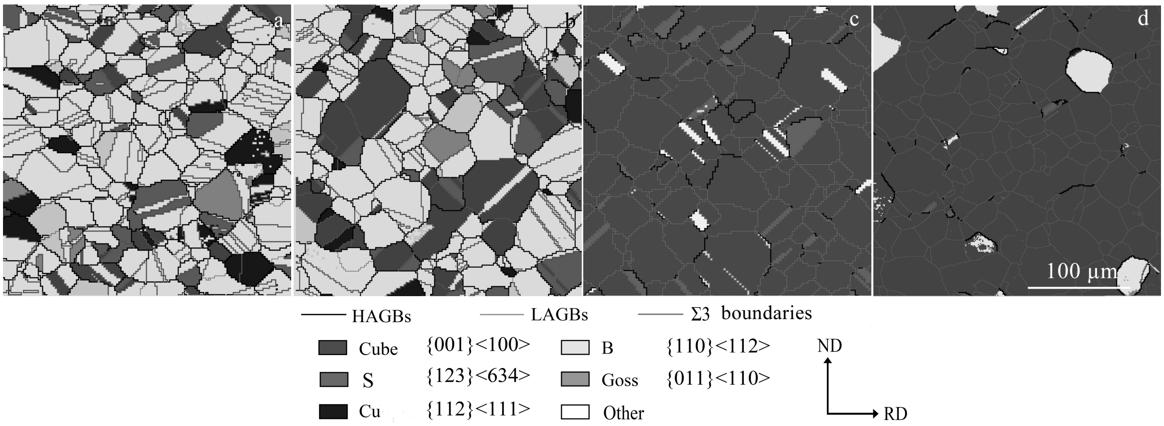

Fig.4 EBSD maps of cold-rolled Cu-44%Ni alloy annealed at 900 °C for 1 h after different cold-rolling reductions: (a) 70%, (b) 80%, (c) 90%, and (d) 99%

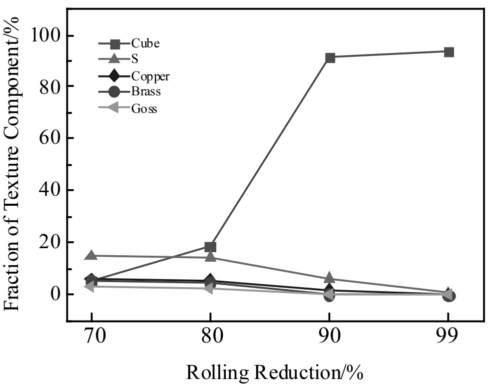

Fig.5 Fraction of main texture components of annealed Cu-44%Ni alloys as a function of rolling reduction

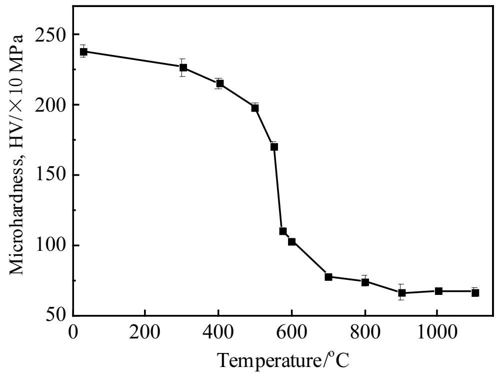

Fig. 6 Microhardness data for Cu-44%Ni alloy substrates annealed at different temperatures after cold-rolling reduction of 99%

2.2 Effect of annealing temperature on the cube texture and microstructure

Fig.6 shows the microhardness curve for the 99% cold-rolled Cu-44%Ni alloys annealed at different temperatures for 1 h. It can be seen that the hardness decreases rapidly when annealing at temperatures between 500 °C and 600 °C, indicating the occurrence of recrystallization. And the hardness does not significantly change after annealing over 700 °C, which suggests that at this stage the recrystallization is complete.

Table 1 Parameters of the microstructure and texture of the Cu-44%Ni alloy determined using the EBSD technique

Rolling reduction/%	f_{cube}/ %	Grain size/ μm	f_{HAGBs}/ %	$f_{\Sigma3}$/ %
70	5.8	28.3	96.0	45.6
80	18.6	27.0	89.0	44.3
90	87.7	21.3	40.0	23.9
99	93.9	18.4	15.1	5.3

During the process of recrystallization (550~700 °C), the deformation texture evolves into the cube texture. This can be seen in Fig.7a~7c, which shows the EBSD maps collected from the rolling plane of the Cu-44%Ni annealed at 550~700 °C. After annealing at 550 °C, a few recrystallization nuclei are observed in Fig.7a. And annealing at 600 °C results in the formation of a mixed deformed/recrystallized microstructure and the emergence of some Σ3 boundaries (Fig.7b). Recrystallized grains dominate the microstructure of the sample annealed at 600 °C, though small non-recrystallized grains are still present after annealing at this temperature. When the temperature is up to 700 °C, the recrystallization is complete. The recrystallized grains are seen with both cube and non-cube orientation (Fig.7c). During this stage, it is clearly seen that the fractions of cube texture, HAGBs and Σ3 boundaries as well as the average grain size increase sharply (Fig.7 and Fig.8).

When the annealing temperature continues to increase, the

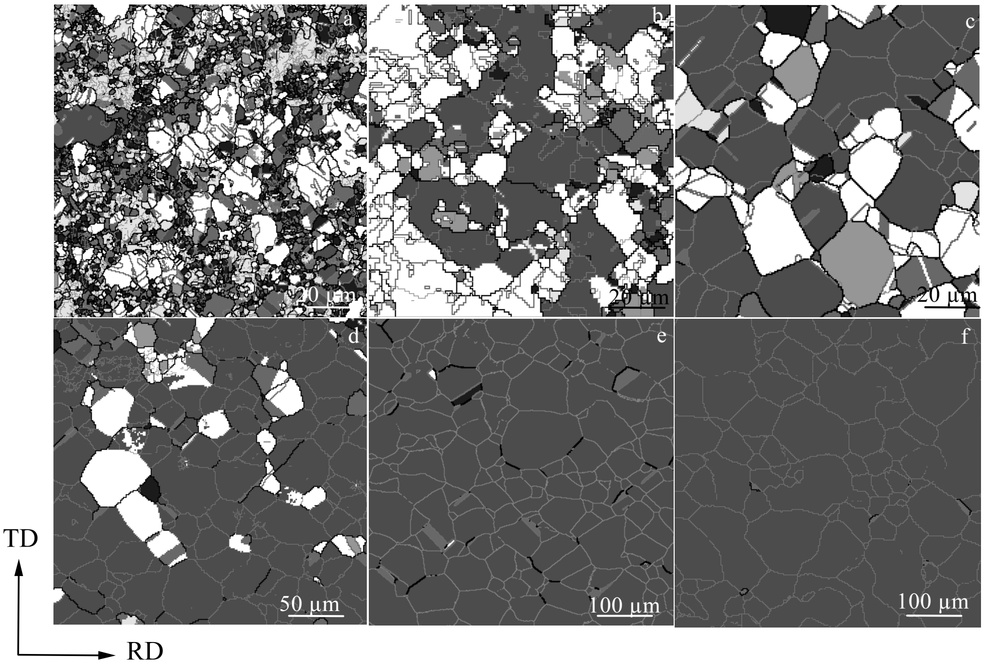

Fig.7 EBSD maps of 99% cold-rolled Cu-Ni alloy annealed at different temperatures for 1 h: (a) 550 °C, (b) 600 °C, (c) 700 °C, (d) 800 °C, (e) 1000 °C, and (f) 1100 °C

fraction of the cube texture and the average grain size increase significantly, but the fractions of HAGBs and Σ3 boundaries decrease rapidly. After annealing at 1100 °C, the fraction of cube texture increases up to 99.8%, and the average grain size increase to 34.0 μm. On the contrary, the fractions of HAGBs and Σ3 boundaries decrease to 2.5% and 1.3%, respectively. Furthermore, by the cumulative value of the area fraction of grains as a function of the tolerance angle (Fig.9), it can be seen that the cube texture gradually sharpens with increasing annealing temperature. After annealing at 1100 °C for 1 h, the cube grains component approaches to 100% when the tolerance angle is 10°.

3 Discussion

The major interest in the present study is to describe the effect of rolling reduction and annealing temperature on the formation of cube texture and microstructural evolution. As shown in Fig.10, it is clearly indicated that the recrystallized cube texture is strengthened by either increasing the rolling reduction or increasing the annealing temperature.

The rolling reduction has long been recognized as an important factor affecting the deformation structure of fcc metals[18]. In the present work, increasing the rolling reduction results in an increased fraction of β-fiber texture (B, Cu, and S) and a suppression of the Goss texture, as can be seen in Fig.3. This is consistent with the findings in other studies[10,18-21]. In all these earlier studies, the β-fiber texture became more pronounced with high rolling reduction.

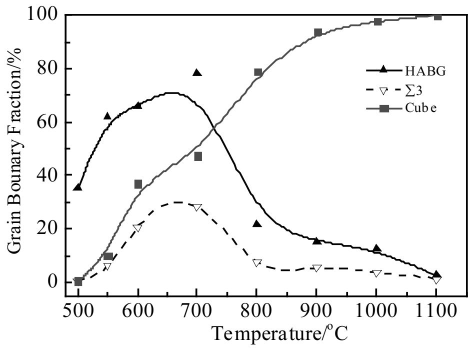

Fig. 8 Fraction of high angle grain boundary and annealing boundary for Cu-44%Ni alloy substrates annealed at different temperatures for 1 h

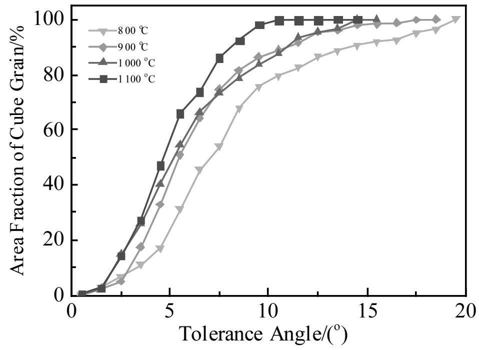

Fig.9 Distribution of the area fraction of cubic grains vs. tolerance angle

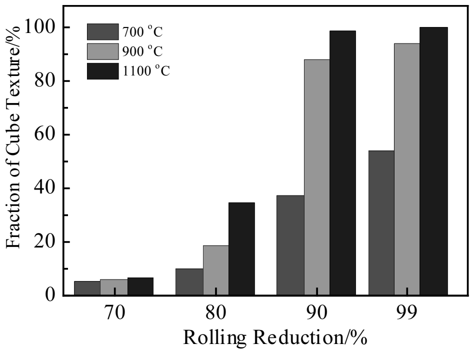

Fig.10 Fraction of cube texture of annealed Cu-44%Ni substrate

The recrystallized cube texture is strongly related to the rolling texture[22]. As illustrated in Fig.4, the textures that form during recrystallization of the different cold-rolled Cu-44%Ni alloys are significantly different. The rolling texture disappears during annealing, leading to a nearly isotropic texture for low reduction or a sharp cube texture for high reduction. It is obvious that the rolling reduction exerts a great influence on achieving a strong cube texture. This has been attributed to the stored energy of deformation associated with the number of defects such as dislocations and dislocation cell walls, which is the driving force for recrystallization[23]. The dislocation interaction is orientation dependent, and thus grains of different orientations are expected to store different amounts of energy during deformation. Etter et al.[20] reported that no distinct stored energy differences between the cube grains and their neighbors (B, Cu, and S grains) can be observed for the lower reduction in Fe-53%Ni alloy. However, all energy differences increase for the cold rolling reduction greater than 80%, which is favored for the bulging of cube grains during the recrystallization annealing. Results obtained on this Fe-Ni alloy are generally applied copper, aluminum or nickel, all of which can moderate high stacking fault energy[24]. In the present work, cold rolling up to 90% reduction could produce high stored energy differences between the cube grains and their neighbors (B, Cu, and S grains), contributing to the development of strong cube orientation. For rolling reduction higher than 90%, there is an evident advantage for the cube grains to grow into the neighboring grains of higher energy. As a result, the grains of higher energy are finally replaced by cube grains. However, for the 80% reduction, lower stored energy gap delays the cube growth and thus favors the development of other orientations. This is probably the main reason for the higher cube texture formed in the annealed sample of heavy rolling reduction. In addition, the increase in the number density of defects in the heavily deformed alloy provides more nucleation sites for recrystallization during annealing, which results in finer recrystallized grains.

In the isochronal annealing experiment, it is apparent that increasing the annealing temperature is advantageous to achieve a good cube texture for the Cu-44%Ni alloy. The average grain size, and boundary populations are also greatly dependent on the annealing temperature. During the process of recrystallization, the formation of cube texture is accompanied by the emergence of a mass of Σ3 boundaries, which is consistent with the trend of HAGBs. It can be explained that the growth of new grains leads to the migration of grain boundaries, and the grain boundary migration results in annealing twins and increases the content of HAGBs. However, further increasing annealing temperatures over 700 °C shows the Σ3 boundaries and HAGBs decrease rapidly at this stage (Fig.9). This could possibly be related to the lower mobility of Σ3 boundaries. The Σ3 boundaries are prone to be consumed by fast moving grain boundary (HAGBs) during the subsequent grain growth process[25]. On the other hand, the cube grains have a growth advantage compared with other non-cube orientated grains[26,27]. Therefore, the majority of non-cube grains are consumed by cube grains, and the growing cube grains will encounter each other and form LAGBs with further increasing annealing temperature. These results agree with previous experiments in Cu-45 at% Ni and pure Ni[25,28]. Besides, it might be important to note that the strengthening of the cube texture can be attributed to a result of normal grain growth at higher annealing temperatures[11].

From the above discussion, the recrystallization texture in the 99% cold-rolled Cu-44%Ni after annealing at 1100 °C can be seen to exhibit a high-quality cube orientation with very few undesirable boundaries, which is suited in ensuring a high critical current density. Thus, the Cu-44%Ni alloy is considered to be a better candidate for potential use as a substrate material for coated conductors.

4 Conclusions

1) A typical pure metal or copper-type rolling texture is obtained in Cu-44%Ni after cold rolling. And with increasing rolling reduction, this rolling texture is gradually strengthened. Furthermore, higher rolling reduction is favorable for forming a sharp cube texture after annealing at high temperature, especially for rolling reduction higher than 90%. Increasing the rolling reduction decreases average recrystallized grain size and HAGBs as well as Σ3 boundaries.

2) During isochronal annealing, increasing the annealing temperature is also advantageous to form a strong cube texture. The cube texture sharpens considerably at higher annealing temperatures due to normal grain growth. The trend of Σ3 boundaries is consistent with that of HAGBs, which firstly increases

during recrystallization process and then decreases with further increasing temperature. However, the average grain size increases with the cube texture content trend during total annealing process.

3) In the case of 99% cold-rolled Cu-44%Ni, after annealing at 1100 °C for 1 h, the fraction of the cube texture reaches 99.8%, the fractions of HAGBs and Σ3 boundaries are 2.5% and 1.35%, respectively. This Cu-44%Ni tape can be considered as a suitable substrate material for coated conductors.

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冷轧形变量和退火温度对 Cu-44%Ni 合金的微观组织及织构演变的影响

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摘 要：研究了冷轧变形量和退火温度对 Cu-44%Ni 合金中立方织构的形成及微观组织演变的影响。结果表明，增大变形量和提高退火温度均有利于立方织构的形成，而且在变形量大于 90%和退火温度高于 900 ℃的条件下，可以得到非常强的立方织构。另一方面，随着变形量的增加，退火孪晶（Σ3 晶界）和大角度晶界降低；但在等温退火中，随着退火温度的增加，Σ3 晶界和大角度晶界先迅速的增加，然后逐渐减少。冷轧变形量 99%的 Cu-44%Ni 合金在 1100 ℃高温退火 1 h 后可以获得了 99.8%的立方织构，并且大角度晶界和退火孪晶界分别为 2.5%和 1.3%。

关键词：冷轧变形量；再结晶；Cu-44%Ni 合金；织构；微观组织

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