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The Influence of Annealing on the Microstructure and Electrical Resistivity of Jelly-Rolled Cu-Nb Composite Wires

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***Abstract*—In this paper, the annealing effects on the microstructure and electrical resistivity of jelly-rolled Cu-x%vol. Nb (x $=$ 25, 33, 50, and 63) composite wires were investigated. During annealing, noticeable changes take place in the microstructure, including recovery and recrystallization of copper and niobium, followed by spheroidization and further coalescence of niobium filaments. With increasing annealing temperature, the diffusion-controlled coalescence of niobium filaments is enhanced due to their proximity. The behavior of the electrical resistivity in the normal state of the Cu-Nb composite shows a noticeable change at a specific value of temperature in the range between 50 K and 70 K. This temperature varies with the niobium volume fraction. Such a behavior is discussed in terms of the decrease of the amount of Cu-Nb interfaces promoted by coarsening and by electron scattering effects at these interfaces.**

***Index Terms*—Electrical resistivity, jelly-rolled Cu-Nb composites, microstructure, spheroidization.**

I. Introduction

THE Cu-Nb composites have been widely investigated due to their high strength, high electrical conductivity and mechanical workability [1]. Due to these properties, these conductors are used in the winding of pulsed high field magnets [2] as well as reinforcing stabilizers in $(Nb,Ti)_3Sn$ wires [3]. In the last application, Cu-Nb composites must be heat treated at about 700°C, which is the temperature necessary to form the A15 superconducting phase [3]. Remarkable changes occur in the microstructure of Cu-Nb conductors during annealing, including recovery and recrystallization of both metals depending on the temperature. It is well known that thermal instability mechanisms such as spheroidization tend to occur in lamellar microstructures when exposed to elevated temperatures [4], [5]. Regarding the jelly-rolled Cu-Nb conductors, the Nb layer evolves to ribbon-shape filaments when the wire is extruded and drawn to large strains. In consequence, the corresponding Cu-Nb interfacial area also increases in a significant manner. Most of the stored energy in the composite is due to the presence of a high density of geometrically necessary dislocations at these Cu-Nb interfaces to compensate strain mismatches. The high energy associated with these interfaces is the driving force for spheroidization of the niobium filaments and, eventually, their coarsening [4], [5]. In this paper, we focus on the microstructural evolution of Nb filaments in jelly-rolled Cu-Nb composite wires during annealing and the corresponding effects on their transport properties.

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II. Experimental

The jelly-rolling process was used to produce Cu-x%Nb conductors (x $=$ 63, 50, 33, and 25) with diameters of 0.8 mm. Throughout this work, the amount of Nb refers to the volume fraction. These conductors have 55 and 151 jelly-rolled elements deformed to a total logarithmic strain of $\eta = 11.8$ and $\eta = 12.8$, respectively. Details about the processing of these composites are given elsewhere [3]. Vacuum annealing of conductors was carried out at 650°C for 200 h, 850°C for 192 h, and 1000°C for 1.5 h and 4 h. The microstructural characterization of deformed and annealed specimens was performed using a XL-30/FEG Philips Scanning Electron Microscope (SEM). Microhardness testing was performed using a 50 g load. The temperature dependence of the electrical resistance, R(T), was determined using the four-point probe technique in the temperature range comprised between 4.2 and 300 K. The excitation current (200 mA) was generated by a Keithley 228 current source and the voltage was measured with a Keithley 2001 multimeter. The R(T) data were taken while warming the sample up to 300 K. The characterization of transport properties was complemented performing the voltage versus excitation current curves at 77 K and 298 K.

III. Results and Discussion

A. Microstructural Characterization

Fig. 1(a) presents the longitudinal section of the as-drawn sample Cu-63%Nb, showing the fine niobium filaments in the jelly-rolled composite. Due to size effects it is not easy to resolve individual filaments, however, an elongated structure parallel to the longitudinal direction is clearly seen.

Fig. 1(b) shows the longitudinal section of the same sample after annealing at 650°C for 200 h. This condition was chosen because many papers report similar thermal cycles during

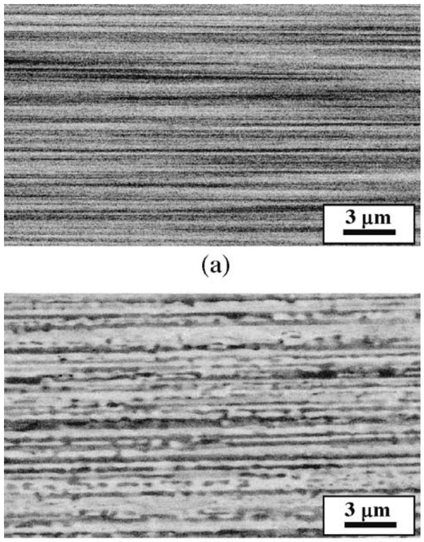

Fig. 1. Longitudinal section of the Cu-63%Nb wire with 55 jelly-rolled elements in: (a) as-drawn condition, and (b) annealed at 650°C for 200 h (FE-SEM,BSE).

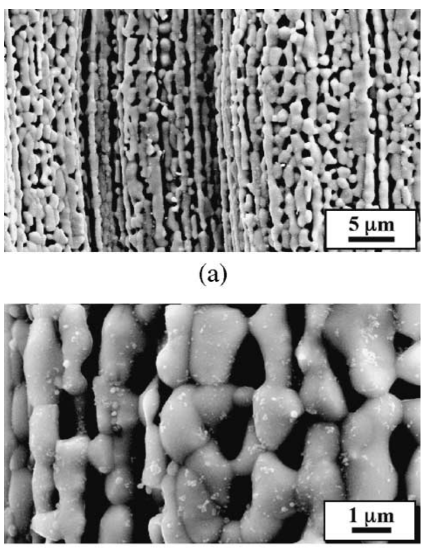

Fig. 2. FEG-SEM micrographs of the composite Cu-63%Nb with 151 jelly-rolled elements: a) niobium filaments after vacuum annealing at 850°C for 192 h. b) Enlarged view of the microstructure showing details of the microstructure.

the synthesis of Cu-Nb reinforced Nb_3Sn wires for superconducting coils [3], [6], [7]. The contrast in the SEM was enhanced to ease visualization of the niobium structures. It was found that spheroidization occurs partially throughout the microstructure. A careful inspection of this figure shows that the niobium layers break-up in many points, however, in a contrasting manner, there are evidences of coalescence of layers in close vicinity resulting in many contact points. It is worth mentioning that despite of filament break-up, the conducting path seems to be preserved.

Fig. 2 shows the microstructure of sample Cu-63%Nb in the annealed state at 850°C for 192 h. As expected, spheroidization at a higher temperature occurs intensively. The structural

TABLE I
VICKERS MICROHARDNESS OF AS-DRAWN AND ANNEALED SAMPLES OF Cu-x%vol. Nb (x = 63, AND 25), WITH 151 JELLY-ROLLED ELEMENTS

Sample	Cu-63%vol. Nb	Cu-25%vol. Nb
as-drawn	261 ± 3	196 ± 4
650°C for 200 h	157 ± 2	123 ± 2
850°C for 192 h	109 ± 3	97 ± 1
1000°C for 1.5 h	117 ± 1	111 ± 2
1000°C for 4 h	112 ± 2	109 ± 3

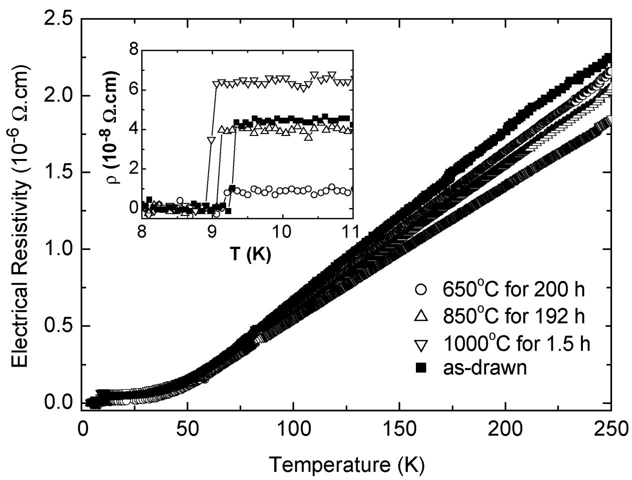

Fig. 3. Temperature dependence of the electrical resistivity for as-drawn and annealed samples of the composite Cu-63%vol. Nb (151 jelly-rolled elements). The inset shows the $\rho(\mathrm{T})$ curves in the low temperature limit.

integrity of the composite is partially preserved and the individual former jelly-rolled components are clearly distinguished. Transverse grain boundaries are noted in the microstructure as well as contact points resulting from coalescence of adjacent Nb layers.

This peculiar morphology consisting of individual grains stacked one after another is known as bamboo structure.

Table I displays the results of microhardness of as-drawn and annealed samples of Cu-x%Nb ($x = 63$, and 25). Results confirm the pronounced softening of the composite during annealing. Regarding the as-drawn samples, the hardness of the sample with $x = 63$ is higher than the observed for $x = 25$ as a consequence of the higher number of Cu-Nb interfaces. In the case of the samples annealed at 1000°C, the results show that softening is more pronounced with increasing annealing time. An additional softening of the composite is not expected for longer annealing times since the observed values are very close, indicating the flattening of the softening curve. A similar trend was found for the composites with 55 jelly-rolled elements. In general, the analysis of the results showed in Table I confirms that hardness decreases monotonically with temperature for all the samples investigated in this work. Oxygen contamination was not significant during annealing.

B. Electrical Characterization

Fig. 3 shows the temperature dependence of the electrical resistivity, $\rho(\mathrm{T})$, of as-drawn and annealed samples of the composite Cu-63%Nb, in the temperature range $4.2\ \mathrm{K} < \mathrm{T} < 250\ \mathrm{K}$. The inset shows the $\rho(\mathrm{T})$ behavior in the

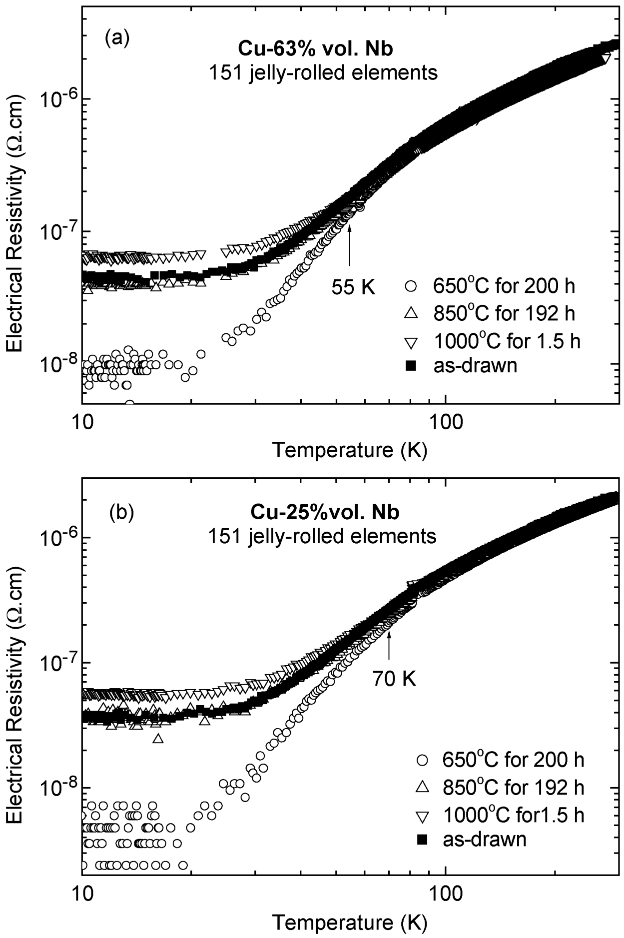

Fig. 4. Temperature dependence of the electrical resistivity for as-drawn and annealed samples of the composite Cu-x%Nb (a) $x = 63$ and (b) $x = 25$.

limit of low temperatures. Compared to the as-drawn condition, the annealed samples show a little drop in superconducting critical temperature (T_c). Regarding the electrical resistivity ρ_N of the Cu-Nb composite above T_c two important features can be identified in these curves:

1) For temperatures just above T_c, we have found that annealing at 650°C for 200 h appreciably depress ρN. For annealed samples at 850°C, ρN is very close to the correspondent one in the as-drawn condition. However, annealing at 1000°C increases the magnitude of ρN. Such a behavior was verified for all Nb volume fractions in the Cu-Nb composite samples.
2) At higher temperatures, there is a noticeable change in the behavior of the $\rho(T)$ curves. Such a behavior is evident in Fig. 4. For instance, this temperature is about 55 K for the composite with 63%Nb. For the lower Nb fraction, this temperature increases to about 70 K. Such a change occurs for all Nb volume fractions in the Cu-Nb composite, at different temperatures. In the high temperature limit, for the Cu-63%Nb, we have found that ρ_N decreases with increasing annealing temperatures. In this limit, for all Nb volume fractions in the Cu-Nb composite, the magnitude of ρ_N for annealed sample at 1000°C is lower than that for as-drawn condition. The opposite behavior was verified in the temperature range just above T_c.

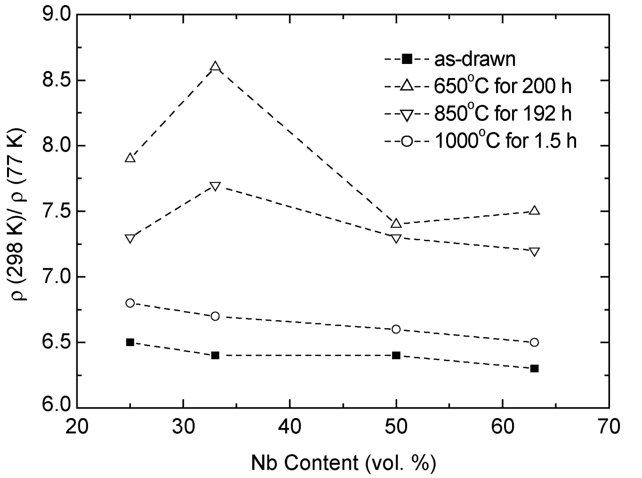

Fig. 5. Variation in the resistivity ratio, $\rho_{298\ K}/\rho_{77\ K}$, for Cu-Nb wires (151 jelly-rolled elements) as a function of Nb content in the as-drawn and annealed states.

It is well known that the electrical resistivity in the Cu-Nb conductor depends mainly on the component associated with electron scattering at Cu-Nb interfaces [8], [9]. The total area provided by these interfaces is strongly decreased due to coarsening [10]. For the Cu-Nb composite investigated in this work, such a behavior is evident from Fig. 2(b).

At light of the results above described, in the high temperature limit (e.g., $T > 70$ K for Cu-25%Nb), coarsening of niobium filaments can explain the lower magnitude of ρ_N for the annealed samples compared to the as-drawn condition. However, in order to understand the behavior of ρ_N in the low temperature limit, another phenomenon has to be taken into account. When the interfacial spacing between two-phase boundaries becomes comparable to or smaller than the mean free path of the electrons, a remarkable drop in conductivity can be expected [11]. The mean free path of electrons in copper at 75 K is about 140 nm [12]. A close inspection on the microstructure displayed in Fig. 2(b) shows a wide distribution of interfilament spacings for the Cu-Nb composite annealed at 850°C. It is worth mentioning that copper has been etched away in this micrograph. However, these values are comparable in magnitude (≈100 nm–500 nm) with the mean free path of electrons reported for copper at 75 K. Therefore, it is likely that the combination of coarsening with the increase of the mean free path of a conduction electron in low temperature limit would be the mechanism responsible for the crossover feature on the ρ_N behavior. Moreover, these phenomena can explain the highest magnitude of electrical resistivity for annealed samples at 1000°C, since the coarsening of the niobium filaments is more significant at this annealing temperature.

Hong and Hill [12] have reported that the changes of the resistivity ratio $\rho_{295\ K}/\rho_{75\ K}$ with annealing temperature show the same trend as that of the electrical conductivity. From the I-V curves taken at room temperature and in liquid nitrogen bath, we have obtained the resistivity ratio $\rho_{298\ K}/\rho_{77\ K}$ of Cu-x%Nb jelly-rolled wires, as shown in Fig. 5. Except by the results observed in the resistivity ratio $\rho_{298\ K}/\rho_{77\ K}$ for annealed samples at 650°C and 850°C for Cu-33%Nb, there is a slight decrease in this resistivity ratio with increasing Nb content. Such a behavior might be associated with the increasing fraction of interfaces with increasing Nb volume fractions in the Cu-Nb composite [12]. Furthermore, from Fig. 5, it is clear that the resistivity ratio $\rho_{298\ \mathrm{K}}/\rho_{77\ \mathrm{K}}$ for the samples annealed at 1000°C is higher than the correspondent ones for the as-drawn condition. Such a result mirrors the $\rho(\mathrm{T})$ behavior for Cu-Nb composite annealed at 1000°C in the high temperature limit, displayed in Fig. 3.

IV. Conclusions

We investigated the microstructure and the electrical properties of a jelly-rolled Cu-x%vol. Nb ($\mathrm{x} = 25$, 33, 50, and 63) conductors in as-drawn and annealed states. Based on this investigation, the following conclusions can be drawn:

1) Partial spheroidization of niobium filaments is evident for samples annealed at 650°C for 200 h. Annealing at higher temperatures leads to spheroidization in a larger extent but new contact points resulting from coalescence of neighboring Nb filaments arise in the microstructure.
2) The $\rho(\mathrm{T})$ behavior shows a noticeable change at a specific temperature in the range comprised between 50 K and 70 K, depending on the Nb content. At temperatures just above $\mathrm{T_c}$, the electrical resistivity for the samples annealed at 1000°C is higher than the observed for the as-drawn condition. The opposite behavior is verified at room temperature. One possible explanation for such a behavior is the temperature dependence of the electron interface scattering.

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