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S. H. Jang
Sungkyunkwan University, South Korea

J. H. Lim
Sungkyunkwan University, South Korea

J. K. Kim
Sungkyunkwan University, South Korea

B. K. Ji
Sungkyunkwan University, South Korea

J. Jo
Sungkyunkwan University, South Korea

See next page for additional authors

Authors
S. H. Jang, J. H. Lim, J. K. Kim, B. K. Ji, J. Jo, W. Nah, J. S. Volf, Hua-Kun Liu, and M. Apperley

Characterization of Thermal Conductivity and Mechanical Properties of Ag-Alloy Sheathed Bi(Pb)-Sr-Ca-Cu-O Superconductor Tape

Seok Hern Jang, Jun Hyung Lim, Jung Ho Kim, Bong Ki Ji, Jinho Joo, Wansoo Nah, John Slavko Volf, Hua Kun Liu, and Miles Apperley

***Abstract*—We evaluated the effect of alloying additions to Ag on thermal conductivity and mechanical properties of Ag-alloy sheathed Bi-2223 (BSCCO) superconductor tape. The tapes were made with combinations of Ag alloys such as Ag-Mg, Ag-Sb, and Ag-Au for inner and outer sheath. Thermal conductivity of the tapes was evaluated by using thermal integral method at 10–120 K.**

It was observed that the addition of alloys reduced remarkably thermal conductivity and improved mechanical strength. The thermal conductivity for Ag-Mg, Ag-Sb, and Ag-Au at 40 K was measured to be 411.4, 142.3, and 109.7 $\mathbf{W/(m \cdot K)}$, respectively, which is approximately 2 to 9 times lower than that of Ag (1004.6 $\mathbf{W/(m \cdot K)}$). In addition, the thermal conductivity of alloy-sheathed tape significantly depended on their thermal conductivity of sheath materials. For Ag-alloy sheathed tapes, the thermal conductivity was much lower (i.e., 5 –18 times lower) than that of the Ag sheathed tape The mechanical property of alloy-sheathed tape was also evaluated and correlated to the microstructural evolution.

***Index Terms*—Alloys, BSCCO tape, thermal conductivity, thermal integral method.**

I. INTRODUCTION

DEVELOPMENTAL techniques for Ag sheathed BSCCO tape made by the powder-in-tube (PIT) process have been well established, and can be applied to various power systems such as motors, transformers, current limiters, and power cables. For these systems, it is necessary to use current leads with high transport properties in order to deliver power at liquid helium. Recently, sheathed HTS current leads made by stacking Ag-sheathed BSCCO tape have received considerable attention.

As a substitute for bulk type HTS current leads, the current leads made of BSCCO tape have several advantages in regard of high critical current density (J_c), good strain tolerance, and ease in making conductors with a long lengths and various geometries. The only weakness is significant helium consumption caused by heat leakage due to the high thermal conductivity of the Ag sheath. In addition, high electrical conductivity of the Ag sheath limits the application of the BSCCO tape for AC applications. Sheath materials with lower thermal and electrical conductivity need to be developed in order to extend the application range of the BSCCO tape, and this can be done by further developing Ag alloys. Furthermore, the addition of alloying elements to Ag is expected to improve the mechanical strength of the tape.

Much research has been performed to develop various Ag alloys as sheath materials [1]–[3]. In these studies, however, the thermal, electrical, and mechanical properties of both the sheath alloys and the Ag-alloy sheathed HTS tapes were not systematically evaluated and related microstructural investigation was not presented. In our study, therefore, we fabricated Ag alloys of Ag-Au, Ag-Sb, and Ag-Mg and characterized the thermal conductivity at 10 to 120 K. 37-multifilament tapes were then made with combinations of these Ag alloys for inner and outer sheath and the thermal and electrical conductivity of tapes were evaluated. In addition, the effect of adding alloying elements on the mechanical strength and microstructural morphology was evaluated.

II. EXPERIMENTAL PROCEDURE

Sheath alloys were fabricated by adding small amounts of Au, Sb, and Mg elements to Ag (99.99% purity) and melting them in a high frequency induction furnace. To make a solid solution, the contents of the alloying-elements were selected to be less than their solubility limits for Ag [4]. The compositions of the Ag alloys were Ag-Au(7.0 at.%), Ag-Sb(1.1 at.%), and Ag-Mg(0.3 at.%) (hereafter, denoted AgAu, AgSb, and AgMg, respectively). Ag and Ag alloy billets were extruded into hollow tubes and heat treated for 8 h in N_2 atmosphere.

$Bi_{1.8}Pb_{0.4}Sr_2Ca_{2.2}Cu_3O_x$ (BSCCO) compound was synthesized by the solid state reaction with constituent powders and 37-filament tape was fabricated using the conventional PIT process. A detailed explanation for the BSCCO powder and tape fabrication process in our study is given in [5], [6]. To evaluate the effect of sheath alloys and their configuration on the properties of tape, various combinations of Ag and Ag alloys were selected as the inner and outer sheath. In our experiment, seven specimens with different sheath configuration were fabricated and their inner and outer sheathes were Ag and Ag, Ag-Au and Ag-Au, Ag and Ag-Sb, Ag-Sb and Ag, Ag-Sb and Ag-Au, Ag-Au and Ag-Sb, and Ag-Au and Ag-Mg,

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S. H. Jang, J. H. Lim, J. H. Kim, B. K. Ji, and J. Joo are with the School of Metallurgical and Materials Engineering, SungKyunKwan University, Suwon, 440-746, South Korea (e-mail: jinho@skku.ac.kr).

W. Nah is with the School of Electrical and Computer Engineering, SungKyunKwan University, Suwon, 440-746, South Korea.

J. S. Volf and H. K. Liu are with the Institute for Superconducting and Electronic Materials, University of Wollongong, Wollongong NSW 2522, Australia.

M. Apperley is with the Australian Superconductors, Engineering and Innovation Education Centre, Coniston NSW 2500, Australia.

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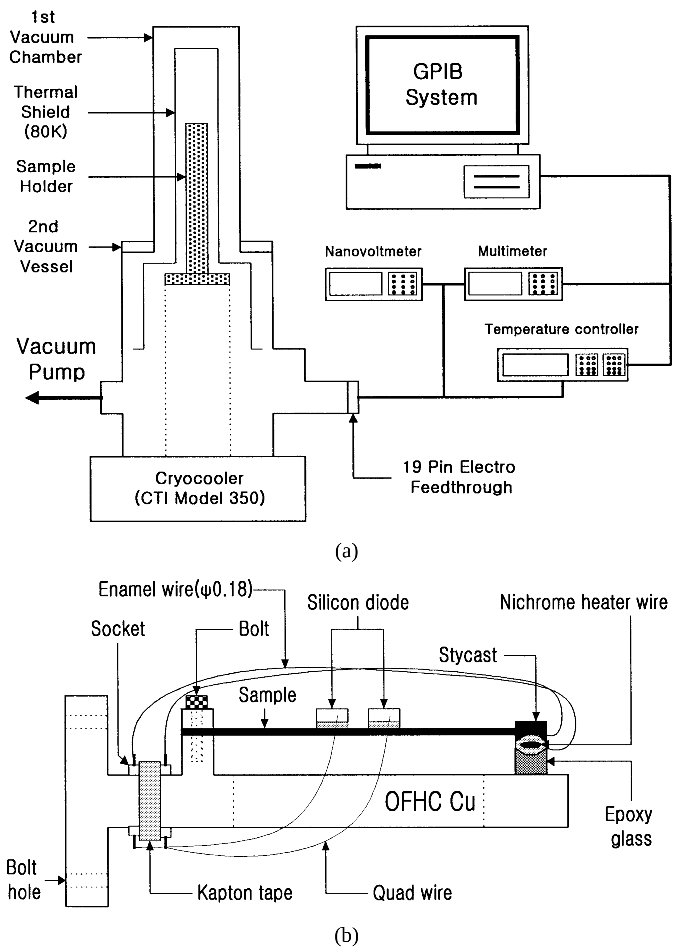

Fig. 1. Schematics of (a) thermal conductivity measurement system and (b) sample holder, sample, diode, heater, and their connections.

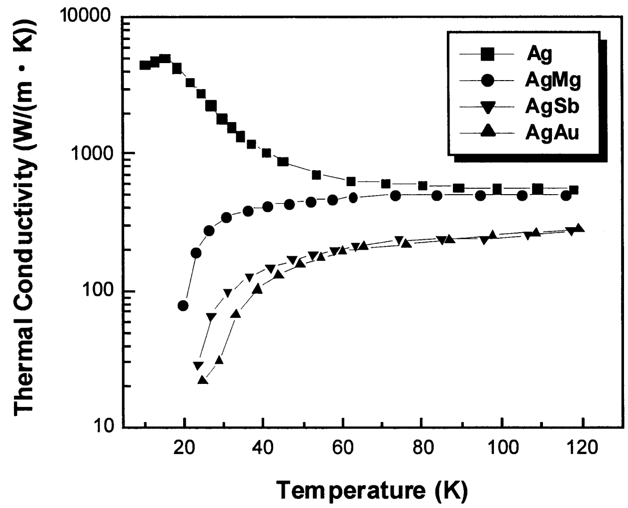

Fig. 2. Thermal conductivity of Ag and Ag alloys from 10 to 120 K.

respectively (hereafter, denoted AgSb-AgAu for specimen with inner sheath of Ag-Sb and outer sheath of Ag-Au alloys, for example). These tapes were sintered at 840°C in an ambient atmosphere for 50 h. The final thickness and width of the tapes were 0.3 mm and 2.8 mm, respectively.

Thermal conductivity was directly evaluated for Ag, Ag alloys, and BSCCO tape using the thermal integral method given by following equation;

$$Q = \frac{A}{L}\int_{T1}^{T2} k dT, \tag{1}$$

where, Q is the heat flux (W), A is the cross-sectional area (m^2), L is the length of the specimen (m), and k is the thermal conductivity ($W/(m \cdot K)$). In the measurement, a cryocooler (CTI model 350) was used to vary the temperature from 10 to 120 K, and the specimen was loaded onto a copper holder in the chamber as shown in Fig. 1. To reduce the heat flow through ambient atmosphere and conduction through the setup, the atmosphere was set to 10^{-6} torr and epoxy glass was inserted between the heater and the copper holder. Once the chamber was cooled down to 10 K, a predetermined current was applied to the heater to induce a temperature gradient (ΔT) of approximately 1 K on the specimen and held until a steady state condition was obtained. The resulting temperature gradient on the specimen was measured using Si-diode temperature sensors. The measurement was done at approximately 20 different temperatures between 10 and 120 K.

Strength was measured in a tension test on a universal tensile testing system (Instron-5655). Electrical conductivity of Ag and Ag alloys was measured by the four-probe method at 10–120 K and room temperature (300 K). Microstructure was characterized by optical and scanning electron microscopy (SEM).

III. Results and Discussion

The measured thermal conductivity of Ag and Ag alloys at 10–120 K is shown in Fig. 2. For pure Ag, the thermal conductivity was almost constant as the temperature decreased from 120 to 80 K. However, the value increased as the temperature decreased further and reached a peak value of 4942.0 $W/(m \cdot K)$ at approximately 15 K, and then decreased slightly with decreasing temperature to 10 K. This variation of thermal conductivity at low temperatures is similar to the typical trend of those for pure materials. The significant increase in thermal conductivity at low temperatures is related to the combined effect of specific heat and mean free path of electron and lattice wave [7].

On the other hand, the thermal conductivity of Ag alloys was lower than that of Ag. It seems that the lower value of Ag alloys is partly due to existence of alloying element as a form of solute or segregated one in matrix, which results in reduction of mean free path of electron and/or lattice wave, i.e., increased thermal resistance due to scattering at impurities and defects. Specifically, there was no sharp peak at low temperature for Ag alloys, which resulted in further decrease of the thermal conductivity with decreasing temperature. The thermal conductivity of AgMg, AgSb, and AgAu at 40 K was measured to be 411.4, 142.3, and 109.7 $W/(m \cdot K)$, respectively, which is approximately 2 to 9 times lower than that of Ag (1004.6 $W/(m \cdot K)$). In general, the thermal conductivity is expected to decrease with increasing amount of the alloying element. Although the contents of Sb (1.1 at.%) is much smaller than that of Au (7.0 at.%), the values of AgSb and AgAu are close to each other, indicating that Sb is more effective in reducing the thermal conductivity of Ag alloys than Au.

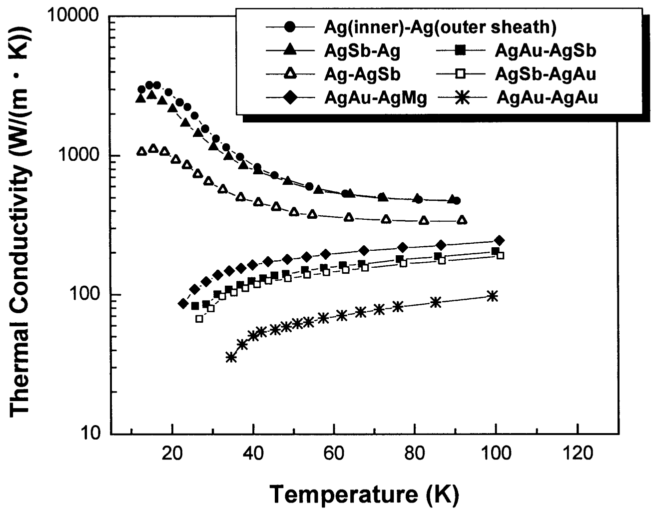

Fig. 3. Thermal conductivity of Ag and Ag alloyed tapes.

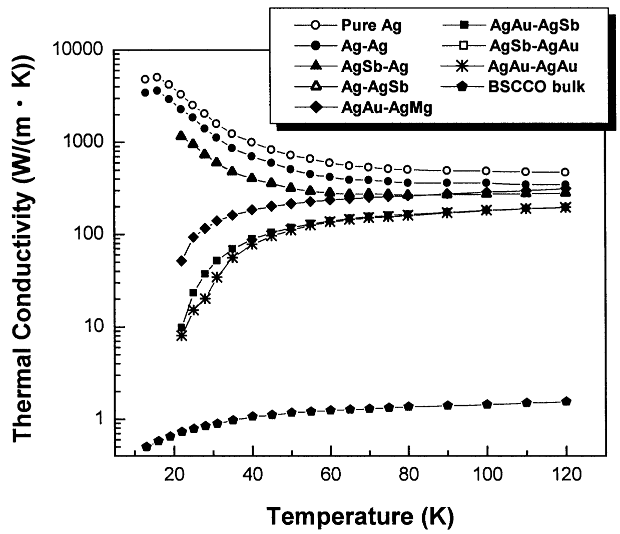

Fig. 4. Calculated thermal conductivity of Ag and Ag alloyed tapes from 10 to 120 K.

The electrical conductivity of Ag and Ag alloys was measured at 300 K and 10–120 K. For Ag, the electrical conductivity at 300 and 40 K was measured to be 0.62 and 8.13 $\mu\Omega^{-1}\text{cm}^{-1}$, which is consistent to literature value (0.63 $\mu\Omega^{-1}\text{cm}^{-1}$ at 300 K) [8]. The corresponding values for Ag alloys at 300 and 40 K were in the range of 0.21–0.41 and 0.40–3.25 $\mu\Omega^{-1}\text{cm}^{-1}$, respectively. As expected, the electrical conductivity for both Ag and Ag alloys increased as temperature decreased. In addition, the electrical conductivity for Ag alloys is several times less than that of Ag. This result is reasonable because the thermal conductivity can be related to the electrical conductivity by Wiedermann-Franz law. It should be kept in mind, however, that Wiedermann-Franz law is not strongly valid for $T \ll \theta_D$ when electrical and thermal relaxation times become incomparable.

To evaluate the effect of sheath alloys on the thermal conductivity of superconductor tape, we used Ag and Ag alloys as inner or outer sheath in various combinations for BSCCO tapes. Fig. 3 shows the thermal conductivity of tapes versus temperature. It can be seen that the thermal conductivities of tapes changed with the various sheath materials. Ag-Ag tape had highest value and showed sharp increase as the temperature decreased. The thermal conductivity at 40 K was measured to 889.6 $\text{W}/(\text{m}\cdot\text{K})$. On the other hand, the thermal conductivity was lower and did not show a peak at lower temperature for Ag-alloy sheathed tapes. The values at 40 K were 783.7, 465.9, 162.7, 125.3, 114.8, and 49.5 $\text{W}/(\text{m}\cdot\text{K})$ for AgSb-Ag, Ag-AgSb, AgAu-AgMg, AgAu-AgSb, AgSb-AgAu, and AgAu-AgAu tape, respectively.

It was also noted that for specimens with both inner and outer sheath made of alloys, the thermal conductivities were much lower (i.e., 5–18 times lower) than that of the Ag-Ag tape. In view of the fact that sheathed HTS current leads are used in the temperature range of 4.2 to 77 K, it is expected that current leads made of Ag-alloy sheathed superconductor tape effectively reduce helium loss due to the lower thermal conductivity.

Assuming the heat flows parallel through both the sheath and core, the thermal conductivity of tape can be calculated from the values of sheath and core. By assuming that thermal conductivity complies with rule of mixture, the thermal conductivity of tape can be calculated as;

$$k_t = k_{\text{BSCCO}} \cdot f_{\text{BSCCO}} + k_{\text{IS}} \cdot f_{\text{IS}} + k_{\text{OS}} \cdot f_{\text{OS}}, \qquad (2)$$

where k_t, k_{BSCCO}, k_{IS}, and k_{OS} are the thermal conductivity of tape, BSCCO core, inner sheath, and outer sheath, respectively. f is the fraction of cross-sectional area for the corresponding sheath. In our specimens, the fraction of BSCCO core and inner and outer sheath was measured to be 0.28, 0.36, and 0.36.

The calculated thermal conductivity of tapes is shown in Fig. 4. The thermal conductivity of pure Ag and bulk BSCCO [9] was inserted for comparison. Because of the same cross-sectional area of inner and outer sheath, the thermal conductivity was the same for AgSb-Ag and Ag-AgSb and AgAu-AgSb and AgSb-AgAu, respectively. The calculated thermal conductivity of tape showed the similar trend and was close to the measured one. The thermal conductivity at 40 K for Ag-Ag, AgSb-Ag, AgAu-AgMg, AgAu-AgSb, and AgAu-AgAu tape was 707.3, 404.2, 185.7, 89.8, and 78.3 $\text{W}/(\text{m}\cdot\text{K})$, respectively, indicating a difference in the range of 13–58%, compared to the thermal conductivity of the measured sample.

Fujishiro *et al.*, also showed that rule of mixture could apply for thermal conductivity of Ag-Au sheathed tape having various BSCCO core fractions [10]. Our result, together with Fujishiro's indicate that the thermal conductivity of BSCCO tape with both arbitrary cross section ratio and various sheaths materials can be practically estimated from the value of its constituents.

Yield strength and ultimate tensile strength were determined from the stress-strain curve for Ag-Ag and selected alloy-sheathed tapes. As shown in Table I, the yield strength and ultimate tensile strength of Ag-Ag was 34.5 and 44.2 MPa, respectively. Corresponding values of alloy-sheathed tapes were higher than those of Ag-Ag; yield strength of 40.2–91.7 MPa and ultimate tensile strength of 49.9–98.9 MPa. In contrast, the elongation of Ag was higher than that of Ag alloys. It is to be noted that mechanical integrity increased but workability

TABLE I
VARIATIONS OF YIELD STRENGTH, ULTIMATE TENSILE STRENGTH, AND ELONGATION OF AG AND AG ALLOYED TAPES AFTER ANNEALING

Specimens	Yield strength (MPa)	Ultimate tensile strength (MPa)	Elongation (%)
Ag-Ag	34.5	44.2	11.0
AgAu-AgAu	40.2	49.9	10.8
AgAu-AgSb	54.3	68.5	10.7
AgAu-AgMg	91.7	98.9	5.9

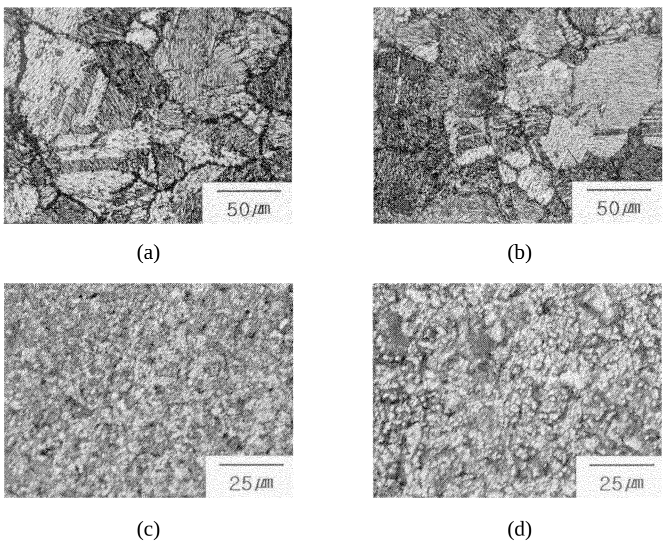

Fig. 5. Optical photographs showing the microstructure of (a) Ag, (b) AgAu, (c) AgSb, and (d) AgMg after annealing.

decreased with a presence of alloying element. It is likely that the improvements of strength for the alloy-sheathed tapes are attributed to strengthening mechanisms such as solid solution hardening, dispersion hardening, or grain size strengthening.

Microstructural investigation showed that the grain size of outer sheath significantly varied with the alloying elements as shown in Fig. 5. It can be observed that the grains in both Ag and Ag alloy sheath are almost equiaxed even though these specimens underwent a substantial amount of deformation. The grain diameter of Ag was measured to be 48.0 μm, while those of the AgAu, AgSb, and AgMg sheath were 29.3, 2.6, and 3.9 μm, respectively. In spite of the content of Sb and Mg being smaller than that of Au, the grain size in AgSb and AgMg sheath was significantly smaller than AgAu. This suggests that Sb and Mg elements restrict grain growth more than Au of the Ag matrix. It is known that the decrease of the grain size in the AgMg alloys is related to the existence of alloying elements in both solute and segregated form such as MgO [3], resulting in smaller grain size. Hence, it is believed that the smaller grain size in the Ag alloys is contributes to the higher strength of alloy-sheathed tape than that of Ag-Ag tape.

IV. CONCLUSIONS

We evaluated the effect of alloying additions to Ag on thermal conductivity and mechanical properties of Ag-alloy sheathed Bi-2223 superconducting tape. It was observed that the addition of alloys significantly reduced the thermal conductivity. The thermal conductivity for AgMg, AgSb, and AgAu at 40 K was measured to be 411.4, 142.3, and 109.7 $\mathrm{W}/(\mathrm{m}\cdot\mathrm{K})$, respectively, which is approximately 2 to 9 times lower than that of Ag ($1004.6\ \mathrm{W}/(\mathrm{m}\cdot\mathrm{K})$).

The thermal conductivity of alloy-sheathed tape significantly depended on their thermal conductivity of sheath materials. For Ag-alloy sheathed tapes, the thermal conductivity was much lower (i.e., 5 –18 times lower) than that of the Ag-Ag tape and did not show a peak at lower temperature. The calculated thermal conductivity of tape from the corresponding value of sheath and core was close to the measured one. Ag alloy-sheathed tape also showed improved mechanical properties probably due to strengthening mechanisms such as solid solution hardening, dispersion hardening, or grain size strengthening.

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