Low Thermal Conductive Bi-2223 Tapes Sheathed with Ag-Au Alloys

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Abstract- With the view of applying to power current leads for superconducting magnet systems and for other cryogenic power handling systems, low thermal conductive Bi-2223 superconducting tapes sheathed with Ag-Au alloy were fabricated and their thermal conductivities were measured from 12 to 260K. The critical current density (*overall-J_c*) was about 1700A/cm² at 77K, 0T and remained nearly constant irrespective of Au concentration up to 11 at.%. The tape sheathed with Ag+11at.%Au alloy, of which the superconductor cross-section ratio f_{sc} was 0.65, had a thermal conductivity value about 0.2W/cmK at 77K. This value is as low as that of Cu-Zn. It was found that the thermal conductivity of the tape was close to the calculated one based on f_{sc} and the independently measured thermal conductivities of the Ag-Au alloy and the Bi-2223 superconductor. The superconducting tapes sheathed with the alloy were confirmed to be suitable for the application as power current leads.

I. INTRODUCTION

Since the discovery of high-T_c oxide superconductors, a great deal of research has been performed for their application to the cryogenic equipment such as high field superconducting magnets (SCM) and SCM energy storage (SMES) systems. For the power current lead use, both bulk oxide superconductors without metal stabilizer and oxide superconducting tapes sheathed with pure silver (Ag) have been proposed in place of conventional copper leads [1]-[3]. Due to the small thermal conductivity of bulk oxide superconductors, heat intrusion through the leads can be kept low. Unfortunately, the relatively low critical current density (J_c) and fragility are disadvantages of the bulk superconductors which must be overcome. Oxide superconducting tapes sheathed with pure Ag have several advantages; the high-J_c characteristics due to the increase of the crystal orientation in the core superconductor, the flexibility in use and an increase in thermal stability, etc. [4],[5]. The high thermal conductivity of the pure Ag sheath, however, causes a serious problem of the heat intrusion into the cryogenic apparatus. For power current leads, it is necessary to develop a new sheathing material to replace pure Ag which has a thermal conductivity as low as possible and which does not cause degradation of the superconducting characteristics such as T_c and J_c.

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A popular method of reducing the thermal conductivity of metals is alloying. As Ag is generally congenial to oxide superconductors, we fabricated the Ag-Au and Ag-Cu alloys as sheathing materials and investigated the dependence of the thermal conductivity and the electrical resistivity on the Au and Cu contents in the alloys [6]. It was found that the thermal and electrical conductivities of the Ag-Au alloy tapes, which were subjected to necessary heat treatment in oxidizing atmosphere to realize the superconducting tape, drastically decreased with increasing Au content up to 11at.%. By contrast, the thermal and electrical conductivities of Ag-Cu alloy tapes hardly decreased with an increase in Cu content because of the oxidation of Cu during the heat treatment. The superconducting characteristics of $Bi_2Sr_2CaCu_2O_X$ (Bi-2212) thick films on these alloy substrates prepared by the doctor blade method [7] and $Bi_2Sr_2Ca_2Cu_3O_X$ (Bi-2223) superconducting tapes sheathed with the same alloys [8] were previously investigated. The Ag-Cu alloys reacted with the Bi-based superconductors and caused degradation of the superconducting characteristics. On the other hand, Ag-Au alloys did not react with the Bi-based superconductors and are very promising materials for the metal stabilizer as a possible replacement for pure Ag.

In the following, we investigate the possibility of applying Bi-2223 superconducting tapes sheathed with Ag-Au alloys to the power current leads. Firstly, we report the thermal conductivities of the Bi-2223 superconducting tapes sheathed with Ag-Au alloys from 12 to 260K. The thermal conductivity is also calculated by use of the thermal conductivities of both the Ag-Au alloy and the Bi-2223 superconductor, and the superconductor cross-section ratio f_{sc} which refers to the ratio of the core superconductor cross section to the total cross section of the tape. The calculated thermal conductivity is compared with the measured one and the estimation of the thermal conductivity is discussed for the current lead design. Secondly, the relation between the

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overall-J_c at 77K, 0T and the Au content in the Ag-Au alloy sheath is investigated. Thirdly, the heat intrusion through a 100cm current lead of 1000A with a stacking structure made by assembling Ag-Au sheathed tapes is theoretically determined using the characteristics of the tape. Lastly, the electrical resistivities of the Ag-Au alloy tapes are measured under magnetic fields up to 12T in order to estimate the effect of the magnetoresistance. The influence of magnetic field on the thermal conductivity of the core superconductor is also discussed.

II EXPERIMENTAL

A. Sample preparation

Ag-Au alloys containing Au from 0 to 30 at.% were prepared using an RF induction furnace in a pure Ar atmosphere. Ag and Au of 4N (99.99%) grade were used as raw materials. For the preparation of the superconducting tapes, the alloy was made into the tape-shape. $Bi_{1.8}Pb_{0.4}Sr_{2.0}Ca_{2.2}Cu_{3.0}O_x$ (Bi-2223) oxide superconducting powder was fabricated as follows; Bi_2O_3, PbO, $SrCO_3$, $CaCO_3$ and CuO raw powder of 4N grade were mixed and calcined at 800°C for 20 hours. After pulverizing the calcined material, the resulting powder was wrapped with alloy tape and then cold-rolled into a tape. Finally, the tape was heat treated at the maximum temperature of 840°C in air which was necessary for Bi-2223 superconducting tape. The finished tape was about 300μm in thickness and about 8mm in width. Sintered Bi-2223 superconducting polycrystal was also fabricated under the same heat treatment conditions using the calcined powder in order to evaluate the thermal conductivity of the core superconductor. Ag-Au alloy tapes without the superconducting core were also fabricated with about 50μm in thickness and about 2mm in width to measure the thermal conductivity and electrical resistivity. The alloy tapes were heat treated under the same conditions as the superconducting tapes.

B. Measurements

The thermal conductivities of the sheathed Bi-2223 superconducting tapes, the alloy tapes and the sintered Bi-2223 polycrystal were measured using a steady state heat flow method from 12 to 260K with an automated measuring system of our own design [9]. The system made use of a Gifford-McMahon (G-M) cycle helium refrigerator as a cryostat. One end of the sample was soldered to the cold head of the refrigerator and a small resistance heater was attached to the other end of the sample by GE7031 varnish. Au+0.07at.%Fe - chromel thermocouples of 73μm in diameter were used differentially to measure the differences in temperature. The electrical resistivity was measured by a conventional four-probe method in the same refrigerator from 12 to 300K, and in liquid helium at 4.2K. In order to investigate the influence of the heat treatment on the sheathing alloy, the electrical resistivity of as-rolled alloy tapes was also measured. To study the effect of the magnetoresistance, the electrical resistivities of the alloy tapes were measured at 4.2K under a magnetic field up to 12 Tesla. The f_{sc} was decided from observations of the tape cross section using an optical microscope.

III. RESULTS and DISCUSSION

A. Thermal conductivity and electrical resistivity of Ag-Au alloys

Fig. 1 shows the temperature dependence of the thermal conductivity of the heat treated Ag-Au alloy tapes (κ_{alloy}) with various concentrations of Au up to 30at.%. κ_{alloy} drastically decreased with increasing Au content at low temperatures. The thermal conductivity of the tape with 11at.%Au was by about three orders smaller than that of the pure Ag tape at 20K. In the Au concentration range from 11 to 30at.%, the further decrease of the thermal conductivity became gradual. This may correspond to the saturation tendency associated with the Au content getting nearer to 50at.%. Thus, the thermal conductivity of the Ag-Au alloy tape with Au content up to about 11at.% is confirmed to decrease drastically with increase in Au content. The thermal conductivity data shown in Fig.1 are fairly consistent with those by Crisp et al [10].

Fig. 2 shows the temperature dependence of the electrical resistivity of the heat treated Ag-Au alloy tapes. The electrical resistivity drastically increased with an increase in the Au content at low temperatures. For example, the electrical resistivity of the Ag+11at.%Au alloy tape was by about three orders larger than that of the pure Ag tape at 20K. As the electronic thermal conductivity and the electrical resistivity are related to one another by the Wiedemann-Franz law, the Au content dependences in Figs.1 and 2 are reasonable.

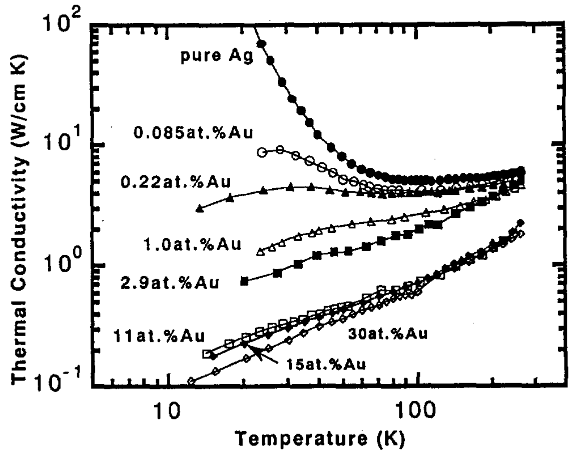

Fig. 1. Temperature dependence of the thermal conductivity of heat treated Ag-Au alloy tapes with the various concentrations of Au.

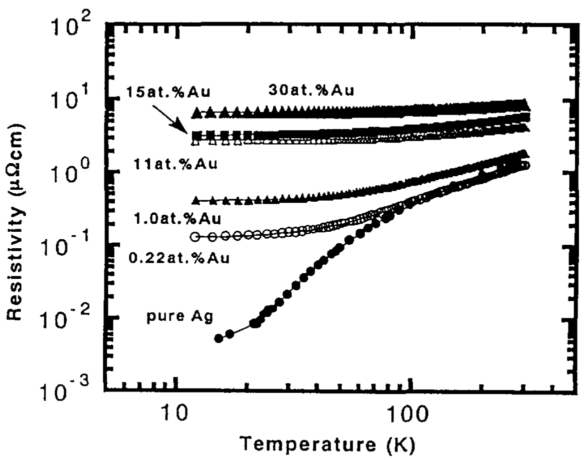

Fig. 2. Temperature dependence of the electrical resistivity of heat treated Ag-Au alloy tapes with the various concentrations of Au.

To investigate the influence of heat treatment in air on the physical properties of the Ag-Au alloys, the electrical resistivity of the as-rolled alloy tapes was also measured. Fig. 3 shows the temperature dependence of the electrical resistivities of both the as-rolled tapes (AR) and the heat treated tapes (HT) with Au content from 0.22 to 11 at.%. It was found that the heat treatment results in a slight decrease of the resistivity values at low temperatures and also a slight increase of the temperature dependence of the resistivity. The decrease of the resistivity at low temperatures may come from the decrease of the dislocation density and the increase of the temperature dependence may come from the softening of the alloy caused by the heat treatment. However, both effects are rather small and, according to the Wiedeman-Franz law, also the thermal conductivity of the alloys should not be much influenced by the heat treatment in air.

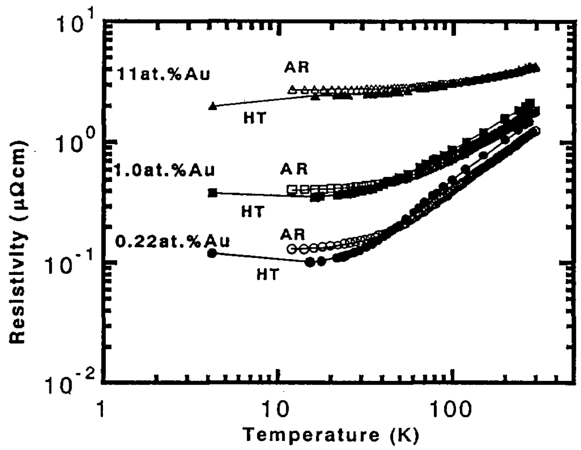

Fig. 3. Temperature dependence of the electrical resistivities of both the as-rolled Ag-Au tapes (AR) and the heat treated tapes (HT) in which Au contains from 0.22 to 11at.%.

B Thermal conductivity of the Bi-2223 superconducting tapes

The thermal conductivities of the Bi-2223 superconducting tapes with various Au contents and superconductor cross-section ratios f_{sc} were investigated. The superconducting transition temperatures of these tapes were about 104K and independent of the Au content and f_{sc}. Figs. 4 and 5 show the temperature dependence of the thermal conductivity of the Bi-2223 superconducting tapes (κ_{AgAuT}) sheathed with Ag+11at.%Au alloy (f_{sc}=0.65) (hereinafter referred to as No.1 sample), and sheathed with Ag+15at.%Au alloy (f_{sc}=0.33) (hereinafter as No.2 sample) with closed circles, respectively. In these figures, the thermal conductivities of the Ag-Au alloy tapes with the same Au content (κ_{alloy} in Fig.1) and the Bi-2223 sintered polycrystal (κ_{poly}) are also shown with open circles and closed triangles, respectively. κ_{poly} was 24 mW/cmK at 200K and decreased monotonically with decreasing temperature. κ_{poly} showed a significant upturn just below T_c(=105K) and took a local maximum around 70K, and then decreased steeply with further lowering temperatures. The value of κ_{poly} is about two orders smaller than that of Ag-Au alloy tape with 11 and 15at.% Au content. It is found that the magnitude of κ_{AgAuT} lies between those of κ_{alloy} and κ_{poly}.

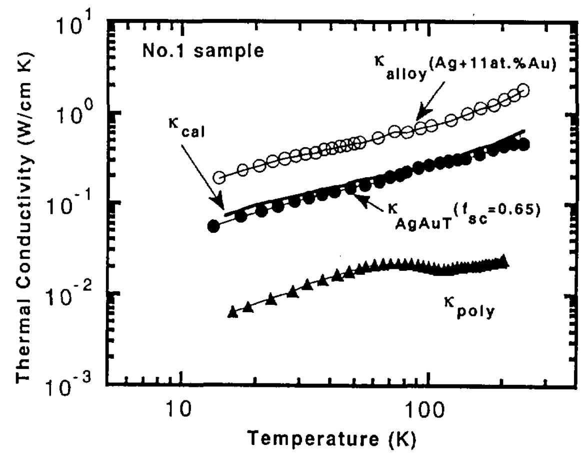

Fig. 4. Temperature dependence of the thermal conductivity κ_{AgAuT} of Bi-2223 superconducting tape (No.1 sample) sheathed with Ag+11at.%Au alloy for ratio f_{sc} 0.65 (closed circles). The thermal conductivity of Ag+11at.%Au alloy tape κ_{alloy}(open circles) and that of Bi-2223 polycrystal κ_{poly} (closed triangles) and the calculated thermal conductivity κ_{cal} (thick line) were also shown.

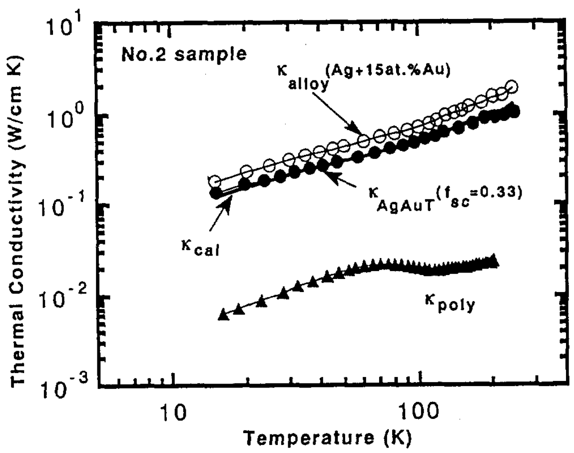

Fig. 5. Temperature dependence of the thermal conductivity of Bi-2223 superconducting tape (No.2 sample) sheathed with Ag+15at.%Au alloy for core ratio f_{sc} 0.33 (closed circles). The thermal conductivity of Ag+15at.%Au alloy tape (open circles), that of Bi-2223 polycrystal (closed triangles) and the calculated thermal conductivity (thick line) are also shown.

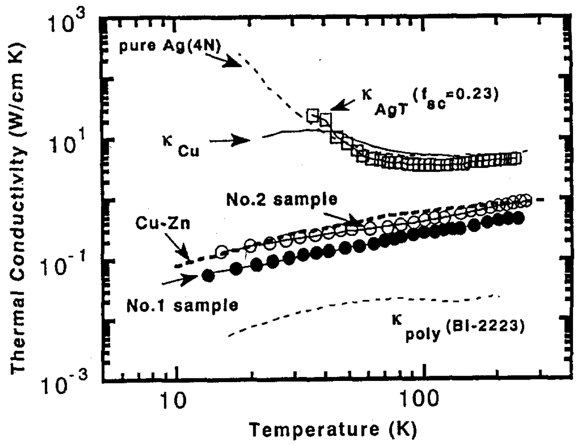

Fig. 6. Temperature dependence of thermal conductivities of both the Bi-2223 superconducting tapes sheathed with pure Ag (f_{sc} = 0.23) and the high purity copper of 4N grade. For comparison, the thermal conductivities of the pure Ag(4N) tape, No.1 and 2 samples, a Cu-Zn alloy and Bi-2223 polycrystal are also shown.

As the superconducting tape consists of the Bi-2223 superconducting core and the Ag-Au alloy sheath, the heat should flow in parallel through both the core and the sheath along the tape. In this case, the thermal conductivity κ_{cal} of the superconducting tape can be calculated using the values of κ_{poly}, κ_{alloy} and f_{sc}, by

$$\kappa_{cal} = f_{sc}\kappa_{poly} + (1-f_{sc})\kappa_{alloy} \quad (1).$$

κ_{cal} was also shown in Figs. 4 and 5 with thick solid lines. The calculated κ_{cal} is very close to the experimentally obtained thermal conductivity κ_{AgAuT}. This confirms that the thermal conductivity of a superconducting tape with arbitrary superconductor cross-section ratio f_{sc} and with some Au concentration in the Ag-Au alloy sheath can be practically estimated by use of Eq.(1). It is useful in power current lead design to be able to estimate the heat flow through the lead rather than to measure it. For comparison, the thermal conductivities of Bi-2223 tapes (f_{sc} =0.23) sheathed with a pure Ag sheath (κ_{AgT}), and the high purity copper (4N) (κ_{Cu}) used in conventional power current leads were also measured. Fig. 6 shows the temperature dependence of κ_{AgT} and κ_{Cu}, together with the thermal conductivities of samples No.1 and 2 , the heat treated pure Ag(4N) tape, a Cu-Zn alloy and κ_{poly}. κ_{AgT} is only slightly smaller than the thermal conductivity of the pure Ag tape because of the low f_{sc}. κ_{Cu} is almost the same as κ_{AgT} from 40 to 200K. The thermal conductivity of No.1 and 2 samples is about 0.2W/cmK at 77K which is comparable to that of the Cu-Zn alloy.

C. Characteristics of overall-J_c

The *overall*-J_c characteristics of the Bi-2223 tape sheathed with Ag-Au alloy for various Au contents were measured [8]. *Overall*-J_c of the tape is defined by

$$overall\text{-}J_c = \frac{I_c}{S} \quad (2),$$

where I_c is the critical current and S is the total cross section of the tape. Fig. 7 shows the relation between the Au content in the Ag-Au alloy sheath and the *overall*-J_c of the tape, in which f_{sc} is kept to 0.65, at 77K and 0T. The *overall*-J_c, remained almost constant at about 1700 A/cm^2, irrespective of Au contents in the Ag-Au alloy sheath up to 11 at.%. As the J_c of Bi-2223 sintered polycrystal was about 500A/cm^2 at 77K and 0T [11], it was found that the *overall*-J_c of the Bi-2223 superconducting tape was about three times larger than that of the Bi-2223 sintered sample. Fig. 8 shows the magnetic field dependence of the *overall*-J_c of the Bi-2223 tape (f_{sc} = 0.65) sheathed with Ag-Au alloy with various Au contents at 77K. The magnetic fields were applied parallel to the tape surface. The *overall*-J_c vs. magnetic field curves of the tapes sheathed with Ag-Au alloys agreed with that of the

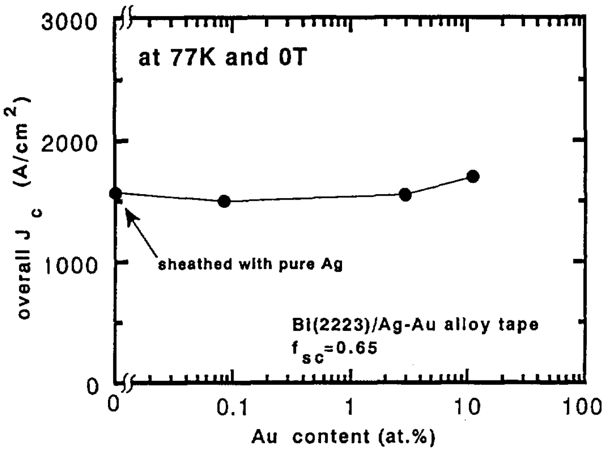

Fig. 7. *Overall-J_c* of the Bi-2223 superconducting tapes with various concentrations of Au at 77K and 0T, in which the superconducting core ratio f_{sc} is 0.65.

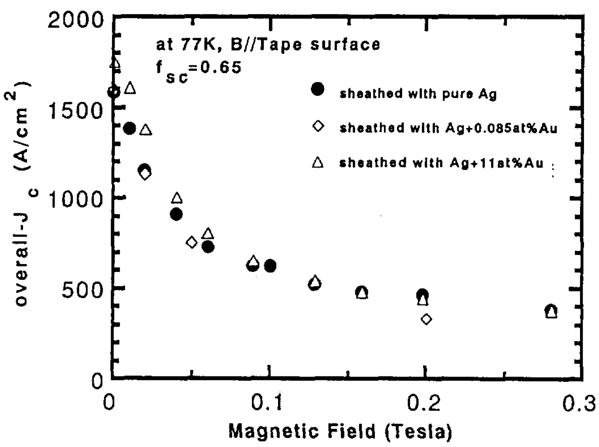

Fig. 8. *Overall-J_c* vs. applied magnetic field curves with various Au contents in the Ag-Au alloy sheathes at 77K. The magnetic field was applied parallel to the tape surface.

pure-Ag sheathed tape. It is recognized that the *overall-J_c* can be somewhat increased by decreasing f_{sc} below 0.65 because of the resultant higher crystal orientation of the superconducting core. However, the heat intrusion through the tape increases at the same time because of the lower f_{sc}. It is necessary to optimize the Au content in the alloy sheath and the f_{sc}, according to the purpose of each cryogenic apparatus.

D. Calculation of heat intrusion through the current lead

As the thermal conductivity of the superconducting tapes can be estimated using Eq.(1) as shown in Figs.4 and 5, the heat intrusion through the current lead using the superconducting tapes was calculated. The heat intrusion Q of the current lead in the area between 77K and 4.2K is given by

$$Q = \frac{S}{L} \int_{4.2}^{77} K_{cal} \, dT \tag{3}$$

where L is the length and S is the cross section of the lead, respectively. We used the Bi-2223 superconducting tapes sheathed with Ag-Au alloy with f_{sc}=0.65 as shown in Fig.7, and calculated the Q value for a 100cm current lead of 1000A using Eqs. from (1) to (3).

Fig. 9 shows the results of the calculation of the relation between the heat intrusion through the lead and the Au content in the Ag-Au alloy sheath. The heat intrusion Q of the current lead decreases with increased Au content. For example, the Q value of a superconducting lead sheathed with the Ag+11at.%Au alloy is about two orders smaller than that of a conventional copper lead without gas cooling or a Bi-2223 superconducting lead sheathed with pure Ag.

E. Influence of magnetic field

In many cases power current leads for cryogenic equipment such as SCMs are used under magnetic fields. The influence of magnetic fields on the electrical resistivity of Ag-Au alloy tapes was investigated. Fig. 10 shows the electrical resistivities of the Ag-Au alloy tapes as a function of magnetic fields up to 12T. The electrical resistivity of tapes with higher Au content depends hardly at all on the magnetic field. This independence is a favorable condition for stable operation of superconducting power current leads with alloy sheathes. Although the intrinsic thermal stability of superconducting tapes sheathed with alloys somewhat deteriorates due to large electrical resistivity in the sheath, the electrical resistivity of bulk oxide superconductors in the normal state is several mΩ • cm, which is three orders larger than that of the Ag-Au alloy tapes. It should still be

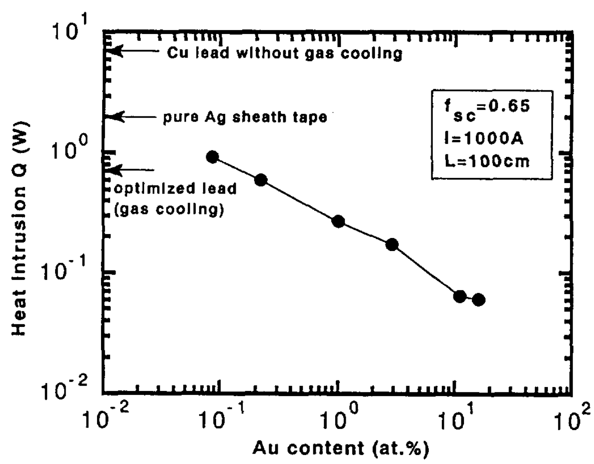

Fig.9. Relation between the heat intrusion through a 100cm superconducting lead of 1000A which is f_{sc}=0.65 and the Au content in the Ag-Au alloy sheath.

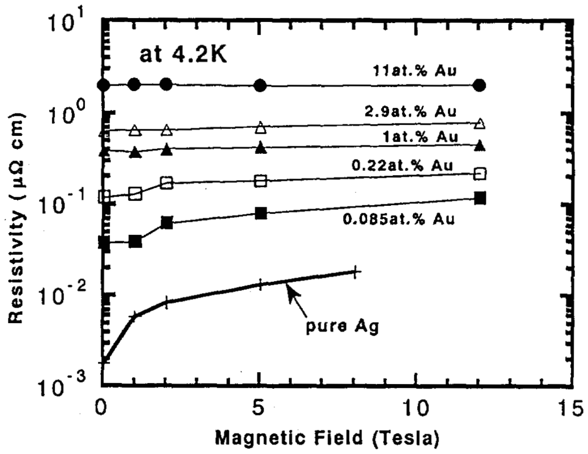

Fig. 10. Magnetic field dependence of the electrical resistivity of Ag-Au alloys with various concentrations of Au at 4.2K.

beneficial for the thermal stability of superconducting power leads to use the Ag-Au alloy sheath. The thermal conductivity of the oxide superconductor is sensitive to the magnetic field and decreases with increasing applied field below T_c [12], and so could cause unstable operation of superconducting power current leads. Because the absolute value of the thermal conductivity of the Bi-2223 superconducting core is about one fiftieth smaller than that of Ag-Au alloy as shown in this investigation, the thermal conductivity of the Bi-2223 superconducting tape is expected to be nearly independent of the applied magnetic field.

IV. SUMMARY

Low thermal conductive Bi-2223 superconducting tapes sheathed with Ag-Au alloys were developed for application to power current leads of cryogenic equipment. The tapes sheathed with the alloy with Au concentration up to 11 at.% showed almost the same superconducting characteristics as those sheathed with pure Ag. *Overall-J_c* of about 1700 A/cm² at 77K and 0T was obtained, independent of the Au content in the Ag-Au alloy sheath. The thermal conductivity of Bi-2223 tape sheathed with Ag+11at.%Au alloy whose superconductor cross-section ratio f_{sc} was 0.65 was about 0.2W/cmK at 77K. This value is almost as low as that of Cu-Zn alloy. The value of measured thermal conductivity can be reproduced by calculation based on the core ratio and the thermal conductivity values of both the Ag-Au alloy and the superconducting core. The Ag-Au alloy sheathed superconducting tapes give full promise for application in power current leads.

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