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AgCu Alloy Sheathed Bi-System Oxide Superconducting Wires

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Abstract: AgCu alloy sheathed Bi(Pb)SrCaCuO oxide wires have been fabricated with respect to improvement in high-J_c reproducibility and mechanical reinforcement. High homogeneity in microscopic composition of oxide core and improved flatness between the core and sheath interface have been focused as important factors for realizing high-J_c reproducibility. Complete elimination of Ag sheath swelling during partial melting for Bi-2212 tapes has also been examined. Remarkable increase in J_c values for Bi-2223 samples prepared using the simultaneous addition of Cu and a small amount of Ti or Au has been obtained, probably due to an increased flux pinning.

1 INTRODUCTION

The Bi(Pb)SrCaCuO superconductors are expected to be used in high-field magnet applications, either as current leads or transmission-cable conductors. Many oxide superconducting coils have generated magnetic fields around 2·2 T at 4·2 K[1–4]–20 K.[5] Most of these coils were pancake-shape coils of Ag-sheathed Bi-2223, Bi-2212 tapes or dip-coated tapes.

In many cases, high critical-current densities (J_c) of 10^4–10^5 A/cm^2 can be obtained at 4·2 K for short samples, but it is very difficult to realize such high J_c values throughout a tape that is longer than several tens of metres. The decrease in or poor reproducibility of high-J_c values has been mainly attributable to inhomogeneous and irregularly oriented microstructures of the core oxide, which are due to the precipitation of other phases, irregular grain growth, and crack of the core oxide in the soft Ag sheath. Alloying the Ag sheath with Cu was found to be effective for reinforcing the tapes with no substantial degradation of J_c.[6] In this paper, improvement in high-J_c reproducibility of Ag- and AgCu-sheathed Bi-2212 tapes has been reported by reducing the microstructural defects and improved wire fabrication process. Complete elimination of Ag sheath swelling during partial melting for Bi-2212 tapes has also been examined. Furthermore, a remarkable increase in J_c for Bi-2223 tapes with a small addition of Ti or Au into the AgCu-sheath has been obtained, probably due to an increased effective flux pinning.

2 EXPERIMENTAL PROCEDURES

The Ag- and AgCu-sheathed tape samples were fabricated by the powder-in-tube technique. Ag–10 at% Cu–x at% M (x = 0–0·5, M = Ti or Au) alloy tubes, 30–50 mm long with 8- and 5·7-mm outer and inner diameters, were used. All the tubes were filled with $Bi_2Sr_2Ca_{0.64}Cu_{1.64}O_z$ calcined powder and then plugged with Ag blocks. The powders were additionally calcined in a vacuum of 50 kPa (0·5 atm) at 660°C for 8 h to avoid sheath swelling. Rods of the BSCCO formed by cold isostatic pressing (CIP) were also used to fill the tubes. These composites were initially swaged to 6 mm in diameter, then drawn to 1·04 mm in diameter, and finally flat-rolled to make tapes about 0·10–0·15 mm thick. The reduction ratios used in drawing or rolling pass were 5–20% at working speeds of 0·5 m/min to 5 m/min. Short samples for

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Bi-2212 were heat-treated by the partial-melting, slow-cooling process. Samples for Bi-2223 were uniaxially pressed with 200 kgf/mm^2 or rolled and then heat treated at 827°C for 50–100 h. This press- or roll-heating process was repeated two or three times.

The powders and tape samples were analysed by a TG-DTA equipped with a mass spectrometer (TD-MS) to obtain thermal information and to determine the optimum sintering condition. Microstructures were examined by an optical and a scanning electron microscope (SEM). The critical current, I_c, at 4·2 K was measured by the four-probe resistive method using the criterion 1 μV/cm. The critical current density, J_c, was calculated for the cross-sectional area of the oxide core.

3 RESULTS AND DISCUSSION

In the present study, factors that reduce the J_c values of tapes were found: (1) swelling of the sheath, (2) rough sheath/core interface of the tape, (3) small crack in the oxide core, (4) precipitation of non-superconducting phases and nonoriented grain growth, (5) irregular thermal transformation of the tape during the partial melting process, and (6) pin-hole of the sheath. All these factors are closely related to the formation of undesirable microstructures or the disturbance of crystalline orientation.

3.1 Elimination of sheath swelling

The swelling of the Ag- or AgCu-sheath is caused by volume expansion of gases that are trapped in the sheath. The gases released out of the oxide powder can be detected by TG-MS analysis as shown in Fig. 1. The CO_2 gas can be introduced during the pulverizing process[7] after calcination or as residue of $SrCO_3$ and $CaCO_3$ due to insufficient calcination treatment. On the other hand, the O_2 gas dissociates from the oxide core during the partial melting process. Similar MS curves were obtained for the Ag- and AgCu-sheathed tape samples, although their partial melting temperatures were 20–30°C less. According to the results shown in Fig. 1, it is suggested that to prevent swelling, one of the best methods is to use powders calcined in a low partial O_2 pressure along with a slow rate of heating so that the O_2 gas is released by diffusion through the covered sheath.

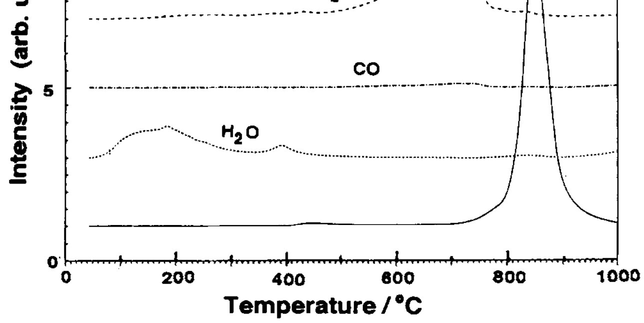

Fig. 1. Mass spectrum curves showing the gas species detected in an exhausted gas from TG-DTA analysis on the calcined $Bi_2Sr_2Ca_{0.64}Cu_{1.64}O_z$ raw powder.

3.2 Tape fabrication with smooth sheath/core interface

Flatness of the interface between the oxide core and the Ag- or AgCu-sheath is very important for the formation of a highly oriented Bi-2212 and Bi-2223 phase microstructure.[8] We found that the interface became rough when the oxide core hardness increased and exceeded a threshold value. The Vickers microhardness number (VHN) of both the oxide core and the Ag- or AgCu-sheath was measured for tape samples fabricated with different initial core densities and working condition and sampled during cold working. The hardness of the oxide core seemed to vary depending on the initial density of the oxide powder fill, the pass reduction ratio, the sheath hardness, and the working speed. The core hardness values of the CIP-core-filled (CIP core) tapes were higher than those of the powder-filled (PWD core) tapes. With respect to the working condition, the oxide core, especially that of the hardest CIP core tape, is likely to become harder as the speed and the pass reduction ratio increase. The core hardness of the hardest CIP core tape, however, decreased rapidly

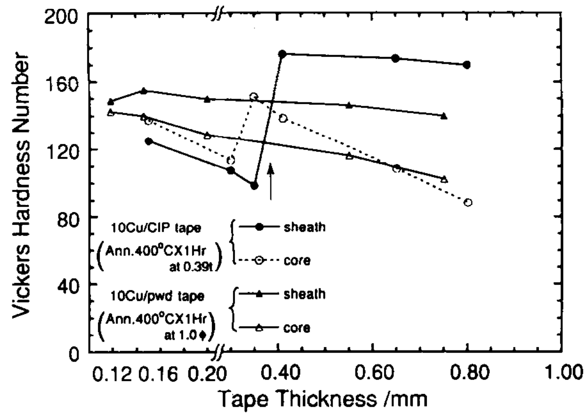

Fig. 2. Vickers microhardness of Ag–10 Cu alloy sheath and oxide core vs tape thickness of the AgCu alloy sheath tape samples fabricated by different oxide core conditions (powder and CIP) with the same working speed of 5 m/min. Intermediate annealing was received by only the 10 Cu/CIP core tape with a thickness of 0·39 mm.

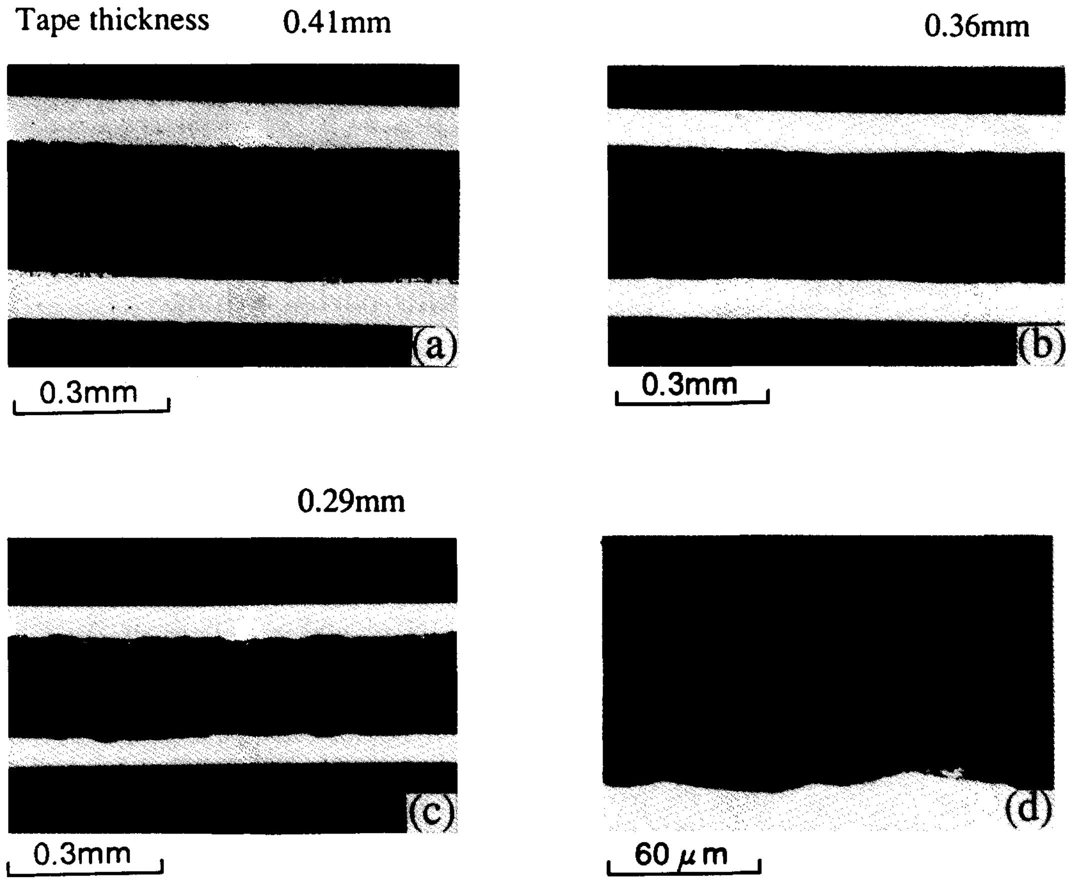

Fig. 3. Optical micrographs showing crack initiation and network formation. The microstructures of the polished longitudinal cross section along the tape, at a thickness of 0·41 mm, before annealing (a); the tape at a thickness of 0·36 mm slightly worked after annealing (b); further worked tape with a thickness of 0·29 mm (c); part (d) is an enlargement of part (c).

for both the Ag/CIP and 10 Cu/CIP tapes. The drop in hardness observed for the 10 Cu/CIP tape is shown in Fig. 2, although, in this case, the tape was annealed at 400°C for 1 h at the 0·39-mm thickness stage. Therefore, it is assumed that the oxide core breaks and the stress stored in the core is released when its hardness exceeds some threshold value, in this case, around 150 VHN, irrespective of sheath hardness.

To investigate why the hardness drops, the microstructures of the tapes were observed by an optical microscope. Micrographs of the cross sections of the 10 Cu/CIP tape samples are shown in Fig. 3: the 0·41-mm thick sample before annealing (a) and, after annealing, the slightly worked 0·36-mm thick sample (b) and 0·29-mm thick sample (c). Cracks that developed at the sheath/core interface propagated into the core, as shown in Fig. 3(b). For tapes that received more roll-working, a network of cracks was formed, and simultaneously, the sheath/core interface became very rough, probably due to the formation of a brick-shaped core, as seen in Fig. 3(c,d). Once a rough surface was created, the surface of the interface could not be smoothed by any further working process. Therefore, to fabricate Ag- or AgCu-sheathed Bi-oxide tapes with a sufficiently flat interface, the core hardness must be kept below 150 VHN.

Critical current versus tape thickness is shown in Fig. 4. The VHN (*Hv* in the figure) values shown in the figure were measured for the thinnest tapes. It is clear that the J_c values of the higher

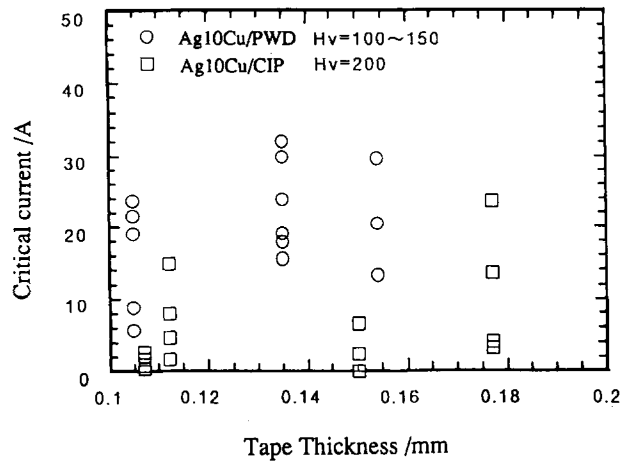

Fig. 4. Critical current vs tape thickness of Ag 10Cu/PWD (powder-filled) tape samples. VHNs (=*Hv*) of the Ag–10Cu sheath were measured for the thinnest tapes. The low-*Hv* tapes were by more frequent annealing than the high-*Hv* tapes.

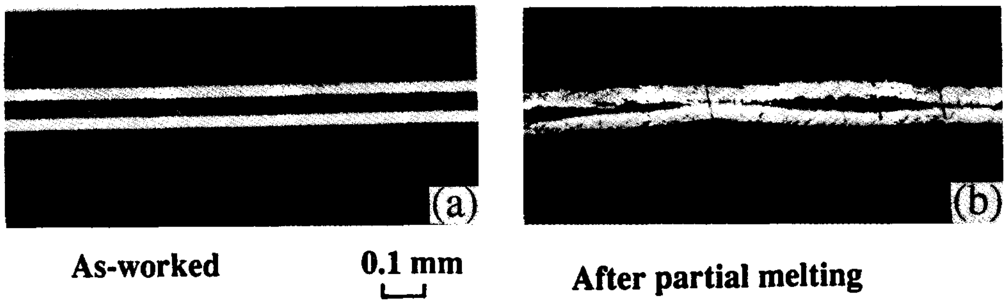

Fig. 5. Optical micrographs showing the longitudinal cross section of the tape samples before (a) and after (b) sintering treatment.

VHN samples are lower than those of the lower VHN samples, and thus the J_c values probably correspond to the interface flatness. There is, however, still a large scatter in the J_c values among tapes of the same thickness, despite the improved tape uniformity observed prior to heat treatment, as seen in the longitudinal cross section shown in Fig. 5(a). The reasons for the large scatter are not clear yet. However, since examination of the microstructure often showed that irregular transformation of the tape occurred during the partial melting process, as shown in Fig. 5(b), the scatter might correspond to difference of the tape cross-sectional area. It is also true that irregular deformation causes severe disturbance of Bi-2212 phase growth and reduces the J_c. This kind of problem is very important from the viewpoint of fabrication of long, very homogeneous tapes with high performance.

3.3 Effect of addition of Ti or Au into AgCu sheath on J_c property

Bi-2223 tapes used AgCu alloy with a small addition of Ti or Au and were prepared by the same procedure as Bi-2212 tapes, the only difference is the final stage repetition of heat treatment and cold-working; namely heat treatment at 827°C for 50–100 h with intermediate cold-rolling or cold-pressing. The highest J_c was obtained by intermediate pressing of 150–200 kgf/mm^2. From a preliminary study, we obtained remarkably high I_c values as shown in Fig. 6, where I_c was shown as a function of additive amount of Ti or Au into the Ag–10 at% Cu sheath. It is clear that even a small amount of addition of Ti or Au into the AgCu sheath gives an increase of the I_c. The increase mechanism of the J_c value has not been determined yet. It is, however, possible to say that Ti or Au atoms, as well as Cu, diffuse from the sheath into the oxide core passing through the sheath/core boundary region and bring about some changes in the microstructure, grain boundary, and/or the sheath/core interface region, resulting in increase of effective pinning force. To know the mechanism of the J_c increase by Ti or Au addition, further studies are being done.

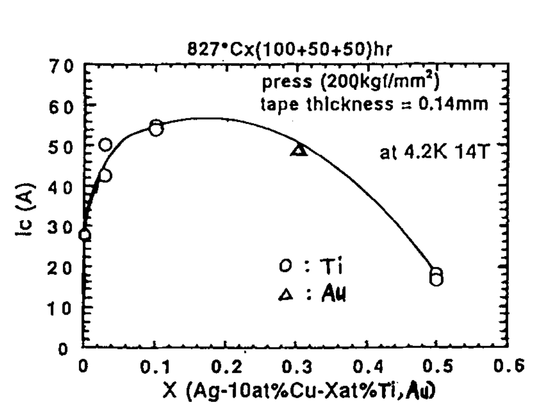

Fig. 6. Critical current vs additional amount of Ti or Au into the Ag–10 at% Cu alloy.

4 CONCLUSIONS

AgCu alloy sheathed Bi(Pb)SrCaCuO oxide tapes have been fabricated with respect to improvement in high J_c reproducibility and mechanical reinforcement. To fabricate tapes with an improved flatness between the core and sheath interface it is essential to keep the core hardness below a threshold value, about 150 in VHN microhardness in the Bi-system. Elimination of Ag sheath swelling during partial melting for Bi-2212 tapes is possible by degassing the calcined powder prior to filling the Ag tube. A remarkable increase in J_c for Bi-2223 samples prepared using the simultaneous addition of Cu and a small amount of Ti or Au has been obtained, probably due to an increased flux pinning.

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