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ADVANCES IN PROCESSING OF Ag-SHEATHED  $(\text{Bi,Pb})_2\text{Sr}_2\text{Ca}_2\text{Cu}_3\text{O}_x$   
SUPERCONDUCTORS\*

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**ADVANCES IN PROCESSING OF Ag-SHEATHED  
(Bi,Pb)<sub>2</sub>Sr<sub>2</sub>Ca<sub>2</sub>Cu<sub>3</sub>O<sub>x</sub> SUPERCONDUCTORS**

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**Abstract**

Advances in the processing and fabrication of Ag-sheathed (Bi,Pb)<sub>2</sub>Sr<sub>2</sub>Ca<sub>2</sub>Cu<sub>3</sub>O<sub>x</sub> (Bi-2223) high-T<sub>c</sub> superconductors by the powder-in-tube technique continue to bring this material closer to commercial applications. Enhancement of the transport critical current density (J<sub>c</sub>) of Ag-sheathed Bi-2223 tapes was achieved by increasing the packing density of the precursor powder, improving mechanical deformation, and adjusting the cooling rate. Long lengths (>150 m) of multifilamentary Bi-2223 tapes have been fabricated and carry critical currents (I<sub>c</sub>) of >50 A (J<sub>c</sub> ≈ 25 kA/cm<sup>2</sup>) at 77 K in self-field. A 1260-m-long tape carried an I<sub>c</sub> of 18 A (J<sub>c</sub> ≈ 12 kA/cm<sup>2</sup>) from end-to-end. Several prototype coils have been assembled from these long-length tapes. Recent progress in the fabrication of Bi-2223 tapes is presented in this paper.

## Introduction

The powder-in-tube (PIT) process continues to be the most promising approach to fabricate long-length superconductors. Critical current density ( $J_c$ ) remains the most important property for practical application of high-temperature (high- $T_c$ ) superconductor tapes. Ag-sheathed Bi-2223 tapes have been incorporated into prototype high- $T_c$  superconductor motors, transmission cables, and fault current limiters; performance has generally been acceptable. However, because high  $J_c$  in magnetic fields is generally necessary, the applicability of such tapes in large electrical equipment has been limited to temperatures  $<30$  K (1). Substantial effort is now focused on addressing this limitation.

Within the past four years, several research groups have reported that, in Ag-sheathed Bi-2223 tapes, the supercurrent is transported through a thin region at the Ag/superconductor interface (2-8). The high-current superconducting layers are generally  $\approx 2-3$   $\mu\text{m}$  thick and have been shown to support a transport current with  $J_c > 10^5$  A/cm<sup>2</sup> at 77 K and zero applied field (4,5). Transport  $J_c$  values of tapes with identical superconductor cross-sectional areas but with differing Ag/Bi-2223 interfacial lengths confirm the importance of the interfacial region (8). The critical current was shown to be proportional to the Ag/Bi-2223 interface perimeter length (IPL); hence,  $J_c$  values can be increased through microstructural design by optimizing the IPL.

Efforts to enhance  $J_c$  by increasing the Ag/Bi-2223 interfacial area continue. Fabrication of multifilamentary tapes achieves this goal, but, in general, the areal fraction of Ag increases in such tapes. An alternative approach is to incorporate Ag wires into a Bi-2223 core. Initial work focused on the use of a single Ag wire [4,9]. In addition to offering the possibility of an improved transport  $J_c$ , significant enhancement of bend-strain tolerance by a wire-in-tube approach has been reported (10). The duplex-core work reported in Refs. 4 and 9 has recently been extended to a two-step process in which many fine Ag wires are coated with Bi-2223 precursor powder and then loaded into a Ag tube. Conventional PIT processing then produces a tape with a very high Ag/Bi-2223 interfacial area (11,12). To date, up to 600 Ag wires coated with Bi-2223 precursor have been loaded into a single Ag tube and processed into tapes. Despite the smaller cross-sectional area of the superconducting core, transport  $J_c$  values are now greater than those of corresponding monofilament tapes (11).

Significant effort is being expended to improve the  $J_c$  of Bi-2223 conductors by tailoring powder stoichiometry, phase assemblage, morphology, mechanical processing, and heat treatment (13,14). Recently, we varied the packing density of the precursor powder, improved the mechanical processing, and modified the heat treatment schedule; the results are described in this paper.

## Experimental Procedure and Results

Multifilament (37-filament) Ag-sheathed Bi-2223 tapes were made by the PIT technique, with precursor powders that exhibit the overall stoichiometry of Bi-2223. Packing density in the Ag tubes was varied by inserting powdered precursor, as well as precursor powder that was prepressed into billets, into the Ag tubes. The precursor powder was packed into the Ag tubes at a density of  $\approx 2.3$  g/cm<sup>3</sup>, whereas the prepressed precursor billets were of two densities:  $\approx 3.5$  g/cm<sup>3</sup> (low packing density) and  $\approx 4.5$  g/cm<sup>3</sup> (high packing density). The powder and prepressed billet Ag tubes were swaged, drawn through a series of dies, and then rolled to a final thickness of  $\approx 200$   $\mu\text{m}$ . Standard mechanical processing that consisted of  $>10\%$  reduction per pass was used to fabricate these tapes. Samples that measured  $\approx 1.2$  m in length

were cut from these three tapes and heat treated in an 8% oxygen atmosphere at 810-825°C. The transport critical currents,  $I_c$  (77 K, self-field, 1  $\mu$ V/cm criterion) that were measured in these tapes are shown in Fig. 1. The tapes with the higher packing density achieved higher  $I_c$  values when they were heat treated at 820°C. These higher  $I_c$  values were maintained uniformly over a length of  $\approx$ 1.2 m. The ratio of Ag to superconductor core cross-sectional area in the tape depends on precursor packing density. Higher initial packing density might lead to an increase in superconductor cross-sectional area and thereby to an increase in  $I_c$ .

In another set of experiments, we varied the mechanical deformation schedule. The Ag tubes packed with the precursor powder and prepressed billets were drawn and rolled according to various reduction ratios per pass. Load cells were mounted on the dies, and the pressure that was exerted on the wires that were being drawn was monitored. Onset of mechanical instability during wire drawing was recorded by monitoring the pressure exerted on the wires. The die pressure measurements formed the basis for optimizing reduction ratios per pass. The cross-sectional area of each sample was observed by scanning electron microscopy (SEM). Figure 2 is a composite of low-magnification SEM images that show the effect of mechanical deformation on the cross sections of two multifilament tapes. Improved mechanical processing of the Ag tube that contained the high-density precursor billet significantly affected the uniformity of the Ag/superconductor interface. The standard mechanical processing included a reduction of  $>10\%$  per pass, whereas the improved mechanical processing reduced the rate of mechanical deformation to  $<10\%$  reduction per pass. Tapes that were processed under improved mechanical deformation conditions exhibited uniform  $I_c$  values of  $\approx 36$  A.

In a parallel experiment, we studied the effect of cooling rate on the  $I_c$  of tapes. Approximately 1-m-long tapes were heated in an 8% oxygen atmosphere to  $\approx 820^\circ\text{C}$  at a rate of  $\approx 2^\circ\text{C}/\text{min}$ , held for 50 h, and then cooled. The standard cooling rate was  $1-2^\circ\text{C}/\text{min}$ , but slower rates were also used. Figure 3 shows the effect of cooling rate on the  $I_c$  values of a

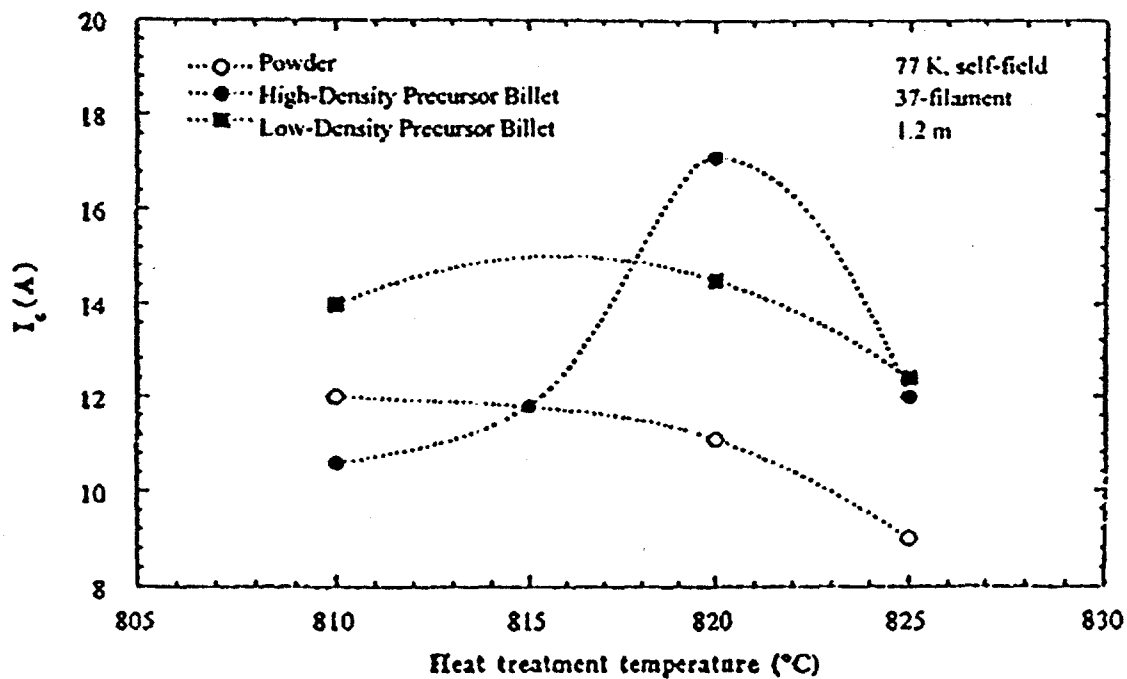
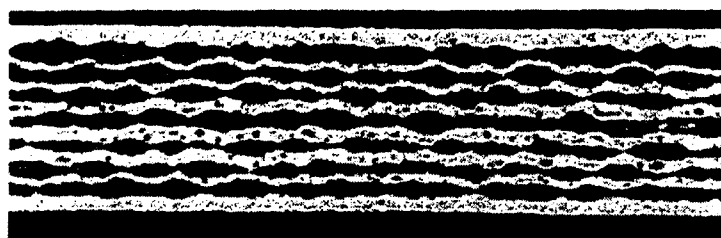
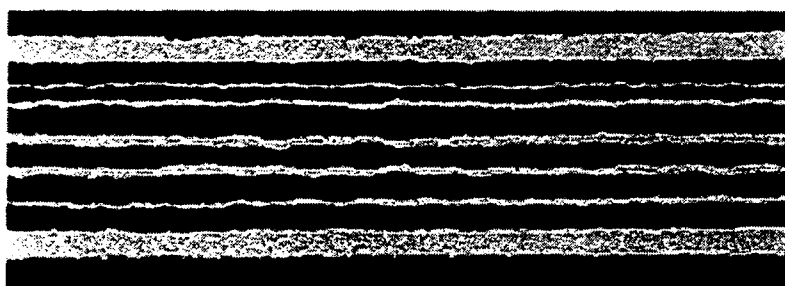


Fig. 1. Transport critical current ( $I_c$ ) as a function of heat treatment temperature for tapes with various precursor powder packing densities.



Consolidated  
Precursor Rod  
Standard  
Mechanical  
Processing  
 $I_c = 18-20$  A



Consolidated  
Precursor Rod  
Improved  
Mechanical  
Processing  
 $I_c = 36$  A

Fig. 2. SEM photomicrograph of multifilament tapes showing effect of improved mechanical processing. Tapes processed by standard mechanical processing are shown for comparison.

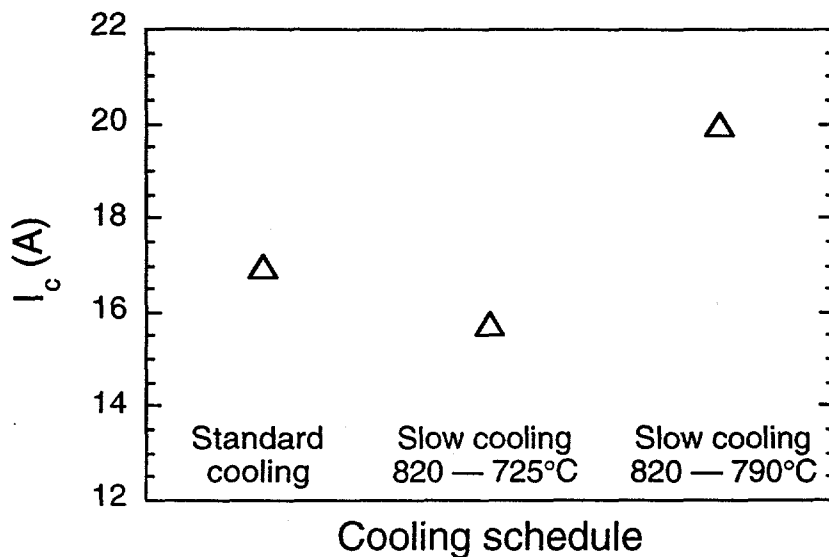


Fig. 3. Transport  $I_c$  of a 1.2-m-long tape vs. cooling conditions.

1.2-m-long tape made from a Ag tube that contained prepressed billets (high packing density). Slow cooling at  $\approx 10^\circ\text{C/h}$  from 820 to 790°C improved  $I_c$  to a level over that achieved with the standard cooling rate. Slow cooling at  $\approx 10^\circ\text{C/h}$  from 820 to 725°C resulted in  $I_c$  values that are similar to those obtained in samples that were cooled at the standard rate. Because of the difference in the thermal conductivity of Ag and the superconducting core, a thermal gradient exists between the core and the Ag in fast-cooled samples. The thermal gradient exerts a stress on the thin layer of Bi-2223 adjacent to the Ag; this stress affects grain alignment and connectivity and thereby influences the  $I_c$ . In extreme cases, the stresses caused by a strong

thermal gradient have induced microcracking in the superconducting layer close to the Ag/Bi-2223 interface (15). Parrel et al. (16) observed that the cooling rate from the sintering temperature had a significant effect on  $J_c$  and attributed the effect to partial decomposition of Bi-2223 during slow cooling. However, other changes in the Bi-2223 also arise from slow cooling, e.g., increase in oxygen content (17). Singh and Vasanthamohan (18) studied the cooling rate effect on Bi-2223 samples sintered at 815 and 825°C in an 8% O<sub>2</sub> atmosphere and noticed that, when the cooling rate was decreased from 100 to 10°C/h, tapes sintered at 815°C showed very little change in  $J_c$ , whereas tapes sintered at 825°C showed an increased  $J_c$ . This difference was attributed to how the Bi-2223 phase is formed at the two temperatures. For the tapes sintered at 825°C, the authors of Ref. 18 noticed an increase in Bi-2223 phase with a decrease in cooling rate; hence,  $J_c$  increases with a decrease in cooling rate. Incorporation of the improvements in packing density of powders, mechanical deformation, and cooling rate has now resulted in  $I_c$  values of  $\approx 55$  A for long lengths ( $>100$  m) of superconductor tapes. We have also fabricated an  $\approx 1260$ -m-long multifilamentary tape that carried an  $I_c$  of 18 A ( $J_c \approx 12$  kA/cm<sup>2</sup>) from end to end at 77 K and self-field.

Poor mechanical properties have seriously hampered the commercial application of high- $T_c$  superconductors. During fabrication and service, the conductors are subjected to axial and bending stresses. During operation, the material is subjected to additional stresses by temperature gradients and magnetic fields. In the presence of large and/or high-field magnets, electromagnetic hoop stresses could even reach the ultimate strength of the material. These stresses can cause microstructural damage in the conductors and thereby degrade current transport properties. Although Ag is widely used as a sheath material, its mechanical properties are not adequate to withstand the stresses developed during fabrication and service. Therefore, techniques, such as adding Ag to the superconductor powder, using alloy sheath material as an alternative to Ag, and fabricating multifilament conductors, have been developed to improve the strain tolerance characteristics of the superconductors.

### Conclusions

Powder-in-tube-fabricated Ag-sheathed Bi-2223 tapes exhibit good transport  $J_c$  values, but transport at 77 K, especially in large applied magnetic fields, must be improved. Good transport of supercurrent seems to be confined to core regions adjacent to the Ag sheath. The packing density of precursor powder, improved mechanical deformation, and cooling rate all exerted a pronounced effect on the critical current of the superconducting tapes.

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### References

1. A. N. Iyer, R. Jammy, U. Balachandran, M. Suenaga, and P. Haldar, *J. Electron. Mater.*, **24** 1873 (1995).
2. S. P. Ashworth and B. A. Glowacki, *Physica C*, **226** 159 (1994).
3. Y. Feng and D. C. Larbalestier, *Interface Sci.*, **1** 401 (1994).

4. M. Lelovic, P. Krishnaraj, N. G. Eror, and U. Balachandran, *Supercond. Sci. Technol.*, **8** 334 (1995).
5. M. Lelovic, P. Krishnaraj, N. G. Eror, A. N. Iyer, and U. Balachandran, *Supercond. Sci. Technol.*, **9** 201 (1996).
6. D. C. Larbalestier, X. Y. Cai, Y. Feng, H. Edelman, A. Umezawa, G. N. Riley, Jr., and W. L. Carter, *Physica C*, **221** 299 (1994).
7. U. Welp, D. O. Gunter, G. W. Crabtree, W. Zhong, U. Balachandran, P. Haldar, R. S. Sokolowski, V. K. Vlasko-Blasov, and V. I. Nikitenko, *Nature*, **367** 44 (1995).
8. B. C. Prorok, M. Lelovic, T. A. Deis, P. Krishnaraj, N. G. Eror, A. N. Iyer, and U. Balachandran, *Adv. Cryo. Eng.*, **42** 739 (1997).
9. J. Schwartz, H. Sekine, T. Asano, T. Kuroda, K. Inoue, and H. Maeda, *IEEE Trans. Magn.*, **27** 1247 (1991).
10. N. Vasanthamohan and J. P. Singh, *Supercond. Sci. Technol.*, **10** 113 (1997).
11. S. E. Dorris, N. Ashcom, T. Truchan, N. Vasanthamohan, D. A. Burlone, and L. D. Woolf, to be published in *Proc. Supercond. Symp.*, 99th Ann. Mtg. Am. Ceram. Soc., Cincinnati, May 4-7, 1997.
12. S. E. Dorris, in *Practical Superconductor Development for Electrical Power Systems*, Argonne National Laboratory Report ANL-95/42, 13 (1995).
13. U. Balachandran, A. N. Iyer, R. Jammy, P. Haldar, J. G. Hoehn, Jr., and M. Suenga, *Adv. Cryo. Eng.*, **42** 753 (1997).
14. Z. Han and T. Freltoft, *Appl. Supercond.*, **2** 201 (1994).
15. M. Lelovic, T. Deis, N. G. Eror, U. Balachandran, and P. Haldar, *Supercond. Sci. Technol.*, **9** 965 (1996).
16. J. A. Parrel, D. C. Larbalestier, and S. E. Dorris, *IEEE Trans. Appl. Supercond.*, **5** 1275 (1995).
17. M. Tetenbaum and V. A. Maroni, *Physica C*, **260** 71 (1996).
18. J. P. Singh and N. Vasanthamohan, *J. Mater. Res.* (1998, in press).