

CONF-960850-18

ANL/ET/CP--89105 RECEIVED

STRENGTH AND FLEXIBILITY OF BULK
HIGH- T_c SUPERCONDUCTORS*

NOV 12 1996

OSTI

K. C. Goretta, Ming Jiang, D. S. Kupperman, M. T. Lanagan, J. P. Singh,
N. Vasanthamohan, D. G. Hinks, J. F. Mitchell, and J. W. Richardson Jr.

Argonne National Laboratory, Argonne, IL 60439 USA

August 1996

The submitted manuscript has been authored by a contractor of the U.S. Government under contract No. W-31-109-ENG-38. Accordingly, the U.S. Government retains a nonexclusive, royalty-free license to publish or reproduce the published form of this contribution, or allow others to do so, for U.S. Government purposes.

DISTRIBUTION OF THIS DOCUMENT IS UNLIMITED

MASTER

Paper submitted to IEEE Trans Appl. Supercond.

*Work supported by the U.S. Department of Energy (DOE), Energy Efficiency and Renewable Energy, as part of a DOE program to develop electric power technology, and Basic Energy Sciences (Materials Sciences), under Contract W-31-109-Eng-38.

DISCLAIMER

This report was prepared as an account of work sponsored by an agency of the United States Government. Neither the United States Government nor any agency thereof, nor any of their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by trade name, trademark, manufacturer, or otherwise does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government or any agency thereof. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government or any agency thereof.

DISCLAIMER

Portions of this document may be illegible in electronic image products. Images are produced from the best available original document.

Strength and Flexibility of Bulk High- T_c Superconductors

K. C. Goretta, Ming Jiang, D. S. Kupperman, M. T. Lanagan, J. P. Singh, and N. Vasanthamohan
Energy Technology Division, Argonne National Laboratory, Argonne, IL 60439, U.S.A.

D. G. Hinks, J. F. Mitchell, and J. W. Richardson Jr.
Materials Science Division, Argonne National Laboratory, Argonne, IL 60439, U.S.A.

Abstract—Strength, fracture toughness, and elastic modulus data have been gathered for bulk high-temperature superconductors, commercial 99.9% Ag, and a 1.2 at.% Mg/Ag alloy. These data have been used to calculate fracture strains for bulk conductors. The calculations indicate that the superconducting cores of clad tapes should begin to fracture at strains below 0.2%. In addition, residual strains in Ag-clad $(\text{Bi,Pb})_2\text{Sr}_2\text{Ca}_2\text{Cu}_3\text{O}_x$ tapes have been measured by neutron diffraction. An explanation is offered for why many tapes appear to be able to tolerate large strains before exhibiting a reduction in current transport.

I. INTRODUCTION

Although engineering critical current density remains the primary consideration for high-temperature superconductor wires, strength and flexibility are also important [1,2]. Large bodies of mechanical-property data have been published for bulk $\text{YBa}_2\text{Cu}_3\text{O}_x$ (Y-123), $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_x$ (Bi-2212) and $(\text{Bi,Pb})_2\text{Sr}_2\text{Ca}_2\text{Cu}_3\text{O}_x$ (Bi-2223), but not Tl-based superconductors. Relevant data for each system will be presented and discussed. Literature values will be used for Y-123, Bi-2212, Bi-2223, $\text{Tl}_2\text{Ba}_2\text{Ca}_2\text{Cu}_3\text{O}_x$ (Tl-2223), Ag, and a 1.2 at.% Mg/Ag alloy. New data will be presented for $\text{Tl}_2\text{Ba}_2\text{CaCu}_2\text{O}_x$ (Tl-2212), and residual-stress data obtained by neutron diffraction will be presented for a Bi-2223/Ag composite tape [2,3].

The goal of this work is to reconcile bulk strength and stiffness data with strain-tolerance reports for Ag-clad tapes [4-8]. It has generally been found that apparent strain tolerances are much higher than would be expected for conductors containing hard brittle cores.

II. EXPERIMENTAL DETAILS

Data for fracture strength, toughness, and elastic modulus were gathered for several superconductors and two Ag alloys.

A. Materials

Two types of Y-123 specimens were used. Strength data were obtained on extruded wires ≈ 0.85 mm in diameter

Manuscript received August 26, 1996.

This work was supported by the U.S. Department of Energy (DOE), Office of Energy Efficiency and Renewable Energy, as part of a DOE program to develop electric power technology, and Basic Energy Sciences (Materials Sciences), under Contract W-31-109-Eng-38.

[9,10]. Other data were obtained from sintered pellets [10,11]. All Y-123 samples were virtually phase pure, with nearly randomly oriented grains less than ≈ 10 μm in size.

All of the mechanical-property data for Bi-2212 and Bi-2223 were obtained from hot-forged bars. Such materials are highly phase pure, exhibit very strong c-axis textures, and are $> 95\%$ dense [12-17]. Data for Bi-2223 composites containing MgO and Ag will also be discussed [18]. Monofilament Ag-clad Bi-2223 tapes [2] for residual strain measurements were fabricated by American Superconductor Corporation (Westborough, MA).

The Tl-2212 and Tl-2223 compounds were chosen for fabrication and characterization because of their stability relative to other Tl-based superconductors [19,20]. Although the $\text{TlBa}_2\text{Ca}_2\text{Cu}_3\text{O}_x$ phase is probably of more interest for bulk applications, we were unable to produce dense specimens of good phase purity [21]. The Tl-2212 was synthesized in highly phase pure form from hydroxide precursors [22,23]. Details of the Tl-2223 have been published [20]. Dense specimens were prepared by wrapping powder compacts in Ag foil and forging at $\approx 835^\circ\text{C}$ in air. Final stresses were 30-40 MPa. The resultant bars were $\approx 90\%$ dense.

The 99.9% Ag alloy was purchased from Johnson-Matthey (Ward Hill, MA) and the 1.2 at.% Mg alloy (referred to henceforth as AgMg) from Vacuumschmelze (Hanau, Germany). Chemical analysis and microstructural details have been published [24,25].

B. Characterization

Microstructures were examined by X-ray diffraction and scanning electron microscopy (SEM). Both fractured and polished surfaces were examined.

Strength (σ) and fracture toughness (K_{IC}) values were measured in three- or four-point bending [26]. Details have been published [10,12-15]. Elastic modulus was measured ultrasonically [16,27,28].

New data were obtained for yielding of the two Ag alloys. Specimens were tested in compression in an Instron universal tester Model 4505 (Canton, MA). Alloys were tested in the soft-annealed state, after having been work hardened $\approx 70\%$, and for the AgMg, after having been heat treated in O_2 at 900°C to induce precipitation hardening through formation of MgO [25].

For the residual strains, a bundle was packed from tape sections 100 μm thick and 20 mm long. The bundle filled a volume ≈ 10 mm in diameter and 20 mm in length. Strains were measured in a direction perpendicular to the long axis of each tape. The diffraction peaks for a variety of crystallographic direction were very sharp. Bi-2223 powder and 99.9% Ag strip were used as references. The error in the average strain measurements was about $\pm 0.01\%$ [3].

III. RESULTS AND DISCUSSION

The microstructures of the Y-123 specimens were nearly randomly oriented. Because they were hot forged, the other specimens exhibited c-axis textures. Textures for the Tl-2212 and Tl-2223 specimens were moderate, those for the Bi-2212 and Bi-2223 specimens were strong. Individual grains for the Y-123, Tl-2212, and Tl-2223 specimens had aspect ratios generally < 3 ; the Bi-2212 and Bi-2223 specimens had grain aspect ratios > 10 (Fig. 1).

The microstructures of the Bi-based specimens are similar to those obtained in powder-in-tube tapes or thick films [1]. They are conducive to good current transport. It would be best if all of the specimens tested had similar microstructures. Bi-2212 and Bi-2223 grains are by nature more platelike than other high-temperature superconductor grains, however, and only they could be aligned effectively by hot forging.

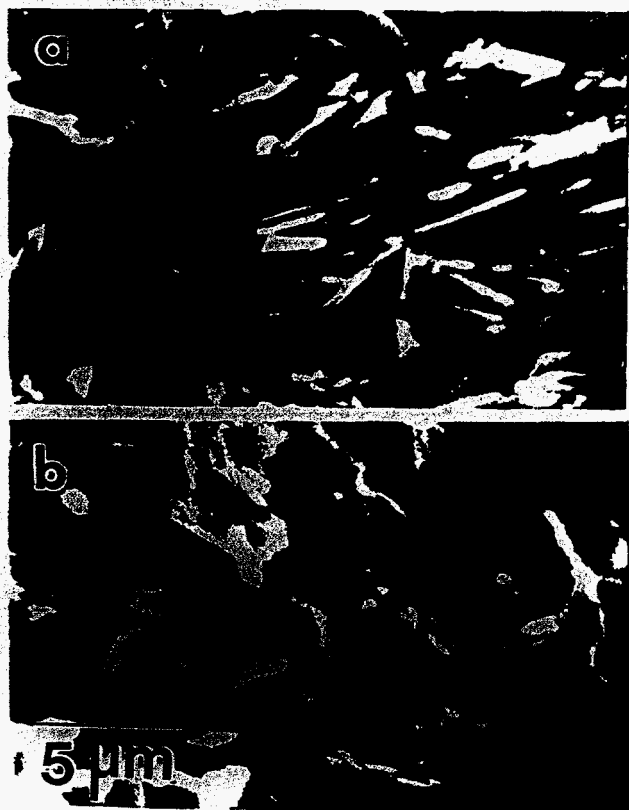


Fig. 1. SEM photomicrographs of fracture surfaces of (a) Tl-2212 test specimen and (b) Bi-2212 test specimen. The grains of the Bi-2212 are much more elongated and strongly textured.

Basic mechanical-property data for these specimens are shown in Table I. The individual grains of all high-temperature superconductors have highly anisotropic properties [16,29-32], and thus comparisons of the data shown in Table I must be made with caution. We have not found any strengths reported that are more than $\approx 15\%$ higher than those listed in Table I. It is noted that the microstructures shown in Fig. 1 are rather good for strength. Large-grained melt-textured materials exhibit significantly lower strengths [33]. Furthermore, additions of Ag particles or ceramic particles or whiskers provide at best only moderate improvements to strength of stiffness [14,18]. By Hooke's law for elastic materials, one can see that none of the superconductors is likely to exhibit a strain to failure $> 0.2\%$.

The Ag data are of interest because in Ag/superconductor composites the alloy sheath will carry some any applied stress and the sheath may impart a residual stress to the superconductor core because of differences in the coefficients of thermal expansion.

The residual stresses for Ag/Bi-2212 or Bi-2223 tapes are particularly complicated because of the configuration and crystal anisotropy. The thermal expansion of Ag [34] is $19-23 \times 10^{-6}$. The thermal expansion of Bi-2223 varies from 14 to 24×10^{-6} , with the larger expansion being in the c-axis direction [29]. It is difficult to predict, even the sign, of the residual stresses in the Ag or superconductor core. Neutron diffraction measurements at the Intense Pulsed Neutron Source at Argonne National Laboratory yielded the average strains in the Ag sheath and Bi-2223 core in various crystallographic directions. It was found that the Ag had an average compressive strain of 0.062% and the Bi-2223 had an average tensile strain of 0.034% .

If one approximates the residual stress by Hooke's law, the residual stresses in both components are $40-50$ MPa, which is a large fraction of the fracture stress for monolithic superconductors. The stress state is favorable, as the superconductor core is in compression. A further residual-stress advantage might be realized if stronger and stiffer alloy sheaths were to be used. Larger favorable stresses might be generated [24].

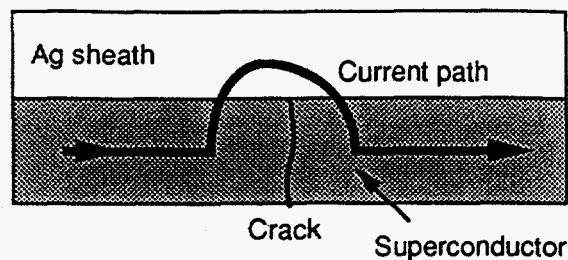


Fig. 2. Schematic diagram of current transport path in an Ag/Bi-2223 tape.

2

TABLE I
SUMMARY OF MECHANICAL-PROPERTY DATA AND APPROXIMATE DENSITIES

Material	Density (g/cm ³)	E (GPa)	σ (MPa)	K _{IC} (MPa√m)
Ag - annealed	10.6	76	65	—
Ag - cold worked	10.6	76	270	—
AgMg - annealed	10.5	74	48	—
AgMg - cold worked	10.5	74	220	—
AgMg - oxidized	10.5	85	385	—
Y-123	6.3	148*	230	1.5-2.0
Bi-2212	6.5	118, 44**	145	2.9
Bi-2223	6.4	82*	145	2.9
Tl-2212	6.4	—	120	2.6
Tl-2223	6.0	101*	—	1.2

*Average value.

**Values for a-b plane and c-direction for highly textured specimens.

All of the data indicate that even within a metallic sheath, a superconductor core should not be able to withstand a strain of more than about 0.2% before cracking. Yet strains to 0.5-1.0% without apparent degradation of superconducting properties are often cited for Ag-clad Bi-2223 conductors. This discrepancy has been convincingly explained recently. Current can flow around cracks by shunting through the metallic sheath, as shown schematically in Fig. 2. So long as there are sufficiently few cracks and they are sufficiently sharp, the critical current density measured with a 1 μ V/cm criterion will not be affected. Details of this analysis can be found in Refs. 35 and 36.

IV CONCLUSION

Mechanical-property data for monolithic high-temperature superconductors and residual-strain data for Ag/Bi-2223 tapes indicate that superconductor cores should crack at strains no greater than \approx 0.2%. The apparent larger strain tolerances of clad conductors is due to current shunting around the crack and passing through the metallic sheath.

ACKNOWLEDGMENT

We thank Y. Xin for supplying Tl-2223 powder and G. N. Riley Jr. for supplying Ag-clad Bi-2223 tapes.

REFERENCES

- [1] E. E. Hellstrom, "Important considerations for processing Bi-based high-temperature superconducting tapes and films for bulk applications," *MRS Bull.*, vol. 17, pp. 45-51, August 1992.
- [2] J. S. Luo, N. Merchant, E. J. Escorcía-Aparicio, V. A. Maroni, B. S. Tani, W. L. Carter, and G. N. Riley Jr., *J. Mater. Res.*, vol. 9, pp. 3059-3067, 1994.
- [3] U. Balachandran, *Practical Superconductor Development for Electrical Power Applications*, Report ANL-94/39, Argonne National Laboratory, 1994.
- [4] T. A. Miller, J. E. Ostenson, Q. Li, L. A. Schwartzkopf, D. K. Finnemore, J. Righi, R. A. Gleixner, and D. Zeigler, "Strain tolerant microfilamentary conductors of Bi₂Sr₂Ca₁Cu₂O_{8- δ} ," *Appl. Phys. Lett.*, vol. 58, pp. 2159-2161, 1991.
- [5] J. W. Ekin, D. K. Finnemore, Q. Li, J. Tenbrink, and W. Carter, "Effect of axial strain on the critical current of Ag-sheathed Bi-based superconductors in magnetic fields up to 25 T," *Appl. Phys. Lett.*, vol. 61, pp. 858-860, 1992.
- [6] J. P. Singh, J. Joo, N. Vasanthamohan, and R. B. Poeppel, "Role of Ag additions in the microstructural development, strain tolerance, and critical current density of Ag-sheathed BSCCO superconducting tapes," *J. Mater. Res.*, vol. 8, pp. 2458-2464, 1993.
- [7] S. Ochiai, K. Hayashi, and K. Osamura, "Improvement of strain endurance of critical current of silver-sheathed superconducting tapes by reducing volume fraction of Bi(Pb)-Sr-Ca-Cu-O oxide core," *Cryogenics*, vol. 33, pp. 976-979, 1993.
- [8] S. Yuhya, M. Hiraoka, T. Jukunaga, and A. Oota, "Mechanical and AC loss properties in mono- and multi-filamentary Bi-2223 Ag-sheathed tapes," *J. Electron. Mater.*, vol. 24, pp. 1839-1842, 1995.
- [9] S. E. Dorris, J. T. Dusek, M. T. Lanagan, J. J. Picciolo, J. P. Singh, J. E. Creech, and R. B. Poeppel, "Extrusion of multilayer superconductor coils," *Ceram. Bull.*, vol. 70, pp. 722-726, 1991.
- [10] K. C. Goretta, N. Chen, M. T. Lanagan, W. Wu, J. P. Singh, R. A. Olson, J. L. Routbort, and R. B. Poeppel, "Sintering and annealing of bulk high-T_c superconductors, Y-Ba-Cu-O to effective properties," *Industrial Heating*, pp. 40-43, February 1993.
- [11] N. Chen, D. Shi, and K. C. Goretta, "Influence of oxygen concentration on processing YBa₂Cu₃O_x," *J. Appl. Phys.*, vol. 66, pp. 2485-2488, 1989.
- [12] C.-Y. Chu, J. L. Routbort, N. Chen, A. C. Biondo, D. S. Kupperman, and K. C. Goretta, "Mechanical properties and texture of dense, polycrystalline Bi₂Sr₂CaCu₂O_x," *Supercond. Sci. Technol.*, vol. 5, pp. 306-312, 1992.

- [13] N. Chen, A. C. Biondo, S. E. Dorris, K. C. Goretta, M. T. Lanagan, C. A. Youngdahl, and R. B. Poeppel, "Sinter-forged $\text{Bi,Pb}_2\text{Sr}_2\text{Ca}_2\text{Cu}_3\text{O}_x$ superconductors," *Supercond. Sci. Technol.*, vol. 6, pp. 674-677, 1993.
- [14] L. J. Martin, K. C. Goretta, J. Joo, J. P. Singh, S. R. Olson, S. Wasylenko, R. B. Poeppel, and N. Chen, "Mechanical properties of BiSrCaCuO/Ag superconductors," *Mater. Lett.*, vol. 17, pp. 232-236, 1993.
- [15] K. C. Goretta, M. E. Loomans, L. J. Martin, J. Joo, R. B. Poeppel, and N. Chen, "Fracture of dense, textured $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_x$ " *Supercond. Sci. Technol.*, vol. 6, pp. 282-286, 1993.
- [16] Chang Faggao, P. J. Ford, G. A. Saunders, Li Jiaqing, D. P. Almond, B. Chapman, M. Cankurtaran, R. B. Poeppel, and K. C. Goretta, "Anisotropic elastic and non-linear acoustic properties of very dense textured $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_x$ " *Supercond. Sci. Technol.*, vol. 6, pp. 484-489, 1993.
- [17] N. Murayama, Y. Kodama, S. Sakaguchi, and F. Wakai, "Mechanical strength of hot-pressed Bi-Pb-Sr-Ca-Cu-O superconductor," *J. Mater. Res.*, vol. 7, pp. 34-37, 1992.
- [18] Y. S. Yuan, M. S. Wong, and S. S. Wang, "Effect of Ag-particulate addition on processing, microstructure, and properties of MgO-whisker-reinforced bulk BPSSCO high-Tc superconductor composites," *J. Mater. Res.*, vol. 11, pp. 1373-1382, 1996.
- [19] T. L. Aselage, E. L. Venturini, S. B. Van Deusen, T. J. Headley, M. O. Eatough, and J. A. Voigt, "Phase stability and properties of near-equilibrium Tl-Ca-Ba-Cu-O superconductors," *Physica C*, vol. 203, pp. 25-36, 1992.
- [20] Y. Xin, B. R. Xu, S. Nasrazadani, W. S. He, D. F. Lu, G. F. Sun, K. W. Wong, and D. Knapp, "A systematic study of the synthesis of useful $\text{Tl}_2\text{Ba}_2\text{Ca}_2\text{Cu}_3\text{O}_{10.8}$ bulk superconductor," *J. Mater. Res.*, vol. 9, pp. 1672-1677, 1994.
- [21] J. L. Routbort, D. J. Miller, E. J. Zamirovski, and K. C. Goretta, "Plasticity of $\text{TlBa}_2\text{Ca}_2\text{Cu}_3\text{O}_x$," *Supercond. Sci. Technol.*, vol. 6, pp. 337-342, 1993.
- [22] R. Mogilevsky, J. F. Mitchell, D. G. Hinks, D. N. Argyriou, and J. D. Jorgensen, "Ternary and quaternary thallium-containing high-Tc superconductors synthesized at low temperature and elevated pressure," *Physica C*, vol. 250, pp. 15-24, 1995.
- [23] J. F. Mitchell, J. A. Garcia, and D. G. Hinks, "Rapid synthesis of 93 K Bi-2212 superconductor from hydroxide precursors," *Physica C*, vol. 245, pp. 126-130, 1995.
- [24] K. C. Goretta, J. L. Routbort, J. P. Carroll, R. L. Thayer, J. Wolfenstine, J. Kessler, and J. Schwartz, "Deformation of Ag/1.2 at.% Mg," *Physica C*, vol. 265, pp. 201-206, 1996.
- [25] K. C. Goretta, W. E. Delaney, J. L. Routbort, J. Wolfenstine, W. Zhang, and E. E. Hellstrom, "Creep of Silver at 900°C," *Supercond. Sci. Technol.*, vol. 9, pp. 422-426, 1996.
- [26] W. F. Brown Jr. and J. E. Srawley, *Plane Strain Crack Toughness Testing of High Strength Metallic Materials*, Philadelphia, PA: Am. Soc. Testing Mater., 1965.
- [27] J. Dominec, "Ultrasonic and related experiments in high-Tc superconductors," *Supercond. Sci. Technol.*, vol. 6, pp. 153-172, 1993.
- [28] Q. Wang, G. A. Saunders, M. Cankurtaran, D. P. Almond, and K. C. Goretta, "Elastic properties as functions of temperature and hydrostatic pressure of sintered $\text{YBa}_2\text{Cu}_3\text{O}_{7-x}$ ceramics with different oxygen concentrations," *Phys. Rev. B*, vol. 52, pp. 3711-3726, 1995.
- [29] R. H. Arendt, M. F. Garbaskas, C. A. Meyer, F. J. Rotella, J. D. Jorgensen, and R. L. Hitterman, "Thermal expansion measurements using neutron diffraction of $(\text{Bi,Pb})_2\text{Ca}_2\text{Sr}_2\text{Cu}_3\text{O}_x$," *Physica C*, vol. 194, pp. 397-402, 1992.
- [30] A. Goyal, P. D. Funkenbusch, D. M. Kroeger, and S. J. Burns, "Anisotropic hardness and fracture toughness of highly aligned $\text{YBa}_2\text{Cu}_3\text{O}_{7.8}$," *J. Appl. Phys.*, vol. 71, pp. 2363-2367, 1992.
- [31] M. Mouallem-Bahout, J. Gaudé, G. Calvarin, J.-R. Gavarrin, and C. Carel, "Anisotropic thermal expansion and Grüneisen coefficients of the bismuth cuprate 2-2-1-2," *Mater. Lett.*, vol. 18, pp. 181-185, 1994.
- [32] K. C. Goretta, J. A. Cluff, J. Joo, M. T. Lanagan, J. P. Singh, N. Vasanthamohan, Y. Xin, and K. W. Wong, "Mechanical properties of high-temperature superconducting wires," in *Proceedings of 4th World Congress on Superconductivity, NASA Conference Publication 3290*, K. Krishen and C. Burnham, Eds. Washington: NASA, 1995, pp. 633-638.
- [33] D. Lee and K. Salama, "Enhancements of current density and mechanical properties of Y-Ba-Cu-O/Ag composites," *Jpn. J. Appl. Phys.*, vol. 29, pp. L2017-L2019, 1990.
- [34] T. Hashimoto, K. Fueki, A. Kishi, T. Azumi, and H. Koinuma, "Thermal expansion coefficients of high-Tc superconductors," *Jpn. J. Appl. Phys.*, vol. 27, pp. L214-L216, 1988.
- [35] Y. Fang, S. Danyluk, Y. S. Cha, and M. T. Lanagan, "Modeling voltage distribution and current limit in $\text{Ag/Bi}_2\text{Sr}_2\text{Ca}_{n-1}\text{Cu}_n\text{O}_{2n+4}$," *J. Appl. Phys.*, vol. 79, pp. 947-952, 1996.
- [36] Y. S. Cha, M. T. Lanagan, K. E. Gray, V. Z. Jankus, and Y. Fang, "Analysis and interpretation of critical current experiments for bismuth-based high-temperature superconductors made by powder-in-tube processing," *Appl. Supercond.*, vol. 2, pp. 47-59, 1994.

4