Doubled critical current density in Bi-2212 round wires by reduction of the residual bubble density

2011 Supercond. Sci. Technol. 24 082001

(http://iopscience.iop.org/0953-2048/24/8/082001)

Supercond. Sci. Technol. **24** (2011) 082001 (5pp) doi:10.1088/0953-2048/24/8/082001

RAPID COMMUNICATION

Doubled critical current density in Bi-2212 round wires by reduction of the residual bubble density

J Jiang, W L Starch, M Hannion, F Kametani, U P Trociewitz, E E Hellstrom and D C Larbalestier

Applied Superconductivity Center, National High Magnetic Field Laboratory, Florida State University, Tallahassee, FL 32310, USA

E-mail: jjiang@asc.magnet.fsu.edu

Received 10 May 2011, in final form 3 June 2011
Published 30 June 2011
Online at stacks.iop.org/SUST/24/082001

Abstract

We have recently shown that the gas present in the only ∼70% dense filaments of as-drawn Bi-2212 wire agglomerates into large bubbles that fill the entire filament diameter during the melt phase of the heat treatment. Once formed, these bubbles never disappear, although they can be bridged by 2212 grains formed on cooling. In order to test the effect of these bubbles on the critical current I_c, we increased the density of the filaments after drawing using 2 GPa of cold isostatic pressure, finding that the bubble density and size were greatly reduced and that I_c could be at least doubled. We conclude that enhancement of the filament packing density is of great importance for making major I_c improvements in this very useful, round superconducting wire.

1. Introduction

$Bi_2Sr_2CaCu_2O_x$ (Bi-2212) is the only cuprate superconductor that can be made into a round-wire conductor with a critical current density (J_c) over 10^5 A cm^{-2} at 4.2 K, and an irreversibility field (H_{irr}(4.2 K)) greater than 100 T [1–3]. This makes Bi-2212 round wire unique and very competitive for high magnetic field (>20 T) applications, especially for nuclear magnetic resonance (NMR) magnets and for accelerators [4–9]. The crossover field with its nearest round-wire competitor, Nb_3Sn, lies in the 18–20 T range. Improving the J_c of Bi-2212 conductors by a factor of 2–3 could thus make a huge impact on high-field magnet applications.

Bi-2212 round wire is fabricated as a multifilamentary conductor by the powder-in-tube (PIT) method, but it must be heat treated at final size by partial melting to develop a high J_c. The starting powder is a nearly single-phase Bi-2212, and in the as-drawn state the filament density is much less than 100%, typically about 70% of the theoretical density of Bi-2212 [10]. Understanding how to further improve J_c in Bi-2212 by process optimization is still quite limited because the relationships between processing, microstructure, and superconducting properties are still poorly understood [11–22]. Under the DOE-HEP sponsored very high-field superconducting magnet collaboration (VHFSMC), we have been investigating about a dozen Bi-2212 round wires with different PIT wire architectures [20–22]. In order to better understand the complex heat treatment process, we quenched samples of many wires just after they entered the melt phase and observed many large gas bubbles, most as big as the filament diameter. Although originally observed using metallographic polishing of transverse and longitudinal cross sections, the gas bubble formation was later confirmed by *in situ* and *ex situ* synchrotron x-ray tomography [22]. The internal gas pressure was also shown to drive molten liquid towards the ends of 1 m long wires and a strong correlation between the local J_c and the local mass density was found [21]. These experiments have convinced us that these melt-state-induced bubbles are a major current limiting mechanism, since they do not disappear in the fully processed wires [22]. The essential problem is that a less than 100% powder packing density is essential to allow the filaments to uniformly deform within their surrounding Ag matrix. In the as-drawn wire,

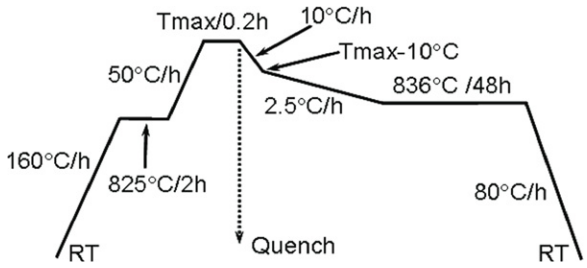

Figure 1. Schematic heat treatment schedule used. The samples were quenched after holding for 12 min at T_{max}.

the residual ∼30% porosity is distributed uniformly and on a scale much smaller than the filament diameter, but on melting the porosity agglomerates into filament-sized, gas-filled bubbles that make the local density of the 2212 highly variable. Although the overall effect of melt processing on J_c is highly positive, it appears that further optimization of J_c must reduce the great density inhomogeneity produced by the bubble formation.

In this study, we employed cold isostatic pressing (CIP) to densify the final size Bi-2212 round wires before melt processing. We found that we could more than double the critical current, I_c, driving the critical current density to values higher than have been reported elsewhere in any Bi-2212 round wire [2, 17, 20].

2. Experimental details

The Bi-2212 round wire was a double-stacked wire made from a 37 filament first stack, 18 such bundles being stacked to yield a 37 × 18 architecture. The wire diameter was 0.8 mm and the filaments were about 15 μm in diameter. The wires were fabricated by Oxford Superconducting Technology (OST) for the VHFSMC using Nexans granulate powder with a composition of $Bi_{2.17}Sr_{1.94}Ca_{0.90}Cu_{1.98}O_x$. The Bi-2212 powder was always surrounded by pure silver but the outer sheath of the 37 × 18 stack was made with a Ag–Mg(0.2 wt%) alloy tube. 8 cm long sections of wire were cut for heat treatment. In order to remove their internal gas, the sample ends were ground with SiC paper to expose the filaments and the samples were then heated at 400 °C for 48 h under a vacuum of 2.7 Pa. After cooling, the furnace was filled with 0.1 MPa oxygen and held for 16 h. When the wires were removed from the furnace, their ends were immediately sealed by dipping in molten silver or Sn. The sealed samples were then CIPped under a pressure of 2 GPa.

Both as-received and CIPped samples were heat treated together using the standard heat treatment schedule shown in figure 1 in 0.1 MPa flowing O_2 [17, 18]. Samples were also quenched into brine after being heated at 887 °C for 12 min to freeze the high-temperature microstructure. Transverse cross sections of quenched and fully processed wires were dry polished using a series of SiC papers with decreasing grit sizes with final polishing conducted in a suspension of 50 nm alumina mixed with ethanol using an automatic vibratory polisher (Buehler Vibromet). The longitudinal cross sections of quenched samples were also prepared with a polish on a 0.5 μm diamond lapping film. They were then deeply etched in a 1:1:2 (volume) mixture of NH_4OH, H_2O_2 and methanol until Bi-2212 filaments were exposed. Microstructures were examined with a Zeiss 1540EsB scanning electron microscope (SEM).

Critical currents of fully processed wires were measured using the four-probe transport method with a 1 $\mu V\,cm^{-1}$ criterion at 4.2 K in a magnetic field of 5 T applied perpendicular to the wire axis. The overall wire critical current density J_E was calculated using the whole wire cross-section, while J_c of the oxide filament area was calculated using the filament area of the as-received and CIPped wires. The quantitative microscopy was done by digital image analysis of the filament cross sections of un-reacted wires with a scanning laser confocal microscope (LEXT).

Magnetic hysteresis was measured for 5 mm long samples using an Oxford Instrument 14 T vibrating sample magnetometer (VSM) at 4.2 and 20 K in fields up to 14 T applied perpendicular to the wire axis. The irreversibility field H_{irr} was approximated by a linear extrapolation of the Kramer function $\Delta M^{1/2}H^{1/4}$ to zero, defining the Kramer irreversibly field H_K. The superconducting transition temperature T_c of these samples was determined using a Quantum Design SQUID magnetometer to measure the magnetic moment as a function of temperature after zero-field cooling the samples to 5 K, then applying a 2 mT field perpendicular to the wire axis, and raising the temperature while measuring the magnetic moment.

3. Results

Figure 2 shows SEM images of transverse cross sections of samples quenched from the melt state after holding at 887 °C for 12 min. The 146 gas bubbles in this quenched, as-received wire cross-section (figure 2(a)) tend to fill the entire filament diameter. In marked contrast, there are only 46 bubbles in the CIPped sample and they are on average much smaller in diameter.

The longitudinal cross sections of inner bundles of the quenched samples are shown in figure 3. The gas bubbles in the as-received wire are big, being 2–3 times as long as their diameter, while the bubbles in the CIPped wire are round and smaller than the filament diameter.

Table 1 compares the values of I_c (4.2 K, 5 T), the resistive transition index n and H_K for the as-received and CIPped wires, which were heat treated together. The I_c (4.2 K, 5 T) values for the CIPped samples were more than doubled, and their n values are also much higher than for the re-received wires, consistent with an increase in the longitudinal uniformity of the I_c [23]. Image analysis showed that the total cross-sectional area of the wire decreased from 0.503 mm^2 for the as-received wire to 0.478 mm^2 for the CIPped wire before reaction. The wire diameters calculated from the cross-sectional area are 0.80 mm and 0.78 mm for as-received and CIPped wires, respectively. The measured filament cross-sectional area decreased from 0.132 mm^2 for the as-received wire to 0.107 mm^2 for the CIPped wire before reaction, a

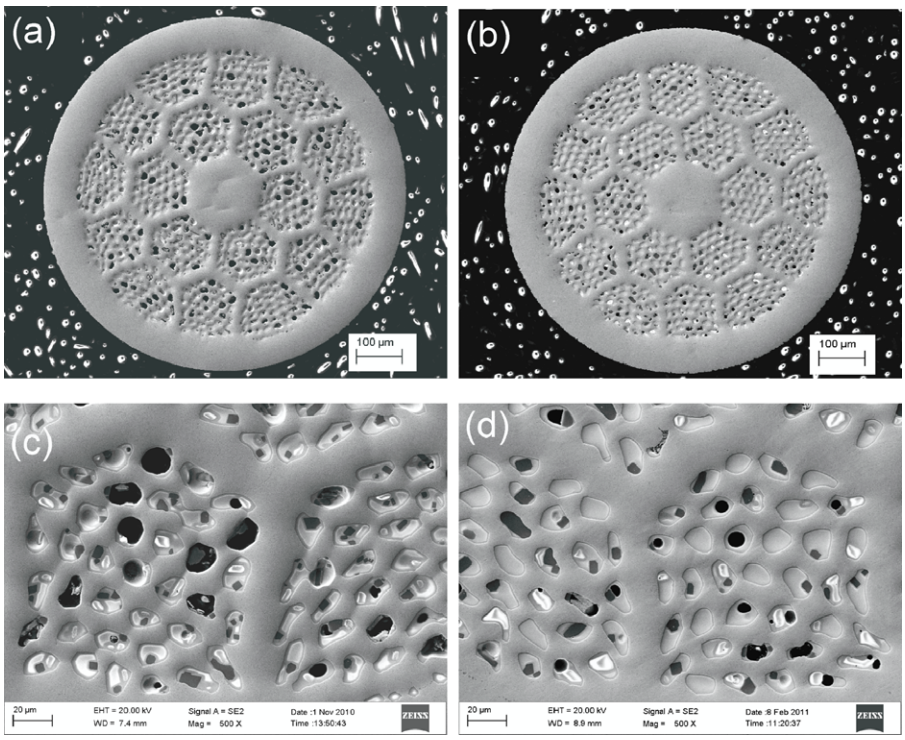

Figure 2. SEM images of transverse cross sections of samples quenched from the melt. (a) and (c) as-received wire; (b) and (d) CIPped wire. The round black spots are the bubbles, the angular dark grey particles are alkaline earth cuprate $(Sr, Ca)_{14}Cu_{24}O_x$, AEC, the light grey regions in the filaments are liquid, and the white particles are the Cu-free phase ($Bi_9(Sr, Ca)_{16}O_x$, CF). The liquid and Ag matrix are almost the same shade of grey. The bubbles in the as-received wire are generally as large as the filaments whereas they are generally smaller than the filaments in the CIPped wire.

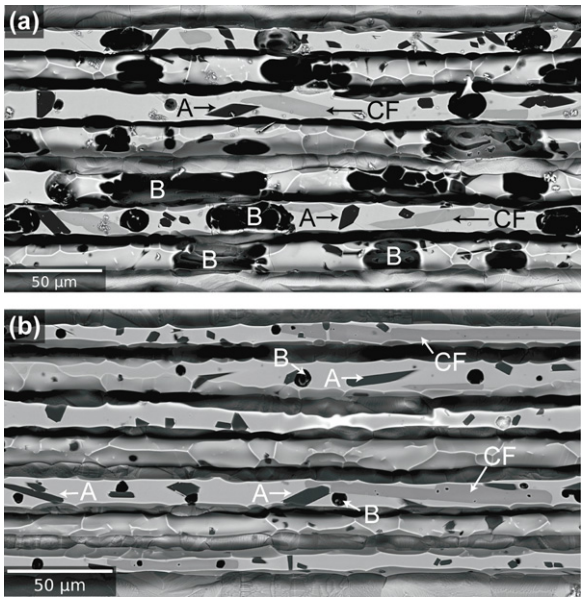

Figure 3. SEM images of longitudinal cross sections of filaments from an inner bundle (see figure 2) of quenched wires. (a) As-received and (b) CIPped wire. All filaments show a continuous amorphous solid structure containing bubbles (B), AEC (A) and CF.

decrease of 19%. This means that CIPping decreased the filament volume by 19%, which increased the powder packing density. Due to the increased I_c and decreased areas in the CIPped wire, J_c (4.2 K, 5 T) was increased from 1667 to 3600 A mm^{-2} for the CIPped wire and J_E (4.2 K, 5 T) increased from 438 to 809 A mm^{-2} for the CIPped wire. Figure 4 shows SEM images of fully processed as-received and CIPped samples. The CIPped sample has obviously denser filaments, but there are still some lower density regions where bubbles had formed in the melt state.

4. Discussion

An overall wire current density J_E (4.2 K, 5 T) of 740 A mm^{-2} was reported in 2005 by Oxford Superconducting Technology (OST) in a 0.80 mm Bi-2212 round wire with 85 × 7 filaments and a filling factor of 27.6% [2], corresponding to a J_c (4.2 K, 5 T) of 2700 A mm^{-2}. Since then, there has been no significant J_c improvement for Bi-2212 round wire. These 2005 wires also experienced serious leaks of Bi-2212 through the outer sheath when used in coil length wires [24]. Since 2007, OST has modified their fabrication process and nearly eliminated the leakage, but the post-2007 wires have always shown lower J_c by more than 30% [20]. The wire used for this study is a 2010 wire, which is rather typical of the post-2007 wires, making the improvement of J_c by CIPping shown in table 1 very valuable. Compared to the previous report [20], CIPping has allowed both better J_c and J_E in the new wires. The advantage of CIPping is that a long length of Bi-2212 round wire or a device made with Bi-2212 round wire can be easily densified before heat treatment if a large pressure chamber is available. We also

Table 1. I_c (4.2 K, 5 T), J_E (4.2 K, 5 T) J_c (4.2 K, 5 T) n value, and Kramer field (H_K) for fully processed as-received (AR) and CIPped samples of 37 × 18 wire.

T_{max} (°C)	AR or CIP	I_c (4.2 K, 5 T) (A)	J_E (4.2 K, 5 T) (A mm^{-2})	J_c (4.2 K, 5 T) (A mm^{-2})	n value	H_K (20 K) (T)
884	AR	155.0	325	1174	9	8.1
884	CIP	385.4	807	3602	21	8.4
885	AR	175.8	350	1332	13	8.2
885	CIP	386.4	809	3611	20	8.5
888[a]	AR	220.0	438	1667	11	8.0

[a] 888 °C gave the maximum I_c for AR wire.

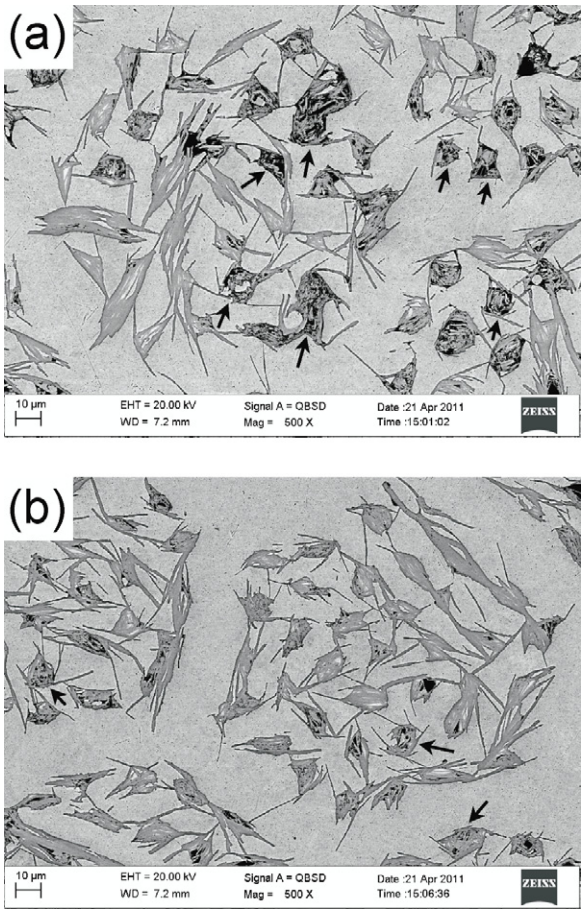

Figure 4. Backscattered electron SEM images of fully processed samples. (a) As-received T_{max} = 885 °C and (b) CIPped T_{max} = 885 °C. The arrows point to low-density regions due to the bubbles present in the melt state. These images were chosen for comparison because these two samples were heat treated together and polished together (see table 1).

tested CIPping for a low filling factor wire with 27 × 7 widely separated filaments. The I_c (4.2 K, 5 T) for this 27 × 7 wire was also doubled from about 50 A in the as-received state to 102 A for the CIPped wire.

We used 2 GPa in this study because that is the highest pressure system we have in our laboratory. 2 GPa is much larger than the yield strength of the wire outer sheath, Ag-0.2 wt% Mg, and we believe it is above the fracture strength of the Bi-2212. Further study is needed to identify the critical pressure for CIPping. A benefit of CIPping is that the high density of the CIPped filaments is preserved during the heat treatment, making denser Bi-2212 filaments in the fully processed wire. The differences in the wire I_c values cannot be ascribed to changes in the vortex pinning because the H_K values are all similar (table 1). The enhanced n values indicate that the enhanced I_c and J_c result from a more uniform conductor [23] and the improved connectivity produced by CIP densification. Even though CIPping at 2 GPa can make the un-reacted Bi-2212 filaments almost fully dense, gas is trapped inside the filaments and we tried to maximize the oxygen in the filaments before densification because oxygen can diffuse through the Ag sheath at elevated temperature, whereas nitrogen and CO_2 cannot and could thus form bubbles and de-densify the filaments. N_2 and CO_2 may be the cause of the small gas bubbles seen in figures 2 and 4(b).

5. Conclusion

We found a significant improvement of J_c in recent Bi-2212 round wires by CIPping them at 2 GPa before melt processing. CIPping densified the Bi-2212 core, resulting in much smaller and fewer bubbles in the melt and doubled I_c (4.2 K, 5 T). J_E (4.2 K, 5 T) rose to over 800 A mm^{-2} and J_c (4.2 K, 5 T) rose to 3600 A mm^{-2}. Densifying the un-reacted wire through approaches like CIPping will be a very effective pathway to achieve the very high J_c and J_E needed for fabricating high current density wind and react Bi-2212 devices.

Acknowledgments

We are very grateful for discussions with Y Huang, H Miao, S Hong, and J A Parrell at OST, with T Shen, A Malagoli, M Dalban-Canassy, D Myers, and P J Lee at FSU, and with members of the Very High Field Superconducting Magnet Collaboration. Technical support from V Griffin, N Craig, and L Sura is also gratefully acknowledged. The work at the NHMFL was supported by an ARRA grant of the US Department of Energy Office of High Energy Physics and by the National High Magnetic Field Laboratory which is supported by the National Science Foundation under NSF/DMR-0084173 and by the State of Florida.

References

[1] Marken K R, Miao H, Meinesz M, Czabaj B and Hong S 2003 *IEEE Trans. Appl. Supercond.* **13** 3335

[2] Miao H, Marken K R, Meinesz M, Czabaj B and Hong S 2005 *IEEE Trans. Appl. Supercond.* **15** 2554
[3] Chen B, Halperin W P, Guptasarma P, Hinks D G, Mitrovic V F, Reyes A P and Kuhns P L 2007 *Nature Phys.* **3** 239
[4] Hasegawa T *et al* 2001 *IEEE Trans. Appl. Supercond.* **11** 3034–7
[5] Weijers H W, Trociewitz U P, Marken K, Meinesz M, Miao H and Schwartz J 2004 *Supercond. Sci. Technol.* **17** 636–44
[6] Markiewicz W D, Miller J R, Schwartz J, Trociewitz U P and Weijers H W 2006 *IEEE Trans. Appl. Supercond.* **16** 1523
[7] Nachtrab W T, Renaud C V Jr, Wong T, Liu X T, Shen T M, Trociewitz U P and Schwartz J 2008 *IEEE Trans. Appl. Supercond.* **18** 1184–7
[8] Godeke A *et al* 2008 *IEEE Trans. Appl. Supercond.* **18** 516–9
[9] Godeke A *et al* 2009 *IEEE Trans. Appl. Supercond.* **19** 2228–31
[10] Karuna M, Parrell J A and Larbalestier D 1995 *IEEE Trans. Appl. Supercond.* **5** 1279
[11] Kumakura H, Kitaguchi H, Togano K and Sugiyama N 1996 *J. Appl. Phys.* **80** 5162
[12] Zhang W and Hellstrom E E 1995 *Supercond. Sci. Technol.* **8** 430
[13] Motowidlo L R, Galinski G, Ozeryansky G, Zhang W, Hellstrom E E, Sumption M and Collings T 1995 *IEEE Trans. Appl. Supercond.* **5** 1162
[14] Matsumoto A, Kitaguchi H, Kumakura H, Nishioka J and Hasegawa T 2004 *Supercond. Sci. Technol.* **17** 989–92
[15] Marken K R, Miao H P, Sowa J M, Parrell J A and Hong S 2001 *IEEE Trans. Appl. Supercond.* **11** 3252–5
[16] Holesinger T G, Kennison J A, Marken K R, Miao H, Meinesz M and Campbell S 2005 *IEEE Trans. Appl. Supercond.* **15** 2562
[17] Shen T, Jiang J, Yamamoto A, Trociewitz U P, Schwartz J, Hellstrom E E and Larbalestier D C 2009 *Appl. Phys. Lett.* **95** 152516
[18] Shen T, Jiang J, Kametani F, Trociewitz U P, Larbalestier D, Schwartz J and Hellstrom E E 2010 *Supercond. Sci. Technol.* **23** 025009
[19] Holesinger T G 2006 Characterization of Oxford Bi2212 conductors *Low Temperature Superconductor Workshop (Tallahassee, FL)*
[20] Jiang J, Myers D, Shen T, Kametani F, Trociewitz U P, Hellstrom E E and Larbalestier D C 2010 Comparative studies of nine new Bi-2212 round wires 2MX-03 *Applied Superconductivity Conf. (Washington, DC, MD)*
[21] Malagoli A, Kametani F, Jiang J, Trociewitz U P, Hellstrom E E and Larbalestier D C 2011 *Supercond. Sci. Technol.* **24** 075016
[22] Kametani F, Shen T, Jiang J, Scheuerlein C, Di Michiel M, Huang Y, Miao H, Parrell J A, Hellstrom E E and Larbalestier D C 2011 *Supercond. Sci. Technol.* **24** 075009
[23] Warnes W H and Larbalestier D C 1986 *Cryogenics* **26** 643–53
[24] Marken K R, Miao H P, Meinesz M, Czabaj B and Hong S 2006 *IEEE Trans. Appl. Supercond.* **16** 992–5