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Significantly enhanced critical current density in Ag-sheathed $(Bi,Pb)_2Sr_2Ca_2Cu_3O_x$ composite conductors prepared by overpressure processing in final heat treatment

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Overpressure (OP) processing otherwise fully treated, commercial Ag-sheathed multifilament $(Bi,Pb)_2Sr_2Ca_2Cu_3O_x$ (2223) composite conductors increased the critical current density J_c (0.1 T, 77 K) by 37% to 30.8 kA/cm^2 and the self-field J_c (SF, 77 K) to 69.6 kA/cm^2. These improvements were obtained on full-size high current conductors such that critical current I_c (0.1 T, 77 K) reached 80.6 A and I_c (SF, 77 K) 181.7 A, even though there was a very strong self-field suppression of I_c. Estimated values for the non-self-field-limited I_c and J_c (0 T, 77 K) reached 235 A and 90 kA/cm^2. Scanning electron microscopy and superconducting quantum inference device measurement revealed that OP processing effectively suppressed cracks, porosity, and the volume fraction of the $Bi_2Sr_2Ca_1Cu_2O_y$ (2212) phase, which are all major current-limiting mechanisms in present 2223 conductors. © *2004 American Institute of Physics*. [DOI: 10.1063/1.1682675]

Even though Ag-sheathed 2223 and 2212 tapes are currently the only first-generation high temperature superconductors available in lengths suitable for large-scale electrical applications, major current-limiting mechanisms limit their performance. Typical critical current density J_c (self-field—SF, 77 K) for the more widely studied 2223 tapes ranges from 30 to 40 kA/cm^2 and J_c (0.1 T, 77 K) from 14 to 20 kA/cm^2 for the best commercial 2223 tape produced by the oxide-powder-in-tube (OPIT) method.[1–3] However, current reconstructions of magneto-optical images show local regions of 2223 tape with J_c as high as 300 kA/cm^2,[4] while J_c up to 1000 kA/cm^2 has been achieved in 2223 thin films,[5,6] suggesting there is still much headroom to improve J_c (SF, 77 K) in 2223 tape.

The currently practiced OPIT fabrication of Ag-sheathed 2223 composites is a complicated process,[7,8] in which a mixture of 2223-precursor powders with nominal composition of (Bi,Pb):Sr:Ca:Cu=2:2:2:3 is packed and sealed in a silver tube, then drawn and rebundled to a multifilamentary composite, and rolled into tape. This as-rolled tape subsequently goes through a thermomechanical cycle that consists of a first heat treatment (HT1), an intermediate rolling (IR), and a final heat treatment (FHT). The FHT includes a second heat treatment (HT2) and a separate low temperature "post anneal" (PA) or a heat treatment combining HT2 and PA.[9,10] About 80% of the precursor oxide powder reacts to 2223 phase in HT1. IR helps densify the superconducting filaments while HT2 and any FHT further increase 2223 formation. However, the IR creates an extensive network of cracks that cannot be fully healed in the subsequent heat treatments.[11,12] A significant amount of porosity, ~20% as indicated by mass density measurement, remains in multifilament 2223 tape after HT2 or FHT at 1 bar.[13,14] These residual cracks and porosity interrupt the grain connectivity and current paths, thus reducing J_c. It is generally accepted that 2223 forms from 2212 and alkaline earth cuprate (AEC) phases in the presence of a transient liquid phase. Residual 2212, present as half-cell intergrowths in 2223 grains and as grains in 2223 colonies, was detected by multiple techniques in 2223 after HT2 or FHT.[2,15,16] Increases in J_c were found to correlate strongly with decreases in the volume fraction of 2212.[2,10,15] A central question in further improving the performance of 2223 tapes is how to eliminate the current limiting mechanisms of porosity, cracks, and residual 2212 to further improve the J_c.

Previously, overpressure (OP) processing was used to heat treat as-rolled 2223 tape, as well as 1 bar processed HT1 and IR tapes.[17] We showed that OP processing reduced the crack density and porosity by 11%–16% and increased J_c (SF, 77 K) by 75% to 59 kA/cm^2 and J_c (0.1 T, 77 K) by 82% to 22 kA/cm^2 compared to samples processed identically at 1 bar. The mass density of OP processed samples reached 97% of 2223 theoretical density. This letter shows that OP processing also works very well when applied only as the final heat treatment (FHT) after all other processing (HT1, IR, and HT2) is done at 1 bar.

Samples up to 8 cm long of a fully processed (FHT), high I_c (159.6 A) multifilament production tape produced by American Superconductor Corp. subsequently received an additional heat treatment at 1 bar or 148 bar overpressure processing (FHT+1b or FHT+OP). The external cross-section dimensions of the tape were ~4.0 mm×0.21 mm. These FHT+1b and FHT+OP samples were annealed using a heat treatment cycle similar to that published elsewhere.[17] The total OP pressure of the Ar/O_2 mixture was 148 bar. The calculated oxygen partial pressure (pO_2) was 0.081 bar

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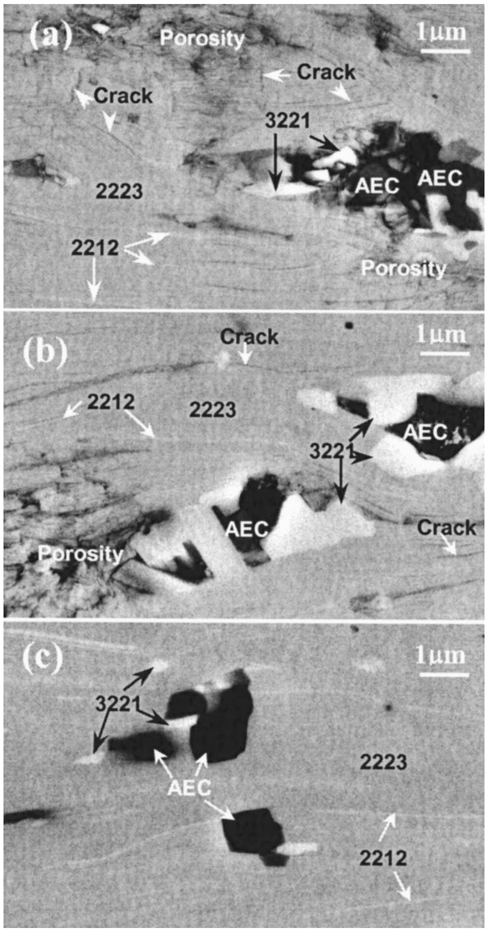

FIG. 1. Backscattered electron SEM images for (a) FHT, (b) FHT+1b, and (c) FHT+OP samples. The dominant gray regions are 2223; the large dark gray or black particles are grains of alkaline earth cuprates (AECs); the white particles are the 3221 phase; and the white streak-like grains are 2212. The irregular and line-like dark gray or black regions are porosity or cracks.

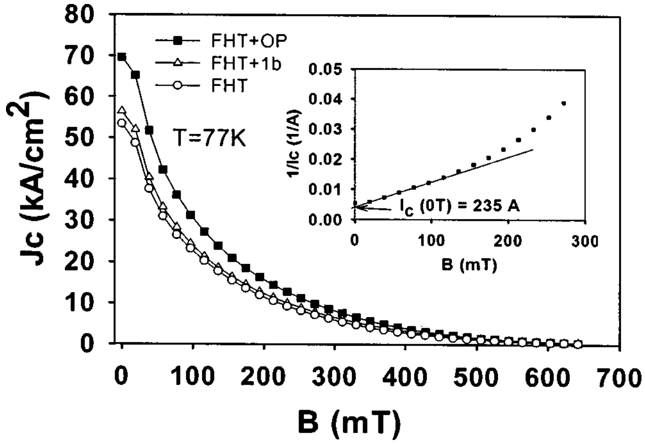

FIG. 2. J_c as a function of magnetic field for FHT, FHT+1b, and FHT+OP samples at 77 K. The field was applied perpendicular to the broad sample surface. The inset shows $1/I_c(B)$ as a function of B for the FHT+OP sample and the linear extrapolation to 235 A for $I_c(0)$.

based on the ideal gas law, and the oxygen fugacity was 0.075 and 0.084 bar at room temperature and 820 °C, respectively, based on the Lewis–Randall law for nonideal gas. The 1 bar samples used a N_2/O_2 gas mixture with pO_2 of 0.077 bar.

Figure 1 shows SEM images of the three samples. The common feature of FHT and FHT+1b samples was a large amount of residual porosity and cracks. In contrast, the FHT+OP sample was dense, the fabrication cracks were well healed, and the porosity was effectively removed. Measurements of the BSCCO cross section showed that the total cross-sectional area of the filaments was reduced from 2.99×10^{-3} cm^2 for FHT and FHT+1b samples to 2.61×10^{-3} cm^2 for FHT+OP samples. The micrographs also show that all three samples contain current limiting phases, including discrete 2212 grains within the 2223 colonies, AECs and $(Bi,Pb)_3Sr_2Ca_2CuO_x$ (3221). The FHT+1b sample [Fig. 1(b)] had the most and largest grains of 3221.

The $V-I$ characteristics of the samples were measured using the four-probe technique in liquid nitrogen with the external magnetic field applied perpendicular to the broad sample surface, which is approximately parallel to the c axis of the sample. Using the 1 μV/cm criterion, the I_c (77 K) values of the FHT+OP sample in self-field and at 0.1 T were 181.7 and 80.6 A, respectively. The corresponding J_c values in self-field and 0.1 T are 69.6 and 30.8 kA/cm^2. Figure 2 shows that these J_c values are 30% and 37% higher than the corresponding J_c values of 53.4 and 22.5 kA/cm^2 for the FHT sample and 23% and 30% higher than those of 56.4 and 23.7 kA/cm^2 for the FHT+1b sample. 30.8 kA/cm^2 is the highest J_c (0.1 T, 77 K) value that has been reported so far for 2223 tape.

The measured I_c at self-field for the FHT+OP sample was 181.7 A. However, Fig. 2 shows the J_c-B curves become flattened at low field due to very large self-field suppression effects on J_c.[18] The inset shows the extrapolation of the critical current for the FHT+OP sample to zero field I_c (0 T, 77 K) based on the model proposed by Kim *et al.*[19] where $1/I_c(B)=B/I_c(0)B_0+1/I_c(0)$. B_0 is the characteristic field and $I_c(0)$ is the critical current at 0 T. The extrapolated I_c (0 T, 77 K) for the FHT+OP sample is 235 A, corresponding to J_c (0 T,77 K) = 90 kA/cm^2.

Figure 3 shows the magnetization moment as a function of temperature taken in a Quantum Design superconducting

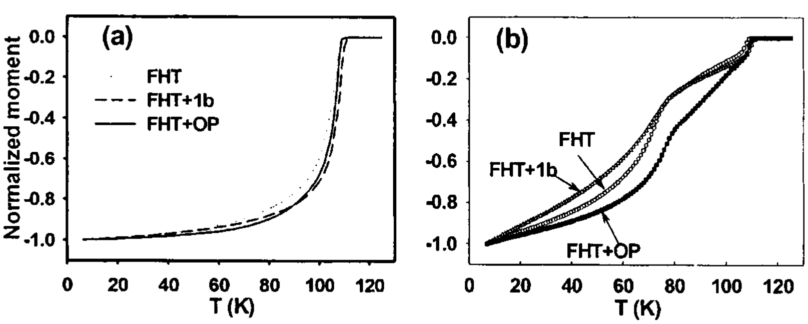

FIG. 3. Magnetization moment as a function of temperature for (a) undeformed and (b) rolled FHT, FHT+1b, and FHT+OP samples obtained by SQUID magnetometer in a 0.5 mT magnetic field that was applied parallel to the long axis of the tape. The rolling reduction is 30%.

quantum interference device (SQUID) magnetometer for the samples before and after they were rolled with a 30% reduction in thickness. As shown in Fig. 3(a), the unrolled samples all had a sharp transition, which is due to the strong shielding by the 2223. After rolling, the magnetization transition was much broader and had a kink at about 80 K [Fig. 3(b)], which is close to the T_c for 2212. The broadening and kink at ~80 K are both thought to be caused by 2212 in the sample that is exposed when the 2223 is crushed in the rolling process.[2,10,20]

A method has been proposed[2,10,20] to evaluate the relative amount of 2212 in the tape, which defines a 2212 SQUID index. As shown in Fig. 3(b), the 2212 SQUID indices are 0.32, 0.29, and 0.11 for FHT, FHT+1b, and FHT+OP samples, respectively. The much smaller value of 0.11 for the OP sample shows that OP processing decreased the 2212 content relative to the FHT and FHT+1b samples. In fact, the value of 0.11 is the smallest 2212 SQUID index that we have measured for 2223 samples from a variety of sources.[2,10,20]

J_c was enhanced by reduction of cross-section area and by improved connectivity. OP processing reduced porosity and cracking from ~20% of the cross-section area in the FHT and FHT+1b samples to less than 5% in the FHT+OP sample, which proportionally enhanced the active cross-section area that carried current. The 30% enhancement of J_c (0.1 T, 77 K) in FHT+OP samples compared to FHT+1b samples was partially due to the 13% reduction in cross-section area, but the remaining 17% enhancement must be due to the improvement in connectivity through eliminating current-blocking cracks and pores, reducing long-range obstructions of 2212 streaks and removing wetting phases at grain boundaries by the precipitation of 3221 seen in Fig. 1. Gurevich and Friesen[21] have shown that nonlinear effects on critical currents are due to removing dissipation obstructions in superconductors, some of which may be pores and grain-boundary wetting phases. 2212 grains and intergrowths can play a very important role in limiting J_c, as pointed out earlier by Umezawa *et al.*[15] Evidently OP processing reduces all these obstructions and thus results in the J_c increase in FHT+OP samples. Present studies using local probes such as magneto-optical imaging are investigating the local current obstructions in exceptionally high J_c samples such as these FHT+OP samples.

In conclusion, we have shown that OP processing 2223 tape after FHT results in a significant increase in transport properties. J_c (0.1 T, 77 K) of 30.8 kA/cm^2 and I_c (SF, 77 K) of 181.7 A, with J_c (SF, 77 K) of 69.6 kA/cm^2 and an extrapolated J_c (0 T, 77 K) of 90 kA/cm^2 have been achieved. Even though J_c (SF, 77 K) of 74 kA/cm^2 was previously reported in a Bi-2223 tape,[8] as commented in the literature,[2] the external cross-section dimensions (1.78 mm× ~0.08 mm) and critical current I_c (~30 A) of that tape were small, so the self-field suppression of I_c was also very small. The increase in J_c in FHT+OP samples is attributed to the OP process decreasing the density of cracks, pores, and residual 2212 content.

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