Overpressure Processing Bi2223/Ag Tapes

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***Abstract*—We report on the effect of overpressure processing on the electromagnetic properties and microstructure of monocore and multifilamentary Bi2223/Ag tapes. Samples at various stages of the usual thermo-mechanical processing for Bi2223/Ag tapes (from as-rolled to fully processed) were subjected to annealing at 815–820°C for 18–108 h in a static Ar + O_2 atmosphere at the overall pressure 17.5 ± 0.5 MPa (pO_2 = 0.003–0.02 MPa). Density measurements, microhardness tests, and SEM examination of polished sections reveal a notable densification of overpressure-processed samples. For partly reacted (so called HT1) samples, we found that overpressure processing for 36 h is sufficient to densify the Bi2223 core from 70 ± 5 to 87 ± 4% theoretical density and increase self-field J_c(77K) from 8 ± 1 to 30 ± 5 kA/cm^2 The results show that one-step processing of Bi2223/Ag tapes is possible.**

***Index Terms*—Bi2223/Ag tapes, Critical currents, Magneto-Optical studies, Overpressure processing**

I. Introduction

Silver sheathed $(Bi,Pb)_2Sr_2Ca_2Cu_3O_x$ (Bi2223/Ag) tapes are presently the only high-temperature superconductors (HTS) available in long lengths for application at liquid-nitrogen temperatures. Multifilamentary tapes with self-field critical current densities $J_c > 30$ kA/cm^2 and excellent J_c homogeneity from the scale of 100 μm to hundreds of meters are routinely produced by several manufacturers [1]. Despite the fact that all prototype devices for power applications are presently based on Bi2223/Ag tapes, there is a widespread concern that Bi2223 has neither the performance nor the low cost needed to make HTS applications generally attractive. Part of this concern is the slow increase in J_c of Bi2223/Ag conductors during the past five years, which is believed to be due to intrinsic limitations of the present processing route.

The Bi2223/Ag conductors are prepared by the oxide-powder-in-tube (OPIT) method and subjected to a thermo-mechanical processing in order to form and align the Bi2223 core. The usual thermo-mechanical processing consists of two heat treatment (HT) steps separated by an intermediate rolling (IR) step. During the first HT step (HT1), about 80% Bi2223 is formed from the precursor powder (Bi2212 + other cuprate phases). The reaction is usually accompanied by retrograde densification (the density decreases by 10–20% as compared to the as-rolled state) [2]. The IR step is needed to densify the Bi2223 core, and the HT2 step is intended to complete Bi2212 to Bi2223 conversion and heal microcracks produced during IR [1]. This thermo-mechanical processing is inherently compromised by its inability to remove porosity without introducing cracks. The result of this compromise is that even the best fully processed tapes contain 10–25% distributed pores [2], [3] and only 5–20% of the cross-section actually carries current [4], [5]. The real potential of Bi2223/Ag tapes was demonstrated by recent magneto-optical (MO) magnetization measurements [6] in monocore Bi2223 tapes that revealed regions with local $J_c \approx 180$ kA/cm^2, a value that is still 5 times smaller than J_c in high-quality Bi2223 thin films [7].

Hot isostatic pressing (HIP) is one of the obvious possibilities to overcome the problem of porosity and cracks in Bi2223 tapes. Earlier work showed that bulk 2223 is stable in its own atmosphere (pellets encapsulated in a PYREX glass) under 150 MPa and 650–870°C and can be densified to 95% theoretical density [8]–[10]. To apply HIP to Bi2223/Ag tapes, one has to solve the problems of isolating the core from the outer atmosphere and controlling very small levels of pO_2 in the HIP system to avoid decomposition of Bi2223. To our knowledge, application of HIP to processing Bi2223/Ag tapes was reported to be successful only at low temperature (700°C and 2 h) for short times under 150 MPa Ar, which restored a self field J_c of 0.5 kA/cm^2 [11]. Pure Ar cannot be used at higher temperature because the Bi2223 will decompose in this low pO_2 atmosphere [12].

In this work, we explore the possibility of using overpressure processing to densify the Bi2223 in Ag-sheathed tapes at temperatures where 2223 forms. Because of the low applied pressure ~20 MPa used in this work, we prefer the term *overpressure* (OP) instead of HIP. Our ultimate goal is to develop a one-step heat treatment that produces a dense, well-aligned Bi2223 microstructure. In this paper, we report the first positive results of our work. We found that applying an overpressure of only 17.5 MPa for 36 h at 815°C is sufficient to densify the Bi2223 core. For HT1 samples subjected to OP annealing, the 77K self-field J_c values are comparable to those of samples produced using the usual thermo-mechanical processing.

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II. EXPERIMENTAL PROCEDURES

Most of the data reported below were obtained using 3–4 cm long samples of 85-filament tapes after different stages of thermo-mechanical processing (as-rolled, after HT1, after IR, and fully processed) made at American Superconductor Corp. We also studied short samples of monocore tape prepared at UW from fully reacted (95% Bi2223) powder with overall composition $Bi_{1.8}Pb_{0.33}Sr_{1.87}Ca_{2.0}Cu_{3.0}O_x$ (Merck).

To isolate the BSCCO core from the high-pressure atmosphere, the ends of the tape samples were sealed by wrapping them in a 40 μm-thick Ag foil, pressing under a pressure of 400 MPa, and diffusion welding by annealing at 800°C for 3 h in 7.5% O_2 (balance N_2).

The overpressure experiments were carried out at ORNL in an overpressure furnace with a static atmosphere [13]. The high-temperature, high-pressure vessel of the furnace was made of Inconel 617 tube (1.7 cm ID, 4.8 cm OD). For safety reasons, the total maximum pressure at the temperatures of interest (815–830°C) was limited to $P_{total} \le 18$ MPa. Samples were put in a closed end of the quartz tube (8 mm ID) that was placed in the Inconel tube. Because of the very limited space in the furnace (a 5 cm long hot zone), only 4–6 tape samples could be stacked together. They were separated from one another by an inert powder. In our first runs, we used $CaZrO_3$ powder. We found, however, that this powder contained 1–2% $CaCO_3$ that decomposes above 600°C, and in the static atmosphere furnace, the trapped CO_2 reacted with Bi2223: $SrCO_3$ particles up to 20 μm in diameter were found by SEM and XRD in the outer filaments of the tapes after OP annealing at 820°C for 64h. We also learned from the nonuniform distribution of $SrCO_3$ particles between the outer and inner filaments of the reacted tape that the outer Ag sheath of multifilamentary tapes might contain microcracks. In the next runs, we wrapped each tape in a 100-μm thick Ag foil and electron-beam welded it. We used $SrZrO_3$ powder to separate the samples from one another.

Overpressure annealing was carried out at 815 and 820°C for 18–108 h at $P_{total} = 17.5 \pm 0.5$ MPa. The initial oxygen partial pressure of $pO_2 = 0.008$–0.021 MPa was achieved in the system by flushing the furnace with pure Ar (99.999%) at ~12 MPa 4–5 times, then adding the necessary amount of an Ar + 20%O_2 mixture, and pressurizing the furnace to ~12 MPa with pure Ar at room temperature. The final pressure ~17.5 MPa was reached during heating. The temperature was measured with a Pt–PtRh thermocouple and is accurate to within ± 2°C.

After the first OP runs at 820°C that began with an initial $pO_2 = 0.008$ MPa, we found that it is very difficult to maintain the proper pO_2 level in the furnace, because of the slow oxidation of the Inconel 617 tube. By putting 3 g of sintered 0.5 Bi2223 + 0.5 Bi2212 pellets close to the hot zone, we were able to maintain pO_2 in the range 0.012–0.005 MPa for 36 h at 815°C. The initial and final pO_2 was measured by mass-spectrometry. Most of the results reported below are obtained using these conditions.

Samples were characterized by I_c measurements in magnetic fields perpendicular to the broad face of the tapes. The I_c was determined from extended voltage–current characteristics using a 1 μV/cm criterion. Microstructure and phase composition were studied using SEM (JEOL JSM6100) of polished sections and XRD (STOE, Cu K_α radiation) of the sample surface after etching off the outer Ag sheath. The same samples were used for plan-view magneto-optical (MO) imaging. The mass density of individual filaments extracted from the samples was calculated from the weight (microbalance Cahn C-35), length, and area of the filament cross section. The Vickers microhardness of the Bi2223 core in monocore tapes was measured using a LECO M-400-G tester (10g, 10s).

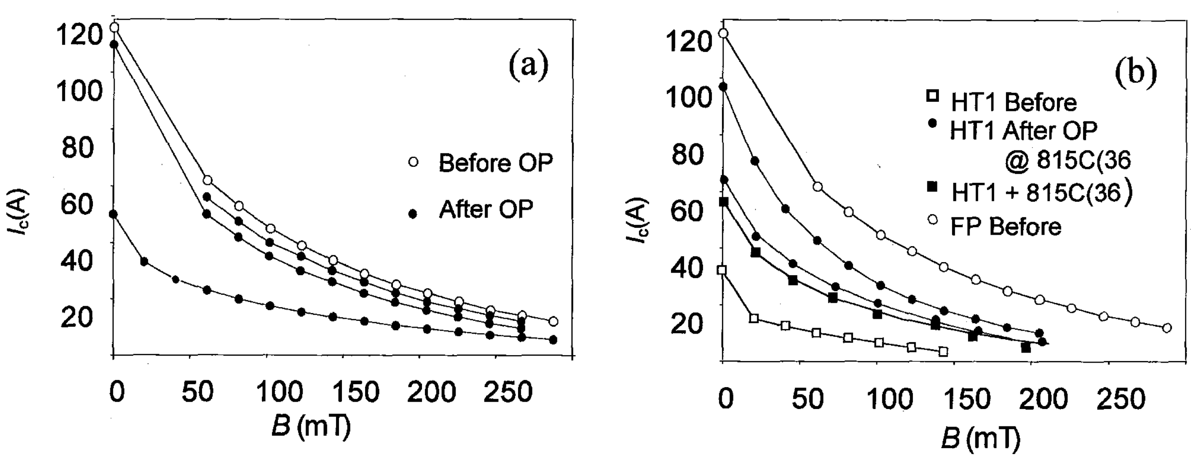

Fig. 1. Magnetic-field dependence of I_c in (a) fully processed Bi2223/Ag tapes and (b) HT1 tapes before and after OP annealing at 815°C for 36 h ($P_{total} = 17.5$ MPa, $pO_2 = 0.012$–0.005 MPa) and after an equivalent heat treatment with $P_{total} = 0.1$ MPa and $pO_2 = 0.008$ MPa. FP = fully processed.

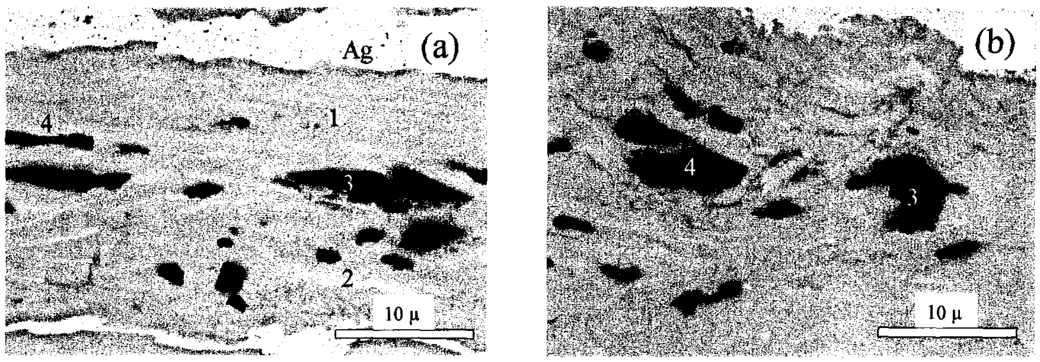

Fig. 2. Microstructure of HT1 sample (a) after OP annealing at 815°C for 36 h (P_{total} = 17.5 MPa, pO_2 = 0.010–0.006 MPa) and (b) after equivalent annealing, but at P_{total} = 0.1 MPa, $pO2$ = 0.008 MPa. Phases: (1) 2223, (2) 2212, (3) 14:24 AEC, (4) 2:1 AEC.

III. Results

A. Multifilamentary Tapes

Fig. 1a compares the $I_c(H)$ curves in fully processed tapes before and after OP annealing, where no improvement is observed. XRD data show that OP processed tapes experience some decomposition (the fraction of Bi2212 increased from 3 ± 1 to 6 ± 2%) due to improper pO_2. The decomposition was also seen in SEM studies: the amount of second phases [$(Ca,Sr)_2CuO_3$ and $(Pb,Bi)_3(Sr,Ca)_5Cu_1O_x$] increased by a factor of 1.5, which together with the remaining 2212 constituted ~10 vol% of the filaments.

In Fig. 1b, we present the $I_c(H)$ data for HT1 samples before and after OP annealing and after a similar annealing but at normal pressure (P_{total} = 0.1 MPa, pO_2 = 0.008 MPa). Although there is a certain irreproducibility in the results, the average I_c values of OP annealed samples are higher. In the best OP sample, the I_c is about 80% of the value for the optimized fully processed sample with self-field J_c(77K) = 35 kA/cm^2. Note that because of the improper pO_2 control, the amount of residual 2212 in this particular sample was reduced to only 7 ± 2%, i.e., twice as large as in the optimized fully processed tape and in the sample annealed 36 h at 815°C without OP. Comparison of microstructures in the HT1 samples processed with and without OP (Fig. 2) shows that OP annealing produces qualitatively denser material.

Measurements of the mass density of individual filaments confirmed the qualitative conclusion of the SEM work. The average mass density of the filaments after OP annealing is 5.5 ± 0.1 g/cm^3 compared to 4.4 ± 0.3 g/cm^3 before OP.

Fig. 3 compares the MO images of HT1 samples after OP annealing and after equivalent annealing, but at P_{total} = 0.1 MPa, pO_2 = 0.008 MPa. The average magnetization level is significantly larger in the sample after OP (cf. Figs. 3a and 3b). However, the flux penetration pattern (Fig. 3c) is very similar to that after annealing at the normal pressure (Fig. 3a).

XRD studies of both HT1 and fully processed samples before and after OP show a noticeable deterioration of grain alignment during OP. The FWHM of the rocking curves of the 00<u>24</u> reflection increased by about 2° (e.g., from ~14 to ~16° for HT1 samples).

B. Monocore Tape with Reacted Bi2223 powder.

Vickers microhardness (HV) was measured in the monocore tape made with fully reacted Bi2223 powder that had been OP processed at 815°C for 18 h and after annealing at 815°C for 36 h at normal pressure (P_{total} = 0.1 MPa, pO_2 = 0.008 MPa). The average HV is dramatically improved from 104 ± 3 to 187 ± 2 kg/mm^2 by applying overpressure. Because the size of the indentation trace (~10 μm) is much larger than the size of second-phase particles (~1–5 μm), this increase can be completely attributed to the higher density of the Bi2223 core.

No supercurrent was measured at 77 K in the tape annealed at normal pressure. The tape subjected to the OP annealing has a very low J_c(77 K, 0T) ≈ 50 A/cm^2. This can be related to the poor texture in these samples. The FWHM of the rocking curve of the 00<u>24</u> reflection was 27.3 ± 0.6°.

IV. Discussion

We processed multifilamentary and monocore Bi2223/Ag tapes under moderate overpressure, and studied the effect of such processing on transport properties, microstructure, and mass density.

Our experiments were carried out in a furnace with a static atmosphere and oxygen-consuming environment. This imposed very rigid constraints on the duration of OP annealing. We were able to maintain $pO2$ in the range 0.012–0.005 MPa, where Bi2223 is stable at 815°C for 36h. Working in this pO_2 range suppressed severe decomposition of 2223, but did not completely eliminate it. In fully processed samples, about 5% of the 2223 decomposed during OP annealing, and in HT1 samples, the formation kinetics of 2223 seems to be slower than for the usual heat treatment with constant pO_2 = 0.008 MPa. This might affect the I_c performance of the samples. Nevertheless, the I_c level reached in these OP experiments is quite encouraging, particularly for HT1 samples. Although, we did not obtain high I_c by OP annealing as-rolled samples, the positive results for HT1

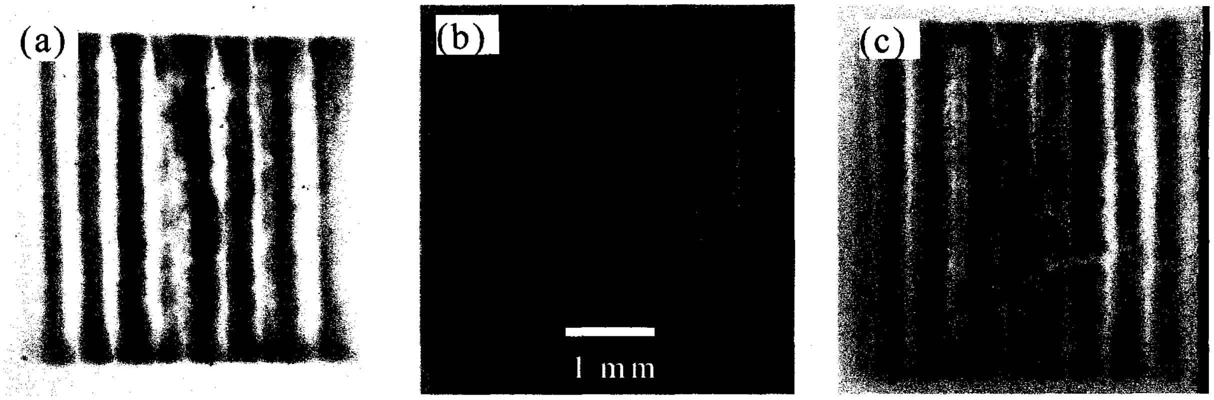

Fig. 3. MO images taken in the ZFC mode at T = 15 K and B = 80 mT of HT1 samples after annealing at 815°C for 36 h (a) without and (b, c) with OP processing. Images (a) and (b) were obtained under exactly the same optical conditions. Image (c) was obtained by changing optical conditions to better resolve the flux-penetration pattern.

samples suggest that one-step processing of Bi2223/Ag tapes is possible.

Microstructural observations, density measurements, and microhardness data give convincing evidence that applying moderate overpressure of 17.5 MPa produces noticeable densification of the Bi2223 core. However, longer annealing time and/or higher external pressures are needed to complete densification.

The same conclusion can be made based on the MO data. Changes in the average magnetization revealed from MO images are consistent with the I_c measurements. However, very similar flux-penetration patterns in the samples annealed with and without overpressure suggest that cracks, which are a key current-limiting defect, still remain in the samples.

Deterioration of 2223 texture during OP annealing was observed, perhaps due to rumpling on densification. Note that the core densification during the IR step of the usual processing is also accompanied by deterioration of texture [14]. Almost zero I_c observed in highly dense Bi2223 monocore tape with very bad texture emphasizes that densification is not sufficient to obtain high I_c. Nevertheless, eliminating porosity is a very necessary step and the results of the present work show that this goal can be attained by OP processing.

V. Conclusion

The main result of our studies is that overpressure processing at 815°C for 36 h under a pressure of only 17.5 MPa is sufficient to densify the Bi2223 core in multifilamentary tapes from 70 ± 5 to 87 ± 4% theoretical density. Longer annealing at higher pressures with better controlled oxygen partial pressure is necessary to fully understand the potential of OP processing for fabricating Bi2223/Ag conductors.

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