Overpressure Processing of Ag-sheathed Bi-2212 Tapes

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***Abstract*—A critical problem for melt-processed silver-sheathed Bi-2212 tapes is gas release during heat treatment which deforms the sheath ("bubbling") and forms voids in the superconducting core. An overpressure furnace is used to study how the extent of bubbling changes with the total pressure (P_{tot}). By processing in P_{tot} up to 9 atmospheres with constant oxygen partial pressure (pO_2 = 1 atm), bubbling in coils is eliminated. The critical current density increases with increasing P_{tot}. Also, the phase assemblage in overpressure-processed tapes is unchanged compared to conventionally processed tapes.**

I. INTRODUCTION

When long lengths of Bi-2212 tapes are processed in pure oxygen at 1 atmosphere (atm) pressure, the silver sheath deforms ("bubbles") at temperatures between 800-900°C [1]. The tape bubbles because the force from the gas pressure inside the tape is greater than the force from the Ag sheath and the external gas pressure. To prevent bubbling, the tapes could be processed in lower oxygen partial pressure (pO_2), but lower critical current densities (J_c) result [2]. Another approach was taken by researchers at Hitachi who developed a "gas pressure melting" method that utilized total pressure (P_{tot}) and pO_2 greater than 1 atm during processing of Bi-2212 wires [3]. They found that increasing P_{tot} resulted in a more homogeneous distribution of small (<0.6 mm) voids in the core.

In our overpressure approach, we use a mixture of O_2 and an inert gas (Ar) with P_{tot} > 1 atm and fixed pO_2 = 1 atm. Since O_2 diffuses rapidly through the Ag, the pO_2 inside and outside the tape quickly equilibrates. In contrast, Ar does not equilibrate during the short time at high temperature. Thus, Ar supplies the external pressure to balance the gas pressure inside the tape. The J_c and microstructure of these overpressure-processed tapes are compared to conventionally processed tapes.

II. EXPERIMENTAL CONDITIONS

The Bi-2212 tapes with powder composition Bi:Sr:Ca:Cu = 2.1:2:1:1.95 were made using the OPIT process and had a final thickness of 140 μm. To ascertain the role of carbon in bubble formation, two tapes with different carbon content were made. The initial carbon content of the powder was 710 ppmwt (parts per million by weight) as measured by high-temperature combustion infrared detection. To lower the carbon content to 220 ppmwt, powder was heated to 700°C in partial vacuum with flowing O_2 for 1 day. Coils were made by sealing the ends of 30 cm of tape and winding the tape around a 3.2 cm diameter MgO coated Inconel mandrel. The two coils with 710 and 220 ppmwt carbon were heated at 5°C/min to 900°C in pure O_2 with P_{tot} = 1 atm, held at 900°C for 15 minutes, and then furnace cooled. This process was repeated for P_{tot} = 2, 4.6, 6, 8, and 9 atm to see what effect P_{tot} had on bubbling in low and high carbon content tapes. The gas pressure was established in the furnace before heating and was maintained until cooling to room temperature. For P_{tot} > 1 atm, an O_2/Ar gas mixture was used to maintain a fixed pO_2 = 1 atm.

For the electromagnetic and microstructural characterization of fully processed tapes, short lengths (4-8 cm) of 162 ppmwt C tape with sealed ends were melt processed in the overpressure furnace using the heating schedule shown in Fig. 1.

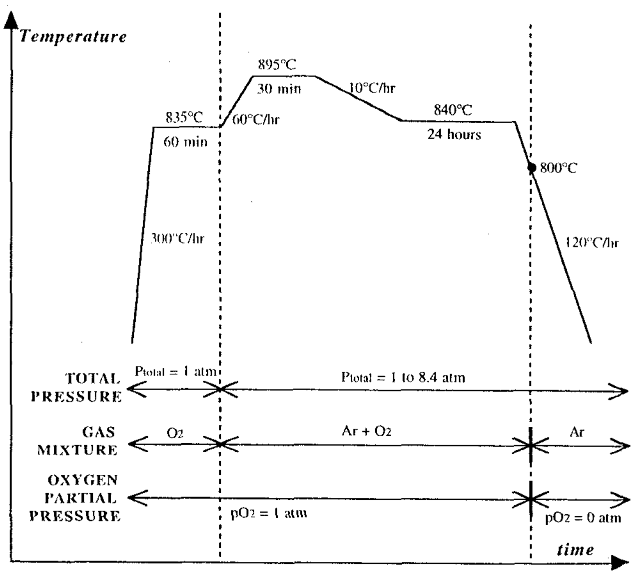

Fig. 1. Heat treatment for Bi-2212 tapes processed in the overpressure furnace. The final cooling rate of 120°C/hr is the fastest rate possible with this furnace design.

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The cooling rate of 120°C/hr during the last step of the heat treatment is the fastest cooling rate we can achieve with our overpressure furnace. In order to see what effect a faster cooling rate has on J_c, one set of samples was processed at $P_{tot} = pO_2 = 1$ atm using an identical heat treatment schedule in a different furnace that is capable of cooling much more quickly (700°C/hr). The transport critical current was measured with multiple voltage taps over 3 cm sections using the standard four point probe technique at 0T, 4.2K, with a 1μV/cm criterion. Backscattered electron imaging was done on the JEOL JEM6100 scanning electron microscope (SEM) using energy dispersive x-ray spectroscopy (EDS) for phase identification.

III. RESULTS AND DISCUSSION

A. Effect of overpressure on bubbling

To illustrate the effect of carbon content and P_{tot} on bubble formation, the length and thickness of the individual bubbles that formed at a lower pressure (2 atm) and a higher pressure (8 atm) were measured. Fig. 2 shows that with increasing P_{tot}, the calculated volume and number of bubbles decreases for both high and low carbon content tapes. Also, at both 2 atm and 8 atm, the high carbon content tape had more bubbles than the low carbon content tape.

The percentage of the tape length that bubbled is plotted versus P_{tot} in Fig. 3. In pure oxygen where $P_{tot} = pO_2 = 1$ atm, the entire coil bubbles. Even 1 atm of overpressure ($P_{tot} = 2$ atm) significantly reduces bubbling. The percentage of tape length that bubbles continues to slowly decrease as the overpressure increases. The P_{tot} needed to eliminate bubbling increases with increasing carbon content. $P_{tot} = 8$ atm and $P_{tot} = 9$ atm eliminates bubbling in the lower and higher carbon content tapes respectively. This result is not surprising, as our previous work has implicated CO_2 as the bubble forming gas [4].

Applying a large enough overpressure during heating to the melting temperature stops bubbling. However, the trapped gas causes further problems. The consequences of having gas still trapped inside the core were seen during the initial investigation we did at Hitachi of the effect of overpressure during the entire heat treatment. One meter of tape with 500 ppmwt C was wrapped into a 3.2 cm diameter coil and processed in the Hitachi overpressure system with the following heat treatment at $P_{tot} = 6$ atm and $pO_2 = 1$ atm: vacuum anneal at 740°C for 16 hours, heat to 890°C at 5°C/min, hold at 890°C for 30 minutes, cool at 10°C/hr to 840°C, hold at 840°C for 24 hours, and furnace cool to room temperature. The silver sheath did not bubble; however, J_c was low. The microstructure of this tape showed large voids in the core. Therefore, even when the outside of the Ag sheath does not bubble, large voids are still present inside the core. These voids degrade the electromagnetic properties of the superconductor. Therefore, in later experiments, low carbon content tape was used in conjunction with overpressure processing in order to minimize bubbling and increase J_c.

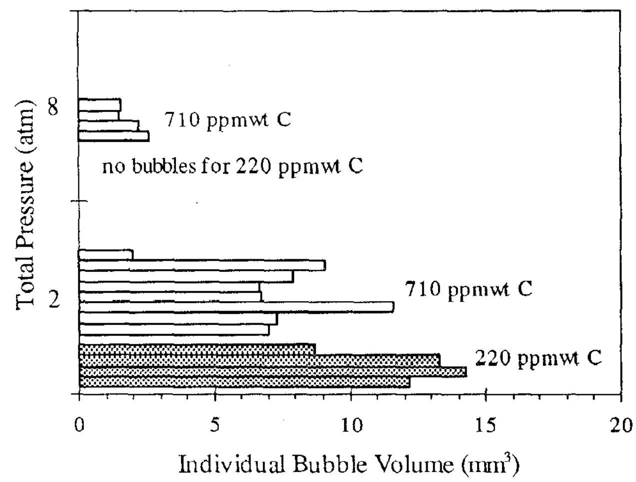

Fig. 2. Histogram of volume and number of bubbles for P_{tot} =2 and 8 atm, pO_2 = 1 atm. At each pressure, more bubbles formed in the high carbon content tape. Additionally, for each carbon content, increasing the pressure decreased the size and number of bubbles that formed.

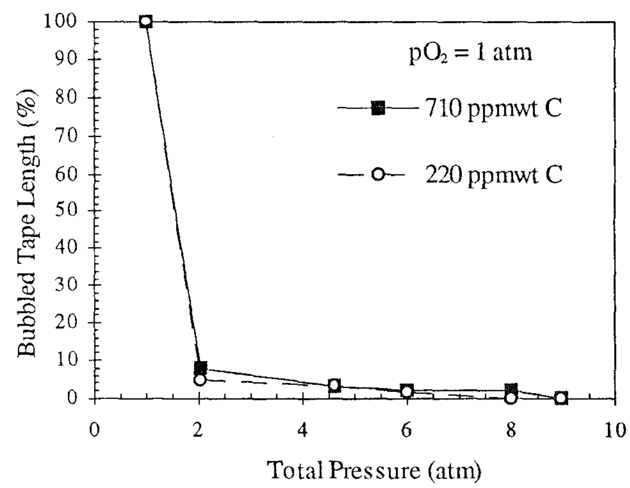

Fig. 3. Graph of percentage of bubbled tape length vs. total pressure. Having just 1 atm of overpressure (P_{tot} = 2 atm) significantly reduces bubbling; bubbling is eliminated by P_{tot} = 9 atm for even the highest carbon content tape.

B. Effect of overpressure on J_c

Fig. 4 shows that increasing P_{tot} from 1 atm to 8.1 atm with pO_2 = 1 atm results in the substantial increase in J_c for the slow cooled (120°C/hr) tapes. Also, there is less scatter in

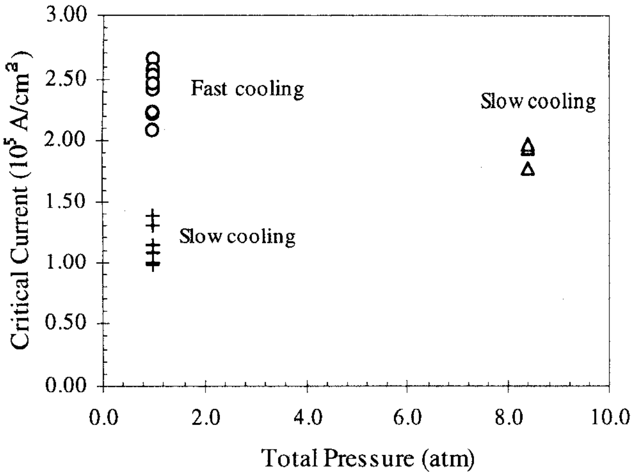

Fig. 4. J_c vs. pressure with fixed pO_2 = 1 atm. The fast (700°C/hr in O_2) and slow (120°C/hr in Ar) cooling rates refer to the last step in the heat treatment shown in Fig. 1. For the slow cooled samples, increasing pressure increases J_c. However, the highest J_c samples are fast cooled.

the J_c values at 8.1 atm than at 1 atm. The overpressure compresses the core during processing, and may encourage Bi-2212 grain alignment, reduce porosity, and thus improve the J_c of the superconductor. If this is indeed the case, overpressure-processed tapes should carry more current than conventionally processed tapes. The data show that the cooling rate has a significant effect on J_c; fast cooling in pO_2 = 1 atm is needed to achieve high J_c. Therefore, incorporating fast cooling into the overpressure processing should improve J_c. However, fast cooling is not feasible for coils that have a large thermal mass, and modifications in the cooling process need to be developed that allow slow cooling while retaining high J_c.

C. Effect of overpressure on microstructure

Fig. 5 shows a representative backscattered electron image of a sample processed in the overpressure furnace with P_{tot} = 8.1 atm, pO_2 = 1 atm, and furnace cooling = 120°C/hr. The microstructure contains the expected phase assemblage for processing in pO_2 = 1 atm: Bi-2212, small grains of $(Sr,Ca)_{14}Cu_{24}O_x$ (14:24 AEC (alkaline earth cuprate)) particles, and very few small grains of $Bi_2(Sr,Ca)_4O_x$ (2:4 CF (Cu free)) particles. Also present are thin long grains of $Bi_2Sr_2CuO_x$ (2201) from the liquid that transformed to 2201 on cooling from 840°C to room temperature and possibly from 2212 that decomposed during cooling. The same phase assemblage is seen in tapes processed at P_{tot} = pO_2 = 1 atm and furnace cooling = 120°C/hr, but there is a large amount of 2201 in this tape(Fig. 6).

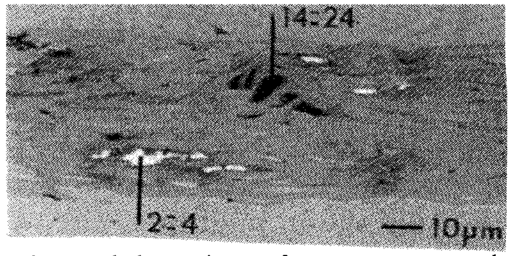

Fig 5. Backscattered electron image of overpressure-processed tape with furnace cooling = 120°C/hr, P_{tot} = 8.1 atm, pO_2 = 1 atm. The non-superconducting phase particles include 14:24 AEC and 2:4 CF.

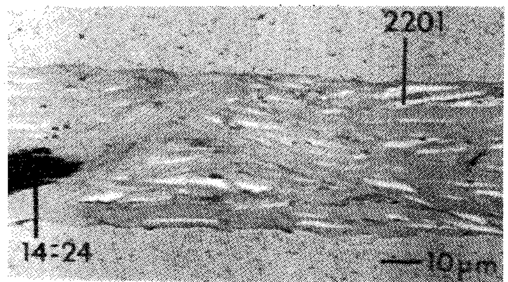

Fig 6. Backscattered electron image of overpressure-processed tape with furnace cooling = 120°C/hr, P_{tot} = 1 atm, pO_2 = 1 atm. The non-superconducting phase particles include 14:24 AEC and Bi-2201.

IV. SUMMARY

While bubbling is still dependent on the starting carbon content of the Bi-2212 powder, overpressure processing, defined as the application of inert gas pressure above the equilibrated pO_2, can eliminate visible bubbling of the silver sheath. Although overpressure processing results in higher J_c than conventional processing, the final cooling rate also has a strong effect on the current-carrying capability of the tapes. The results suggest that while overpressure processing may decrease porosity in the core and encourage grain alignment, slow cooling rates may cause the Bi-2212 to decompose, thus degrading the current paths and decreasing the J_c. Either incorporating fast cooling or modifying the slow cooling process may result in even higher J_c.

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