Transport Critical Current Density of Bi-Sr-Ca-Cu-O/Ag Superconductor Tapes with Addition of Fe$_3$O$_4$ as Flux Pinning Center

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Abstract. This paper reports on the flux pinning capability of micron size Fe$_3$O$_4$ in Bi-Sr-Ca-Cu-O superconductor tapes. Ag sheathed high temperature superconductor tapes with starting compositions (Bi,Pb)$_2$Sr$_2$Ca$_2$Cu$_3$O$_{10}$ (2223) and (Bi,Pb)$_2$Sr$_2$Ca$_2$Cu$_3$O$_{10}$-(Fe$_3$O$_4$)$_{0.01}$ were fabricated using the powder in tube method. The Bi-Sr-Ca-Cu-O powders were prepared by using the co-precipitation technique. The effects of Fe$_3$O$_4$ addition on the microstructure, phase formation, critical temperature and transport critical current density, $J_c$, were studied. The $J_c$ value of the Fe$_3$O$_4$ added tapes is higher (6,090 A/cm$^2$ at 77 K and 24,500 A/cm$^2$ at 30 K, in zero field) than the non-added tapes (3,730 A/cm$^2$ at 77 K and 13,318 A/cm$^2$ at 30 K, in zero field). A sudden decrease of $J_c$ in low magnetic fields ($B < 0.12$ T) when applied parallel ($B_||$) and perpendicular ($B_\perp$) to the tapes surface was observed. The destruction of weak links played an important role in the early $J_c$ suppression. The rate of decrease of $J_c$ was observed to decrease when the magnetic field was increased further. Improvement in the flux pinning was observed in (Bi, Pb)$_2$Sr$_2$Ca$_2$Cu$_3$O$_{10}$-(Fe$_3$O$_4$)$_{0.01}$ tapes. This study shows that magnetic particles such as Fe$_3$O$_4$ can act as effective pinning centers leading to the enhancement of $J_c$ in the system.

1. Introduction

The (Bi,Pb)$_2$Sr$_2$Ca$_2$Cu$_3$O$_{10}$ (Bi-2223) superconductor is one of the most promising materials for tape or wire applications. The poor performance especially under magnetic fields which arises from the weak pinning of flux lines, limits the application of this material [1]. High critical current density ($J_c$) is the requirement to meet practical applications. The strong increase of $J_c$, up to the theoretical limit can be achieved when the flux lines are pinned and their movement completely prevented [2]. The flux lines could be pinned by introducing effective artificial pinning centers so as to sustain the current density at higher fields and higher temperatures. This method includes the introduction of magnetic impurities as pinning centers. Magnetic dots are successfully used as artificial pinning arrays in superconducting film [3].

In this study we introduced Fe$_3$O$_4$ magnetic particle in Bi-Sr-Ca-Cu-O/Ag tapes prepared by powder in tube (PIT) method. Previous theoretical results indicate that magnetic particle can lead to frozen flux superconductor [4]. In contrast with conventional system, magnetic field induced by the
non-uniform magnetization penetrates into the superconductor provides strong coupling between the flux lines and magnetic textures.

2. Experimental Details

Sample with nominal composition Bi$_{1.6}$Pb$_{0.4}$Sr$_2$Ca$_2$Cu$_2$O$_{10}$ (BPSCCO) were prepared using the metal acetates of bismuth, strontium, lead, calcium and cooper (purity $\geq 99.99\%$), oxalic acid, deionized water and 2-propanol. The coprecipitation method was used due to advantages such as good homogeneity, low reaction temperature, fine and uniform particle size. The coprecipitation precursors were prepared by pouring the solution containing the metal ions into another containing 0.5 M oxalic acid dissolved in deionized water:2-propanol (1:1.5) and a uniform, stable, blue suspension was obtained. The slurry was filtered after 5 min of reaction time followed by drying stage in temperature of 80°C for 12 hr. The blue precipitate powders were heated up to 730°C in air to remove the remaining volatile materials.

The calcined powders were reground and heated again at 845°C in air for 24 hr followed by cooling at 2°C/min. After the sintering process, Fe$_3$O$_4$ ultra-fine particles (with average grain size of greater than 1 µm) were added with composition (Bi$_{1.6}$Pb$_{0.4}$Sr$_2$Ca$_2$Cu$_2$O$_{10}$(Fe$_3$O$_4$)$_x$ with $x = 0$ and 0.01. The powders were ground and packed into Ag tube with outer diameter of 6.12 mm and inner diameter of 4.43 mm. This wt % is chosen based on our initial studies on bulk Bi-2223 samples where 0.01 wt% of Fe$_3$O$_4$ optimized the superconducting properties of the system.

The tube was grooved rolled, drawn into wire and then flat rolled into tape form. All deformation processes were carried out using a rolling cylinder 20 mm in diameter and rolling speed of about 0.6 m/min. The tapes were cut into 2 cm long sections and divided into six groups. Each group was sintered for 100 hr at different sintering temperature. The FF 201 sample was sintered at 840 °C, FF 211 at 845 °C, FF 221 at 850 °C sintered for 100 h. FN 201, FN 211 and FN 221 are tapes without Fe$_3$O$_4$ addition sintered at various temperatures for 100 hr.

The transition temperature was determined using the standard four-point probe method in conjunction with a closed cycle refrigerator. The transport critical current density measurements were done at 77 K using the four-point probe method with the 1 µVcm$^{-1}$ criterion. In this criterion as the current is varied, the voltage (V) across the tape with known cross section area was measured and divided by the distance between the voltage probes. The phases in the samples were determined by X-ray diffraction analysis (using Philips X’Pert Pro diffractometer) equipped with a monochromat on at the diffracted beam side. The volume fractions of the Bi-2223 and Bi$_2$Sr$_2$CaCu$_2$O$_8$ (Bi-2212) phase were estimated by assuming that the amounts of those phases are proportional to the strongest diffraction line of each phase [5]. Microstructure of sample was recorded using a Philips XL-30 scanning electron microscope. A Philips energy dispersive X-ray (EDX) analyzer PV99 was used to determine the distribution of Fe$_3$O$_4$ in the tapes.

3. Results and Discussion

Table 1 shows $J_c$ for different sintering temperature. It is clearly seen that $J_c$ is strongly correlated with the sintering temperature with optimum value at 845 °C. The volume fraction of the Bi-2223 phase versus sintering temperature is also given in Table 1. The Bi-2223 phase increases with sintering temperature up to 845 °C followed by a decrease at higher temperature. These results are similar to the Bi-2223 added with TiO samples [6].

Figure 1 and 2 show the effect of sintering time and sintering temperature on $J_c(B)/J_c(B = 0$ T) in a semi log scale for the tapes in applied magnetic field. $J_c$ is higher when the applied field is parallel to the surface and is smaller when $B$ is perpendicular to the tapes’ surface [7]. $J_c$ is higher in the Fe$_3$O$_4$ added (FF 211) tape compared to non-added tape. $J_c$ decreased drastically when a small applied field. This effect is due to the decoupling of large amounts of Josephson junctions weak links. Weak links act as major supercurrent links in the low $J_c$ tapes and are easily destroyed in low fields. At low magnetic field, the $J_c$ of the Fe$_3$O$_4$ added tapes are larger than that of the non-added tape. Addition of Fe$_3$O$_4$ improved the $J_c$ below 0.1 T. At fields higher than 0.12 T, a lower rate of decrease is
observed and is attributed to flux pinning within the grains. The results indicate that Fe$_3$O$_4$ particles enhance the transport properties of Bi-2223/Ag tapes and may act as effective flux pinning centers. These figures show a sudden drop in the $J_c$ as soon as the magnetic field is applied (from 0 – 0.12 T) due to the destruction of the weak links. However, the Fe$_3$O$_4$ added samples show a smaller rate of decrease in $J_c$ as the field is increased from 0.12 T to 0.8 T compared to non-Fe$_3$O$_4$ added tape. This indicates that the magnetic particles enhance the strong link in the tapes. It is also interesting that the FF 221 tape with lower Bi-2223 volume fraction has higher $J_c$ at 30 K compared to FF 201 and FF 211.

Table 1. Sintering temperature, $J_c$ and volume fraction of 2223:2212 phase of the tapes

| Samples       | Sintering Temp (ºC) | $J_c$ at 30 K (A/cm$^2$) | $J_c$ at 77 K (A/cm$^2$) | 2223: 2212 (%) |
|---------------|---------------------|--------------------------|--------------------------|----------------|
| Bi$_{1.6}$Pb$_{0.4}$Sr$_2$Ca$_2$Cu$_3$O$_{10}$-$\delta$(Fe$_3$O$_4$)$_{0.01}$ | FF 201 | 840 | 17270 ± 180 | 5180 ± 280 | 70 : 30 |
|               | FF 211 | 845 | 24550 ± 220 | 6090 ± 340 | 76 : 24 |
|               | FF 221 | 850 | 25540 ± 170 | 5650 ± 190 | 57 : 43 |
| Bi$_{11.6}$Pb$_{0.4}$ Sr$_2$Ca$_2$Cu$_3$O$_{10}$ | FN 201 | 840 | 12270 ± 180 | 2640 ± 140 | 72 : 28 |
|               | FN 211 | 845 | 13180 ± 230 | 3730 ± 270 | 74 : 26 |
|               | FN 221 | 850 | 8360 ± 150 | 2550 ± 180 | 70 : 30 |

Figure 1. Effects of sintering temperature on normalized $J_c$ versus $B$ for $B_{||}$ and $B_{⊥}$ to the tape plane at 77 K for Bi$_{1.6}$Pb$_{0.4}$Sr$_2$Ca$_2$Cu$_3$O$_{10}$-$\delta$(Fe$_3$O$_4$)$_{0.01}$ sample. FF201 is sample sintered at 840 ºC, FF 211 at 845 ºC and FF 221 at 850 ºC.

Figure 2. Effects of sintering temperature on normalized $J_c$ versus $B$ for $B_{||}$ and $B_{⊥}$ to the tape plane at 77 K for Bi$_{11.6}$Pb$_{0.4}$ Sr$_2$Ca$_2$Cu$_3$O$_{10}$ sample. FN201 is sample sintered at 840 ºC, FF 211 at 845 ºC and FN 221 at 850 ºC.

Figure 3 shows the effect of sintering temperature on the temperature dependence of $J_c$ of the Fe$_3$O$_4$ added tape and Figure 4 shows $J_c$ of the non-Fe$_3$O$_4$ added tapes. A change in the $J_c$ versus $T$ curve is observed. The Fe$_3$O$_4$ added tape showed a parabolic-like $J_c$ which curved downwards. The differences in the temperature dependent $J_c$ indicate a different pinning mechanism in both cases [8].

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Figure 3. Effects of sintering temperature on the temperature dependence of critical current density $J_c$ to the tape for Bi$_{1.6}$Pb$_{0.4}$Sr$_2$Ca$_2$Cu$_3$O$_{\delta-(Fe_3O_4)_{0.01}}$ sample. FF201 is sample sintered at 840 °C, FF 211 at 845 °C and FF 221 at 850 °C. Solid lines are for eye guide. (FF 201 (♦), FF 211 (■), FF 221 (▲)).

Figure 4. Effects of sintering temperature on the temperature dependence of critical current density $J_c$ to the tape for Bi$_{1.6}$Pb$_{0.4}$Sr$_2$Ca$_2$Cu$_3$O$_{\delta}$ sample. FN201 is sample sintered at 840 °C, FN 211 at 845 °C and FN 221 at 850 °C. Solid lines are for eye guide. (FN 201 (♦), FN 211 (■), FN 221 (▲)).

Figure 5 shows the SEM micrograph of the Fe$_3$O$_4$ used in this study. The average grain size is slightly greater than 1 µm. The microstructure of the Fe$_3$O$_4$ added tape (Figure 6(a)) shows well-aligned and elongated grain, with high apparent density compared to non-added tape (Figure 6(b)). Better grain connectivity and higher degree of grain alignment reducing the weak link lead to higher $J_c$ values.

In order to evaluate the distribution of Fe$_3$O$_4$ in the tape, mapping analysis has been performed. Figure 6(a) shows the distribution of Fe$_3$O$_4$ of the cross section of the tape and the white dots corresponds to Fe$_3$O$_4$ from EDX analysis. It is found that $J_c$ is enhanced in Fe$_3$O$_4$ added tape where the size of the grains decreased. This is an indication that the energy loss is higher in samples having large grains than in samples with small grains [8].

Figure 5. SEM micrograph of the Fe$_3$O$_4$ used in this study.
The current carrying capability is directly related to the bulk pinning force density $F_p$. For spherical pinning centers with radius $R$, Ullmaier showed that $F_p = F_p[B(T)]^m$ where $m = 3/2$ for $R > \xi$ and $m = 5/2$ for $R < \xi$, where $\xi$ is the coherence length [9]. From the SEM result (Figure 5), the average size of Fe$_3$O$_4$ is more than 1µm. The coherence length of the Bi-2223 system is around 2.9 nm [10], i.e. the diameter of pinning center is larger than the coherence length ($R > \xi$), consistent with $m = 3/2$. This is also consistent with the result of self-field effect on $J_c$ versus temperature curve [8].

Using a self-field approximation together with $J_c$ dependence on $T$, between 30 K and 80 K a change in the characteristic length ($L_c$) associated with the pinning force is observed and is attributed to the change in the microstructure [7, 11] in the tapes. The $J_c$ versus $T$ curve of the Fe$_3$O$_4$ added Bi-2223 tapes (Figure 3) is consistent with $R_g < L_c$ while the non-added tapes (Figure 4) is consistent with $R_g > L_c$ [8] where $R_g$ is the grain size.

Conclusion
The addition of Fe$_3$O$_4$ in Bi-2223/Ag tapes effectively enhances the flux pinning at low magnetic field and consequently increased $J_c$ of the tapes. This enhancement is proposed to be due to the interaction which bounds the flux line and the magnetic Fe$_3$O$_4$. Magnetic impurities generally suppress superconductivity, however, our result shows that magnetic particles improved the transport current in line with previous calculation on frozen flux superconductor with magnetic particles as pinning centers.

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