ORIGINAL PAPER

Enhanced Transport Critical Current Density in Ag-Sheathed ($\text{Bi}_{1.6}\text{Pb}_{0.4}$) $\text{Sr}_2\text{Ca}_2\text{Cu}_3\text{O}_y$ Superconductor **Tapes with Different Nano-Sized Co3O4 Addition**

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Received: 3 April 2014 / Accepted: 28 November 2014 / Published online: 16 December 2014 © Springer Science+Business Media New York 2014

Abstract Ag sheathed $(Bi_{1.6}Pb_{0.4})Sr_2Ca_2Cu_3O_y(C_0a_4)_x$ superconductor tapes with addition of $Co₃O₄$ nanoparticles (with size 30 and 50) were fabricated using the powder-intube (PIT) method. The structure and microstructure were studied along with critical temperature (T_c) and transport critical current density (J_c) . The nanoparticle added tapes showed a higher J_c value compared with the non-added tapes. The 30-nm Co₃O₄ added tapes showed a higher J_c value compared with the 50 nm $Co₃O₄$ added tapes. This study showed that $Co₃O₄$ nanoparticles could act as effective pinning centers leading to enhancement of J_c in the Bi-2223/Ag sheathed tapes. The addition of smaller $Co₃O₄$ nanoparticles (30 nm) results in stronger pinning. The full vortex magnetic energy due to addition of $Co₃O₄$ magnetic nanoparticles led to the enhancement of *J*c.

Keywords Co-precipitation · Critical current density · Critical temperature · Nanoparticles

1 Introduction

Large-scale applications of high-critical temperature (T_c) superconductors at liquid nitrogen temperature under magnetic fields require high transport critical current density. The flux pinning capability is one of the basic parameters that is important for the application of superconductors. In order to

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enhance the flux pinning properties and improve the transport current density, many attempts have been made by introducing artificial defects of nanometer size as possible pinning centers [\[1](#page-5-0)[–6\]](#page-5-1). The improvement in transport critical current density (J_c) can be obtained by introducing an efficient pinning center with size that matches the coherence length. Magnetic nanoparticles have been suggested as effective pinning centers [\[7,](#page-5-2) [8\]](#page-5-3). The use of magnetic nanoparticles with various shapes such as nanorod has been suggested to increase the pinning potential and thus enhance J_c [\[9\]](#page-5-4).

The two important characteristic lengths in superconductors are the coherence length (*ξ)* and penetration depth (*λ)*. Generally, the critical current density will increase when the size of magnetic particles is increased, and *L* is larger than *ξ* but smaller than *λ* [\[10\]](#page-5-5). In another report however, pinning is expected to be optimized when the size of the defects approaches the coherence length. The coherence length of $Bi₂Sr₂Ca₂Cu₃O₁₀$ is 2.9 nm [\[11\]](#page-5-6) and penetration depth is in the range of 60–1000 nm. Provided that the particle size is carefully controlled i.e. between 2.9 and 60 nm and the dopant level is kept below a critical threshold, the addition of nanoparticles may improve the flux pinning [\[9,](#page-5-4) [12\]](#page-5-7).

The J_c of the nano-sized Fe₃O₄ added tapes is 5130 A/cm² at 77 K and 23130 A/cm² at 30 K in zero fields. Fe₃O₄ can act as an effective flux pinning center leading to enhancement of J_c in the bulk as well as the tape form $[13]$. The J_c (77 K) of the 6-nm ZnOadded sample was 46 times larger than that of the non-ZnOadded sample [\[14\]](#page-5-9). In nano-sized MgOadded tapes, the temperature and magnetic field dependence of J_c exhibited significant enhancement compared with the non-added tapes [\[15\]](#page-5-10). The nanoparticle with size closer to the coherence length was suggested to be more effective in enhancing J_c [\[13](#page-5-8)[–15\]](#page-5-10).

Most studies on enhancing J_c only used one size nanoparticles. It is interesting to investigate the effect of

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different sized magnetic nanoparticles on the transport current properties of the Bi-2223 superconductors. In this work, $Co₃O₄$ magnetic nanoparticles with an average size of 30 and 50 nm were added into $Bi_{1.6}Pb_{0.4}Sr₂Ca₂Cu₃O₁₀$ (Bi-2223) Ag-sheathed tapes. These sizes were chosen because according to a previous theoretical study [\[9\]](#page-5-4), when the magnetic nanoparticle size is larger than the coherence length but smaller than the penetration depth, enhanced flux pinning is expected. Nano- $Co₃O₄$ is superparamagnetic above 45 K, ferromagnetic between 25 and 45 K, and antiferromagnetic below the transition temperature of 27 K which is less than that of the bulk value $T_N = 33$ K [\[16\]](#page-5-11). The effect of these nanoparticles on the microstructure and the electrical transport properties of Bi-2223/Ag tapes are reported.

2 Experimental Details

Superconductor powders with nominal starting composition $(Bi_{1.6}Pb_{0.4})Sr_2Ca_2Cu_3O_y(C_{0.3}O_4)_x$ ($x = 0-0.05$ *wt* %) were synthesized using the co-precipitation method. The average particle size of $Co₃O₄$ was 30 and 50 nm. Using the results from the polycrystalline samples that showed that J_c was optimized with 0.02 wt % in 30-nm Co₃O₄added pellets and 0.01 wt % in 50-nm Co₃O₄added pellets, Ag-sheathed superconductor tapes were fabricated using the powder-in-tube method. The $(Bi₁₆Pb₀₄)Sr₂Ca₂Cu₃O₁₀$ powders were added with 0.02 wt % $Co₃O₄$ (30 nm) and 0.01 wt % $Co₃O₄$ (50 nm) nanoparticles before being packed into a Ag tube with an outer diameter of 6.03 mm and an inner diameter of 4.43 mm. The tube was drawn into a 1 mm wire by extrusion process and then pressed into tape of 0.30 mm in thickness and 1.53 mm in width. The tapes were then sintered at 845◦ C for 50 h and cut into 3 cm sections for J_c measurements.

The electrical resistance–temperature measurements were carried out using the four-point probe technique in conjunction with a CTI cryogenics closed-cycle refrigerator (model 22). The four-point probe method using the $1-\mu$ V/cm criterion was used to measure the transport critical current density (J_c) between 30 and 77 K. J_c was also measured in a magnetic field of 0–0.75 T at 77 K.

The phase was determined using the X-ray diffraction (XRD) method by employing a Bruker D8 Advance diffractometer. The size of $Co₃O₄$ was determined using a Philips transmission electron microscope (TEM) (model CM12).

3 Results and Discussion

shows the TEM micrograph of the 30 -nm Co₃O₄ particles. Figure [2](#page-2-1) shows the XRD patterns of the tapes with $x = 0$ *wt* %, $x = 0.02$ *wt* % of Co₃O₄ (30 nm), and $x = 0.01$ *wt* % of Co₃O₄ (50 nm). X-ray diffraction patterns show that the Bi-2223 is the major phase, while the Bi-2212 is the minor phase in all tapes.

Figure [3](#page-3-0) shows the microstructure of the $x = 0$ wt %, $x = 0.02$ wt % of Co₃O₄ (30 nm), and $x = 0.01$ wt % of $Co₃O₄$ (50 nm) tapes. Plate-like grains are observed in the non-added sample that is normally observed in the Bi-based system. The non-added sample showed a larger grain size than the sample with nano- $Co₃O₄$. The grain morphology of all the added samples was almost similar, except for minor variations in texture and porosity.

Figures [4](#page-3-1) and [5](#page-3-2) shows the critical current density versus Co₃O₄ content in polycrystalline Bi_{1.6}Pb_{0.4}Sr₂Ca₂Cu₃O₁₀ at 30 and 77 K. The highest J_c was observed in the $x = 0.02$ wt % $Co₃O₄$ (30 nm) sample and in the $x = 0.01$ wt % Co₃O₄ (50 nm) sample [\[17\]](#page-5-12). Figure [6](#page-4-0) shows the temperature dependence of J_c in zero magnetic fields for $x = 0$ wt %, $x = 0.02$ wt % of Co₃O₄ (30 nm), and $x = 0.01$ $x = 0.01$ wt % of Co₃O₄ (50 nm)-added tapes. Table 1 shows the J_c of $Bi_{1.6}Pb_{0.4}Sr_2Ca_2Cu_3O_{10}$ tapes with and without the addition of $Co₃O₄$ nanoparticles at 30 and 77 K in zero fields. J_c for the added samples was higher than that for the non-added tape. The highest J_c at 30 K (22800 A/cm2*)* and 77 K (6840 A/cm2*)* was shown by the sample with $Co₃O₄$ (30 nm). This result showed that $Co₃O₄$ nanoparticles enhanced the transport critical current density in the bulk and tape samples. In the intermediate temperature region (between 35 and 55 K), the J_c of the 50-nm $Co₃O₄$ -added tapes was higher than that of the 30-nm $Co₃O₄$ -added tapes. This indicated the possible different flux pinning mechanisms in this region for the two tapes.

The magnetic field dependence of the critical current density at 77 K with fields parallel and perpendicular to the tapes' surface is shown in Fig. 7 . The J_c decreased when the magnetic field was applied parallel to the surface of the tape due to the strong anisotropy of Bi-2223. The J_c of tape with $x = 0.01$ wt % of Co₃O₄ (50 nm) tape decreased faster than that with $x = 0.02$ wt % of Co₃O₄ (30 nm) tape. The nano-sized particles introduced into the granular networks are believed to act as possible pinning centers, and this improved the microstructure and strengthens the weak links between grains.

In conclusion, the effect of nano-sized $Co₃O₄$ addition on flux pinning capability of $(Bi_{1.6}Pb_{0.4})Sr_2Ca_2Cu_3O_{10}$ tape was investigated. The highest J_c in the tapes was observed in the sample with $x = 0.02$ wt % Co₃O₄ (30 nm). All the nanoparticleadded samples showed a higher J_c value compared with the non-added samples. This study showed that the magnetic nanoparticles could act as effective pinning centers leading to enhancement of J_c . The preparation **Fig. 1** TEM micrograph of $Co₃O₄$ showing an average grain size of 30 nm

method can be further optimized to improve the electrical transport properties. The flux line network and the magnetic texture can interact effectively if their characteristic scales have the same order of magnitude. Generally, when the size of a pinning center is larger than *ξ*gut smaller than *λ*, the

Fig. 2 XRD patterns of the tape sample for $x = 0$ *wt* %, $x = 0.02 \text{ wt } \%$ of Co₃O₄ (30 nm), and *x* = 0*.*01 *wt* % of $Co₃O₄$ (50 nm). *H* denotes the high- T_c phase (Bi-2223), and L denotes the low- T_c phase (Bi-2212)

critical current density will increase. The average size of the Co₃O₄ used in this study is $L = \sim 30-50$ nm, and this satisfies the requirement $\xi < L < \lambda$ for a frozen flux superconductor, which is likely an explanation for the enhanced *J*^c in the system. The full vortex magnetic energy from the

 (a) (b)

Fig. 3 SEM micrographs of the tape sample for $\mathbf{a} \times \mathbf{a} = 0$ wt %, $\mathbf{b} \times \mathbf{a} = 0.02$ wt % of Co₃O₄ (30 nm), and $\mathbf{c} \times \mathbf{a} = 0.01$ wt % of Co₃O₄ (50 nm)

1 $T = 77 K$ 30 nm 50 nm 0.8 J_{c} (Mcm^{2})
 $_{\circ}$
 $_{\circ}$ 0.4 0.2 Þ 0 0 0.01 0.02 0.03 0.04 0.05 $Co₃O₄$ (wt. %)

Fig. 4 Critical current density (J_c) versus $Co₃O₄$ content in Bi_{1.6}Pb_{0.4}Sr₂Ca₂Cu₃O₁₀ pellets at 30 K

Fig. 5 Critical current density (J_c) versus Co₃O₄ content in Bi_{1.6}Pb_{0.4}Sr₂Ca₂Cu₃O₁₀ pellets at 77 K

Table 1 Critical current density (J_c) of Bi_{1.6}Pb_{0.4}Sr₂Ca₂Cu₃O₁₀ tapes with and without the addition of the Co3O4 nanoparticles at 30 and 77 K in zero fields

Fig. 7 *J*^c dependence on magnetic field applied both parallel and perpendicular to the tape surface, at 77 K, for the tape samples for $x = 0$ wt %, $x = 0.02$ wt % of Co₃O₄ (30 nm), and *x* = 0.01 wt % of $Co₃O₄$ (50 nm)

addition of $Co₃O₄$ magnetic nanoparticles played an important role in enhancing J_c in Bi_{1.6}Pb_{0.4}Sr₂Ca₂Cu₃O₁₀/Ag apes.

Acknowledgments This research was supported by the Ministry of Education, Malaysia, under grant no. FRGS/2/2013/SG02/UKM/01/1 and the Universiti Kebangsaan Malaysia under grant no. UKM-DIP-2012-032.

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