

THE EFFECT OF MAGNETIC NANOPARTICLE ADDITION ON THE SUPERCONDUCTING PROPERTIES OF $\text{Bi}_{1.6}\text{Pb}_{0.4}\text{Sr}_2\text{Ca}_2\text{Cu}_3\text{O}_8$ SUPERCONDUCTORS

H. Baqiah¹, S.A.Halim¹, M.I.Adam¹, S.K.Chen¹, S.S.H.Ravandi¹, M.A.M.Faisal¹,
M.M.Kamarulzaman¹ and M.Hanif²

¹*Superconductors and Thin film laboratory, Department of Physics, Faculty of Science, University Putra Malaysia*

²*Department of Chemistry, Faculty of Science, University Putra Malaysia
43400 UPM Serdang, Selangor*

ABSTRACT

Sm_2O_3 nanoparticle was added to Bi-2223 superconductor prepared by solid state reaction technique with intermediate grinding. A stoichiometric precursor of $x=0.00-0.05$ Sm_2O_3 nanoparticle is systematically added to the well balanced $\text{Bi}_{1.6}\text{Pb}_{0.4}\text{Sr}_2\text{Ca}_2\text{Cu}_3\text{O}_8$ in order to trace the effect of nanoparticles addition to the system. Microstructure, resistive transitions, phase volume, and cell parameters were hence investigated. Addition of Sm_2O_3 nanoparticle is found to slowly decrease the Bi-2223 phase volume and the resistive transitions for $x=0-0.02$ samples whereas accelerated formation of the Bi-2212 phase is detected for further additions. Changes in superconducting properties of Sm-added Bi-2223 system were discussed and the findings were further compared with available literature

INTRODUCTION

Since the discovery of BSCCO superconductors with $T_c = 105$ K by Meada et al. [1], extensive research in this system still continues. BSCCO system has layer structure with three identical phases, particularly, $\text{Bi}_2\text{Sr}_2\text{CuO}_6$, $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_8$, and $\text{Bi}_{1.6}\text{Pb}_{0.4}\text{Sr}_2\text{Ca}_2\text{Cu}_3\text{O}_8$. Chemical doping and addition are known to affect and control the superconducting and physical properties of layered structure superconductors. The superconducting properties of Bi2223 system are highly related to its hole concentration, pinning ability and its phases structure [2-5]. Hence, research interest in BSCCO system is mainly concentrated in the enhancement of overall property of the Bi2223 phase in order to prepare the materials for practical applications. C. Terzioglu et al. reported the doping of Sm_2O_3 in Bi2223. The critical temperature T_c and microstructure were found to degrade with this rare earth doping in this system [2]. The addition of Nd [3] and doping of Pr [6] however found to enhance the flux pinning property of the Bi2223 system.

The addition of element in the form of nanoparticle may cause different effects in the microstructure and pinning properties of high temperature superconductors. It has been reported that the addition of ZrO_2 [7], SiC [8], MgO [9], Al_2O_3 [10, 11], Cr_2O_3 [12] in

Bi2223 enhance the flux pinning of this system. Yahya et al. [13] has mixed Bi2223 powder thoroughly with rod-like $\gamma\text{Fe}_2\text{O}_3$ nanoparticle before the final step sintering process. Increase in the critical current J_c is noticed despite the fact that $\gamma\text{Fe}_2\text{O}_3$ inhibit the growth of Bi2223 phase [13] and decreasing the critical temperature T_c .

In this work, we report on the effect of Sm_2O_3 nanoparticle addition before the final steps of heat treatment of $\text{Bi}_{1.6}\text{Pb}_{0.4}\text{Sr}_2\text{Ca}_2\text{Cu}_3\text{O}_8$ superconductor. Resistive-superconducting transition, phase structure and morphology of samples were discussed in details.

METHODOLOGY

Samples with chemical composition $\text{Bi}_{1.6}\text{Pb}_{0.4}\text{Sr}_2\text{Ca}_2\text{Cu}_3\text{O}_8$ were prepared by solid state reaction technique. High purity powders of Bi_2O_3 , PbO , SrCO_3 , CaCO_3 and CuO were weighed using 0.0001g accuracy balance, mixed and ground using mortar and pestle. The powders were calcined and ground two times at 800°C and 830°C for 24 hours each. The calcined powder then was pressed into pellets with 13mm diameter and 2mm thickness using 5 tones pressure then sintered at 850°C for 120 hours with one intermediate grinding and pressing at the same force. Fine powder of $(\text{Bi}_{1.6}\text{Pb}_{0.4}\text{Sr}_2\text{Ca}_2\text{Cu}_3\text{O}_{10})_{1-x}(\text{nano Sm}_2\text{O}_3)_x$, with $x=00, 0.005, 0.01, 0.02, 0.03$ and 0.05 , were mixed with Sm_2O_3 nanoparticles, stirred for the same time including unadded sample in order to insure identical physical properties. The mixture powder were then pressed at 5 tons and sintered for 30 hr at 850°C .

XRD measurement was carried out by Philips X'Pert Pro diffractometer. Phase analysis of XRD data were done by X'pert high score software. Lattice parameters were calculated and refined from XRD data using software based nonlinear least square method. Resistivity measurements were carried out by four point probe technique with silver paint contact and used to determine $T_{c(\text{onset})}$, where the resistivity start to decrease and $T_{c(R=0)}$, where the materials lose resistivity completely. Scanning electron microscopy micrographs were done by using a JEOL 6400 SEM with 4500 magnification

RESULTS AND DISCUSSION

XRD pattern of samples with $x=00-0.05$, Sm_2O_3 nanoparticle addition are shown in figure 1. The volume fractions of high and low phase were determined using (1) and (2) equations respectively.

$$V_{\text{Bi2223}} = \frac{\sum I_{\text{Bi2223}}}{\sum I_{\text{Bi2223}} + \sum I_{\text{Bi2212}}} \times 100\% \quad (1)$$

$$V_{Bi2212} = \frac{\sum I_{Bi2212}}{\sum I_{Bi2212} + \sum I_{Bi2223}} \times 100\% \quad (2)$$

I_{Bi2223} and I_{Bi2212} are the intensity of peaks related to high T_c (Bi2223) and low T_c (Bi2212) phases respectively. V_{Bi2223} and V_{Bi2212} are the volume fractions of Bi2223 and Bi2212 phases respectively.

The initial addition of Sm_2O_3 nanoparticle causes decreasing of peaks related to high T_c phase (Bi2223) and those related to low T_c phase (Bi2212) phase to increase slightly. This results in decreasing of the volume fraction of Bi2223 phase and that of Bi2212 to decrease slightly as shown in Figure 2. Some Small peaks of low T_c phase which are absent in the pure are observed and few small peaks related to high phase are disappeared by increasing Sm_2O_3 nanoparticles concentration. The strong intensity of peaks related to Bi2223 phase in pure remains at $x=0.05$ sample.

Lattice parameters of high T_c phase (Bi2223), calculated by software based on nonlinear least square method, are presented in Table 1. It is shown that the **a**-axis increases whereas the **c** decreases slightly with increasing of Sm_2O_3 nanoparticles concentrations. Incorporation of oxygen atoms in the Bi2223 unit cell causes an increase in the a-axis while decreasing the c-axis parameter [2, 14, 16].

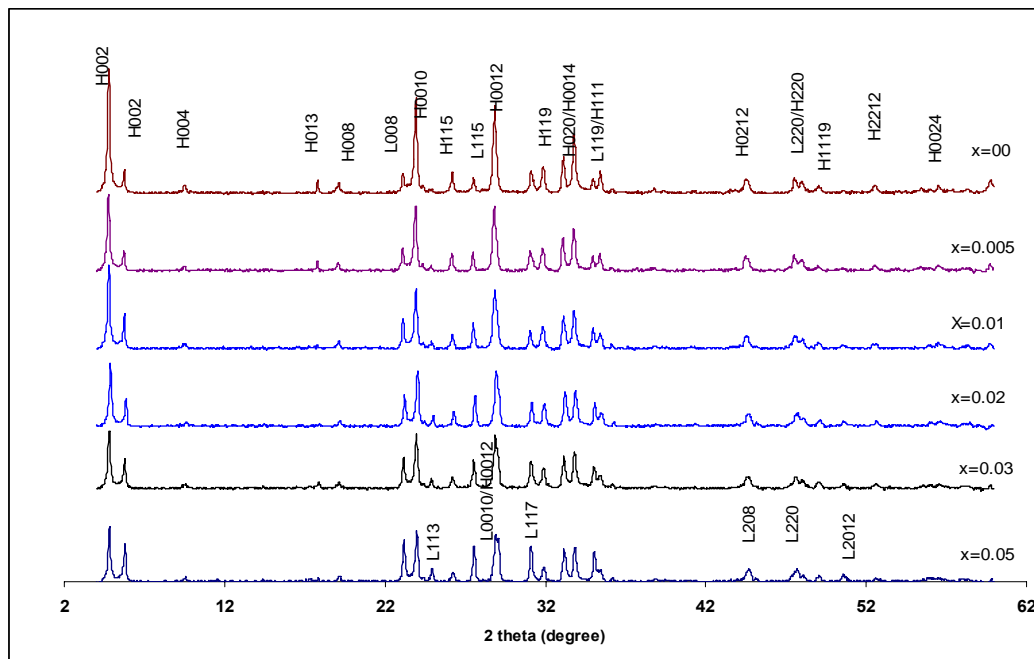


Figure 1: XRD patterns of the $(Bi_{1.6}Pb_{0.4}Sr_2Ca_2Cu_3O_{10})_{1-x}(nanoSm_2O_3)_x$ samples with $x=00-0.05$. Peaks of the high- T_c (Bi2223) and low- T_c (Bi2212) phases are indexed by H and L, respectively.

The addition of Sm_2O_3 nanoparticle causes XRD peaks to shift to large angle. Figure 3 show the shift of H(002) peak belonging to Bi2223 phase. This result may be related to the decrease of grain size. Maximum shift was found for sample with $x=0.02$ as obtained from SEM micrograph calculation to have smallest average grain size.

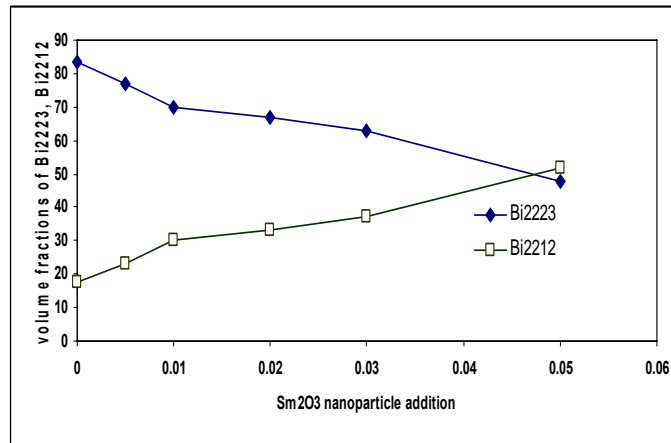


Figure 2: Volume fraction of Bi2223 and Bi2212 phases volume as a function of Sm_2O_3 nanoparticles addition.

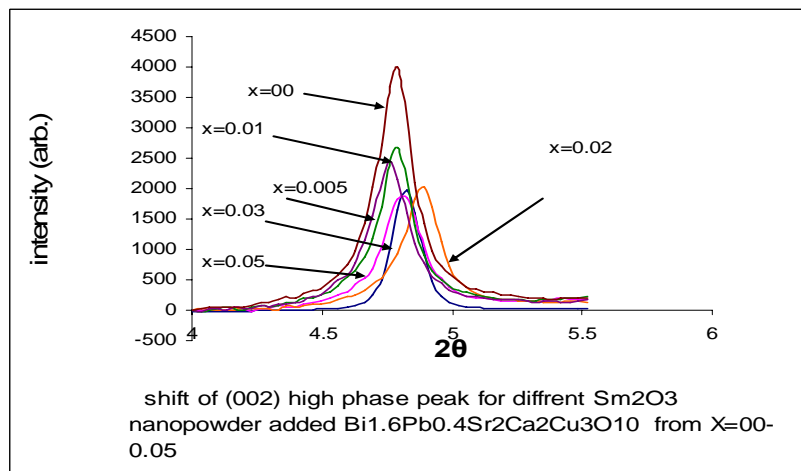


Figure 3: Shift of H (002), Bi2223 phase peak, for $(\text{Bi}_{1.6}\text{Pb}_{0.4}\text{Sr}_2\text{Ca}_2\text{Cu}_3\text{O}_{10})_{1-x}(\text{nanoSm}_2\text{O}_3)_x$ samples with $x=0.00$ - 0.05

The measurements of electrical resistance were done using four point probe technique in the temperature ranges between 60 and 280 K. Normalized resistivity versus temperature were plotted in Figure 4. All samples exhibit metallic behavior, which means that resistivity decreases with decreasing temperature. In granular

superconductors, the $T_{c(R=0)}$ can be associated with the flow of supercurrent in the intergrain region whereas the $T_{c\text{ onset}}$ associated with intrinsic property of grains. Table 1 summarize the value of critical temperature $T_{c(R=0)}$, $T_{c\text{ onset}}$, transition width ΔT_c for samples with $x=00-0.05$. The increase of Sm_2O_3 nanoparticle concentrations decrease the $T_{c(R=0)}$ of Bi2223 system. The result could relate to the suppression of superconductivity by the increase of Sm ion concentration [14].

Table 1: Summary of critical temperature $T_{c(R=0)}$, $T_{c\text{ onset}}$, transition width ΔT_c and cell parameters of Bi2223 phase for different Sm_2O_3 addition.

Sm_2O_3 Addition	$T_{c(R=0)}$(K)	$T_{c\text{ onset}}$(K)	ΔT_c(K)	$a(\text{Å})$	$c(\text{Å})$
00	102	119	17	5.4113	37.1327
0.005	98	117	19	5.132	37.1322
0.01	94	117	23	5.417	37.1296
0.02	93	117	24	5.4313	37.09
0.03	82	111	29	5.4345	37.0868
0.05	78	109	36	5.4143	37.086

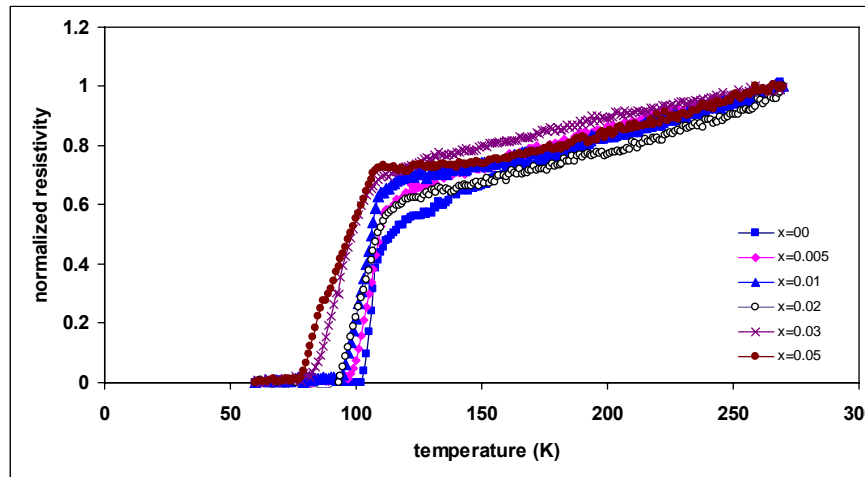


Figure 4: Normalized resistivity-temperature dependence of $(\text{Bi}_{1.6}\text{Pb}_{0.4}\text{Sr}_2\text{Ca}_2\text{Cu}_3\text{O}_{10})_{1-x}(\text{nanoSm}_2\text{O}_3)_x$ samples with $x=00-0.05$ measured from 60K to 280 K.

The critical temperature $T_{c(\text{onset})}$ of the sample with $x=0.005-0.02$ is seen at 117 K. Although the Sm_2O_3 content have increased the weak link and Bi2212 phase in the system, circulation of supercurrent may found its way through grains of Bi2223 phase. The $T_{c(\text{onset})}$ is further decreased in the samples with $x=0.03$ an $x=0.05$. This result could be related to structure change in Bi2223 phase that remain up to $x=0.05$ sample as extracted from XRD result analysis.

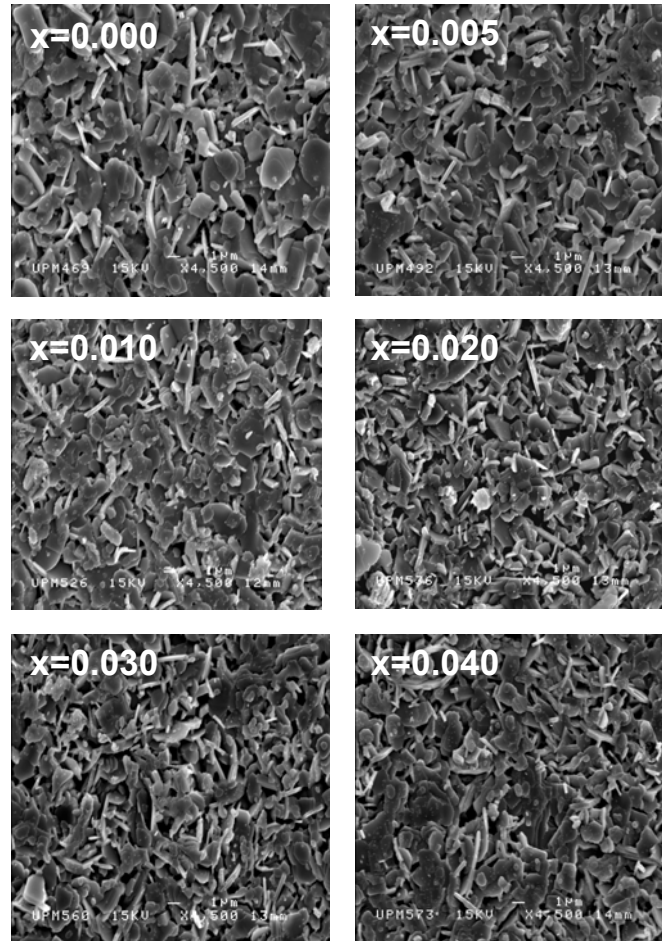


Figure 5: SEM micrograph for $(\text{Bi}_{1.6}\text{Pb}_{0.4}\text{Sr}_2\text{Ca}_2\text{Cu}_3\text{O}_{10})_{1-x}(\text{nanoSm}_2\text{O}_3)_x$ samples with $x=00-0.05$.

Surface morphology micrographs taken by SEM for all samples are shown in Figure 5. The free- Sm_2O_3 sample surface is dominated by large platelet grain that oriented in the C axis direction. XRD result for sample confirms the presence of (00L) peak along the 2θ . The size of these platelet grains seems to decrease in the samples with $x=0.005-0.02$. Increases of the porosity with more random distribution of grains are seen at samples with $x \geq 0.01$. The estimated average grain size from SEM picture are 2.4, 2, 1.8, 1.3, 1.4 and 1.5 μm for samples with $x=00-0.05$ respectively. A slight increase in

grain size is noticed for $x=0.03-0.05$ samples, this indicates accelerated favorability of Bi2212 grain phase in these samples. Addition of rare earth element is known to promote the Bi2212 phase formation [15, 16].

CONCLUSION

Addition of Sm_2O_3 nanoparticle to $\text{Bi}_{1.6}\text{Pb}_{0.4}\text{Sr}_2\text{Ca}_2\text{Cu}_3\text{O}_{10}$ powder before the final stage sintering was investigated. Bi2223 phase was found to decrease by increasing addition and transform slowly to Bi2212 phase. The **c**-axis of Bi2223 decreases slightly and **a**-axis increases by increasing the Sm_2O_3 addition. Critical temperature decreases by increasing Sm_2O_3 addition. Superconducting transition curve has small double transition at $x=0.03$ and $x=0.05$. The addition of Sm_2O_3 nanoparticle reduce the grain growth of Bi2223 and the grain size become smaller up to $x=0.02$.

ACKNOWLEDGMENT

This research has been supported by the Malaysian Ministry of Science, Technology and Innovation under grant number 5523132 and the saga grant 5486510

REFERENCES

- [1]. H. Maeda, Y. Tanaka, M. Fukutomi and T. Asano (1988). A New High- T_c Oxide Superconductor without a Rare Earth Element. *Jpn. J. Appl. Phys.* part 2 **27**, L209.
- [2]. Cabir Terzioglu, Mustafa Yilmazlar, Ozgur Ozturk and Ekrem Yanmaz (2005). Structural and physical properties of Sm-doped $\text{Bi}_{1.6}\text{Pb}_{0.4}\text{Sr}_2\text{Ca}_{2-x}\text{Sm}_x\text{Cu}_3\text{O}_y$ superconductors. *Physica C*, **423**, 119–126
- [3]. R.P. Aloysius, P. Guruswamy and U. Syamaprasad, (2005). Enhanced critical current density in (Bi,Pb)-2223 superconductor by Nd addition in low percentages. *Physica C*, **426–431**, 556–562
- [4]. Huseyin Sozeri, Nader Ghazanfari, Husnu Ozkan and Ahmet Kilic, (2007). Enhancement in the high- T_c phase of BSCCO superconductors by Nb addition. *Supercond. Sci. Technol.*, **20**, 522–528
- [5]. S.A. Halim, S.B. Mohamed, H. Azhan, S.A. Khawaldeh and H.A.A. Sidek (1999). Effect of barium doping in Bi–Pb–Sr–Ca–Cu–O ceramics superconductors. *Physica C*, **312**, 78–84
- [6]. MHPu, W. H Song, B. Zhao, X. C. Wu1, T. Hu, Y P Sun1 and J.J.Du (2001). Enhanced flux pinning in (Bi, Pb)-2223/Ag tapes by slight Pr substitution, *Supercond. Sci. Technol.*, **14**, 305–310
- [7]. Z.Y. Jia, H. Tang, Z.Q. Yang, Y.T. Xing, Y.Z. Wang and G.W. Qiao (2000). Effects of nano- ZrO_2 particles on the superconductivity of Pb-doped BSCCO. *Physica C*, **337**, 130–132
- [8]. Y.C. Guo, Y. Tanaka, T. Kuroda, S.X. Dou and Z.Q. Yang (1999). Addition of nanometer SiC in the silver-sheathed Bi2223 superconducting tapes. *Physica C*, **311**, 65–74

- [9]. W. Wei, J. Schwartz, K.C. Goretta, U. Balachandran and A. Bhargava (1998). Effects of nanosize MgO additions to bulk Bi Sr CaCu O. *Physica C*, **298**, 279–288
- [10]. M. Annabi, A. M_chirgui, F. Ben Azzouz, M. Zouaoui and M. Ben Salem (2004). Addition of nanometer Al₂O₃ during the final processing of (Bi,Pb)-2223 superconductors. *Physica C*, **405**, 25–33
- [11]. A. Ghattas, M. Annabi, M. Zouaoui, F. Ben Azzouz and M. Ben Salem (2008). Flux pinning by Al-based nano particles embedded in polycrystalline (Bi,Pb)-2223 superconductors. *Physica C*, **468**, 31–38
- [12]. Kong Wei and R. Abd-Shukor (2007). Superconducting and Transport Properties of (Bi-Pb)-Sr-Ca-Cu-O with Nano-Cr₂O₃ Additions. *Journal of Electronic Materials*, **36**, No. 12, 2007
- [13]. S. Y. Yahya, M. H. Jumali, C. H. Lee and R. Abd-Shukor (2004). Effects of γ -Fe₂O₃ on the transport critical current density of (Bi_{1.6}Pb_{0.4})Sr₂Ca₂Cu₃O₁₀ superconductors. *Journal of Material Science*, **39**, 7125–712
- [14]. O. Ozturk, M. Akdogan, H. Aydin, M. Yilmazlar, C. Terzioglu and I. Belenli (2007). Substitution of Sm at Ca site in Bi_{1.6}Pb_{0.4}Sr₂Ca_{2-x}Sm_xCu₃O_y superconductors. *Physica B*, **399**, 94–100
- [15]. Berdan Ozkurt, Ahmet Ekicibil, M. Ali Aksan, Bekir Ozçelik, M. Eyüphan Yakıncı, Kerim Kiymaç (2007). Structural and Physical Properties of Nd Substituted Bismuth Cuprates Bi_{1.7} Pb_{0.3-x}Nd_xSr₂Ca₃Cu₄O_{12+y}. *J. Low Temp Phys.*, **149**, 105–118
- [16]. G. Ilonca, A.V. Popa, T.R. Yangb, I.Gr. Deaca, C. Lunga, R. Stiufluca and G. Stiufluca (2001). Effects of rare earth ion substitution for Ca in (Bi,Pb):2223 Superconductors. *International Journal of Inorganic Materials*, **3**, 769–772