

Effect of Co_3O_4 Nanoparticles Addition on (Bi,Pb)-2223 Superconductor

A. N. Jannah, R. Abd-Shukor, and H. Abdullah

Abstract—The effect of nano Co_3O_4 addition on the superconducting properties of (Bi, Pb)-2223 system was studied. The samples were prepared by the acetate coprecipitation method. The Co_3O_4 with different sizes (10-30 nm and 30-50 nm) from $x=0.00$ to 0.05 was added to $\text{Bi}_{1.6}\text{Pb}_{0.4}\text{Sr}_2\text{Ca}_2\text{Cu}_3\text{O}_y(\text{Co}_3\text{O}_4)_x$. Phase analysis by XRD method, microstructural examination by SEM and dc electrical resistivity by four point probe method were done to characterize the samples. The X-ray diffraction patterns of all the samples indicated the majority Bi-2223 phase along with minor Bi-2212 and Bi-2201 phases. The volume fraction was estimated from the intensities of Bi-2223, Bi-2212 and Bi-2201 phase. The sample with $x=0.01$ wt% of the added Co_3O_4 (10-30 nm size) showed the highest volume fraction of Bi-2223 phase (72%) and the highest superconducting transition temperature, T_c (~102 K). The non-added sample showed the highest T_c (~103 K) compared to added samples with nano Co_3O_4 (30-50 nm size) added samples. Both the onset critical temperature $T_c(\text{onset})$ and zero electrical resistivity temperature $T_c(R=0)$ were in the range of 103-115 \pm 1K and 91-103 \pm 1K respectively for samples with added Co_3O_4 (10-30 nm and 30-50 nm).

Keywords—Bi(Pb)-Sr-Ca-Cu-O superconductor, coprecipitation, nano Co_3O_4 , transition temperature T_c .

I. INTRODUCTION

Bi-Sr-Ca-Cu-O-based high temperature superconductors, either in bulk form such as wires and tapes or as components of more complex composites, have shown some potentials for commercialization. There have been many studies to improve the superconducting properties, in particular the critical current density (J_c). Many research groups have investigated the effects of adding different elements or nano-particles in order to improve J_c and T_c [1]-[10]. These studies helped in the understanding of the mechanism of superconductivity in high T_c cuprates. The investigation of the influence of nano particles on superconductors is very important from the viewpoint of practical application. In this work, Co_3O_4 nano-particles was added to Bi(Pb)-Sr-Ca-Cu-O as magnetic impurities in the superconductor system. In this work, the structure, phase formation and electrical properties of Bi(Pb)-Sr-Ca-Cu-O with nano Co_3O_4 addition were investigated. The samples were

characterized by SEM, XRD and electrical resistance measurement.

II. EXPERIMENTAL DETAILS

Samples of $\text{Bi}_{1.6}\text{Pb}_{0.4}\text{Sr}_2\text{Ca}_2\text{Cu}_3\text{O}_y(\text{Co}_3\text{O}_4)_x$ with 0.01 wt%, 0.02wt%, 0.03wt%, 0.04wt% and 0.05 wt% were prepared by the acetate coprecipitation method. Two sets of precursor powder were prepared; one set was added with nano Co_3O_4 (10-30 nm size) and the other set was added with nano Co_3O_4 (30-50 nm size). The non-added sample was used as a reference. The mixture were thoroughly mixed and ground and calcined at 845 $^\circ\text{C}$, with the heating rate of 2 $^\circ\text{C}/\text{min}$ and cooling rate of 2 $^\circ\text{C}/\text{min}$ for 24 h. Then, the samples were reground and pressed into pellets and heated at 850 $^\circ\text{C}$ in an air for 48 h. Morphological and structural analyses of the samples were performed by an SEM and X-ray diffractometer, respectively. The transport properties were measured by the standard four point probe method.

III. RESULTS AND DISCUSSION

Fig. 1(a) and (b) present the electrical resistance versus temperature in zero magnetic field of the superconducting samples with pure Bi(Pb)-Sr-Ca-Cu-O and nano Co_3O_4 addition with different sizes (10-30 nm and 30-50 nm) at $x=0.01\text{wt}\%$, 0.02wt%, 0.03wt%, 0.04wt% 0 and 0.05wt%. The dc electrical resistivity measurements show a well defined metallic behavior and superconducting transitions for all the compositions. All the samples show the zero electrical resistivity $T_c(R=0)$ within the range 70-103 \pm 1K, while onset temperatures $T_c(\text{onset})$ are 83-115 \pm 1K (Table I). The highest superconducting transition temperature, T_c is shown by the sample with nano Co_3O_4 (10-30 nm size) addition at $x=0.01\text{wt}\%$, with $T_c\sim 102$ K and the normal-state resistivity lower than the pure sample. The high T_c for the $x=0.01$ wt% sample may be due to homogeneity in the sample. Excessive Co_3O_4 degraded the superconductivity of Bi2223, which can affect the transport properties in this type of material [1].

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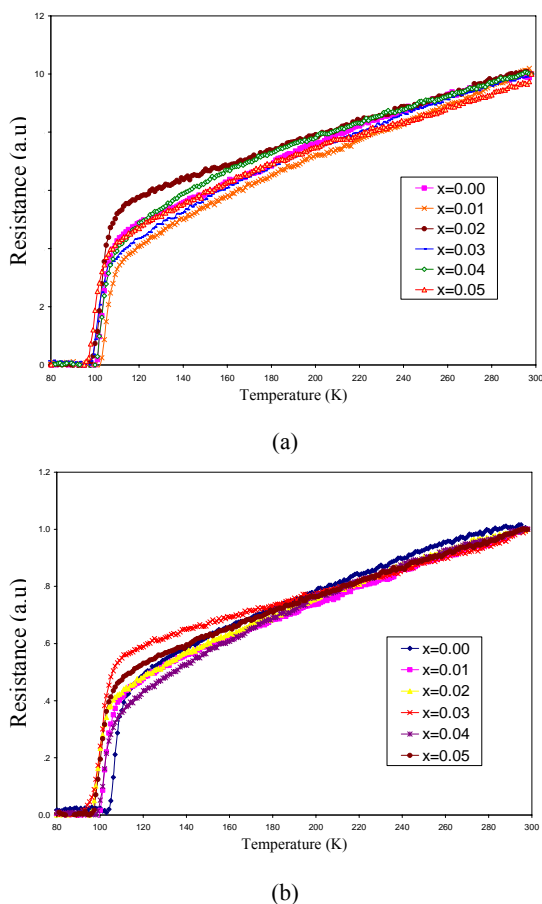


Fig. 1 Variation of normalized resistivity as a function of temperature for sample added with (a) Co_3O_4 (10-30 nm size) and (b) Co_3O_4 (30-50 nm size)

The lower zero resistance transition temperature with increasing Co_3O_4 content could be interpreted as a result of the suppression of superconductivity by the Co_3O_4 . It was observed that Co_3O_4 added samples showed a broader transition, which indicates weak links between superconducting grains and lower percentage of Bi2223 phase compared to the non-added sample. The structural phase transformation is supported by XRD measurements.

Fig. 2 shows the X-ray diffraction patterns for non-added and Co_3O_4 added samples. In this work, all the peaks of Bi-(2223), Bi-(2212) and Bi-(2201) phases have been used for the estimation of the volume fraction of the phases. The volume fraction of the Bi-(2223) phase relative to the Bi-(2212) phase and Bi-(2201) phase could be estimated from the intensities according to

$$Bi(2223) = \frac{\sum I(2223)}{\sum I(2223) + \sum I(2212) + \sum I(2201)} \times 100 \quad (1)$$

$$Bi(2212) = \frac{\sum I(2212)}{\sum I(2223) + \sum I(2212) + \sum I(2201)} \times 100 \quad (2)$$

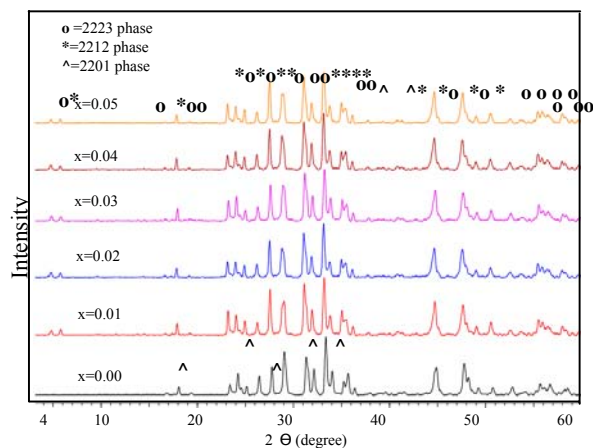
$$Bi(2201) = \frac{\sum I(2201)}{\sum I(2223) + \sum I(2212) + \sum I(2201)} \times 100 \quad (3)$$

where I is the intensity of the number of peaks corresponding to the respective phase present.

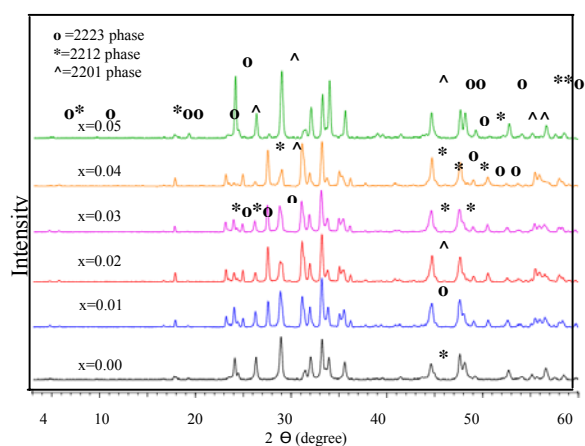
Fig. 2 shows the X-ray diffraction patterns of all samples, where the o, * and ^ indicate the peaks due to the Bi-(2223), Bi-(2212) and Bi-(2201) phases, respectively. The majority of the peaks for all the samples correspond to the Bi-(2223) and the minority belongs to Bi-(2212) and Bi-(2201) phases. All samples with nano Co_3O_4 addition contain the 2212 phase. Several peaks were shifted toward either higher or lower value of 2θ angle as the concentration of Co_3O_4 increase. Some of the added samples have peaks belonging to the 2201 phase. However the crystallographic structure remains in the tetragonal form.

Tables I and II show the T_{conset} , T_{czero} and volume fraction for non-added Bi(Pb)-Sr-Ca-Cu-O and nano Co_3O_4 addition at $x=0.01\text{wt}\%$ 0.02wt% 0.03wt% 0.04wt% and 0.05wt%. From the table, sample with nano Co_3O_4 addition that exhibited the highest T_c showed a slight decrease of the Bi 2212 phases and an enhancement of the 2223 phase volume fraction [7]. It can be seen from Table I that the Bi-(2223) phase in sample with $x=0.01\text{wt}\%$ addition of Co_3O_4 (10-30 nm size) has the highest percentage of Bi-2223 phase and reaches maximum value of 72%. With further increase in Co_3O_4 the percentage of the 2223 phase decreased and the percentage of 2212 phase increased. A decrease of 2223 phase volume fraction with an increase of 2212 phase was clearly seen for both Co_3O_4 addition (10-30 nm and 30-50 nm sizes). This indicates that a small amount of nano Co_3O_4 addition plays a certain role in the phase formation of the BSCCO system and may enhance the low T_c phase (2212) formation and destabilize the high T_c phase (2223) [5]. It showed also that $x=0.01\text{wt}\%$ of nano Co_3O_4 can be the appropriate amount of magnetic impurities to enhance the critical temperature and the phase formation of 2223 phase. The size of the nano Co_3O_4 also played a significant role in decreasing the T_c in addition to the effect of secondary phase particles.

The SEM micrographs of the cross sectional view of the samples for Bi(Pb)-Sr-Ca-Cu-O and Co_3O_4 added samples are shown in Fig 3. Superconducting grains are seen to be connected with each other strongly and the surface morphology of the sample comprises of platelets and layered grains with uniform and homogenous microstructure. The platelet-type features are also found in nano Co_3O_4 added samples but the size of the grains was larger than the pure one. For sample with $x=0.01\text{wt}\%$, a compact platelets were observed and this may be able to establish connectivity between grains. Therefore, the increase in T_c with an the appropriate nano Co_3O_4 addition in the sample is associated with the enhancement in 2223 phase formation and the improved links between grains.



(a)



(b)

Fig. 2 X-ray diffraction pattern for sample added with (a) Co_3O_4 (10-30 nm size) and (b) Co_3O_4 (30-50 nm size)

TABLE I

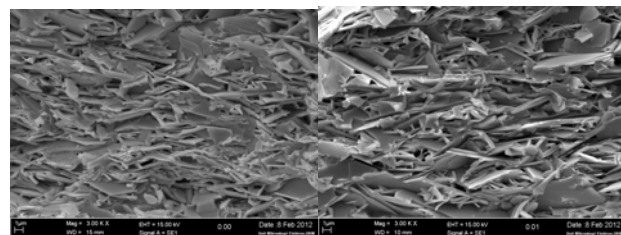
THE RESULTS OF XRD AND LATTICE AND RESISTIVITY MEASUREMENTS OF SAMPLES ADDED WITH Co_3O_4 (10-30 NM SIZE)

x (wt%)	$T_{C\text{onset}}$ ($\pm 1\text{K}$)	$T_{C(R=0)}$ ($\pm 1\text{K}$)	Volume fraction(%)		
			2201	2212	2223
0.00	112	100	7	9	16
0.01	115	102	7	21	72
0.02	110	101	0	39	61
0.03	108	97	0	39	61
0.04	113	100	3	34	63
0.05	111	95	3	46	51

TABLE II

LATTICE AND RESISTIVITY MEASUREMENTS OF SAMPLES ADDED WITH Co_3O_4 (30-50 NM SIZE)

x (wt%)	$T_{C\text{onset}}$ ($\pm 1\text{K}$)	$T_{C(R=0)}$ ($\pm 1\text{K}$)	Volume fraction(%)		
			2201	2212	2223
0.00	110	103	0	29	71
0.01	106	99	0	29	71
0.02	105	95	3	23	74
0.03	103	91	0	33	67
0.04	105	98	3	44	63
0.05	105	96	26	27	47



(a)

(b)

Fig. 3 SEM surface images for the samples with a) pure Bi(Pb)-Sr-Ca-Cu-O and b) nano Co_3O_4 addition(10-30 nm size) at $x=0.01\text{wt}\%$

IV. CONCLUSION

In conclusion, nano Co_3O_4 addition to Bi(Pb)-Sr-Ca-Cu-O can improve the superconducting properties of the samples. The sample with 0.01wt% Co_3O_4 (10-30nm size) addition had the highest T_c of 102 K as against 100 K for the non-added samples. The same sample also exhibited the highest volume fraction of the 2223 phase. As the amount of Co content increased the amount of secondary phases also increased which obstructed the flow of supercurrent in the system. The size of the nano Co_3O_4 also played a significant role in decreasing the T_c in addition to the effected of secondary phase. Even though magnetic impurities suppress superconductivity, these results showed that an appropriate amount of nano Co_3O_4 can improve the superconducting properties of the Bi-based samples, while excessive Co_3O_4 addition led to the degradation of the superconductivity of Bi2223.

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