# PREPARATION OF Bi(Pb)-Sr-Ca-Cu-O SUPERCONDUCTOR WITH NANO Co<sub>3</sub>O<sub>4</sub> ADDITION

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#### Abstract

The effect of nano  $Co_3O_4$  addition (0-0.05 wt%) on the Bi(Pb)-Sr-Ca-Cu-O (Bi-2223) precursor powder has been investigated in terms of X-ray diffraction, scanning electron microscopy and dc electrical resistivity. The samples were prepared by the conventional nitrate coprecipitation operation. The X-ray diffraction patterns of all the samples indicated the presence of large amount of Bi-2223 phase along with minor amount of Bi-2212 and Bi-2201 phases. The volume fraction was estimated from the intensities of Bi-2223, Bi-2212 and Bi-2201 phases. The sample with x=0.01 wt% of added  $Co_3O_4$  shows the highest volume fraction of Bi-2223 phase formed (71%) and the highest superconducting transition temperature,  $T_C$  (~102 K). Both the onset critical  $T_c$ (onset) and zero electrical resistivity  $T_c(R=0)$  were determined from the dc electrical resistivity which the temperature range 83-115 ±1K and 70-102 ±1K respectively.

### **1** 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 impressive progress and great promise for commercialization and reached a level where they are ready for use as components in various devices such as power cables, high-field magnets, magnetic resonance imaging system, and magnetic sensors. They have several advantages properties that make them attractive when considered for engineering application. In order to enhance their transport capabilities, many research groups have investigated the effects of adding of different elements or nano-particles [1,2,3,4,5,6,7]. The investigation of the influence of nanometer particles on the superconductors is very important from the viewpoint of practical application. Here, nano-particles of  $Co_3O_4$  was added to the samples and it act as the magnetic impurities in the superconductor system [5]. In this work, the structure, phase formation and electrical properties of the pure Bi(Pb)-Sr-Ca-Cu-O sample and the mixture of Bi(Pb)-Sr-Ca-Cu-O with nano  $Co_3O_4$  powder sample will be investigated. These samples will be characterized by SEM, XRD and resistance measurement.

### **2** Experimental Details

The precursor powders were prepared by the conventional nitrate coprecipitation operation with nominal chemical compositions of  $Bi_{1.6}Pb_{0.4}Sr_2Ca_2Cu_3O_y$ . Two kinds of precursor powder were prepared; one was a pure Bi(Pb)-Sr-Ca-Cu-O and the other was a mixture of Bi(Pb)-Sr-Ca-Cu-O with nano Co<sub>3</sub>O<sub>4</sub> powder with 0.01 wt%, 0.02wt%, 0.03wt%, 0.04wt% and 0.05 wt% of Co<sub>3</sub>O<sub>4</sub> nanoparticles with 10-30 nm size was added to the samples. The Co non-added powder (0.00wt%) was used as a reference when comparing the effects of Co<sub>3</sub>O<sub>4</sub> on the phase formation and the microstructure. The mixture were thoroughly mixed and ground for 2 hours. All of the precursor undergone for calcinations and sintering process, with the heating rate of  $2^{0}$ C/min and cooling rate of  $2^{0}$ C/min. Morphological and structural analyses of the samples were performed by an SEM and X-ray diffractometer. The transport properties were measured by the standard four point probe method.

## **3** Results and Discussions

Figure 1 presents the curve of electrical resistance versus temperature in zero magnetic field of the superconducting samples with pure Bi(Pb)-Sr-Ca-Cu-O and nano Co<sub>3</sub>O<sub>4</sub> addition at x=0.01wt%, 0.02wt%, 0.03wt%, 0.04wt% 0 and 0.05wt%. The dc electrical resistivity measurements show a well defined metallic behaviour and superconducting transitions for all the compositions. T<sub>C</sub> transition curve for pure Bi(Pb)-Sr-Ca-Cu-O as comparison. All the samples show the zero electrical resistivity  $T_c(R=0)$  within the range 70-102 ±1K, while onset temperatures  $T_c$ (onset) are 83-115 ±1K, and these are tabulated in Table 1. The highest superconducting transition temperature,  $T_c$  is given by sample with nano Co<sub>3</sub>O<sub>4</sub> addition at x=0.01wt%, with  $T_C \sim 102$  K and the normal-state resistivity lower than the pure sample. This sample showed an improvement compared to the pure sample, and it is believed that the decrease in normal-state resistivity is related to the structural phase transformation (from 2212 to 2223) and weak-links improvement between the grains in the samples. Samples with x=0.02wt%, x=0.03wt%, x=0.04wt% and 0.05wt% showed decrease in critical temperature. That was due to the decrease in the high  $T_c$  phase and increase in the formation of low  $T_c$ phase [2]. However the resistance dropped with single-step features in all samples implying that these samples consisting mainly of 2223 phase as the Co<sub>3</sub>O<sub>4</sub> concentration was increased. Hence, x=0.01wt% nano Co<sub>3</sub>O<sub>4</sub> can be the appropriate amount at low concentration of addition that exhibits the highest  $T_c$ . The decreasing of critical temperature value for samples with Co<sub>3</sub>O<sub>4</sub> addition up to 0.05wt% indicate that the strength of coupling between the grains of mixed phases has been decreased resulting in weaker-link.



Figure 1. Resistivity as a function of temperature for samples.

Figure 2 shows the indexed X-ray diffraction patterns for pure 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 fraction amount of the Bi-(2223) phase relative to the Bi-(2212) phase and Bi-(2201) phase could be estimated from the intensities according to

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

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

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

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



2θ (degree)

Figure 2. X-ray diffraction pattern for all samples.

Figure shows 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. Majority of the lines for all the samples correspond mainly to Bi-(2223) and minority belongs to Bi-(2212) and Bi-(2201) phases. All samples with nano  $Co_3O_4$  addition contain low  $T_c$  peaks which correspond to the 2212 phase. Several peaks have been shifted toward either higher or lower value of 20 angle as the concentration of  $Co_3O_4$  increase. Only samples with x=0.00wt% and x=0.05wt% have peaks belonging to 2201 phase. However the crystallographic structure remains in the tetragonal form. The percentage of the 2223, 2212 and 2201 phases calculated using Eqs. (1), (2) and (3). Table 1 shows the  $T_{CZero}$ ,  $T_C$ onset and volume fraction for pure Bi(Pb)-Sr-Ca-Cu-O and nano  $Co_3O_4$  addition at x=0.01wt% 0.02wt% 0.03wt%0.04wt% and 0.05wt%. From the table, sample with nano  $Co_3O_4$  addition that exhibits the highest  $T_C$  show a slightly decrease of the intermediate phases and an enhancement of the 2223 phase's volume fraction than the pure sample.

X	T <sub>Conset</sub>	$T_{Czero}$	Volume fraction(%)		
(wt%)	(±1K)	(±1K)	2201	2212	2223
0.00	112	100	7	23	60
0.01	115	102	0	29	71
0.02	110	97	0	34	66
0.03	108	97	0	37	63
0.04	83	75	0	42	58
0.05	86	70	9	35	56

**Table 1.** *T<sub>C</sub>*zero, *T<sub>C</sub>onset* and volume fraction for all samples.

It can be seen from Table 1 that the Bi-(2223) phase in sample with x=0.01wt% has the highest percentage of Bi-2223 phase and reaches maximum value of 71%, that is maximum volume formation compared to other samples. And with further increasing Co<sub>3</sub>O<sub>4</sub> the percentage of the 2223 phase decreases and the percentage of 2212 phase increases. A decrease of 2223 phase volume fraction with an increase of 2212 phase was clearly seen due to nano Co<sub>3</sub>O<sub>4</sub> addition at x=0.02wt% to x=0.05wt% . This indicates that small amount of nano Co<sub>3</sub>O<sub>4</sub> 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 at x=0.01wt% of nano Co<sub>3</sub>O<sub>4</sub> can be the appropriate amount of magnetic impurities to enhance the critical temperature and the phase formation of 2223 phase of Bi(Pb)-Sr-Ca-Cu-O superconductor.



(a)

(b)



(c)

(d)



Figure 3. SEM surface images for the samples with a) pure Bi(Pb)-Sr-Ca-Cu-O and nano  $Co_3O_4$  addition at b) x=0.01wt% % c) x=0.02wt% % d) x=0.03wt% e) x=0.04wt% and f) 0.05wt%.

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

### Conclusion

In conclusion, nano  $\text{Co}_3\text{O}_4$  addition to Bi(Pb)-Sr-Ca-Cu-O can improve the superconducting properties of the samples. The sample with 0.01wt% addition had the highest  $T_C$ . The same sample also exhibited the highest volume fraction of the 2223 phase (71%). Eventhough the magnetic impurities tend to suppress the superconductivity, these results show that an appropriate amount of nano  $\text{Co}_3\text{O}_4$  can improve the superconducting properties of the Bibased samples, while excessive  $\text{Co}_3\text{O}_4$  addition leads to degradation of the superconductivity of Bi2223.

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