

Research Article

# Improvement of Superconducting Properties of (Bi, Pb) -2223 added with Nano-Particles SnO<sub>2</sub>

Bushra A. Aljurani<sup>†</sup> and Mohammed N. ALdulaimi<sup>\*†</sup>

<sup>†</sup>College of Science, University of Baghdad, Iraq

Accepted 28 Feb 2015, Available online 22 April 2015, Vol.5, No.2 (April 2015)

## Abstract

Solid state reaction method was used to prepare samples of (SnO<sub>2</sub>)<sub>x</sub> Bi<sub>1.7</sub> Pb<sub>0.3</sub> Sr<sub>2</sub> Ca<sub>2</sub> Cu<sub>3</sub> O<sub>10+δ</sub> with x=0,0.1,0.2, 0.4,0.6 and 0.8 wt%. Two sets of sample with different particle size of SnO<sub>2</sub>, have been prepared. The first one was prepared by addition SnO<sub>2</sub> normal particle (120-200nm), the second was prepared by added SnO<sub>2</sub> nano- particle (20-40nm). X-ray analysis and electrical resistivity measurements were carried out to study the performance of these samples. Phase examination by XRD indicated that SnO<sub>2</sub> nano particle enhanced the (Bi,Pb)-2223 phase formation by increasing volume fraction of high T<sub>c</sub> phase up to x=0.4 wt%. On the other hand, high concentrations of SnO<sub>2</sub> nano-particles retarded the phase formations by increasing of low T<sub>c</sub> phase. The crystal structure for all superconductor samples was orthorhombic structure. Also it has been observed that sample prepared by added nano particle of SnO<sub>2</sub> display a sharp superconducting transition and a higher-T<sub>c</sub> than that of the sample prepared by added SnO<sub>2</sub> normal- particle

**Keywords:** Bi-based superconductors, SnO<sub>2</sub> addition, nano particle

## 1. Introduction

Considerable efforts have been made in the synthesis, processing, and characterization of Bi-Sr-Ca-Cu-O superconducting ceramics because these materials have a higher T<sub>c</sub> and good chemical durability (resistance to moisture), and also they contain no rare earth elements (Bilgili O. Y. Selamet *et al*, 2012). But it is difficult to prepare single high T<sub>c</sub> phase Bi-2223. The low T<sub>c</sub> phase Bi-2212 was frequently observed as a major impurity phase because of its greater thermodynamic stability. It was found that the partial substitution of Bi by Pb is quite an effective solution to solve this problem (Tarascon, J.M *et al*, 1998). The substitution of Pb into Bi-Sr-Ca-Cu-O system increases the volume fraction of (Bi,Pb)-2223 phase by forming partially melted liquid phases, which is important for improving both T<sub>c</sub> and the transport critical current density (Hatano, T. *et al*, 1998). The other limitation for using Bi-2223 phase for applications is the intergrain weak links and weak flux pinning capability (Jia, Z.Y. *et al*, 2000). Recently, the addition of nano-oxides to (Bi,Pb)-2223 superconducting phase plays an important role for enhancing the flux pinning and critical current density (Ghattas, A. *et al*, 2008; Guilmeau E *et al*, 2003).

Ghattas *et al*. prepared the superconducting system with the addition of nano-size Al<sub>2</sub>O<sub>3</sub> (40 nm) with 0

and 0.2 wt% concentration on the polycrystalline (Bi,Pb)-2223. Samples were synthesized in air by solid state reaction. Nanometre Al<sub>2</sub>O<sub>3</sub> particles were added during the final sintering cycle of a multi-step preparation process. They studied the onset temperature, effective energy U, critical current density J<sub>c</sub> behaviour in applied magnetic field and volume pinning force density which were enhanced by the addition of 0.2 wt% Al<sub>2</sub>O<sub>3</sub>.

Baqiah. *et al*. prepared Bi-2223 superconductor system by solid state reaction technique with intermediate grinding by added with nano particle of Sm<sub>2</sub>O<sub>3</sub> with x=0.00-0.05 wt%. They found that addition of Sm<sub>2</sub>O<sub>3</sub> nano particle slowly decreased the volume fraction of Bi-2223 phase and accelerated the formation of the Bi-2212 phase for x= 0-0.02 samples. Changes in superconducting properties of Sm-added Bi-2223 system were discussed.

Jousoh *et al*. synthesized phase formation of Bi<sub>1.6</sub>Pb<sub>0.4</sub>Sr<sub>2</sub>Ca<sub>2</sub>Cu<sub>3</sub>O<sub>x</sub> (Bi-2223) with nano- Al<sub>2</sub>O<sub>3</sub> addition processed via co-precipitation method. This method was introduced to produce ultra fine and homogeneous powders. XRD data showed the enhancement of 2223 phase as sintering time increased. Furthermore, the single step transition observed by resistivity measurement was the evidence to show the dominance high -T<sub>c</sub> phase. The results showed the increment around 1K to 5K of critical transition temperature when resistivity is zero.

Jannah *et al*. studied the effect of different nano sized CO<sub>3</sub>O<sub>4</sub> (10, 30, and 50nm) addition on the

\*Corresponding author: Mohammed N. ALdulaimi

Bi<sub>1.6</sub>Pb<sub>0.4</sub>Sr<sub>2</sub>Ca<sub>2</sub>Cu<sub>3</sub>O<sub>10</sub>(Co<sub>3</sub>O<sub>4</sub>)<sub>x</sub> Superconductor with x=0-0.05 wt. %. The samples were prepared by the conventional solid state reaction method. They found that samples with x=0.01 wt% CO<sub>3</sub>O<sub>4</sub> (10 nm) showed the highest T<sub>c</sub> at 102 K. The highest J<sub>c</sub> was observed in the x=0.03 wt %CO<sub>3</sub>O<sub>4</sub>(10 nm) and x= 0.02 wt % CO<sub>3</sub>O<sub>4</sub> (30 nm ). The X-ray diffraction patterns of all the samples indicated the majority Bi-2223 phase along with minorBi-2212 and Bi-2201 phases. The sample with x=0.01 wt% of the added Co<sub>3</sub>O<sub>4</sub>(10-30 nm size)showed the highest volume fraction of Bi-2223 phase (72%) and the highest superconducting transition temperature.

In this paper, the effect of SnO<sub>2</sub> nanoparticles addition on superconducting properties of (Bi,Pb)-2223 phase was studied. For this study ,superconducting samples of (SnO<sub>2</sub>)<sub>x</sub> Bi<sub>1.7</sub> Pb<sub>0.3</sub>Sr<sub>2</sub> Ca<sub>2</sub>Cu<sub>3</sub> O<sub>10+δ</sub>with different particle size and different concentration x=0,0.1,0.2,0.4,0.6 and 0.8 wt % were prepared using solid state reaction technique. X-ray Diffraction (XRD) and electrical resistivity are the basics tools for characterizing these samples.

### 2. Experimental Part

(SnO<sub>2</sub>)<sub>x</sub> Bi<sub>1.7</sub> Pb<sub>0.3</sub>Sr<sub>2</sub> Ca<sub>2</sub>Cu<sub>3</sub> O<sub>10+δ</sub> with x=0,0.1,0.2,0.4,0.6 and 0.8 wt %were prepared by conventional solid-state reaction method. Appropriate amounts of Bi<sub>2</sub>(CO<sub>3</sub>)<sub>3</sub>(99.9%), Pb<sub>3</sub>O<sub>4</sub>(99.9%), SrCO<sub>3</sub>(99.9%), CaO(99.9%), CuO (99.9%) and SnO<sub>2</sub> (99.9%) powders were used as starting materials. .The powder of precursor was mixed together by using agate mortar. The mixture homogenization takes place by adding a sufficient quantity of 2-propanol to form a past during the process of grinding from about (1 h). In the second step , the materials were grounded to a fine powder and then calcined in air at 800 °C for (24)h, the mixture was then pressed into pellets (1.3 cm) in diameter and (0.2) cm thick, using hydraulic type (SPECAC), under pressure of 0.7, GPa.. The pellets were sintered in air at 830 °C for 140 h.

Four probe dc method at temperature range (77-300) K was used to measure the resistivity (ρ) and to determine the critical temperature (T<sub>c</sub>).

### 3. Results and Discussion

XRF techniques in this study are adopted to determine the real elemental content of the prepared samples. The XRF analysis of (SnO<sub>2</sub>)<sub>x</sub> Bi<sub>1.7</sub> Pb<sub>0.3</sub>Sr<sub>2</sub> Ca<sub>2</sub>Cu<sub>3</sub> O<sub>10+δ</sub>with x=0.0 and x=0.1 wt% samples that were sintered at 830 °C for 140h are shown in Fig.( 1,2).It should be mentioned that there are no peaks corresponding to the Sn based compound that were detected in the (XRF) patterns displayed in Fig(1).

However, the XRF analysis for the bulk sample with nano SnO<sub>2</sub> with x=0.1 wt%, shows the existence of the Sn element as shown in Fig.( 2). The result demonstrated that there is no unwanted element in the sample and no contaminations are observed in the

spectra, which verify the high purity of the investigated samples.

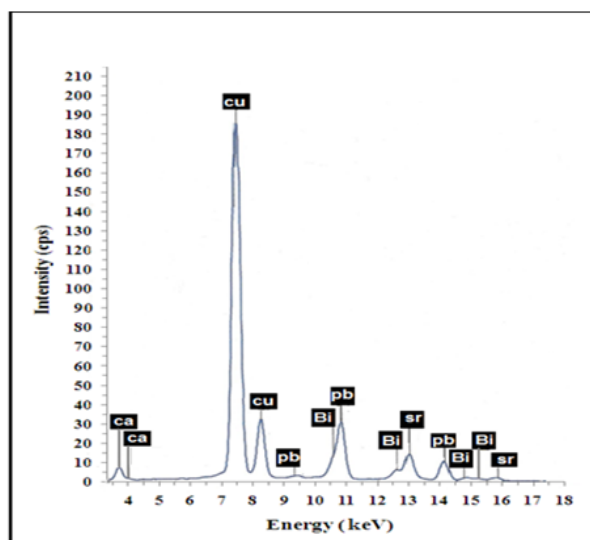


Fig.1 XRF patterns of (SnO<sub>2</sub>)<sub>x</sub> Bi<sub>1.7</sub> Pb<sub>0.3</sub>Sr<sub>2</sub> Ca<sub>2</sub>Cu<sub>3</sub> O<sub>10+δ</sub>samples with x=0.0 wt%

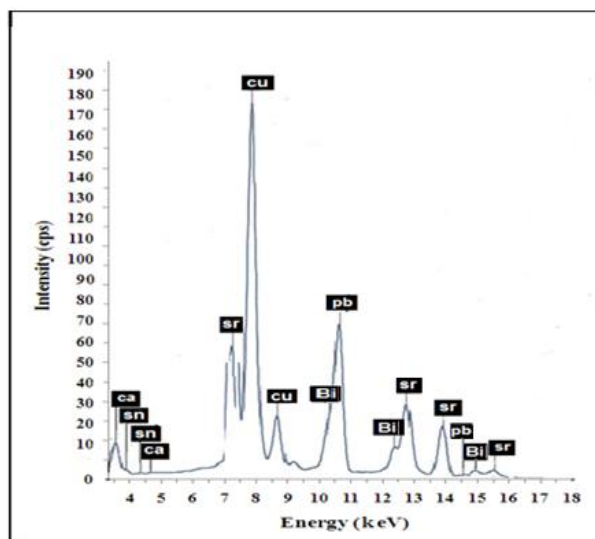


Fig.2 XRF patterns of (SnO<sub>2</sub>)<sub>x</sub> Bi<sub>1.7</sub> Pb<sub>0.3</sub>Sr<sub>2</sub> Ca<sub>2</sub>Cu<sub>3</sub> O<sub>10+δ</sub>samples with x=0.1 wt%

Room temperature XRD patterns of samples(SnO<sub>2</sub>)<sub>x</sub> Bi<sub>1.7</sub> Pb<sub>0.3</sub>Sr<sub>2</sub> Ca<sub>2</sub>Cu<sub>3</sub> O<sub>10+δ</sub> with x=0.1,0.2,0.4,and 0.6wt% (of SnO<sub>2</sub>) normal and nano particle addition are shown in fig (3.3-4) . The volume fractions of high and low phase were determined using (1) and (2) equation (Baqiah, H. et al, 2009)respectively.

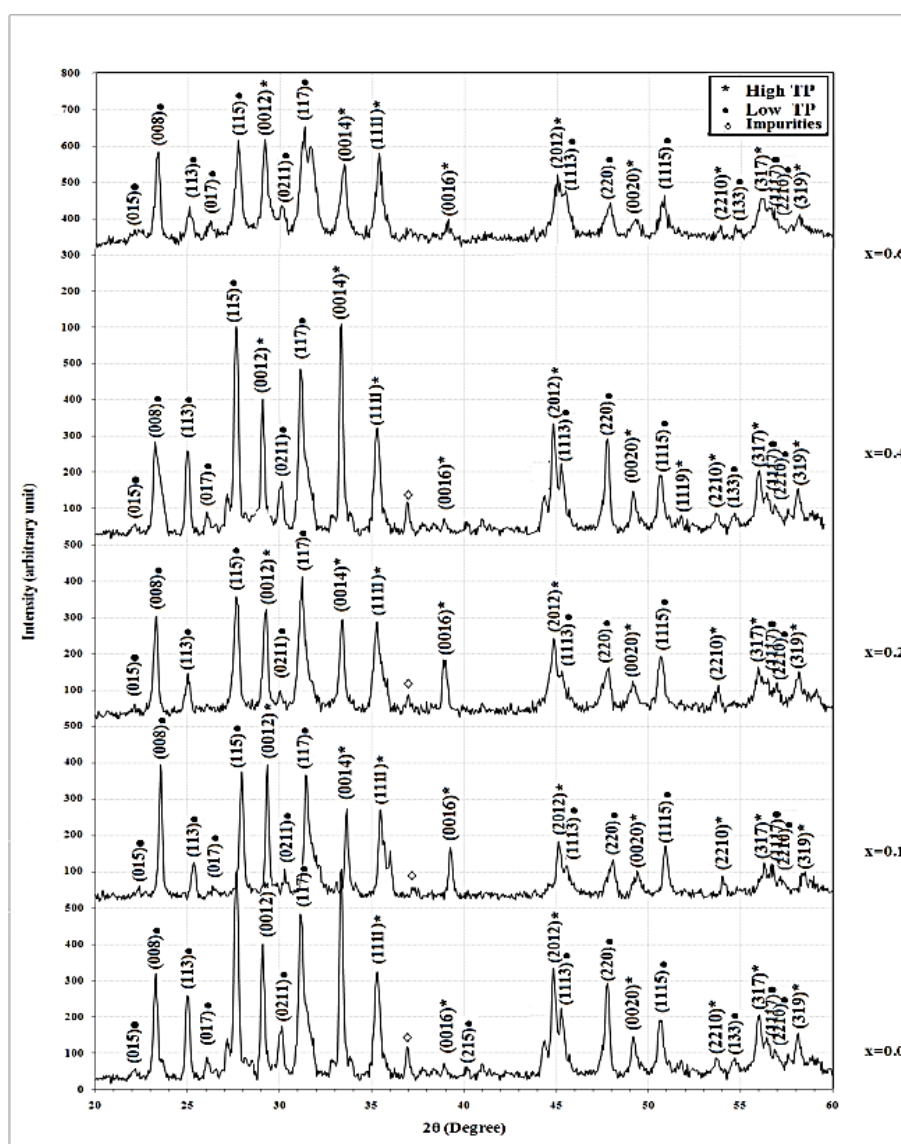
$$\text{Bi}(2223)\% = \frac{\sum I(2223)}{\sum I(2223) + \sum I(2212)} \times 100 \tag{1}$$

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

It is found from the XRD patterns when the concentration of SnO<sub>2</sub> increase the volume fraction of

**Table 1** Variation in lattice parameters, c/a, density of unit cell ρ<sub>m</sub> and volume fraction of Bi-3222 and Bi-2122, for (SnO<sub>2</sub>)<sub>x</sub> (Bi<sub>1.7</sub> Pb<sub>0.3</sub> Sr<sub>2</sub> Ca<sub>2</sub> Cu<sub>3</sub> O<sub>10+δ</sub>) system for different of particle size and different nominal composition

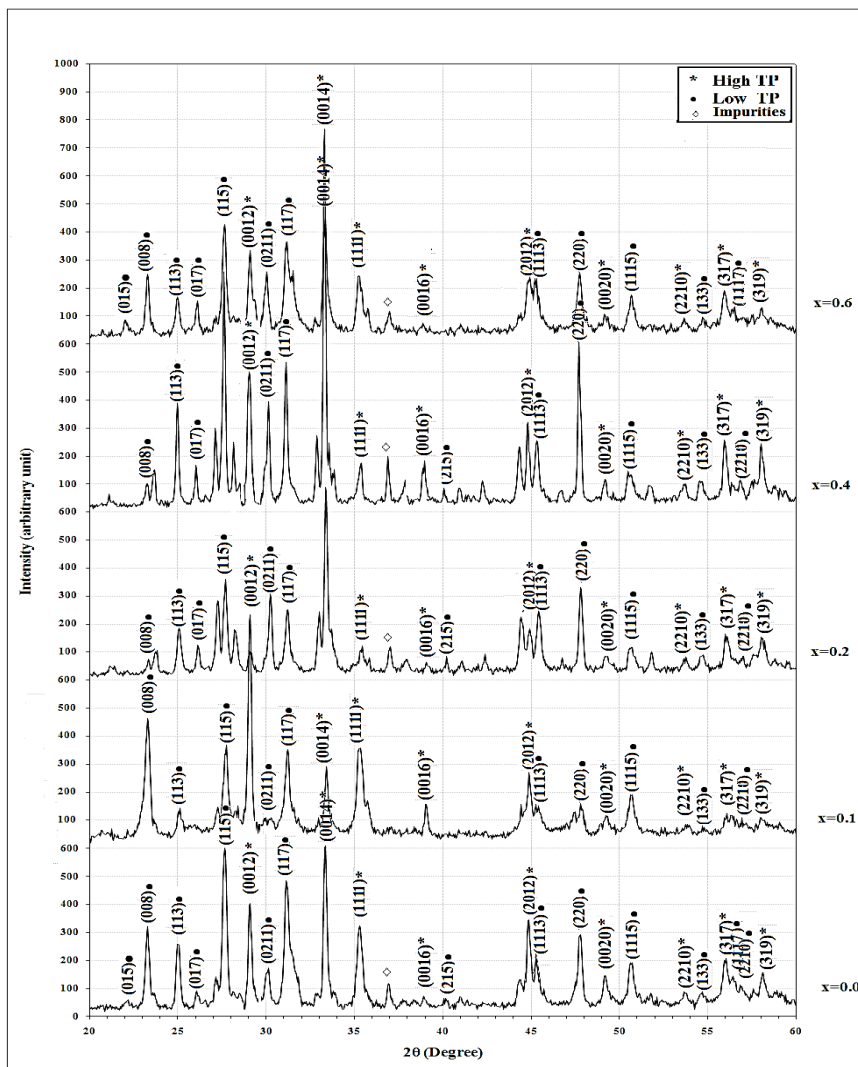
Preparation method	Xwt %	a Å	b Å	c Å	c/a	V	ρ <sub>m</sub> (g/cm <sup>3</sup> )	volume fraction Bi-2223	volume fraction Bi-2122
Normal	0.0	4.982	5281	37.236	7.474	979.677	1.613	69.3	30.7
	0.1	5.389	5.430	37.378	6.936	1093.765	1.573	70.28	29.72
	0.2	5.385	5.448	37.056	6.881	1087.13	1.587	71.55	28.45
	0.4	5.541	5.365	37.192	6.712	1105.624	1.468	75.3	24.7
	0.6	5.302	5.601	37.123	7.001	1102.42	1.470	68.9	31.1
Nano	0.1	5.417	5.324	37.320	6.889	1076.313	1.579	79.8	20.2
	0.2	5.462	5.321	37.253	6.82	1082.695	1.589	80.4	19.6
	0.4	5.085	5.343	37.372	7.349	1015.366	1.591	84.8	15.2
	0.6	5.77	5.421	37.132	6.435	1161.458	1.444	82.5	17.5



**Fig.3** X-ray diffraction patterns for (SnO<sub>2</sub>)<sub>x</sub> (Bi<sub>1.7</sub> Pb<sub>0.3</sub> Sr<sub>2</sub> Ca<sub>2</sub> Cu<sub>3</sub> O<sub>10+δ</sub>) sample. sintered at 830°C for 140h added with normal SnO<sub>2</sub>

high T<sub>c</sub> phase Bi-2223 increase up to x= 0.4 wt% for both normal and nano SnO<sub>2</sub> addition , while the volume fraction of low T<sub>c</sub> phase Bi-2122 is decreasing up to x=0.4 as shown in Figs.(5,6) Similar result were

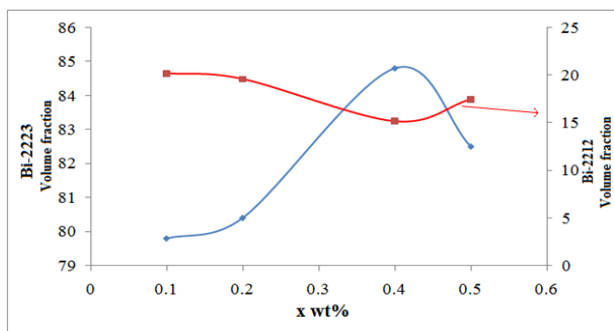
pointed out by Abou- Aly *et al.* It is showed be mentioned that the relative intensity of diffraction peaks varies slightly from samples prepared in different x for both partials size. Figs. (3,4) show an



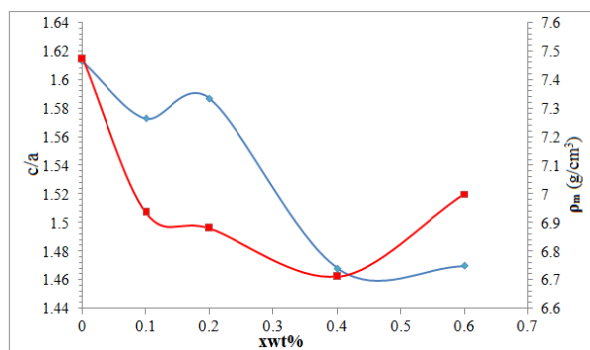
**Fig.4** X-ray diffraction patterns for (SnO<sub>2</sub>)<sub>x</sub>(Bi<sub>1.7</sub>Pb<sub>0.3</sub>Sr<sub>2</sub>Ca<sub>2</sub>Cu<sub>3</sub>O<sub>10+δ</sub>)sample sintered at 830°C for 140h added with nano SnO<sub>2</sub>

increasing of the intensities for all peaks up to x=0.4, and these peaks were lower with increase of SnO<sub>2</sub>concentration. On the other side a present of H (0012) at 2θ =28.8 for all concentration of SnO<sub>2</sub> is very important to prove the superconductivity for Bi-compound.

Lattice parameters of high T<sub>c</sub> phase (Bi-2223), are listed in Table (3-1). It is shown that the c/a decreases slightly with increasing of SnO<sub>2</sub> for normal and nano particles concentration. The variation of lattice parameters a,b,c effect on the volume of the unit cell and then causes variation of the density.

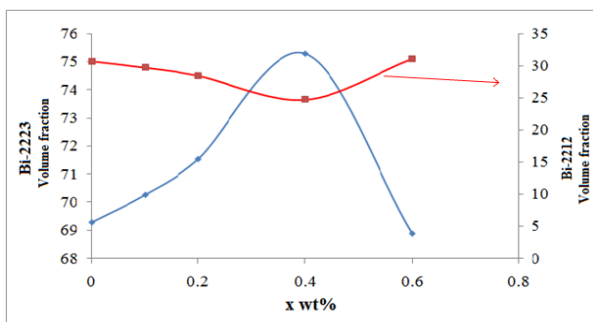


**Fig.5** Variation of Volume fraction for Bi-2223 and Volume fraction for Bi-2122 as a function of SnO<sub>2</sub> nano concentration of (SnO<sub>2</sub>)<sub>x</sub>(Bi<sub>1.7</sub>Pb<sub>0.3</sub>Sr<sub>2</sub>Ca<sub>2</sub>Cu<sub>3</sub>O<sub>10+δ</sub>) samples

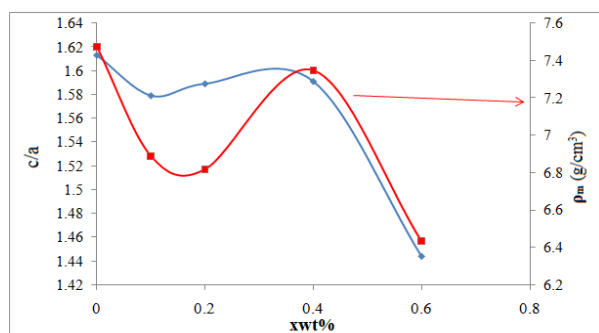


**Fig.7** Variation c/a ratio and density of unit cell (ρ<sub>m</sub>) as a function of SnO<sub>2</sub> norml of (SnO<sub>2</sub>)<sub>x</sub>Concentration Bi<sub>1.7</sub>Pb<sub>0.3</sub>Sr<sub>2</sub>Ca<sub>2</sub>Cu<sub>3</sub>O<sub>10+δ</sub>

The ratio  $c/a$  and the density  $\rho_m$  of the unit cell for all the prepared sample have also been calculated as shown in Table (1) and Figs. (7,8). An increases of the density could be attribute to the reduction of the porosity and vacancies in the system.



**Fig.6** Variation of Volume fraction for Bi-2223 and Volume fraction for Bi-2122 as a function of SnO<sub>2</sub> normal concentration of (SnO<sub>2</sub>)<sub>x</sub> (Bi<sub>1.7</sub>Pb<sub>0.3</sub>Sr<sub>2</sub>Ca<sub>2</sub>Cu<sub>3</sub>O<sub>10+δ</sub>) samples



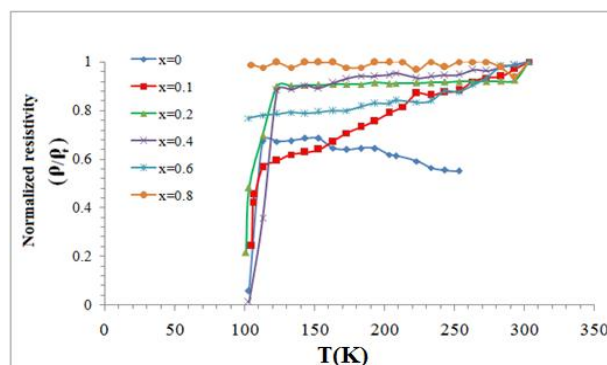
**Fig.8** Variation  $c/a$  ratio and density of unit cell ( $\rho_m$ ), as a function of SnO<sub>2</sub> nano of (SnO<sub>2</sub>)<sub>x</sub> Concentration Bi<sub>1.7</sub>Pb<sub>0.3</sub>Sr<sub>2</sub>Ca<sub>2</sub>Cu<sub>3</sub>O<sub>10+δ</sub>

The variation of the electrical normalized resistivity with temperature for normal and nano particles of (SnO<sub>2</sub>)<sub>x</sub> Bi<sub>1.7</sub> Pb<sub>0.3</sub>Sr<sub>2</sub> Ca<sub>2</sub>Cu<sub>3</sub> O<sub>10+δ</sub> with different concentration (x) of SnO<sub>2</sub> are shown in Figs.(9-10). All samples show a metallic behaviour at high temperature followed by superconducting transition as the temperature is lowered, except for the samples with  $x=0.8$  wt% for both normal and with nano (SnO<sub>2</sub>) which showed a semiconductor behaviour. For the sample with nano SnO<sub>2</sub>, the magnitude of the normal state resistivity decreases up to  $x=0.4$  wt%, above which the normal state resistivity increases. Also, the transition to  $T_c$  seems to be a one-step transition (nearly sharp), indicating the formation of nearly (Bi,Pb)-2223 single phase. This is in agreement with the XRD results which showed an increase in the volume fraction of the prepared sample up to  $x=0.4$ . This means that the addition of low concentration of nano SnO<sub>2</sub> enhances the volume fraction of (Bi,Pb)-2223 phase by reducing the impurity phases and then enhance  $T_c$  as it is clear from Table (2). For higher concentrations above  $x=0.4$  wt%, the grains of (SnO<sub>2</sub>)

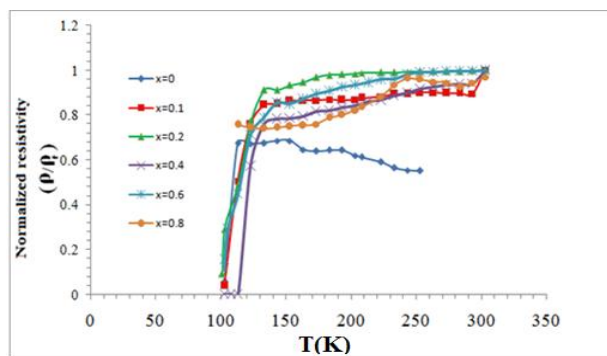
act as a barrier and may hinder the grain growth, and then destroy the superconducting phase. Or the decrease of  $T_c$  can be attributed to the formation of amorphous grain boundaries due to the created liquid phases during the sintering process. Barik *et al.* reported that the higher concentration of nano-particles supports the cooper pair- breaking mechanism. A similar result was observed by Terzioglu *et al.* in studying the effect of Sm<sub>2</sub>O<sub>3</sub> nano particles on (Bi,pb)-2223 phase.

**Table 2** Transition temperature ( $T_c$ ) for different percent weight (x wt%) Of (SnO<sub>2</sub>)<sub>x</sub> Bi<sub>1.7</sub> Pb<sub>0.3</sub> Sr<sub>2</sub> Ca<sub>2</sub> Cu<sub>3</sub> O<sub>10+δ</sub> different particle size of of (SnO<sub>2</sub>) addition

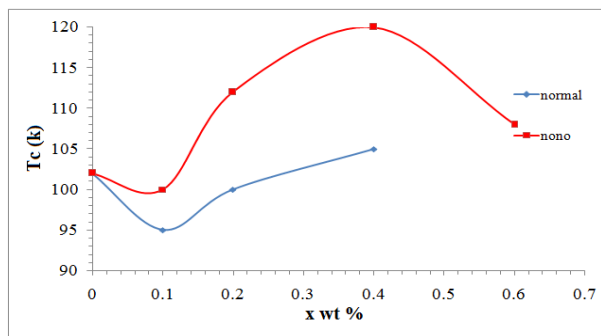
Preparation method	Xwt%	Tc
Normal Size of SnO <sub>2</sub> addition	0.1	95
	0.2	100
	0.4	105
	0.6	Semi
	0.8	Semi
Nano partical size of SsnO <sub>2</sub> addition	0.1	103
	0.2	112
	0.4	120
	0.6	108
	0.8	semi



**Fig.9** Normalized resistivity ( $\rho/\rho_t$ ) as a function of a temperature for (SnO<sub>2</sub>)<sub>x</sub> Bi<sub>1.7</sub> Pb<sub>0.3</sub> Sr<sub>2</sub> Ca<sub>2</sub> Cu<sub>3</sub> O<sub>10+δ</sub> samples for different concentrations of normal (SnO<sub>2</sub>)



**Fig.10** Normalized resistivity ( $\rho/\rho_t$ ) as a function of a temperature for (SnO<sub>2</sub>)<sub>x</sub> Bi<sub>1.7</sub> Pb<sub>0.3</sub> Sr<sub>2</sub> Ca<sub>2</sub> Cu<sub>3</sub> O<sub>10+δ</sub> samples for different concentrations of nano (SnO<sub>2</sub>)



**Fig. 11** Variation of the critical temperature  $T_c$  as a function of SnO<sub>2</sub> concentration  $x$ (wt%) of  $(\text{SnO}_2)_x (\text{Bi}_{1.7} \text{Pb}_{0.3} \text{Sr}_2 \text{Ca}_2 \text{Cu}_3 \text{O}_{10+\delta})$  for different particle size

Fig. (11) shows the variation of  $T_c$  with SnO<sub>2</sub> normal and nano- particles addition concentrations ( $x$ ) for  $(\text{SnO}_2)_x \text{Bi}_{1.7} \text{Pb}_{0.3} \text{Sr}_2 \text{Ca}_2 \text{Cu}_3 \text{O}_{10+\delta}$ .  $T_c$  increases until  $x=0.4$ wt% for both particle size and then it decreases with further increase in SnO<sub>2</sub> nano-particle concentrations while, in normal particles of SnO<sub>2</sub> degrades superconducting phase and behavior as semiconductor.

### Conclusion

It was found that the addition of SnO<sub>2</sub> nano-particle to (Bi,Pb)-2223 superconducting phase increased the volume fraction, and the superconducting transition temperature up to 0.4 wt %.

The higher concentrations of SnO<sub>2</sub> nano-particles  $x > 0.4$  wt % reduced phase formation and superconducting transition temperature of (Bi,Pb)-2223. This was attributed that high concentration of SnO<sub>2</sub> nano-particle, induced large agglomeration between superconducting grains and hence reduced superconducting grain connectivity and deteriorated of intergranular current density.

### References

- Bilgili O. Y.Selamet. K.Kocabas (Nov 2012), J Supercond, Magn.21, P449.
- Tarascon, J.M., Lepage, Y., Greene, L.H., Bagley, B.G., Barboux, P., Hwang, D.M.H., Hull, G.W., Makinnon, W., Giroud, R.M (1988), Phys. Rev. 38, 2504.
- Hatano, T., Aoto, K., Ikeda, S., Nakamura, K., Ogawa, K. (1988), Jpn. J. Appl. Phys. 27, L2055
- Jia, Z.Y., Tang, H.Z., Yang, Q.Y., Xing, T., Wang, Y.Z., Qiao, G.W (2000), Physica C 337, 130.
- Ghattas, A., Annabi, M., Zouaoui, M., Ben Azzouz, F., Ben Salem, M (2008), Physica C 31, 468.
- Guilmeau, E., Andrzejewski, B., Noudem, J.G. (2003), Physica C 387, 382.
- Ghattas, A., Zouaoui, M., Annabi, M., Madani, A., Ben Azzouz, F., Ben Salem, M (2008), J. Phys. Conf. Ser. 97, 012179.
- Baqiah, H., Halim, S.A., Adam, M.I., Chen, S.K., Ravandi, S.S.H., Faisal, M.A.M., Kamarulzaman, M.M., Hanif, M. (2009), Solid State Sci. Technol.17, 81
- M.M Jousoh, S.Y.Yahya, I.Humadneh and R.Abd-Shukor (2012)
- Jannah .A.N. R.Abd-Shukor, and H.Abdullah (2013), Word Academy of Science and Technology.75, P. 367.
- Abou-Aly ,A.I.M.M.H.Abdel Gawad .R.awad. (2011), phys.,24, 2077
- Barik, H.K., Ghorai, S.K., Bhattacharya, S., Kilian, D., Chaudhuri, B.K (2000), J. Mater. Res. 15, 1076
- Terzioglu, C., Yilmazlar, M., Ozturk, O., Yanmaz, E. (2005), Physica C 423, 119