Journal of Applied Sciences

ISSN 1812-5654

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Journal of Applied Sciences 9 (4): 783-788, 2009
ISSN 1812-5654

Study of $Bi_{1.65-x}Pb_{0.35}Sb_xSr_2Ca_2Cu_3O_y$ Superconductor

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Abstract: In this study, the effect of annealing time and Sb doping on Bi-based superconductor properties have been investigated. Partial substitution of Sb for Bi in the $Bi_{1.65-x}Pb_{0.35}Sb_xSr_2Ca_2Cu_3O_y$ (BPSSCCO) superconductor has been found to increase the volume fraction of Bi (2223) phase. The BPSSCCO superconductor is made by solid state reaction method, using Bi_2O_3, PbO, CuO, $CaCO_3$, $SrCO_3$ and Sb_2O_3 powders as starting material. This study, reports $Bi_{1.65-x}Pb_{0.35}Sb_xSr_2Ca_2Cu_3O_y$ compound properties with x = 0.0, 0.03, 0.05 and 0.07 as variable. For that the effect of annealing time and amount of Sb doping on BPSSCCO microstructure, critical temperature and critical current density have been investigated. The structural analysis was carried out using XRD, SEM and EDX instruments. Both the critical temperature (T_c) and critical current density (J_c) were measured at 77 K. The critical current density was performed via V-J measurement in liquid nitrogen. The X-ray diffraction patterns show that low amount of Sb beside long annealing time enhance the fraction of Bi (2223) phase as well the critical current density. In order to study the effect of Sb on resistivity at room temperature, the V-I curves in the samples were measured. The resistivity of Sb doped samples at room temperature increases directly with increases of Sb. On the contrary, it is decreases by annealing time increase.

Key words: $Bi_{1.65-x}Pb_{0.35}Sb_xSr_2Ca_2Cu_3O_y$, superconductivity, critical temperature, critical current density, microstructure, XRD, SEM, EDX, susceptibility

INTRODUCTION

Since the discovery of high T_c superconductivity in Bi- Sr-Ca-Cu-O system, three superconducting phases of $(Bi, Pb)_2Sr_2Ca_{n-1}Cu_nO_y$ have been identified (Rao *et al*., 1993). They include Bi (2201) phase (n = 1, $T_c \approx 20K$), Bi (2212) phase (n = 2, $T_c \approx 85K$) and Bi (2223) phase (n = 3, $T_c \approx 110K$). Here after, we used the abbreviation Bi (2201), Bi (2212) and Bi (2223) phases for n = 1, 2 and 3, respectively. However, the Bi (2223) Phase is difficult to obtain and its formation is influenced by many preparation conditions, such as composition, annealing time and temperature, atmosphere and pressure at during sintering, the doped ion and quantity, heat-treatment method and operational procedure. Numerous studies in doping have been made in order to synthesize and stabiles the Bi (2223) phase in Bi- Sr-Ca-Cu-O system and to raise the T_c (Halim *et al*., 1999; Zhu and Nicholson, 1992). Therefore, T_c value alone could not define the relationship between the doping and the formation of high T_c Bi (2223) phase. In order to study the doping effect on the T_c and on the formation of Bi (2223) phase, the high T_c phase development must be studied under specified condition. Takano *et al*. (1988) showed that the incorporation of Pb in the nominal composition of this system is very effective in increasing the volume fraction of the high T_c phase (Zargar Shoushtari *et al*., 2002). In the electric field applications it is necessary to fabricate superconducting materials with high critical current densities J_c. It has been realized that the polycrystalline superconductors can be described as arrays of superconducting grains weakly coupled by josephson junctions. These weakly coupled grains are known to limit the J_c values of superconductors (Safar *et al*., 1995; Yoshida and Endo, 1993). A tremendous effort has been done to improve the links between the grains and the properties of the Bi-based superconductor such as doping of them by Sm (Yilmazlar *et al*., 2006), Sn (Seyoum *et al*., 1990), Cd (Zargar Shoushtari *et al*., 2008; Sasakura *et al*., 2001), Ag (Oota *et al*., 1995; Zargar Shoushtari *et al*., 2006), Zr, Hf, Mo and W (Makarova *et al*., 2005) and MgO (Zhao *et al*., 2001). In this study by substituting of Antimony for Bi in $Bi_{1.65-x}Pb_{0.35}Sb_xSr_2Ca_2Cu_3O_y$ superconductor, in which x = 0.0, 0.03, 0.05 and 0.07, the critical temperature, critical current density and the microstructure of samples were studied.

MATERIALS AND METHODS

The $Bi_{1.65-x}Pb_{0.35}Sb_xSr_2Ca_2Cu_3O_y$ superconductor was studied in Physics Department, Shahid Chamran University, since 2005. In this study, samples with starting

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composition of $Bi_{1.65-x}Pb_{0.35}Sb_xSr_2Ca_2Cu_3O_y$, with x = 0.0, 0.03, 0.05 and 0.07, were prepared by the solid state reaction method. Powders of Bi_2O_3, PbO, $SrCO_3$, $CaCO_3$, CuO and Sb_2O_3 with high pure degree are mixed. In order to prevent the growth of additional phases during the process, the powders were ground and milled for an hour. The mixed powders were calcined at 820°C for 15 h in air. Some bars and pellets with 20 mm in diameter and 3 mm thick were prepared. Then, they put in an Alumina crucible and placed in the furnace and the procedure of sintering was done in the air. All of the samples were shown the Misener effect experiment at liquid nitrogen well. The critical temperature and critical current density of samples were measured by the standard four-probe method at constant DC current of 40 mA. The AC susceptibility measurements were performed using a Lake Shore AC susceptometer, Model 7000. The X-ray diffraction (XRD) patterns of samples were taken by a Philips X-ray diffractometer Model PW1840. The microstructure characterization was performed using SEM and EDX and the images obtained by a scanning electron microscope Model 1455VP of the LEO Company.

RESULTS AND DISCUSSION

Figure 1 shows the results of XRD measurements patterns of the samples. Based on present XRD measurements, we observed that by substitution of Sb for Bi up to the amount of 0.05, the Bi (2223) phase increases in the sample and Sb acts as a phase stabilizer. As one can see that, the samples consist of a mixture of Bi (2223), Bi (2212) and Bi (2201) phases as the major constituents. From the XRD results one will notice that the volume of Bi (2223) phase decreases with increasing the amount of Antimony (sample d). The results of XRD show that the optimum amount of Antimony is about x = 0.05 (sample c), which has the highest volume fraction of Bi (2223) phase. The volume fraction of the phases can be estimated using various methods (Yurchenko *et al*., 2003). We have used all the peaks of the Bi (2223), Bi (2212), Bi (2201) Bi (21.8112) = $Bi_{1.6}Pb_{0.04}Sr_{1.81}CaCu_2O_{8.39}$ and Bi (21.8612) = $Bi_{1.6}Pb_{0.04}Sr_{1.86}CaCu_2O_{8.716}$ phases for the estimation of the volume fractions of the phases and ignored the voids.

The percentage of each phases in the samples were calculated as follows:

$$Bi(2223)(\%) \approx \frac{\sum I[Bi(2223)]}{A} \times 100$$

$$Bi(2212)(\%) \approx \frac{\sum I[Bi(2212)]}{A} \times 100$$

$$Bi(2201)(\%) \approx \frac{\sum I[Bi(2201)]}{A} \times 100$$

$$Bi(21.8112)(\%) \approx \frac{\sum I[Bi(21.8112)]}{A} \times 100$$

$$Bi(21.8612)(\%) \approx \frac{\sum I[Bi(21.8612)]}{A} \times 100$$

Where:

$$A \equiv \sum I[Bi(2223)] + \sum I[Bi(2212)] + \sum I[Bi(2201)] + \sum I[Bi(21.8112)] + \sum I[Bi(21.8612)]$$

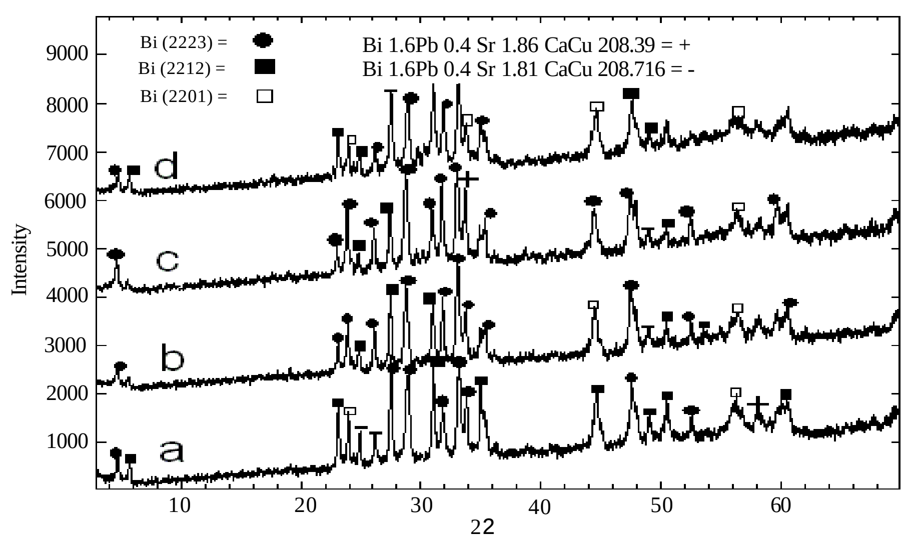

Fig. 1: XRD patterns of samples with different Antimony amounts: (a) 0.0, (b) 0.03, (c) 0.05 and (d) 0.07 with annealing time of 270 h

Table 1: Relative volume fractions of Bi (2223), Bi (2212), Bi (2201), Bi (21.8612) and Bi (21.8112)

Sample	a	b	c	d
Amount of Sb	0.00	0.03	0.05	0.07
Bi (2223) (%)	47.24	71.27	78.52	32.50
Bi (2212) (%)	37.81	21.64	12.24	41.64
Bi (21.8612) (%)	0.96	-	6.44	-
Bi (21.8112) (%)	6.41	1.63	1.08	8.74
Bi (2201) (%)	7.56	5.45	1.72	17.11

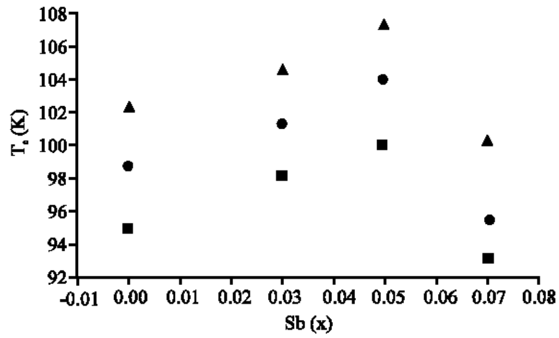

Fig. 2: The critical temperature of doped samples versus the amount of Antimony for annealing times of 150 (■), 210 (●) and 270 (▲) h

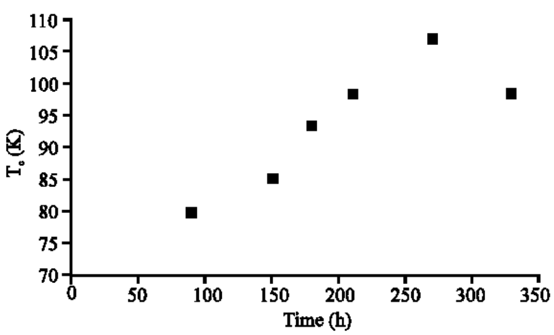

Fig. 3: The critical temperature versus annealing time for sample of 0.05 Antimony

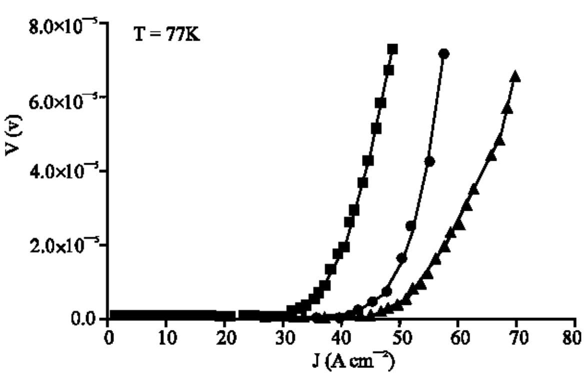

Fig. 4: The V-J curves for different annealing times of 150 (■), 210 (●) and 270 (▲) h, for the sample with Sb content of 0.05

where, I is the intensity of the present phases. The volume fractions of the phases for all the samples are given in the Table 1. As it shows, the maximum volume fraction of Bi (2223) phase obtained for sample with x = 0.05 (sample c). By increasing the amounts of more than 0.05, the volume fraction of Bi (2223) phases decreases. Also by decreasing x to the amounts less than 0.05, the volume fraction Bi (2223) phase decreases too.

Figure 2 shows the T_c versus the Sb content for different sintering times. One can see that the critical temperature of samples increases by increasing the amount of Sb up to x = 0.05 and then decreases for x > 0.05. Also, by increasing sintering time up to 270 h, the critical temperature of the sample increases. We attribute the increase of T_c, to enhancement of Bi (2223) phase in the sample. Also, the XRD data show that by increasing of Sb content more than 0.05, the other phases of Bi-Sr-Ca-Cu-O system are grown, which cause a decrease in T_c. So, the maximum T_c was observed for the sample with Sb content of 0.05 and sintering time of 270 h. The Fig. 2 also shows that by increasing of sintering time up to 270 h, the T_c of all samples increases. The results of this investigation are well compatible with the results of XRD patterns which are shown in Fig. 1.

For the sample with x = 0.05, we tried a longer sintering time up to 330 h, but we did not observe any improvement in T_c, instead T_c was decreased. The results of the critical temperature versus annealing time are shown in Fig. 3. Critical temperature for the sample with x = 0.05 enhances with increasing annealing time. The results show that by increasing the annealing time, the amount of Bi (2223) phase in the sample enhances. In fact, by increasing the annealing time, the amounts of unwanted phases are reduced and therefore the percentage of Bi (2223) phase increased which is well compatible with the results of XRD patterns in Fig. 1.

Figure 4 shows the V-J curves, which are taken at liquid nitrogen temperature, of the sample with x = 0.05 at different annealing times. One can observe that by increasing the annealing time, the critical current densities of the samples increase. As annealing time increases, the pinning energy will increases too and the entered average force to fluxoids will increase to being separated from the pinning centers, so that, the critical current density increases.

Figure 5 shows the measured J_c versus the amount of Sb doped in the samples for different annealing times. Present observations suggest that not only Sb acts as a Bi (2223) phase stabilizer, but also either causes some improvement between the grains or increases the flux pinning centers. Figure 5 shows that if we increase Sb content, J_c increases

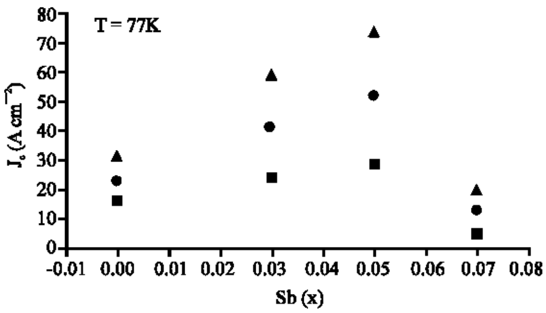

Fig. 5: The critical current density of doped samples as a function of the Antimony amounts for annealing times of 150 (■), 210 (●) and 270 (▲) h

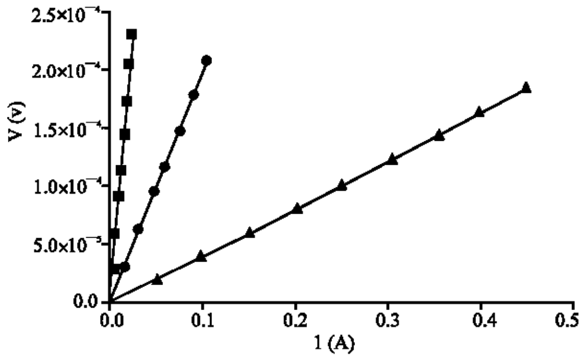

Fig. 6: The V-I curves for different annealing times 150 (■), 210 (●) and 270 (▲) h for the sample with 0.05 Antimony at room temperature

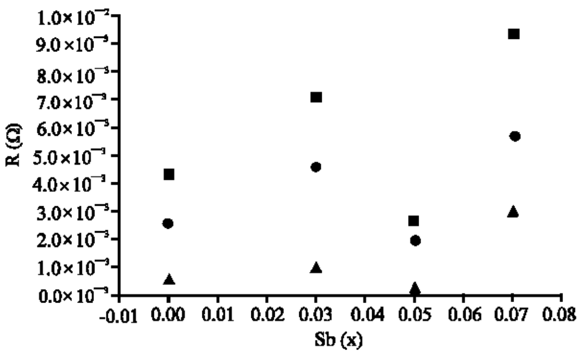

Fig. 7: The resistance of doped samples as a function of the Antimony amounts for annealing times of 150 (■), 210 (●) and 270 (▲) h at the room temperature

too. Also increasing annealing time up to 270 h, the J_c increases as well. So, the maximum J_c was found for the sample of $Bi_{1.6}Pb_{0.35}Sb_{0.05}Sr_2Ca_2Cu_3O_y$, annealed for 270 h. When the amount of Antimony gets more than 0.05, the unwanted phases such as Bi (2212) and Bi (2201) will increase in the sample. These unwanted phases play the

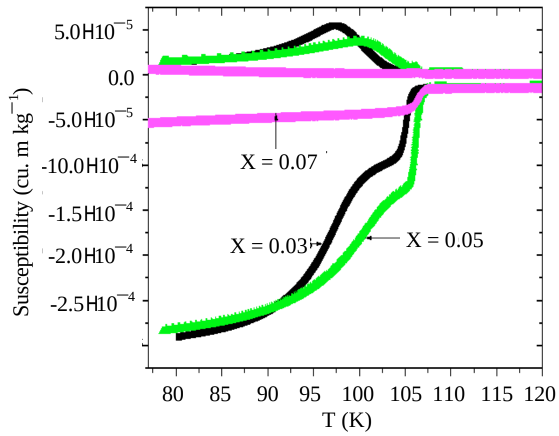

Fig. 8: Temperature dependence of the susceptibility for samples with Antimony content of 0.03, 0.05 and 0.07

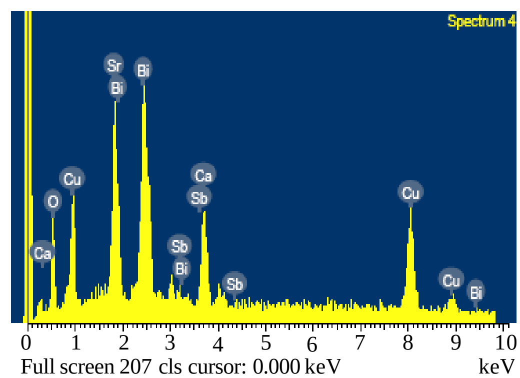

Fig. 9: The EDX image of the sample with the amount of Antimony 0.05 with annealing time of 270 h

role of the weak links and decrease the critical current density. It is shown that the Bi (2201) phase locates mainly between the superconducting grains, preventing the super current flow (Guo *et al*., 1998). It was also shown that J_c was raised as the Bi (2201) phase decreased in Bi (2223) tapes (Guilmeau *et al*., 2002).

Figure 6 shows the room temperature V-I curves of the sample with x = 0.05 at different annealing times. One can see that the normal state resistance of the samples will decrease by increasing the annealing time. This means that the junctions between the grains have been improved in the sample by increasing the annealing time.

In Fig. 7 we have plotted the resistance of doped samples as a function of the Antimony amounts for different annealing times at the room temperature. Also, one can see that increasing the period of annealing time up to 270 h, the resistance (R) decreases as well. So, the minimum R was found for $Bi_{1.6}Pb_{0.35}Sb_{0.05}Sr_2Ca_2Cu_3O_y$ sample, annealed for 270 h. When the amount of

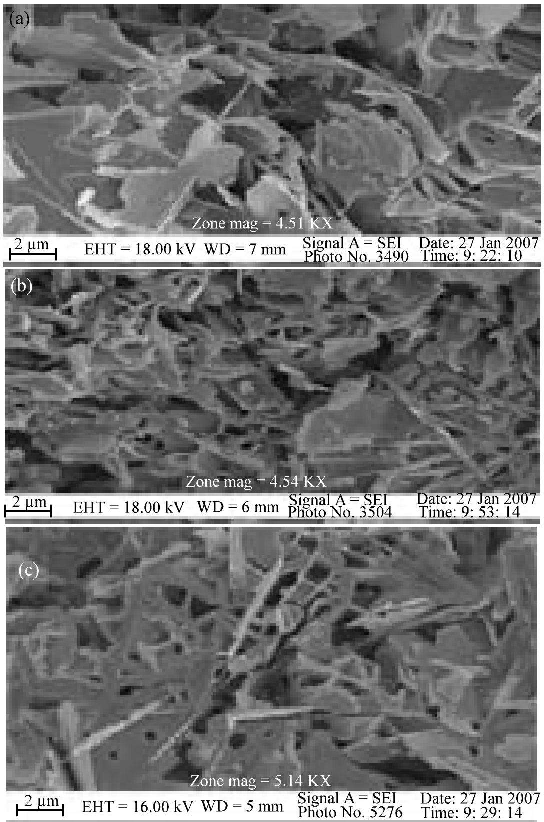

Fig. 10: The SEM micrographs of samples with different amounts of Antimony (a) 0.03, (b) 0.07 and (c) 0.05 with annealing time of 270 h

Antimony gets more than 0.05, the resistance of the normal state of the samples will increase. Because, entering higher amount of Antimony oxide in the samples, other unwanted phases will be grown. These phases will weaken the junctions between the grains; therefore the electric resistance of the normal state will increase.

Figure 8 shows the results of susceptibility measurements for samples with x = 0.03, 0.05 and 0.07 for Antimony. The temperature dependencies of the real χ' and imaginary χ" parts of Ac susceptibility for all of the samples were measured in Ac field of 50 A/m with a frequency of 333 Hz which was applied parallel to the long dimension of the samples. In particular, the imaginary component of the AC susceptibility has been widely used to probe the nature of the weak links in polycrystalline superconductors. It is also employed to estimate some of the important physical properties like the critical current density and the effective volume fraction of the superconducting grains (Sedky and Youssif, 2001). The real part of the AC susceptibility χ' in polycrystalline samples shows two drops as the temperature is lowered, below the onset of diamagnetic transition and correspondingly the derivative of the χ' displays two peaks. The first sharp drop at the critical temperature is due to transition within the grains and the second gradual change is due to the occurrence of the superconducting coupling between the grains. The imaginary part χ" shows a peak which is a measure of the dissipation in the sample. It can be seen from the curves that the intergranular coupling between grains in sample x = 0.05 is better than sample x = 0.03 and sample x = 0.03 is much better than sample x = 0.07.

The results of EDX images show that there is not unwanted element in the samples, that means, the samples are not contaminated during the synthesis process. One of the EDX image is shown in Fig. 9.

Figure 10a-c show the SEM micrographs for the samples with amounts of Antimony 0.03, 0.07 and 0.05 with annealing time of 270 h. It seems that the increase in the amount of Antimony more than x = 0.05 leads to the formations of more pores. It seems that decreasing the amount of Antimony the values of x less than 0.05 leads to the formation of large grains. These pores and large grains act as weak links between the grains, destroying grains connectivity and disrupting current flow. These results are in agreement with the results of x-ray diffraction patterns and the critical current densities.

CONCLUSION

The role of Sb in the synthesis of $Bi_{1.65-x}Pb_{0.35}Sb_xSr_2Ca_2Cu_3O_y$ with x = 0.0, 0.03, 0.05 and 0.07 has been investigated. The samples were prepared by solid state reaction using different annealing times. The results of susceptibility measurements for the samples show that diamagnetic fraction of the sample with x = 0.05 in more than the others. The result of this investigation is well compatible with the results of XRD measurements. Doping of the sample with Antimony x = 0.05 and annealing time of 270 h, can increase the percentage of Bi-2223 phase. Present results also show that the maximum value of the critical current density and the critical temperature were obtained for the sample with annealing time of 270 h and Sb content of 0.05.

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