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Fabrication and characterization of $\text{Bi}_{1.6}\text{Pb}_{0.4}\text{Sr}_2\text{Ca}_2\text{Cu}_3\text{O}_y$ superconductors prepared by solid state reaction technique

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Abstract

In this work three sets of superconducting samples with composition $\text{Bi}_{1.6}\text{Pb}_{0.4}\text{Sr}_2\text{Ca}_2\text{Cu}_3\text{O}_y$ were prepared by the solid state techniques and resistivity measurements were carried out on these samples in the temperature range 77 K – 175 K. The standard four-probe technique was used to measure the resistivity of the samples. The appropriate amount of Bi_2O_3 , PbO , SrCO_3 , CaCO_3 and CuO of 99.99 % purity were used as starting raw material and were mixed. The mixed powder was initially calcinated at constant temperature of 820°C and the pallets of diameter 14 mm and thickness 1-2mm were prepared under the pressures 200 kg/cm² (sample A), 300 kg/cm² (sample B) and 250 kg/cm² (sample C). Then the pallets were subjected to several cycles of sintering at constant temperature in the furnace and allowed to furnace cool to room temperature in air. The resistivity measurements were carried out on these samples using locally made resistivity probe. Rh-Fe resistance thermometer was used to measure the temperatures of the sample. The samples A, B and C showed the onset superconducting transition temperatures at 103 K, 111 K, and 110 K respectively. Samples A and B showed broad transitions while sample C showed a sharp drop indicating the presence of only one superconducting phase Bi 2223 in the sample.

1. INTRODUCTION

It is reported in the literature [1,2] that there are three different superconducting phases in the (Bi,Pb)SrCaCuO superconducting system with number of CuO layers in the crystal structure. The $(\text{Bi,Pb})_2\text{Sr}_2\text{Ca}_2\text{Cu}_3\text{O}_y$ (2223 phase) system exhibits a highest superconductive transition temperature T_c at 110 K and has three CuO layers. Similarly $(\text{Bi,Pb})_2\text{Sr}_2\text{CaCu}_2\text{O}_y$ (2212 phase) system has two CuO layers and the $(\text{Bi,Pb})_2\text{Sr}_2\text{CuO}_y$ (2201 phase) system has a single copper oxide plane. These two systems exhibit superconductive transition temperatures T_c of 60-85 K and 20 K respectively [3].

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Since the discovery of superconductivity in BiSrCaCuO ceramics, the preparation of single phase 2223 materials has proven difficult because the 2212 phase is thermodynamically more favorable at elevated temperatures than the 2223 phase. The partial substitution of PbO for Bi₂O₃ has since been found to help stabilize the high T_c composition, thereby increasing the volume fraction of this phase. By stabilizing the high T_c phase in BiSrCaCuO ceramics, higher process yields have become possible for products utilizing these compositions [3].

The conventional solid state reaction technique is used to prepare the samples, because it requires very long heat treatment with several intermediate grindings in order to produce almost a single 2223 phase [4]. In this paper preparation, and fabrication of Bi based superconductors of nominal composition Bi_{1.6}Pb_{0.4}Sr₂Ca₂Cu₃O_y and their resistance measurements in the temperature range 77 K to 175 K are reported.

2. EXPERIMENTAL TECHNIQUE

2.1 Sample preparation

Oxide powders with compositions Bi₂O₃, PbO, SrCO₃, CaCO₃ and CuO of 99.99 % purity were used as starting raw materials. The weights of these chemicals were selected according to nominal cation ratio Bi:Pb:Sr:Ca:Cu is 0.8:0.2:1:1:1.5.

The starting raw materials of 1/70 mole mostly pre dried oxides and carbonates of purity greater than 99.99 % were weighed to an accuracy of 0.1 mg. After thoroughly mixing, the mixture was ground to a fine powder using the mortar and pestle. The ground powder sample was divided into three portions with the labels as sample A, sample B and sample C. Then the samples A and C were fired at constant temperature of 820⁰C in the furnace for 24 hours and the sample B was fired at the same temperature for 12 hours. Then the fired powder samples were reground.

After that three sets of pallets of diameter 14 mm and thickness 1-2 mm were prepared under the pressure of 200 kg/cm², 300 kg/cm² and 250 kg/cm² out of samples A, B and C respectively.

Then the set of sample A was subjected to a preliminary sintering at constant temperature of 860⁰C in the furnace for 60 hours and was allowed to furnace cool to room temperature in air.

The set of sample B was subjected to preliminary sintering at constant temperature of 830⁰C in the furnace for 60 hours and was allowed to furnace cooling to room temperature in air. Then the pallets were reground and palletized under the pressure of 300 kg/cm². After that, the pallets were subjected to a second cycle sintering cycle at 860⁰C in the furnace for 60 hours and were allowed to furnace cool to room temperature in air.

Similarly, the set of sample C was subjected to preliminary sintering at constant temperature of 820⁰ C in the furnace for 24 hours and was allowed to furnace cool to room temperature in air. Then the pallets were reground and palletized under the pressure of 250 kg/cm². The above steps were repeated and the pallets were subjected to final sintering at 860⁰ C in the furnace for 100 hours and were allowed to furnace cool to room temperature in air.

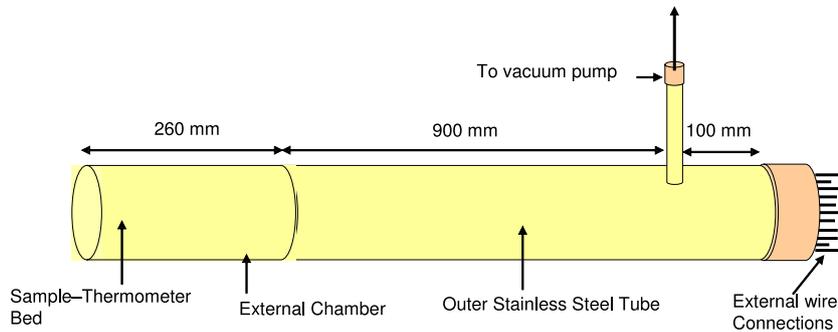


Fig. 1 Schematic diagram of the Resistivity Probe

2.2 Resistivity Probe

The diagram of the Resistivity Probe [5] used in this experiment is shown in the Fig. 1. Initially both sides of the sample bed were well cleaned with sand paper until they become smooth. Then the resistance Rh-Fe thermometer was mounted on one side of the bed and the sample was mounted on the other side of the bed. After that power supplies, voltmeters and ammeters were connected to the thermometer and the sample as shown in Fig. 2.

2.3 Electronic circuit and experimental procedure

The circuit diagram of the experiment is presented in Fig. 2. The temperature of the sample was measured by Rh-Fe thermometer and controlled by electrical heating.

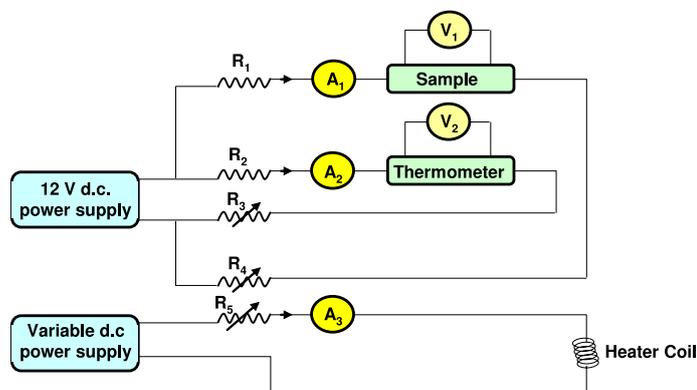


Fig. 2 Electrical Circuit of the experiment

The Resistivity Probe was pumped out to a good vacuum and was immersed in liquid Nitrogen. Constant currents of 1 mA and 50 mA were applied to the thermometer and the sample respectively. The voltages across the thermometer and the sample were measured by the digital voltmeters. While the sample was gradually warmed by electrical heating, the resistances of the sample and the corresponding temperatures of the sample were recorded. These measurements of resistance were repeated while the sample was cooling.

3. PRESENTATION OF RESULTS AND DISCUSSION

3.1. Resistivity measurements on $\text{Bi}_{1.6}\text{Pb}_{0.4}\text{Sr}_2\text{Ca}_2\text{Cu}_3\text{O}_y$ Superconductor (Sample A)

Resistivity of $\text{Bi}_{1.6}\text{Pb}_{0.4}\text{Sr}_2\text{Ca}_2\text{Cu}_3\text{O}_y$ (sample A) was measured in the temperature range from 77 K to 175 K, while the sample was cooling and warming, and the data are presented in Fig. 3.

The data points shown in circles, obtained when the sample A was cooling, exhibit an onset superconducting transition temperature $T_{c,\text{onset}}$ of approximately 103 K while the data points shown in triangle, obtained when the sample warming, exhibit a transition temperature $T_{c,\text{onset}}$ of approximately 101 K. The superconducting transition widths of the sample ΔT_c were found to be 22 K and 20 K, when cooling and warming respectively. It is clear that this sample shows a considerably broad superconducting transition and this may be due to the presence of impurities or non superconducting regions or multi superconducting phases in the sample. The most probable reason may be the existence of multi superconducting phases. Also a small difference is observed in the data during cooling and warming and this may be due to the temperature equilibrium problems in the experiment.

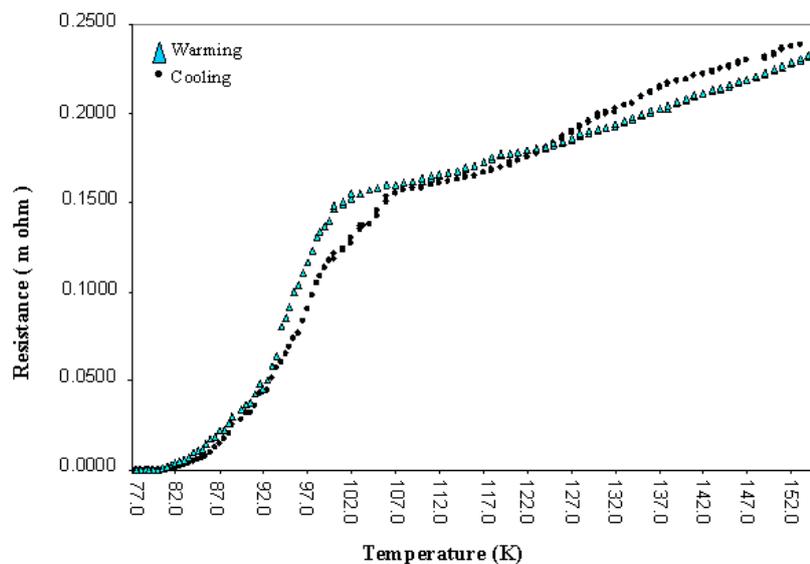


Fig.3 The temperature dependence of the electrical resistance of $\text{Bi}_{1.6}\text{Pb}_{0.4}\text{Sr}_2\text{Ca}_2\text{Cu}_3\text{O}_y$ superconductor (Sample A) when Cooling and Warming.

3.2. Resistivity measurements on $\text{Bi}_{1.6}\text{Pb}_{0.4}\text{Sr}_2\text{Ca}_2\text{Cu}_3\text{O}_y$ Superconductor (Sample B)

Resistivity of $\text{Bi}_{1.6}\text{Pb}_{0.4}\text{Sr}_2\text{Ca}_2\text{Cu}_3\text{O}_y$ (sample B) was measured in the temperature range from 77 K to 175 K, while the sample was cooling and warming, and the data are presented in Fig. 4.

It is apparent that the sample B possesses an onset superconducting transition temperature $T_{c,\text{onset}}$ of approximately 111 K and 112 K when cooling and warming respectively. The superconducting transition widths of the sample ΔT_c were found to be 30 K and 31 K respectively while cooling and warming. This sample also shows a broad superconducting transition and this may be due to the presence of impurities or non superconducting regions or multi superconducting phases in the sample. These two data are almost identical confirming reproducibility of the sample.

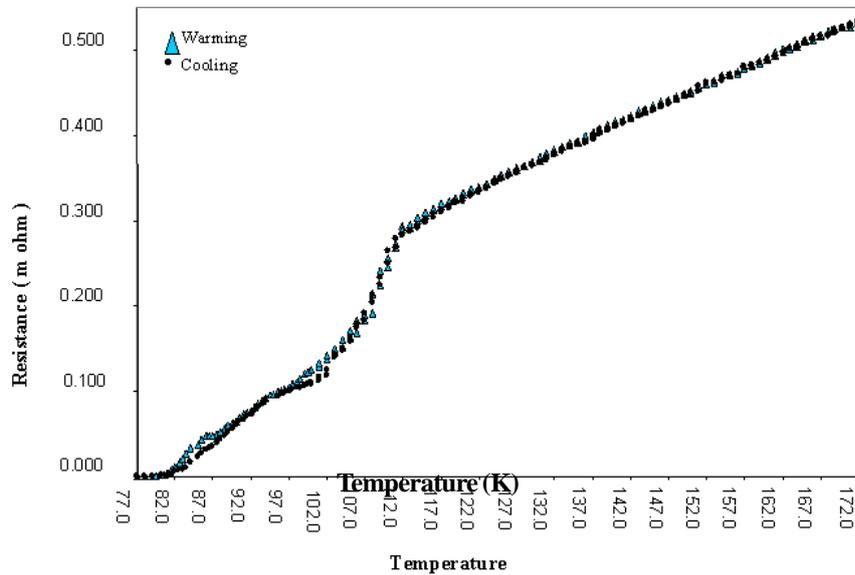


Fig. 4 The temperature dependence of the electrical resistance of $\text{Bi}_{1.6}\text{Pb}_{0.4}\text{Sr}_2\text{Ca}_2\text{Cu}_3\text{O}_y$ superconductor (Sample B) when Cooling and Warming.

3.3. Resistivity measurements on $\text{Bi}_{1.6}\text{Pb}_{0.4}\text{Sr}_2\text{Ca}_2\text{Cu}_3\text{O}_y$ Superconductor (Sample C)

Resistivity of $\text{Bi}_{1.6}\text{Pb}_{0.4}\text{Sr}_2\text{Ca}_2\text{Cu}_3\text{O}_y$ (sample C) was measured in the temperature range from 77 K to 153 K, while the sample was cooling and warming, and the data are presented in Fig. 5.

As shown in Fig. 5, this sample shows a sharp drop with a very small transition width in its superconducting transition and exhibits onset superconducting transition $T_{c,\text{onset}}$ of approximately 110 K and 110 K, when cooling and warming respectively. The superconducting transition widths of the sample ΔT_c when cooling and warming were found to be at 28 K and 26 K respectively.

The sharp drop with a small width in the superconducting transition of this sample indicates that mainly, there is only one superconducting phase Bi 2223 in the sample. However, the small width in

the transition indicates that a very small admixture of some other phases exist. Also a small difference is observed in these data and this may be due to an electronic problem in the experiment.

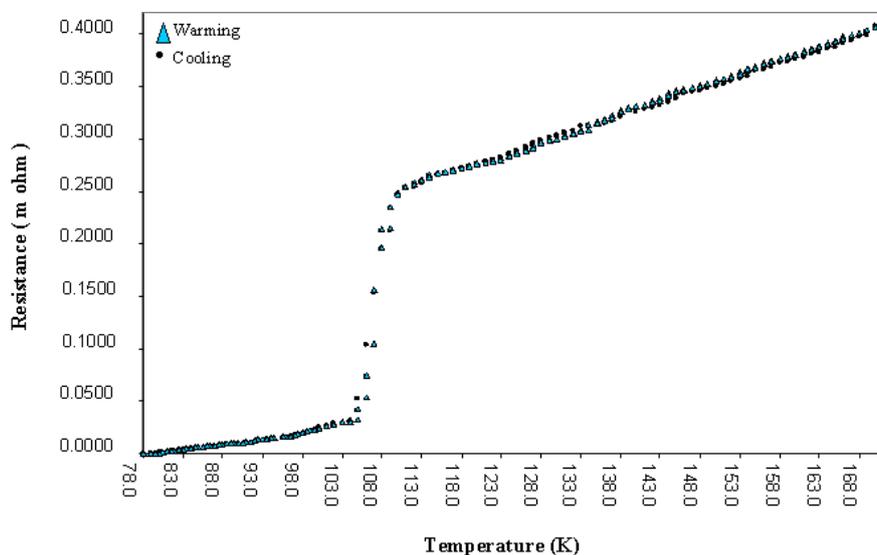


Fig. 5 The temperature dependence of the electrical resistance of $\text{Bi}_{1.6}\text{Pb}_{0.4}\text{Sr}_2\text{Ca}_2\text{Cu}_3\text{O}_y$ superconductor (Sample C) during Cooling and Warming.

4. CONCLUSION

Three Bi based superconductors with composition $\text{Bi}_{1.6}\text{Pb}_{0.4}\text{Sr}_2\text{Ca}_2\text{Cu}_3\text{O}_y$ samples A,B and C were prepared under different conditions shown in Table 1.

Table 1 The conditions under which the samples were prepared.

Sample	Initial firing temperature and time	Final firing temperature and time	Pressure
A	820°C (24 hrs)	860°C (60 hrs)	200 kgcm ⁻²
B	820°C (12 hrs)	830°C (60 hrs), 860°C (60 hrs)	300 kgcm ⁻²
C	820°C (24 hrs)	820°C (24 hrs), 860°C (100 hrs)	250 kgcm ⁻²

Sample A, B and C show superconducting transition temperatures (onset) 103 K, 111 K and 110 K while cooling and 101 K, 112 K and 110 K while warming respectively. Samples A and B show broad transitions indicating presence of non superconducting regions, impurities or more than one superconducting phase in the sample. Sample C shows single drop transition indicating only one Bi 2223 phase existing in the sample. This suggests that sample C which was sintered continuously longer time (100 hrs) shows single stage transition at 110 K confirming the presence of only high

purity Bi 2223 phase. The conventional solid state reaction method requires very long heat treatment with several intermediate grinding stages in order to produce single Bi 2223 superconducting phase.

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