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Katı Hal Reaksiyonu ile Hazırlanan Kurşun Konsantrasyonunun Değişiminin Bi-223 Süperiletken Özellikleri Üzerindeki Etkisi

Moeen GHAFOOR*, Jameel-Un NABI and Jibran ABBASI

[1]*Faculty of Engineering Sciences, Ghulam Ishaq Khan Institute of Engineering Sciences and Technology, Topi, Pakistan*

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Özet. Bu çalışmada, hem kurşun bulundurmayan süper iletken tabletler hem de farklı kurşun kombinasyonları bulunduran tabletler katı hal reaksiyonuyla hazırlandı. Hazırlanan örnekler X ışını toz dağılımları kullanarak (XRD), elektron mikroskobu (SEM) ve enerji dağıtıcı X ışını spektroskopisi ile taranarak karakterize edildi. Süper iletken özellikleri elektrik direnci ve kritik akım yoğunluğu transferi Jc kullanılarak incelendi. Sonuçlarımıza göre 48 saat orta derecede öğütülen kalsinasyonla hazırlanan örneğimiz ideal süper iletken geçiş ısısı Tc ve kritik akım yoğunluğu transferi Jc değerlerine sahiptir.

Anahtar Kelimeler: Süperiletken, Bi-2223, electric direnci, XRD, SEM, EDS, Kurşun

Effects of change in concentration of lead on the properties of Bi-2223 superconductor prepared through solid state reaction

Abstract. In this work, superconducting pellets without lead (Pb) as well as with different concentrations of lead are prepared by solid state reaction. The prepared samples are then characterized using X-ray powder diffraction (XRD), scanning electron microscope (SEM), and energy-dispersive X-ray spectroscopy (EDX). The superconducting properties are examined using electrical resistivity and transport critical current density Jc. Our results show that the sample prepared with calcination of 48 hours with intermediate grinding has the ideal value of the superconducting transition temperature Tc and transport critical current density Jc.

Keywords: Superconductor, Bi-2223, electrical resistivity, XRD, SEM, EDS, Lead

1. INTRODUCTION

Among all of the Type II superconductors, the Bi (Pb)-Sr-Ca-Cu-O is considered to be the most promising, because of its high critical temperature [1-2] and current density. This material has a nominal composition of Bi2Sr2Cam–1CumO2m+4+δ, [3], where m is the number of Cu-O layers. Now, among different phases of this system the three most common superconducting phases are: Bi-2201 (m = 1), Bi-2212 (m = 2) and Bi-2223 (m = 3), with Tc ~ 20, 80 and 110 K, respectively. The coexistence of these different phases and their dependence on parameters like temperature, time of sintering and calcination makes this useful system very complex [4]. It is desirable to synthesize pure 2223 phase, but this is very difficult because of its undergrowth with

2212. This undergrowth is due to the high complexity of the reaction and the small difference in the thermodynamic stabilities of 2212 and 2223 [5-6].

Many different methods like sol-gel method [7-8], aerosol pyrolysis technique [9-10], and polymer-matrix method [11-14] are used today for the preparation of superconductors but the simplest one is the solid state reaction [15-19]. At present, solid state reaction has been used to synthesize Bi-2223 type-II superconductor. Furthermore, it is known that parameters such as calcination time [20] and the concentration of Lead [21] strongly influence the grain size and phase concentrations, and hence, the critical temperature Tc and critical current density J¬c. In this research, our objective has been to find the optimized concentration of lead needed to improve superconducting properties. It can be seen that the products obtained as a result of solid state reaction are not always stoichiometric and homogeneous on microscopic scale and are usually non reproducible [22]. This non-stoichiometry is because of varying timings of grinding and due to the high heating temperatures that are involved [23].

Solid state reaction can synthesize the product from off the shelf materials and also does not require sophisticated apparatus. However, the formation kinetics of the superconducting phase is low due to slow diffusion rates [24].

It is essential that the grains of every superconductor should have a specific orientation, a large size and a desired direction; this gives rise to high current density. As the critical temperature Tc and critical current density Jc depend on the hole concentration [25] and the oxygen content [26] in the sample, so it can be assumed that with the application of uniaxial pressure one can increase the current density of the bulk material. Moreover, partial substitution of Bi by Pb changes the formation of 2223 phase [27].

In this research, solid state reaction between Bi2O3, SrCO3, CaCO¬3, PbO and CuO powders in the cation ratio of Bi:Sr:Ca:Cu = 2:2:2:3 creates a mixture of 2212 and 2223 phases. To control the powder grain size and phase composition, two important parameters in the formation of Bi-2223, it is desirable to synthesize the product oneself rather than using the ones available commercially.

Study has shown that the formation of (Bi-Pb) 2223 phase is significantly enhanced by partial substitution of Bi by Pb. Further investigation also shows that the best stoichiometric amounts of both Bi and Pb are Bi2.5 and (Bi + Pb) 2.2 for both Bi-2223 and (Bi, Pb)-2223 phases [28], respectively. The research related to the effect of calcination time has led the scientists to the result that the kinetics of formation and grain size of Bi-2223 are strongly dependent on the calcination conditions, and as a result, also on the phase assemblages formed at the end of the calcination process, which are important in determining the reactivity during sintering.

The pelletization pressure also plays an important role in the synthesis process as the amount of 2212 phase can be decreased by applying appropriate pressure [29]. Hence, after making every process optimum, the best critical temperature that we achieved was 103.051 K.

2. EXPERIMENTAL TECHNIQUE

It has already been studied by scientists that the method of introduction of Pb greatly affects the properties of the sample [30]. Therefore, in this research, we will draw results regarding the effects of change in concentration of Pb on superconducting properties. To achieve this, four different samples are prepared in open air by classical solid state reaction. The stated materials are 99.9% pure commercially available PbO, Bi2O3, SrCO3, CaCO3 and CuO. One pellet is prepared without Pb while the other three pellets are prepared by varying the amount of Pb while keeping the calcination time constant, that is, 24 hours.

All samples are grounded in an agate mortar with an agate pestle for about 1 hour 30 minutes in the required stoichiometric ratios. After that, the first heat treatment technique is applied on all samples, after which they are calcinated in air in a silver crucible at 845˚C for specified times followed by further grounding for about 30 minutes. As the phase assemblage is affected by the calcination conditions, so to obtain the proper precursor phases necessary for the formation of Bi-2223, the above mentioned conditions are crucial. After this, all samples are compressed into pellets of diameter 1.5 cm and thickness of 0.3 cm by using a Hydraulic Press under a uniaxial pressure of 410MPa for 24 hours. Once the pellets are obtained, they are subjected to the second heat treatment process, and hence, are sintered at 835˚C for 48 hours; studies reveal that raising the sintering temperature up to 845˚C results in good intergranular coupling. Finally, all the samples are annealed at a rate of 2˚C/min. The details about the composition of each and every pellet along with its heat treatment details are present in table 1. The masses of each tablet before and after heat treatment are also recorded in order to find any changes in the mass as a result of the heat treatment. This sums up the preparation of the pellets.

Table 1. Chemical formulae of pellets and preparation conditions.

Pellet	Chemical Formula	Calcination Parameters	Pressure	Sintering	Annealing Parameters
1	$(Bi_{1.6}Pb_{0.4})Sr_2Ca_2Cu_3O_{10+x}$	845˚C/24 hours	410MPa/24 hours	845˚C/48 hours	2˚C/min
2	$Bi_2Sr_2Ca_2Cu_3O_{10+x}$	845˚C/24 hours	410MPa/24 hours	845˚C/48 hours	2˚C/min
3	$(Bi_{1.7}Pb_{0.3})Sr_2Ca_2Cu_3O_{10+x}$	845˚C/24 hours	410MPa/24 hours	845˚C/48 hours	2˚C/min
4	$(Bi_{1.5}Pb_{0.5})Sr_2Ca_2Cu_30_{10+x}$	845˚C/24 hours	410MPa/24 hours	845˚C/48 hours	2˚C/min

Next, for the characterization, all the samples are characterized by X-ray powder diffraction using X'Pert Graphics and identified with Cu Kα-radiation at 40 KV and 20 mA with a step size of 0.002˚C /s (I= 1.5406 Å) in the range $4^\circ \leq 2\theta \leq 70^\circ$. These scanning restrictions are used as they are found to provide precise areas of diffraction peaks for calculating phase concentrations. The grain size and microstructure morphology of the samples is studied using a Jeol Scanning Electron Microscope JSM-5300, operated at 30kV. For Compositional Analysis, Energy Dispersive Spectroscopy (EDS) is used on the prepared samples using Oxford X-ray micro-probe analyzer connected to a Jeol scanning microscope JSM-5300.

The most important parameters of a superconductor are its critical temperature Tc and critical current density Jc. The critical temperature is measured by using the standard four point probe method. Semicircular slices are cut from the samples and gold probes are attached to these samples one by one with the help of good quality silver pastes. The sample is cooled down in the absence of magnetic field with the help of liquid nitrogen; excitation current is applied to the sample plane perpendicular to the pressing direction. The transport critical current density is measured by the conventional four-probe technique at 77 K using liquid nitrogen.

3. RESULTS and DISCUSSION

After continuous research, researchers have learnt that we can achieve zero resistance if the grains of 2223 exist in ceramic matrix generated by 2212 phase. SEM analysis of the samples shows that the grain size and granularity of the samples is greatly affected by the change in the Pb content. SEM micrographs of the samples are shown in figures 1(a) to 1(d).

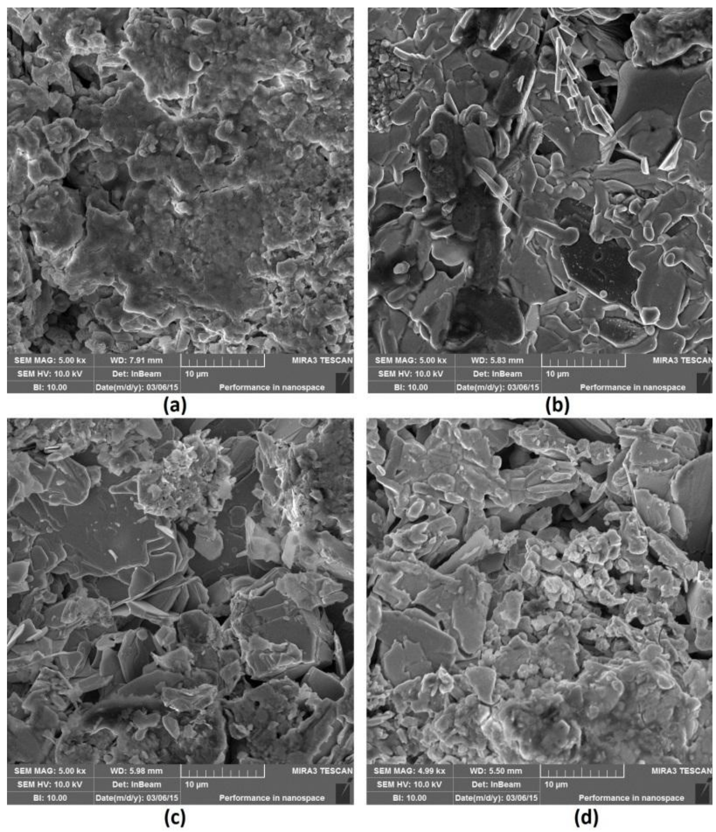

Figure 1. SEM Images for (Bi-2223) phase with different Pb concentration: (a) Pellet 1; (b) Pellet 2; (c) Pellet 3; (d) Pellet 4.

One of the most important tests is the energy dispersive x-ray (EDS). It is used to confirm if the real compositions of phases exist or not. To make this point clear, EDS spectra of our samples are taken from the same region as that of the SEM. Figures 2(a) to 2(d) show all the spectra of our samples.

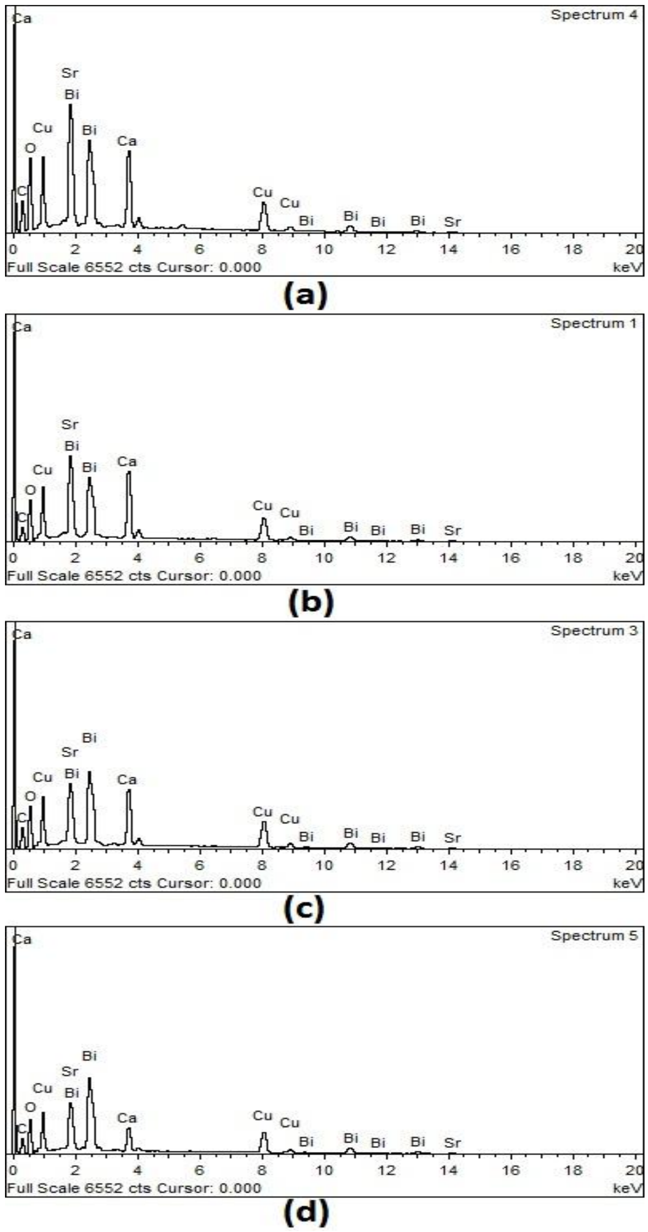

Figure 2. EDS Images for (Bi-2223) phase with different Pb concentration: (a) Pellet 1; (b) Pellet 2; (c) Pellet 3; (d) Pellet 4.

XRD patterns are taken from the surfaces of the pellets which show that the samples are of pure phase. XRD patterns reveal significant shifts in the positions of the peaks, their intensities and their hkl values. The phases of the pellets are directly affected by the change in calcination time and temperature. In Figure 3 we have shown the XRD patterns of all the samples.

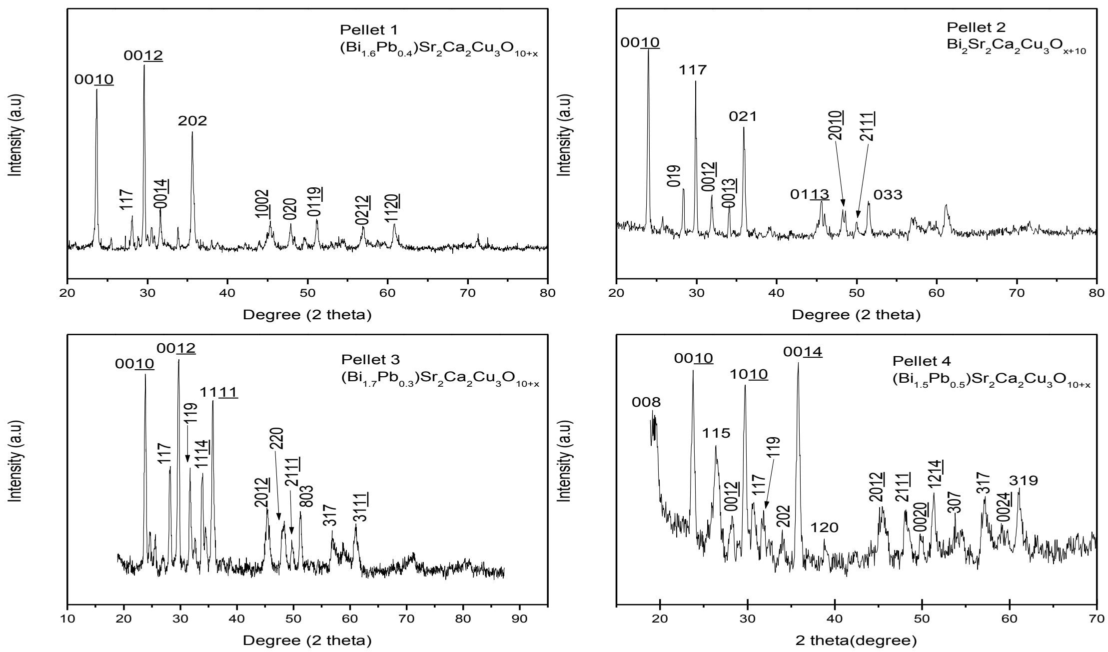

Figure 3. XRD analysis of different samples of Bi-2223 with different Pb concentration.

The Meissner effect was checked before taking resistivity measurements. The sample temperature is brought down to that of liquid nitrogen after which the sample shows strong magnetic repulsion in the vicinity of a magnet. This confirms that all of our samples are superconductors. Then resistance is measured both at the room temperature and at the liquid nitrogen temperature. This shows that the resistance of each sample reduces to zero at a different temperatures (see Figure 4).

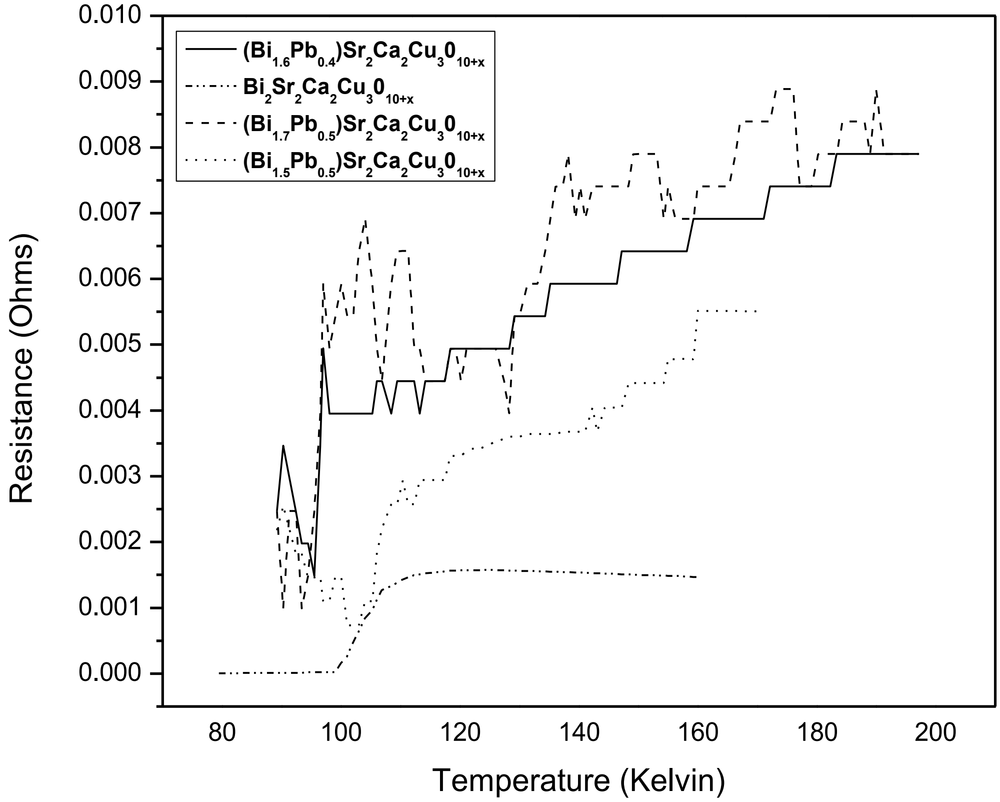

Figure 4 Resistance measurements of different samples of Bi-2223 with different Pb concentration.

This change in critical temperature is due to the different preparatory conditions. Table 2 sums up all our findings regarding the change in critical temperature (Tc) with the change in the concentration of Lead (Pb).

Table 2. Critical temperatures of pellets with chemical formulas.

Pellet	Chemical Formula	Critical Temperature
1	$(Bi_{1.6}Pb_{0.4})Sr_2Ca_2Cu_3O_{10+x}$	95.474 K
2	$Bi_2Sr_2Ca_2Cu_3O_{10+x}$	100.000 K
3	$(Bi_{1.7}Pb_{0.3})Sr_2Ca_2Cu_3O_{10+x}$	93.407 K
4	$(Bi_{1.5}Pb_{0.5})Sr_2Ca_2Cu_30_{10+x}$	103.051 K

4. CONCLUSION

In earlier research, researchers have shown that introduction of Pb in the 2212 phase results in sharper transition temperatures and higher critical current densities. In this research, however, we have tried to find the optimum concentration of Pb to be added as PbO. When all the samples with varying concentration of Pb are prepared, it is found that change in concentration of Pb greatly affects the Critical Temperature as shown in Table 2. It is found that with the increase in Pb concentration, the critical temperature also increases and in our case the maximum Tc which is achieved is 103.051K with a concentration ratio of (Bi1.5Pb0.5) Sr2Ca2Cu3010+x. Also this has resulted in better granularity of the samples which has improved the critical current density Jc.

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