Increasing critical temperature on BPSCCO superconductors with addition of Al\textsubscript{2}O\textsubscript{3}

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Abstract. We have investigated the effect of addition of Al\textsubscript{2}O\textsubscript{3} in the manufacture of the BPSCCO superconductors using solid state reaction method. Precursors with consist of Bi\textsubscript{2}O\textsubscript{3}, PbO\textsubscript{2}, SrCO\textsubscript{3}, CaCO\textsubscript{3}, and CuO powders were ground using mortar agate for 3 h. The samples were heated at 300°C for 6 h to remove the moisture in the powders, and then ground again for 6 h. Then the samples were calculated at 820°C for 20 h and then Al\textsubscript{2}O\textsubscript{3} (1wt% and 2wt%) were added and ground again. The powders were compacted with a diameter of 10 mm and a thickness of 2 mm and then sintered at 850°C for 30 h. The characterizations were carried out by using Cryogenic Magnet to determine the superconductivity properties, SEM-EDX to analyze the morphology and XRD to determine the phase formed. The resistivity versus temperature results showed the increase of T\textsubscript{c} with the addition of Al\textsubscript{2}O\textsubscript{3}. Sample with 1wt% Al\textsubscript{2}O\textsubscript{3} has T\textsubscript{c(onset)} of 90 K and T\textsubscript{c(0)} of 66 K, when the sample with 2wt% Al\textsubscript{2}O\textsubscript{3} has T\textsubscript{c(onset)} of 94 K and T\textsubscript{c(0)} of 73 K. The morphology formed looks less homogeneous, the crystalline granules are melt-shaped, clumpy and there are porosity.

1. Introduction

Until today, bismuth-based superconductors (BSCCO) are among the most attractive and the most promising in the industry. For example, in advanced technologies in electromagnetic such as transportation, machinery, electricity, and diagnostic tools [1]. The manufacture of superconductors is carried out following technological developments that require sophisticated materials. Many researchers are interested in superconductors because they have the unique characteristic of critical temperature (T\textsubscript{c}) [2]. Some of the common forms of HTSCs include BiSrCaCuO (BSSCO), HgBaCaCuO, HgTlBaCaCuO, YBaCuO (YBCO), and TlBaCaCuO (TBCCO). Many efforts are being made to reinforce the transition temperature of HTSCs. Some researchers modified the fabrication techniques of the materials [3]. Bi2223 superconducting granules have a very complex structure thanks to their high antisotropy [4]. The Bi-based superconductor involves three superconductors stages with the composition Bi\textsubscript{2}Sr\textsubscript{2}Ca\textsubscript{2-n}Cu\textsubscript{n}O\textsubscript{2m+4} with n = 1, 2 and three having T\textsubscript{c} values up to 20, 80, and 110 K [5]. To achieve a higher critical temperature (T\textsubscript{c}) of superconducting research continuously was carried out. Superconducting materials have grown since the invention of ceramic superconductors. BSCCO superconducting base material system with a superconductivity critical temperature of about 110 K, to increase the critical temperature to be applied to the technology well [6]. The advantage of the BPSCCO superconducting is that a higher magnetic field can be achieved compared to conventional sensors, and
it is easy to manufacture. The electric resistance of a superconductor increases with the appliance of an external magnetic flux. However, the rise in electric resistance continues even after the discharge of the magnetic flux [7]. In the formation of the Bi-2223 phase within the addition of Pb can produce single-phase samples, because Pb enlarges the formation domain of Bi-2223 [8]. The use of other dopants such as Al$_2$O$_3$ has been widely used in the fabrication of Bi superconductors by some research. The utilization of a little amount of Al$_2$O$_3$ dopant can increase the pinning flux and critical current density (Jc) [9], increase the value of Jc [10], and increase phase formation [11].

The method that is widely used in the manufacture of superconductors is the solid reaction method because this method has the advantage of being simple and easy to manufacture and inexpensive costs in making superconductors [12]. Therefore, in this research, the solid reaction method was used in the fabrication of the superconducting Bi$_{1.6}$Pb$_{0.4}$Sr$_2$Ca$_2$Cu$_3$O$_{10}$ with the addition of (x = 1wt% and 2wt%) Al$_2$O$_3$, then characterization was carried out by SEM testing, critical temperature test and XRD. The purpose of this study was to work out the wt% value of addition in the Bi$_{1.6}$Pb$_{0.4}$Sr$_2$Ca$_2$Cu$_3$O$_{10}$ superconducting which affects the good morphological results, critical temperature value, and phase formation.

2. Method
The method utilized in this research is that the solid state reaction method. The precursors utilized in this research were Bismuth III Oxide (Bi$_2$O$_3$) with a PA of 98%, Lead Oxide (PbO$_2$) with a PA of 97%, Strontium Carbonate (SrCO$_3$) with a PA of 96%, Calcium Carbonate (CaCO$_3$) with a PA of 99%, Copper Oxide (CuO) with a PA of 99% and powder of Aluminum Oxide (Al$_2$O$_3$) with a PA of 99%. The equipment used in the manufacture of Bi$_{1.6}$Pb$_{0.4}$Sr$_2$Ca$_2$Cu$_3$O$_{10}$ superconductors material with doping of 1wt% and 2wt% Aluminum Oxide (Al$_2$O$_3$) in this research are spatula, (mortar agate and pastel), digital scales, pellet mold, crucible, press machine, combustion boat and muffle furnace. The characterization test equipment in this study is the SEM-EDX type Jeol JSM-6390A, Cryogenic Type Instrumen Teslatron Oxford, and XRD type Shimadzu X-Ray Diffractometer 7000. The initial stage of making Bi$_{1.6}$Pb$_{0.4}$Sr$_2$Ca$_2$Cu$_3$O$_{10}$ superconducting material with the addition of (x = 1wt%, 2wt%) Aluminum Oxide (Al$_2$O$_3$) is the preparation of tools and materials, then weighing Bi$_2$O$_3$, PbO$_2$, SrCO$_3$, CaCO$_3$, CuO, and Al$_2$O$_3$ with stoichiometric calculations BPSCCO 2223 phase. The powder was mixed and ground using a mortar agate for 3 h. BPSCCO samples were calcined at 300°C with a hold time of 6 h to remove the moisture in the powders. After that, the specimen was ground again with mortar agate for 6 h. And then, the specimen were calculated at 820°C with a hold time of 20 h. The specimen were ground again for an hour with the addition of (x = 1 wt%, 2 wt%) Al$_2$O$_3$. After adding Al$_2$O$_3$ dopant, the sample were pressed (p = 250MPa) into pellets with a thickness of 2 mm and diameter of 10 mm. Then the pellets were sintered at 850°C and sintering time 30 h. Superconducting material Bi$_{1.6}$Pb$_{0.4}$Sr$_2$Ca$_2$Cu$_3$O$_{10}$ with the addition of (x = 1wt%, 2wt%) Aluminum Oxide (Al$_2$O$_3$) were characterized using a SEM-EDX to analyze sample morphology, Cryogenic Magnet to determine the superconductivity properties, and XRD to determine the phase formed.

3. Results and Discussion
Some the results of SEM (Scanning Electron Microscope), Cryogenic Magnets, and X-Ray Diffraction (XRD) analyse are presented in Fig. 1 to 3, respectively.

3.1 Characterization Using SEM (Scanning Electron Microscope)
The results of sample a magnification of 1000X testing using SEM type JEOL JSM-6390A are point in Fig. 1. The morphology of the P1 sample (1wt% Al$_2$O$_3$) in Fig. 1(a) shows that the morphology that is formed looks not homogeneous, the crystalline grains are melt-shaped, agglomerated and the amount of porosity is visible. It is possible that the porosity that occurs is due to the compacting process, which is because the Bi$_{1.6}$Pb$_{0.4}$Sr$_2$Ca$_2$Cu$_3$O$_{10}$ pellet, which is a ceramic-based superconductor, has easy brittle properties and low density. Whereas Fig.1(b) also shows that the morphology formed looks not
homogeneous, the crystalline grains are melt-shaped, clumped and the amount of porosity is low. So, the morphology of sample P2 looks more homogeneous than P1.

Figure 1. Material Morphology Superconductor BPSCCO with magnification 1000X (a) 1wt% Al₂O₃ (P1) (b) 2wt% Al₂O₃ (P2).

3.2 Characterization Using Cryogenic Magnets

The relationship of resistivity to temperature in the manufacture of superconductors Bi₁.₆Pb₀.₄Sr₂Ca₂Cu₃O₁₀ with the addition of (x = 1wt% and 2wt%) Al₂O₃ using a Cryogenic Magnet shown in Fig. 1(a) and Fig. 2(b).

Figure 2. Relationship of Resistivity to Temperature (a) 1wt% Al₂O₃ (P1) (b) 2wt% Al₂O₃ (P2).

Figure 2(a) point that the graph of the characterization of the resistance to temperature relationship in the Bi₁.₆Pb₀.₄Sr₂Ca₂Cu₃O₁₀ superconductors material with the addition of 1wt% Al₂O₃ is formed, it can be seen from the results of the test Tc where the critical temperature for sample P1 has Tc onset = 90 K and Tc zero = 66 K. Figure 2(b) shows that the graph of the characterization of the relationship of resistance to temperature in the Bi₁.₆Pb₀.₄Sr₂Ca₂Cu₃O₁₀ superconductors material with the addition of 2wt% Al₂O₃ is also formed, it can be seen from the results of the crystalline temperature test using Cryogenic Magnet where the critical temperature (Tc) of sample P2 has Tc onset = 94 K and Tc zero = 73
K. The addition of 2wt% Al$_2$O$_3$ causes an increase in critical temperature ($T_c$) compared to the addition of 1wt% Al$_2$O$_3$. The results of the critical temperature test for the superconducting material Bi$_{1.6}$Pb$_{0.4}$Sr$_2$Ca$_2$Cu$_3$O$_{10}$ with the addition of (x = 1wt% and 2wt%) Al$_2$O$_3$ are summarized in Table 1.

### Table 1. The $T_c$\text{onset}$ and $T_c$\text{zero}$ of sample P1 and P2.

| Sample Code | % addition | Sintering | $T_c$\text{onset}$ (K)$ | $T_c$\text{zero}$ (K)$ |
|-------------|------------|-----------|-------------------------|-------------------------|
| P1          | 1 wt% Al$_2$O$_3$ | 850 | 30 | 90 | 66 |
| P2          | 2 wt% Al$_2$O$_3$ | 850 | 30 | 94 | 73 |

3.3 Characterization Using XRD

XRD test results to identify the phases formed in the superconducting material Bi$_{1.6}$Pb$_{0.4}$Sr$_2$Ca$_2$Cu$_3$O$_{10}$ with the addition of dopant (x = 1 wt% and 2 wt%) Al$_2$O$_3$ is shown in Figure 3.

![Figure 3](image.png)

**Figure 3.** Diffraction Pattern of BPSCCO Superconducting Material (a) 1wt% Al$_2$O$_3$ (P1) and (b) 2wt% Al$_2$O$_3$ (P2).

From Figure 3(a) it can be seen that the phases formed in sample P1 are Bi$_{0.04}$Sr$_{3.2}$Ca$_{5.54}$Cu$_{15.158}$O$_{28.312}$, Bi-2212 and Bi-2223 with SrCuBiO and SrO as an impurity. The Bi-2212 phase is optimal at intensity of 486.81 count second with an 2θ of 31.07, the Bi-2223 phase is optimal at intensity of 244.71 count second with an 2θ of 33.23, and the Bi$_{0.04}$Sr$_{3.2}$Ca$_{5.54}$Cu$_{15.158}$O$_{28.312}$ phase is optimal at intensity of 973.82 count second with an 2θ of 32.10. Whereas in Figure 3(b) it can be seen that the phases formed in P2 are Bi$_{0.31}$Sr$_{4.05}$Ca$_{5.64}$Cu$_{17}$O$_{29}$, Bi-2212 and Bi-2223 with SrCuBiO as impurities. The Bi-2212 phase is optimal at intensity of 559.22 count second with an 2θ of 31.10, the Bi-2223 phase is optimal at intensity...
of 323.31 count second with an 2θ of 33.19 and the Bi$_{0.31}$Sr$_{1.05}$Ca$_{5.64}$Cu$_{7.29}$O$_{29}$ phase optimal at intensity of 1107.85 count second with an 2θ of 32.11.

The results of the above analysis prove that the addition of wt% Al$_2$O$_3$ adds impurity to the superconductor Bi$_{1.6}$Pb$_{0.4}$Sr$_2$Ca$_3$Cu$_4$O$_{10}$. This is indicated by the presence of an impurity phase in the addition of Al$_2$O$_3$, namely SrO and SrCuBiO. As a result of the formation of the impurity phase, the critical temperature of the Bi$_{1.6}$Pb$_{0.4}$Sr$_2$Ca$_3$Cu$_4$O$_{10}$ materials is reduced [13]. Sample P1 and sample P2 were identified by producing the same phase, namely the Bi-2212 phase, but the difference between the two samples can be seen in the wt% addition of Al$_2$O$_3$ and sample P2 resulting in an intensity of Bi-2223 phase higher than sample P1. The percentage of purity of the Bi-2212 phase can be determined by calculating the volume fraction ratio. The value of the volume fraction and crystal size of each sample is shown in Table 2.

### Table 2. The crystal size and volume fractions of Bi2212 and Bi2223.

| Sample Code | Crystal Size (nm) | Volume Fraction (%) |
|-------------|-------------------|---------------------|
|             | Bi2212            | Bi2223              |
| P1          | 93.12             | 59.21               | 40.79               |
| P2          | 90.80             | 52.11               | 47.89               |

Table 2 of crystal size analysis pointed that addition of 2wt% Al$_2$O$_3$ to the superconductor Bi$_{1.6}$Pb$_{0.4}$Sr$_2$Ca$_3$Cu$_4$O$_{10}$ can reduce the crystal size when compared to the addition of 1wt% Al$_2$O$_3$. The smallest crystal size was obtained in the manufacture of Bi$_{1.6}$Pb$_{0.4}$Sr$_2$Ca$_3$Cu$_4$O$_{10}$ superconductors by adding 2wt% Al$_2$O$_3$ at sintering temperature of 850°C and sintering time of 30 h. And the table point that comparison of volume fraction Bi-2212 and Bi-2223 in the P1 sample with the addition of dopant 1wt% Al$_2$O$_3$ is 59.21% and 40.79%, while in P2 with the addition of 2wt% Al$_2$O$_3$ dopant is 52.11% and 47.89%. Based on the volume fraction, the making of the Bi$_{1.6}$Pb$_{0.4}$Sr$_2$Ca$_3$Cu$_4$O$_{10}$ superconductor with the addition of 2wt% Al$_2$O$_3$ can increase the Bi-2223 phase although the dominant phase remains in Bi-2212 phase.

### Conclusions

From the results of research on increasing the Tc in Bi$_{1.6}$Pb$_{0.4}$Sr$_2$Ca$_3$Cu$_4$O$_{10}$ superconductors with the addition of Al$_2$O$_3$ it can be concluded that addition of 2wt% Al$_2$O$_3$ in the manufacture of Bi$_{1.6}$Pb$_{0.4}$Sr$_2$Ca$_3$Cu$_4$O$_{10}$ superconductors produces a higher critical temperature (Tc) than the addition of 1wt% Al$_2$O$_3$. In addition, the more addition of Al$_2$O$_3$ can reduce grain size and the morphology is more homogeneous. The optimal critical temperature (Tc) was produced, namely $T_{c_{\text{room}}} = 94$ K and $T_{c_{\text{zero}}} = 73$ K in the addition of 2wt% Al$_2$O$_3$ with the formed phase Bi-2212. The crystal size analysis showed that the addition of 2wt% Al$_2$O$_3$ to the superconductor Bi$_{1.6}$Pb$_{0.4}$Sr$_2$Ca$_3$Cu$_4$O$_{10}$ can reduce the crystal size when compared to the addition of 1wt% Al$_2$O$_3$.

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