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The Preparation of $(Tl_1Pb_{1-x}M_x)Sr_2Ca_2Cu_3O_{9-\delta}$ Superconductor via Stoichiometric Reaction

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Abstract

The $(Tl_1Pb_{1-x}M_x)Sr_2Ca_2Cu_3O_{9-\delta}$ superconductor with M=Bi and Cr has been prepared by solid state reaction method. The preparation of the precursor material using metal oxides powder of high purity(>99.99%) Strontium Oxide, Calcium Oxide and Copper Oxides. The powder were mixed completely and heated at 900^{o}C for 48 hours with several intermediate grindings. An appropriate amounts of Tl_2O_3 and Cr_2O_3/Bi_2O_3 (mole%) were then added to the precursor and completely mixed before being pressed into pellets form of 1.3 cm in diameter and 0.2 cm in thickness under 7 ton/cm^2 of pressure and then first heated at 900°C in the flowing oxygen for 4 minutes, followed by furnace cooling to room temperature. The dc electrical resistance measurement in a range of 300K-50K were carried out using the four-point method with silver paste contact in conjunction with a closed cycle refrigerator from CTI Cryogenic Cycle Refrigerator Model 22. We observed that $T_{c\ zero}$ was changed from 69K to 70K and $T_{c\ onset}$ from 88K to 90K with addition of 0.1mole-% Cr_2O_3. When 0.2 mole-% Cr_2O_3 was introduced, the $T_{c\ zero}$ decresed to 65K. It has been noted the addition of Bi_2O_3 could not enhanced the $T_{c\ zero}$ as well as $T_{c\ onset}$. All samples showed metallic characteristic at normal state.

Keywords: superconductor; cryogenic; $T_{c\ onset}$; $T_{c\ zero}$, and normal state.

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1. Introduction

Starting from the discovery of high Tc superconductivity by Bednorz and Muller until now, a great amount of researches contribute to superconductivity. Several workers around the word not only explored several precursor materials but also have studied several systems. It has been known that Thallium system is the most promising material in the improvement of critical temperature. Sheng and Hermann [2] discovered high temperature superconductivity (HTSC) in the Tl-Ba-Ca-Cu-O system. As a consequently, many new superconducting systems and phases have been reported [1]. The Tl-Ba-Ca-Cu-O system high temperature superconductor is one of the most promising candidates in terms of critical temperature since its high Tc [1-7]. Several techniques such as: the preparation of precursor [2-5], the role of the doping [6,7,8,11,12] and the modification of sintering temperature [13,14] have been applied to the superconducting material in order to increase the transition temperature.

The basic structure of Tl-Ba-Ca-Cu-O superconducting phases contains $(Tl\text{-}O)_2$ bilayers or (Tl-O) monolayer separated by BaO-CuO_2-Ca- CuO_2-BaO layers [3]. The transition temperature for Tl-Ba-Ca-Cu-O system is in the range of 85K-125K [1] has been reported. In this paper we study the $(Tl\text{-}O)_2$ trilayers with the formula of $(Tl_1Pb_{1-x}M_x)Sr_2Ca_2Cu_3O_{9-\delta}$ (x= 0.0 - 0.3) by solid state reaction. Chen and his colleagues [14] and Sheng and his colleagues [15] reported that Cr atom has an ability to improve the phase formation. All of them reported the enhancement of formation Tl-1201 phase and Tl-1212 phase respectively by Cr substitution. However, little information about Tl-1223 phase. Cr-substituted $TlSr_2CaCu_2O_{7-\delta}$ compounds are commonly synthesized via the conventional solid-state method, as the reaction proceeds by diffusion in the solid state, it requires mixing and grinding of high purity powder of oxides and/or carbonates for long hours before heating the mixtures at high temperature [13]. The high temperature processing of the Tl superconductors is further complicated by the volatility of Tl_2O_3, which as a result produced superconducting oxides that are highly porous and of low density [4]. Whereas M ASubramaniam and his colleagues [16] reported that the highest transition temperatures was observed when the samples were partially melted at > 900^{o}C. They also reported that Tc zero of 110K and Tc onset of 112K were achieved.Faizah and his colleagues [8] reported that Tc zero=95K and Tc onset 112K and samples showed existence of dominant Tl-2212 phase along with small impurity phases. They also reported that the addition of Ag showed little effect on Tc values and showed a lowering of Tc to around 70K. Critical temperatures around 100K [14] and 104K [15] have been achieved for the doped Tl-1212 and Tl-1223 phases. A Tc of 100K of the Bi-substituted sample in $(Tl,Bi)Sr_2CaCu_2O_7$ has also been reported [16]. Pb-doped $(Cu_{0.5-x}Pb_xTl_{0.5})Ba_2Ca_2Cu_3O_{10-\delta}$ superconductor was reported with a Tcof 115K [23]. It can be seen that the Rare Earth elements were found to favor the formation of the Tl-1223 [17]. It is interesting to compare the effect of Bi and Cr on the superconductivity and formation of the Tl-1223 phase. In this research, we prepared Bi substitued Tl-site to produce $(Tl_1Pb_{1-x}Bi_x)Sr_2Ca_2Cu_3O_{9-\delta}$ and $(Tl_1Pb_{1-x}Cr_x)Sr_2Ca_2Cu_3O_{9-\delta}$ with x=0, 0.1and 0.2 respectively.

2. Experiment Procedure

Samples with the nominal starting composition of $(Tl_1Pb_{1-x}M_x)Sr_2Ca_2Cu_3O_{9-\delta}$ (Tl-1223) with x = 0.0, 0.1, and 0.2 were prepared by the solid state reaction method. An appropriate amounts of high purity (>99.99%) Strontium Oxide, Calcium Oxide and Copper Oxides were mixed completely using an agate mortal to obtain a

homogeneous mixture. The precursor powder were heated at 900°C for 24h with twice intermittent grinding. An appropriate amounts of Tl_2O_3 and Cr_2O_3according to mole% were then addedto the precursor and completely mixed before being pressed into pellets form of 1.3 cm in diameter and 0.2 cm in thickness under 7 ton/$cm^2$of pressure and then first heated at 900°C in the flowing oxygen for 4 minutes, followed by furnace cooling to room temperature. In order to compensate thallium loss during heating, excess of 10%Tl_2O_3 were added in.The dc electrical resistance measurements in a range of 300K-30K were carried out using the four-point method with silver paste contact in conjunction with a closed cyclerefrigerator from CTI Cryogenic Cycle Refrigerator Model 22 and a temperature controller from Lake Shore Temperature Controller Model 340 for temperature-dependent measurements. A constant current source between 1 and 100mA was used throughout the measurements. The powder X-ray diffraction (XRD) pattern were recorded using a Bruker model D8 Advance diffractometer with CuK_αradiation.

Theoretically, the stoichiometric reaction can be written as:

$$2SrO_3 + CaO + 2CuO \longrightarrow Sr_2CaCu_2O_5 + 2CO_2$$

The sintering step can be presented as follows:

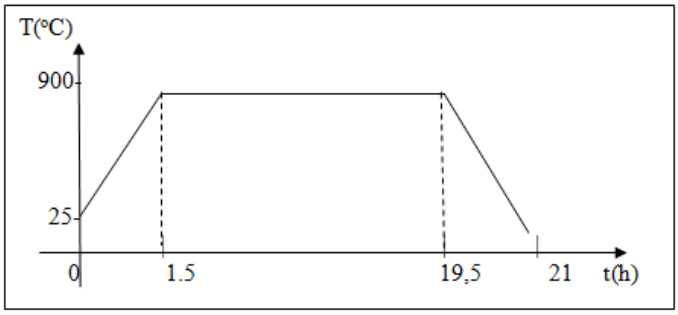

Figure 1: Sintering process of precursor material $Sr_2CaCu_2O_5$

The next step is graining process for 1 hour ,sintering process is applied to the powder for second time for 6 hours at 900°C. The cooling process is allowed to room temperature with addition of Tl_2O_3, PbO and Bi_2O_3/Cr_2O_3:

$$Tl_2O_3 + 0.9PbO + Sr_2CaCu_2O_5 + 0.05Bi_2O_3 \longrightarrow (Tl_1Pb_{0.9}Bi_{0.1})Sr_2Ca_2Cu_3O_{9-\delta}$$

$$Tl_2O_3 + 0.9PbO + Sr_2CaCu_2O_5 + 0.05Cr_2O_3 \longrightarrow (Tl_1Pb_{0.9}Cr_{0.1})Sr_2Ca_2Cu_3O_{9-\delta}$$

3. Results and Discussion

The electrical resistance versus temperature curve of $(Tl_1Pb_{1-x}M_x)Sr_2Ca_2Cu_3O_{9-\delta}$ samples for x=0.0-0.2 are shown in Figure 2 and Figure 3 respectively. From the Figure 2 it can be seen that the addition 0.1 mole-% Bi_2O_3 has increased the resistance of the superconductor. Thus, the sample has a lower critical temperature compared to that the pure sample. When 0.1 mole-% Cr_2O_3 was introduced, we observed that the resistance

decreased. The lower resistance lead to the increasing of critical temperature. The resistance graph of $TlPbSr_2Ca_2Cu_3O_{9-\delta}$ shows $T_{c\ onset}$ around 88K and $T_{c\ zero}$ at 69K. It is also clearly be seen that the sample without addition of Bi/Cr shows a metal characteristic at normal state. The addition of 0.1 mole-% Bi as well as 0.1 mole-% Cr also show a metal characteristic at normal state. From the Figure 2, we noted that the pure sample has two $T_{c\ onset}$, the critical temperature where the resistance decreased gradually. For this sample, the $T_{c\ zero}$, where the resistance abruptly drop to zero was observed at 69K. This temperature increased to 70K when 0.1 mole% Cr_2O_3 was introduced. This composition is in high temperature superconductor [9]. The resistance history of the sample with addition of 0.2mole-% Bi_2O_3 as can be seen in Figure 3.

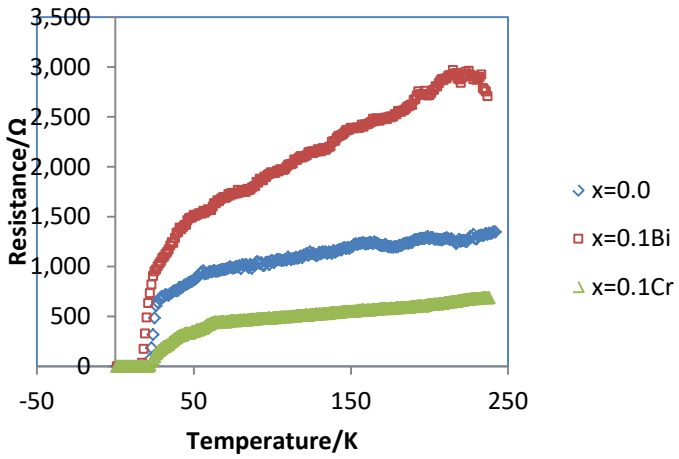

Figure 2: The dc electrical resistance towards the temperature

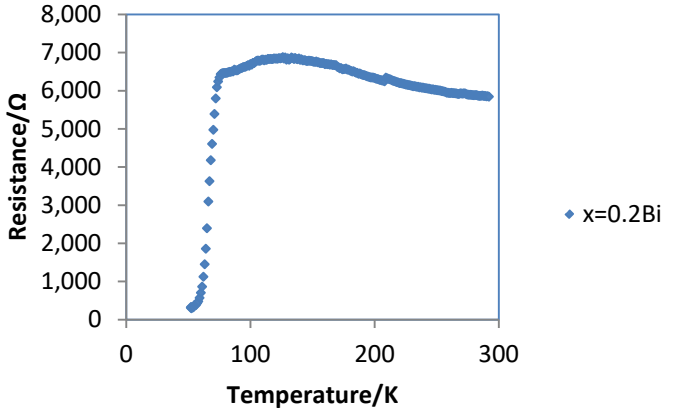

Figure 3: The dc electrical resistance of $Tl_1Pb_{0.8}Bi_{0.2}Sr_2Ca_2Cu_3O_{9-\delta}$

Figure 3 shows the decreasing of $T_{c\ zero}$ from 69K to 65K in the composition of $Tl_1Pb_{0.8}Bi_{0.2}Sr_2Ca_2Cu_3O_{9-\delta}$. This due to the phase unstability with higher ammount of Bi_2O_3. As it can be seen this sample also shows the over doped condition, we could not increase the content of Bi_2O_3 furthermore. The resistance of the sample increases at this composition. Figure 4 shows the resistance history with addition of 0.3mole-% Cr_2O_3. The tail occurrence at normal state might be due to the ohmic contant. At normal state, the sample shows a metal-like characteristic.

The decreasing of T_c in this composition due to the presence of the other phase as a result of sintering process. However, we need XRD-analysis software to calculate the major phase and minor phase occurrence in each composition. The results will be reported elsewhere.

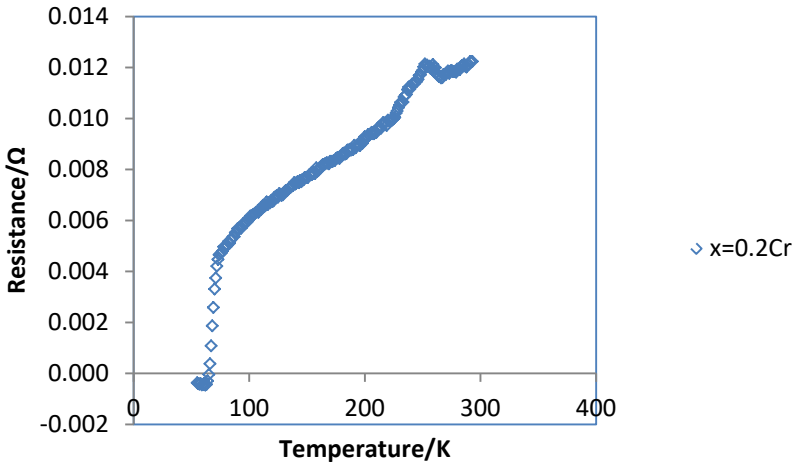

Figure 4: The dc electrical resistance of $Tl_1Pb_{0.8}Cr_{0.2}Sr_2Ca_2Cu_3O_{9-\delta}$

We can observe the $T_{c\ zero}$ for this composition around 64K and the $T_{c\ onset}$ at 72K. It means that the addition of 0.2mole% Cr_2O_3 could not enhance the $T_{c\ zero}$ and $T_{c\ onset}$. The resistivity of the samples at room temperature versus Cr_2O_3/ Bi_2O_3 content as presented in Figure 5. As it can be seen the optimum resistivity occurred when 0.1mole% Cr_2O_3 was introduced. As such, this composition is reasonable to be developed in the future work.

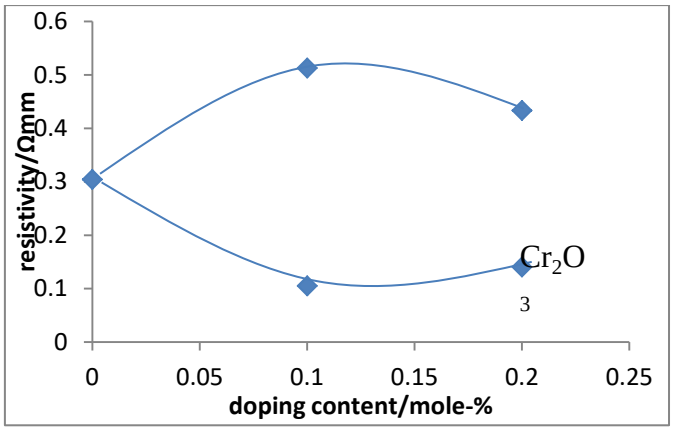

Figure 5: Graph of resistivity at room temperature

4. Conclusion

In conclusion, samples with nominal starting composition $(Tl_1Pb_{1-x}M_x)Sr_2Ca_2Cu_3O_{9-\delta}$ with M=Bi and Cr have been prepared via solid state reaction. The results showed that the addition of 0.1 mol% Cr_2O_3 has changed the

$T_{c\ zero}$ from 69K to 70K, but the addition of 0.2mole% Cr_2O_3 decreased it to 65K. The addition of 0.1mole% and 0.2mole% Bi_2O_3 did not show any influence to the $T_{c\ zero}$ as well as $T_{c\ onset}$.

5. Recommendation

The composition of $Tl_1Pb_{0.9}Cr_{0.1}Sr_2Ca_2Cu_3O_{9-\delta}$ is recommended as a starting material in the future work. A higher purity of the precursor powder is also to be considered in the sample preparation.

Acknowledgement

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References

[1] Faizah M Salleh, A K Yahya, I Hamadneh and R Abd-Shukor, " Effects of Ag Addition and Properties of Tl-2212 Superconductors",Solid State Science and Technology, Vol.10 No 1 & 2 pp 30-36, 2002

[2] Jorda, J L, HopfingerlTh, Couach M, Pugnat P, Bertrand C &Galezl, Ph, Formation of Thallium-1223 Superconductors.Journal of Superconductivity, **11**(1):87-89, 1998

[3]Kaneko.,Tetsuyuki., Hamada., Kazuyuki., Adachi., Seiji., Yamauchi, H., Tanaka & Shoji, Synthesis of Tl based "2234" superconductors, Journal of Applied Physics **71** (5):2347-2350, 1992

[4] Kikuchi, M., Ohshima, E., Ohnishi, N., Muraoka, Y., Nakajima, S., Aoyagi, E., Ogawa, M., Akimitsu, J., Oku, T., Hiraga, K &Syono, Y., Synthesis and superconductivity of oxycarbonates of the Tl-1201 phase. Physica C **219** (1-2):200-204, 1994

[5] Kolesnikov, N. N., Korotkov, V. E., Kulakov, M. P., Shibaeva, R. P., Molchanov,V. N., Tamazyan, R. A & Simonov, Structure of superconducting single crystal of 2201 Thallium Cuprate ($Tl_{1.85}Cu_{0.15}Ba_2CuO_6$), Tc = 110 K, Physica C **195** (3-4):219-224, 1992

[6] Maignan A, Martin C, Hardy V, Simon Ch, Hervieu M &Raveau B, A crystal study of the 128K superconductor $Tl_2Ba_2Ca_2Cu_3O_{10-\delta}$ Synthesis and anisotropic magnetic properties. Physica C **157**: 460-468, 1994

[7] Mega Fatmasari, KesuperkonduksiandanPembentukanFasa $(Tl_{0.5}Pb_{0.5-x}M_x)Sr_2CaCu_2O_7$ dengan M=Bi dan Cr untuk x = 0, 0.1, 0.2, 0.3, 0.4 dan 0.5, Thesis, FST, UKM, 2011

[8] Abd-Shukor R, BaharuddinYatim& Lau R S L, Formation of Tl-1212 phase in $(Tl_{0.8}M_{0.2})Sr_2(Ca,Cr)Cu_2O_7$ (M: Cd and Sc) High-Tc Superconductor, Journal of Materials Science Letters **16**: 818-820, 1997

[9] Abd-Shukor R, Introduction to Superconductivity: in Metals, Alloys & Cuprates, Penerbit Universiti

Pendidikan Sultan Idris, Tanjong Malim, 2004

[10] Sheng Z Z& Hermann A M, Superconductivity in the rare earth free Tl-Ba-Cu-O system above liquid nitrogen temperature, Nature **332**: 55-58, 1998

[11] Syahrul Humaidi, Achiruddin and Tua Raja Simbolon, The Development of the superconductor Critical Temperature (Tc), Jurnal Reintek, Volume 7 no.1: 28-32, 2012

[12] Syahrul Humaidi, Awan Maghfirah and Tua Raja Simbolon, The Preparation of Thallium Cuprum Oxide superconductor precursor ($Tl_2Ba_2CaCu_2O_{8-\delta}$), Research report, Dana PNBP FMIPA USU 2012, SK No. 45/ UN5.2.3.1/ PPM/SKP/2013, LP USU, 2012

[13] Syahrul Humaidi, The Effect of Cr_2O_3 Addition on the Critical temperature development and Phase Stability of ($Tl_2Ba_2CaCu_2O_{8-\delta}$) high Temperature Superconductor, Final Report PDD, DIKTI RI-LP USU, 2014

[14] Gritzner, G Sudra, H eder, M., J. Phys. Conf. Ser. 43, 462, 2006

[15] Sundradesan, A Asada H, Crisan , A, Nie, J C, Kito H, Iyo, A, Tanaka, Y Kusunoki, M Ohsima S, Physica C 388-389, 473, 2003

[16] Abd Shukor, R Tee, K S, J Mater. Sci, let. **17**, 103, 1998

[17] Eder, M E, Gritzner, G,Supercond. Sci. Technol. **18**, 87, 2005