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Formation of Tl-1212 Phase in CR Substituted $(Tl_{1-x}Cr_x)$ $Sr_2CaCu_2O_7$ Superconductor

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ABSTRACT

Superconducting samples of $(Tl_{1-x}Cr_x)$ $Sr_2CaCu_2O_7$ for x = 0.0-0.7 have been prepared by a solid state reaction technique at normal pressure using high purity elements. The powder X-ray diffraction patterns have been studied for samples. All samples showed the Tl-1212 as the major phase and Tl-1201 as the minor phase. The superconducting transition temperature Tc has been determined from the electrical resistivity measurements. The data of Tc, for $(Tl_{1-x}Cr_x)$ $Sr_2CaCu_2O_7$ show enhancement in its value from 104 K to112 K as x (Cr-content) increases from 0.7 to 0.1. Our results shows the highest onset temperature for x = 0.2-0.3 and the lowest temperature for x = 0.7.

Keywords: Phase Formation, Tl-1212 Superconductor, Cr-Substitution, High Tc, Superconducting Transition, Diffraction Patterns, Reaction Technique, Solid State, Powder X-Ray

1. INTRODUCTION

The Tl-based cuprate high temperature superconductor continues to be an interesting family of material because it can form many phases. It is an ideal system to study the role of metal oxide and copper oxide layers in high temperature superconductivity. The studies of high temperature materials have been necessary to develop conductors for practical and also the studies of the phase formation can be useful to find out the crucial role of the oxygen partial pressure during the sintering process. The TlBaCaCuO (Tl/Ba) and TlSrCaCuO (Tl/Sr) make up the Tl-based cuprate superconductor. The Tl/Ba system can be written as $Tl_2Ba_2Ca_{n-1}Cu_nO_z$ with n = 1-4 and $TlBa_2Ca_{n-1}Cu_nO_z$ with n = 1-5. The Tl/Sr system can be written as $TlSr_2Ca_{n-1}Cu_nO_z$ with n = 1-3 (Lee, 2011). The three phases in the Tl/Sr system are $TlSr_2CuO_5$ (Tl/Sr-1201), $TlSr_2CaCu_2O_7$,(Tl/Sr-1212), $TlSr_2Ca_2Cu_3O_{10}$ (Tl/Sr-1223). But still not too many reports on Tl-1234 phase. There are some elements for raising the transition temperature the Tl-1212 phase. Substitution of rare earth cations into the C^{a2+} sites (Eder and Gritzner, 2005; Yip and Abd-Shukor, 2009) and Sr^+ sites (Eder and Gritzner, 2005; Lee and Huang, 1997) and partial replacement of Tl^{3+} by either Pb^{4+} (Subramanian *et al*., 1988), Bi^{3+} (Li and Greenblatt, 1988) or Cr^{3+} (Sheng *et al*., 1989; Li *et al*., 1995; Abd-Shukor and Jaafar, 1999) may assist the formation of Tl-1212 phase. Critical temperature around 100 K of the Bi-substituted sample in Tl-1212 (i.e., $(Tl,Bi)Sr_2CaCu_2O_7$) has been reported (Abd-Shukor and Tee, 1998), Pb-doped $(Cu_{0.5-x}Pb_xTl_{0.5})$ $Ba_2Ca_2Cu_3O_{10-\delta}$ superconductor was reported with a Tc of 115 K (Nawazish and Ullah, 2010). The effect of rare earth elements was found to favor the formation of the Tl-1212 phase (Eder and Gritzner, 2005).

And several researches were studied on the effect of partial substitution of Ca with R (R = Y and most rare-earths) in $TlSr_2C_aCu_2O_7$ (Kondo *et al*., 1991). Also transition temperature of $TlSr_2CaCu_2O_7$ phase was found to increase up to 110 K when Cr was substituted at different metal sites (Sheng *et al*., 1991). Therefore Tl-1234 superconducting phase was prepared by (Ihara *et al*., 1997) under the high pressure of 3.5 GPa at 850-9500°C for 2 h in a gold capsule (Iyo *et al*., 2001). But sintering technique in the high-pressure is not a suitable practical application.

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Later on (Nawazish and Mumtaz, 2007) were prepared $Cu_{0.5}Tl_{0.5}Ba_2Ca_3Cu_{4-y}Zn_yO_{12-\delta}$ by using two step of solid state reaction method with ZnO, Ba(No$_3$)2, $Ca(No_3)_2$ and Cu(CN) as starting compound. Recently, was studied effect of Mg nano-oxide addition on superconductivity of $Cu_{0.25}Tl_{0.75}Ba_2Ca_3Cu_4O_{12-\delta}$ by using a single step solid state reaction method (Mohammed, 2012). The samples in this study were synthesized under normal pressure with Cr substitution at the Tl site.

It is interesting to studied the effect of Cr on the superconductivity of $(Tl_{1-x}Cr_x)$ $Sr_2Ca_3Cu_4O_{11}$ phase. Also Cr was substituted to Tl-1234 due to never any researcher was studied on $(Tl_{1-x}Cr_x)$ $Sr_2Ca_3Cu_4O_{11}$ phase. Although in this research was attempted to prepare a Tl-1234 phase from base materials, but the final samples are shown a Tl-1212 phase.

In this study superconductivity of samples by using a solid state reaction method was studied for x = 0.0-0.7.

2. MATERIALS AND METHODS

2.1. Materials

Samples with nominal starting composition of $(Tl_{1-x}Cr_x)$ $Sr_2Ca_3Cu_4O_{11}$ with x = 0.0, 0.1, 0.2, 0.3, 0.4, 0.5, 0.6 and 0.7 were mixed by the solid state reaction method using metal oxides powders. Appropriate amounts of high purity (>99.99%) CuO nano size, Sr_2O_3 and CaO were mixed completely using an agate mortal to obtain a homogeneous mixture. The precursor powders were heated at 900°C for 24 h with several intermittent grindings. Appropriate amounts of Tl_2O_3 and Cr_2O_3 were then added to the precursor powders, completely mixed and then pressed into pellets form of 1.3 cm diameter and 0.2 cm thickness. These pellets were then wrapped in a silver foil to reduce possible volatilization of Tl. The wrapped materials were first heated at 870°C in the flowing oxygen for 1 hour, followed by furnace cooling to room temperature. In order to compensate thallium loss during heating, excess 10% of Tl_2O_3 were added.

2.2. Characterization Methods and Instrumentation

The powder X-ray diffraction method using a Bruker D8 Advance diffractometer with Cukα source was used to identify the resultant phases. The volume fraction of superconducting phases was estimated by assuming that the amount of the phases are proportional to the strongest diffraction line of each phase, i.e., the (110) reflection of the 1212 and the (200) reflection of the 1201 phase.

The electrical resistance versus temperature measurements was carried out using the four-point method with silver paste contacts in conjunction with closed cycle refrigerator from CTI Cryogenic (Model 22) and a temperature controlled from Lake Shore (Model 330). A constant current source between 1 and 100mA was used throughout the measurements. The Tc onset is defined as the temperature where there is a sudden drop in the resistance.

3. RESULTS AND DISCUSSION

The room temperature Powder X-Ray Diffraction (PXRD) patterns for $(Tl_{1-x}Cr_x)$ $Sr_2CaCu_2O_7$ superconducting samples (x = 0.0-0.7) are shown in **Fig. 1 and 2**. The diffraction lines of the patterns are well indexed by a tetragonal unit cell of Tl-1212 with space group P4/mmm. It is interesting to note that although for all samples were used from base materials to prepare Tl-1234 but formation of PXRD patterns shows the Tl-1212 as a major phase and Tl-1201 as a minor phase. It is due to Sr_2O_3 in the precursor powder. For improving the formation of Tl-1234 in the $(Tl_{1-x}Cr_x)$ $Sr_2Ca_3Cu_4O_{11}$ samples should substituted Sr_2O_3 by other elements such as BaO_2 (Mohammed *et al*., 2010).

The amounts of Tl-1212 phase are assumed to be proportional to the intensity of their strongest diffraction lines the (103, 110 and 111 reflection of the 1212 phase and the 200 reflection of the 1201 phase) contained in the samples.

The comparison between the PDXD patterns determined that intensity of samples was decreased with increasing of x. In addition the strongest diffraction lines of Tl-1212 phase in the PXRD were appeared sharply for x = 0.2-0.3.

The electrical resistance versus temperature curves of $(Tl_{1-x}Cr_x)$ $Sr_2CaCu_2O_7$ samples for x = 0.0-0.3 are shown in **Fig. 3** and for x = 0.4-0.7 are shown in **Fig. 4**. The highest Tc (onset and zero resistance) are determine for x = 0.2-0.3 at (112 K and 93 K) respectively and then Tc onset are decreased to 103 K for x = 0.7.

As shown in **Fig. 3** the x = 0.0 sample shown an insulator behavior and samples for x = 0.1-0.6 are shown a metallic normal state behavior. Also samples with x = 0.2 and x = 0.3 are shown the highest Tc onset (112 K). Meanwhile, sample with x = 0.7 shown the lowest Tc onset (103 K) and shown a semiconductor behavior **Fig. 4**.

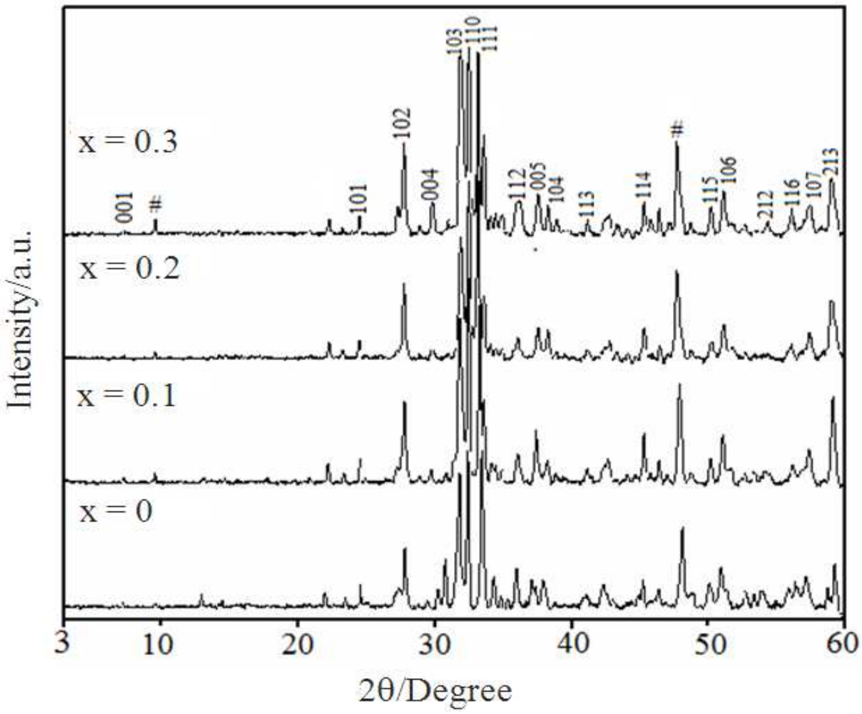

Fig. 1. X-ray powder diffraction patterns of $(Tl_{1-x}Cr_x)\ Sr_2CaCu_2O_7$ for *x* = 0.0-0.3. (#) indicat the Tl-1201 phase

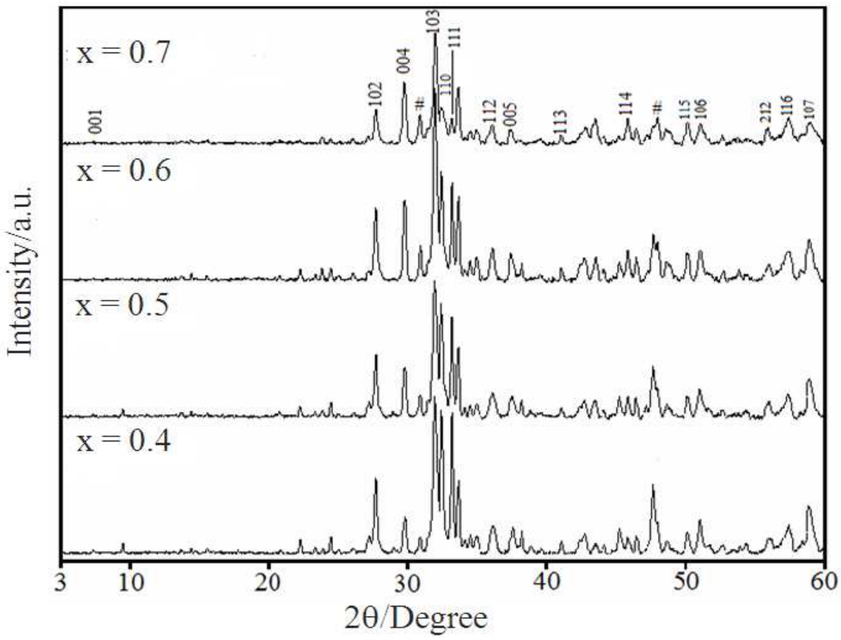

Fig. 2. X-ray powder diffraction patterns of $(Tl_{1-x}Cr_x)\ Sr_2CaCu_2O_7$ for *x* = 0.4-0.7. (#) indicat the Tl-1201 phase

Figure 5 shows the variation of Tc onset with Cr content(x) for all samples. As shown in (**Fig. 5**) the Tc onset was increased from 103K to 112 K for x = 0.7 to x = 0.2-0.3 respectively.

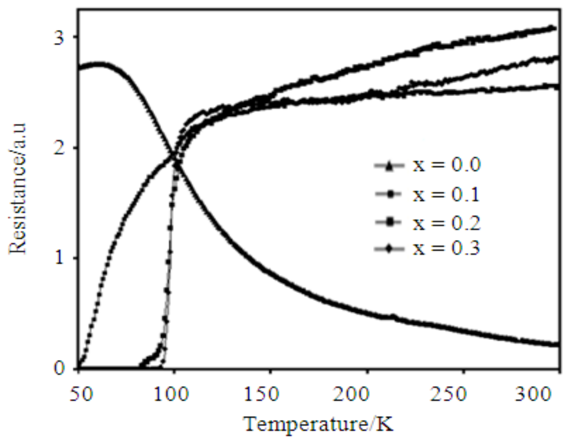

Fig. 3. Resistance versus temperature curves of $(Tl_{1-x}Cr_x)$ $Sr_2CaCu_2O_7$ for *x* = 0.0-0.3

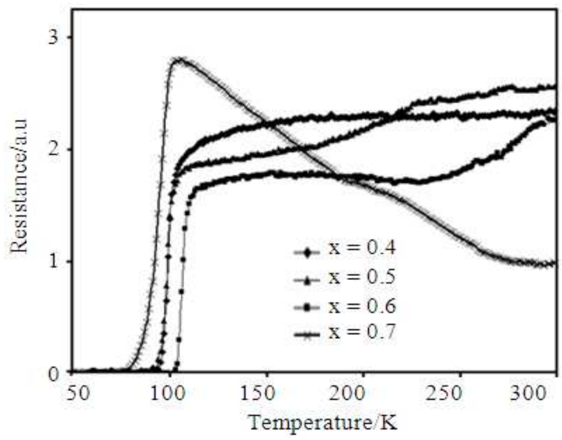

Fig. 4. Resistance versus temperature curves of $(Tl_{1-x}Cr_x)$ $Sr_2CaCu_2O_7$ for *x* = 0.4 -0.7

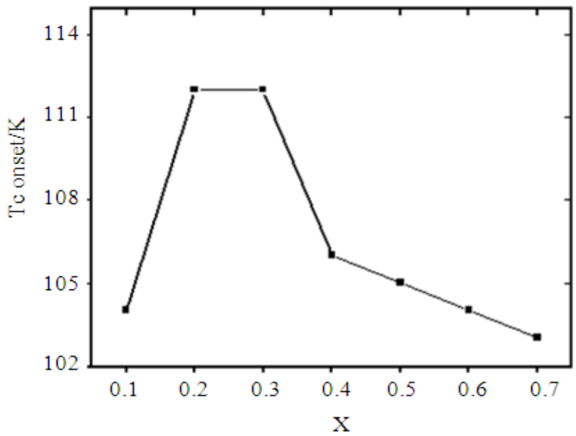

Fig. 5. Tc onset as a function of *x* in $(Tl_{1-x}Cr_x)$ $Sr_2CaCu_2O_7$

Table 1. Tc (onset and zero) of $(Tl_{1-x}Cr_x)$ $Sr_2CaCu_2O_7$ for samples *x* = 0.0-0.7

x	$T_{c\ onset}$ (K)	$T_{c\ zero}$ (K)
x = 0.0	Insulator	Insulator
x = 0.1	104	47
x = 0.2	112	92
x = 0.3	112	93
x = 0.4	106	92
x = 0.5	105	93
x = 0.6	104	93
x = 0.7	103	78

The Tc (onset and zero resistance) of $(Tl_{1-x}Cr_x)$ $Sr_2CaCu_2O_7$ samples are shown in **Table 1.**

4. CONCLUSION

In this present study, we have synthesized $(Tl_{1-x}Cr_x)$ $Sr_2CaCu_2O_7$ phase from the starting oxide elements by a solid state reaction technique at normal pressure and studied the effect of Cr substitution on it. Also it was successfully to find out a high Tc for all samples. The superconducting transition temperature enhanced as x increased from 0.1-0.3 and then decreased when x increased from 0.4-0.7. The PXRD patterns indicated a Tl-1212 as a major phase and Tl-1201 as a minor phase. To improve the formation of $(Tl_{1-x}Cr_x)$ $Sr_2CaCu_2O_7$ phase have to substitute Sr_2O_3 by other elements such as BaO_2 to can obtain a $(Tl_{1-x}Cr_x)$ $Sr_2Ca_3Cu_4O_{11}$ phase.

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6. REFERENCES

Abd-Shukor, R. and K.S. Tee, 1998. Effectiveness of Bi versus Pb for superconductivity in the $TlSr_2CaCu_2O_7$ (1212) phase. J. Mater. Sci. Lett., 17: 103-106. DOI: 10.1023/A:1006522530907

Abd-Shukor, R. and N.A.N. Jaafar, 1999. Formation and superconductivity of Pr and Nd-substituted Tl-1212 phase $(Tl_{0.85}Cr_{0.15})$ $Sr_2CaCu_2O_7$. J Mater. Sci.: Mater. Elect., 10: 677-681. DOI: 10.1023/A:1008994704163

Eder, M.H. and G. Gritzner, 2005. Thallium-based cuprate superconductors doped with rare earth oxides. Supercond. Sci. Technol., 18: 87-91. DOI:10.1088/0953-2048/18/1/014

Ihara, H., K. Tokiwa, K. Tanak, T. Tsukamoto and T. Watanabe *et al*., 1997. $Cu_{1-x}Tl_xBa_2Ca_3Cu_4O1_{2-y}$ ($Cu_{1-x}Tl_x$-1234) superconductor with Tc=126 K. Physica C: Superconductivity, 282: 957-958. DOI: 10.1016/s0921-4534(97)00592-3

Iyo, A., Y. Aizawa, Y. Tanaka, M. Tokumoto and K. Tokiwa *et al*., 2001. High-pressure synthesis of $TlBa_2Ca_{n-1}Cu_nO_y$ (n = 3 and 4) with Tc = 133 K (n = 3) and 127 K (n = 4). Physica C.; Superconductivity, 357: 324-328. DOI: 10.1016/S0921-4534(01)00238-6

Kondo, T., Y. Kkubo, Y. Shimakawa, T. manako and H. Igarashi, 1991. Oxygen nonstoichiometry and Tc -variation in $TlSr_2$ ($Ca_{1-x}Lu_x$) Cu_2O_y (x = 0-1.0). Physica C.: Superconductivity, 185: 669-670. DOI: 10.1016/0921-4534(91)92137-z

Lee, H.K., 2011. Effect of Ba substitution on the formation of (Tl,Bi)-1234 superconducting phase. J. Supercond. Nov. Magnetism, 24: 2183-2187. DOI: 10.1007/s10948-011-1179-4

Lee, W.H and B.C. Huang, 1997. Changes in doping state of Tl $(Sr_{2-x}R_x)CaCu_2O_{7-\delta}$ (R = Sm, Eu, Dy) system. Physica C., 289: 114-122. DOI: 10.1016/s0921-4534(97)01581-5

Li, S. and M. Greenblatt, 1988. Preparation and superconducting properties of (Tl, Bi)$Sr_2CaCu_2O_y$. Physica C.: Superconductivity, 157: 365-369. DOI: 10.1016/0921-4534(89)90029-4

Li, Y.F., O. Chmaissem and Z.Z. Sheng, 1995. Crystal structure and Tc of 1212-type cuprate $(Tl,Cr)Sr_2(Ca,Tl)Cu_2O_7$. Physica C.: Superconductivity, 248: 42-48. DOI: 10.1016/0921-4534(95)00163-8

Mohammed, N.H., 2012. Effect of MgO Nano-oxide Additions on the Superconductivity and Dielectric Properties of $Cu_{0.25}T_{l0.75}Ba_2Ca_3Cu_4O_{12-\delta}$ Superconducting Phase. J. Supercond. Nov. Magnetism, 25: 45-53. DOI: 10.1007/s10948-011-1207-4

Mohammed, N.H., M. Roumie, H.A. Motaweh, R. Awad and D. El-said Bakeer *et al*., 2010. Determination of stoichiometry and superconducting properties of Tl-1234 and ($Cu_{0.25}Tl_{0.75}$)-1234 phases substituted by Erbium. J. Supercond. Nov. Magn., 23: 465-474. DOI: 10.1007/s10948-009-0599-x

Nawazish, A.K. and M. Mumtaz, 2007. $Cu_{0.5}Tl_{0.5}$ $Ba_2Ca_3Cu_{4-y}ZnyO_{12-\delta}$ (y = 0, 1.0, 2.0, 3.0, 3.5): superconductor with four ZnO_2 planes. J. Low. Tempr. Phys., 149: 97-103. DOI: 10.1007/s10909-007-9495-6

Nawazish., A.K. and S. Ullah, 2010. Effect of Pb doping on the superconducting properties of $(Cu_{0.5x}Pb_xTl_{0.5})Ba_2Ca_2Cu_3O_{10-\delta}$. J. Supercond. Nov. Magnetism, 23: 1281-1287. DOI: 10.1007/s10948-010-0770-4

Sheng, Z.Z., D.X. Gu, Y. Xin, O.D. Pederson and L.W. Finger *et al*., 1991. A new 1212-type phase: Cr-substituted $TlSr_2CaCu_2O_7$ WITH T_c UP TO ABOUT 110 K. Mod. Phys. Lett. B., 5: 635-642. DOI: 10.1142/S0217984991000770

Sheng, Z.Z., L. Sheng, X. Fei and A.M. Herman, 1989. Superconductivity above 77 K in the R-Tl-Sr-Ca-Cu-O system (R represents rare earths). Phys. Rev. B., 39: 2918-2920. DOI: 10.1103/PhysRevB.39.2918

Subramanian, M.A., C.C. Torardi, J. Gopalakrishnan, P.L. Gai and J.C. Calabrese *et al*., 1988. Bulk Superconductivity up to 122 K in the Tl-Pb-Sr-Ca-Cu-O System. Science, 242: 249-252. DOI: 10.1126/science.242.4876.249

Yip, W.Y. and R. Abd-Shukor, 2009. Phase formation and transition temperature variation of $TlSr_2$ ($Ca_{1-x}R_x$) Cu_2O_7 (R = Dy, Ho and Tb) superconductor. J. Supercond. Nov. Magn., 22: 705-709. DOI: 10.1007/s10948-009-0484-7