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Effect of Simultaneous Doping of Rhenium and Tellurium on T_c and Phase Formation of Tl- based Cuprate Superconducting Materials

Prafulla Ch. Kalita
Department of Electronics, Sibsagar College, Sivasagar, India

Abstract:

Preparation of new High Temperature Superconductor (HTS) materials is the important factors for development of efficient and lossless energy systems. Here the effect of tellurium (Te) and Rhenium on the critical temperature (Tc(0)) of Tl- based Cuprate Superconductors is presented. $Tl_{1.85}Re_{0.12}Te_{0.03}Ba_2Ca_2Cu_3O_x$ *and* $Tl_{1.88}Re_{0.12}Ba_2Ca_2Cu_3O_x$ *have been prepared with nominal starting composition by solid-solid reaction method. Ba-Ca-Cu-O precursor was made for this purpose by mixing and grinding of stoichiometric amounts of* $BaCO_3$, $CaCO_3$ *and CuO followed by sintering at* 850^oC. *Finally, appropriate amounts of* Re_2O_7, *and* (Re_2O_7 - TeO_2) *with* Tl_2O_3 *were added to the precursor and the whole mixture was annealed at* 950^oC *under oxygen flow. Doping of Re produced a chemical stabilization of the precursor against barium carbonation under ambient air. The critical temperatures Tc(0) of Re-doped compounds were found 86K. Simultaneous doping of Te and Re increases the value of Tc(0) (123K) of the superconductor. X-ray diffraction revealed the presence of some additional phases in ((Te-Re) - doped compound including the high Tc Tl-1223 and Tl-2223 phases . The gain homogeneity of the prepared specimens were excellent as obtained through SEM analysis. Simultaneous doping of tellurium and rhenium in Tl- cuprates induced pairing mechanisms to occur outside the* CuO_2 *layers, thus enhancing the high Tc phase formation and hence the Tc(0) value.*

Keywords: *Th-based superconductor, HTS materials, sinterin*

Introduction

Since the discovering of superconductivity, a Continuous and enormous efforts have been made by different groups of researchers for searching new high Tc superconducting materials .These approaches lead to them to discover new high Tc superconducting compounds La-Ba-Cu-O , Hg-Ca-Ba-Cu-O, Tl-Ca-Ba-Cu-O etc. The Thallium based high temperature superconducting compounds with critical temperatures as high as ~120K were first reported by Sheng and Hermann ,in 1988, by sintering Ba-Ca-Cu-O precursor oxide with Tl_2O_3 powder at 880-910^0 C in oxygen flow [1]. The importance of bulk thallium based cuprates as practical high-temperature superconductors has already been established by the works of different researchers worldwide. Especially, the Tl-1223 and Tl-2223 phases gain utmost importance due to relatively high critical temperature shown by them. Unlike the rare-earth-based compounds, the Tl- based copper oxides are thermally unstable phases, and rapid loss of thallium takes place above 875^{0}C. Hence it is difficult to prepare pure single phase[2]. Recently, doped Tl-based cuprates have been extensively studied due to their stability and phase purity. Eder and Gritzner reported the formation of high quality Tl-1223 and Tl-1212 materials with well connected grains by doping with rare earth oxides, viz., Y, La, Pr, Sm, and Dy [3]. Small amounts of a dopant like Te (less than 0.05) strongly favoured the formation of the Tl-1223 phase in bulk samples [4].

But the Tl-2223 phase is difficult to prepare in pure form [5, 6]. It requires the reaction conditions such as temperature, time of sintering and the thallium partial pressure given by the amount of Tl_2O_3 to be fine-tuned. This can be done only if the precursor remains almost stable during the fabrication of the superconductor. Doping with Re makes the thallinated precursor highly resistant to air degradation [7,8]. This is partly because of the fact that rhenium efficiently prevents the Ba carbonation of the $Ba_2Ca_2Cu_3O_x$ precursors [9, 10]. Moreover, it was found that the rhenium has a significant influence on the magnetic, microwave and structural properties of the Tl-superconductor in thin film form [11, 12]. The motivation behind the present work is to see the effect of doping of rhenium and rhenium-tellurium on the phase formation and critical temperature of Tl-based cuprate superconductors. X-ray powder diffraction reveals a multiphase mixture in the two samples.

Experimental Method : Preparation Of The Bulk Superconductor

The Tl-containing superconducting oxide powders were synthesized by the conventional solid-state reaction route [13]. For the preparation of each thallium free precursor, stoichiometric amounts (2:2:3) of $BaCO_3$ (99.9%-Ba, Sr<0.03%, Strem Chemicals), $CaCO_3$ (99.95%-Ca, Strem Chemicals) and CuO (99.999%-Cu, Strem Chemicals) were taken. Then, these chemicals were mixed and finely grinded in an agate mortar. The mixture was calcined in an open platinum crucible at 850^oC under oxygen flow for a total period of 16 h. The mixture was subjected to intermittent grindings after every 4 hours to avoid agglomeration formation. The undoped Tl-based cuprate was synthesized by sintering a mixture of appropriate amount of Tl_2O_3 (99.99%-Tl, Strem Chemicals) and one of the precursor samples in a platinum crucible at 950^oC for 8 hours. Intermittent grindings were carried out after every 2 hours.

For the preparation Re-doped Tl-based cuprate superconductor, mixing ratio of Tl_2O_3 to Re_2O_7 (99.99%-Re, Strem Chemicals) was kept at 1.88 : 0.12 and for that of (Te, Re)-doped Tl-based cuprate, the ratio of Tl_2O_3, TeO_2 and Re_2O_7 was maintained at 1.85: 0.03: 0.12. These were added to another prepared precursors and sintered under the same experimental procedures and conditions as that of undoped Tl- cuprate.

However, this carries some difficulties inherent in the synthesis of pure phase samples due to volatility of the thallium component above its melting point of $\sim 710^0C$ and severe decomposition of the products at high reaction temperature [2]. Prevention of Thallium loss is most important factor not only for its toxicity but also for maintaining the stoichiometry of the products to ensure the formation of the desired phase. In order to prevent severe thallium loss during the reaction, samples have been wrapped in Ag foil as this foil is less expensive substitute for Au and Pt.

The formed compounds were pelletized for resistivity measurements at a pressure of $0.0283 tonne/mm^2$ using polymer press (PF-M15, Technosearch Instruments, India).The pellets were of 15 mm in diameter and approximately 1 mm thick. The pellets were annealed at 600^0C for 6 hours under oxygen flow.

Results And Discussion

The x-ray diffractograms of the compounds obtained by using a Philips PW1710 diffractometer with Cu $K\alpha 1/K\alpha 2$ ($K\alpha 2/K\alpha 1=0.5$) radiation are shown in figures 1(A)-1(C). From the diffractograms, it is seen that some peaks are common for all the three compounds. These are approximately at $2\theta = 21.3^o$, 24.06^o, 28.24^o, 29.8^o, 30.8^o, 35.2^o, 36.3^o, 38.6^o, 43.6^o, 48.5^o, 49.6^o, 54.5^o, and 58.5^o. The (006) and (008) peaks appeared for the undoped Tl-compound. But, they vanish completely for the Re-doped compound. On the other hand, the (006) peak appeared in the XRD (Te, Re)-doped compound. Again, one of the (101) peaks of the Tl-2212 phase (at 2θ=24.06) is prominent in both undoped and (Te, Re)-doped compound whereas, it is much diminished in the Re-doped Tl-based cuprate. Similarly, one of the (110) peaks (at 2θ=32.2) of the Tl-1223 phase appeared for undoped and (Te, Re)-doped Tl-cuprates, but absent in the Re-doped one. The disappearance of peaks at 42^o and 46.8^o in the diffractogram for the Re-compound is another important observation. Peaks after 60^o could not be assigned for any particular phases. On examining the XRD of the (Te, Re)-doped sample (figure 1 (C)), numerous new peaks were found (indicated by arrows). These were totally absent in the XRD of the Re-doped compound.

The new peaks obtained in the XRD of the (Te, Re)-doped sample can be explained from the fact that some additional pairing interactions may occur outside the CuO_2 layers in high-T_c superconductors, especially those with T_cs higher than 100 K. It can be said that simultaneous doping of rhenium and tellurium induces these additional pairing mechanisms by forming charge reservoir layers with Tl, which is known to be a negative U-centers. On analyzing the FTIR spectra of figure 4 (a) and 4 (b)), we found that at the positions 3233.2 cm^{-1}, 2159.2 cm^{-1}, 1079.4 cm^{-1} and 1016.7 cm^{-1}, some additional distinct peaks appeared for the doubly doped compound $Tl_{1.85}Re_{0.12}Te_{0.03}Ba_2Ca_2Cu_3O_z$. Also, peaks after 700 cm^{-1} appeared at dissimilar positions for these two compounds. All these indicate the formation of some new bonds of Tl with either of the dopants, leading to the charge reservoir layers and hence enhancement of Tc in high temperature cuprates [14].

The $T_{c(0)}$ value was found to be 123 K, as indicated in figure 3 (c). The Re doping protects the sample from air degradation, but it promotes the growth of Tl-1212 phases and suppresses other phases. The $T_{c(0)}$ values of the Re-doped and undoped Tl-based superconductors are 86 K and 114 K respectively, as indicated by figures 3 (b) and 3(a). Re dopants may disturb the local oxygen distribution in both in the 2212 and 1212 lattice, which lowers the T_c of the material [8].

Figures 2 (a)-2 (c) show the scanning electron micrographs of all the above mentioned superconducting compounds. The surface morphology of the compound $Tl_2Ba_2Ca_2Cu_3O_x$ reveals small plate-like structures connected to each other (figure 2 (a)). Partially separated and relatively large plate-like structures are seen in the SEM of the Re-doped cuprate (figure 2 (b)).

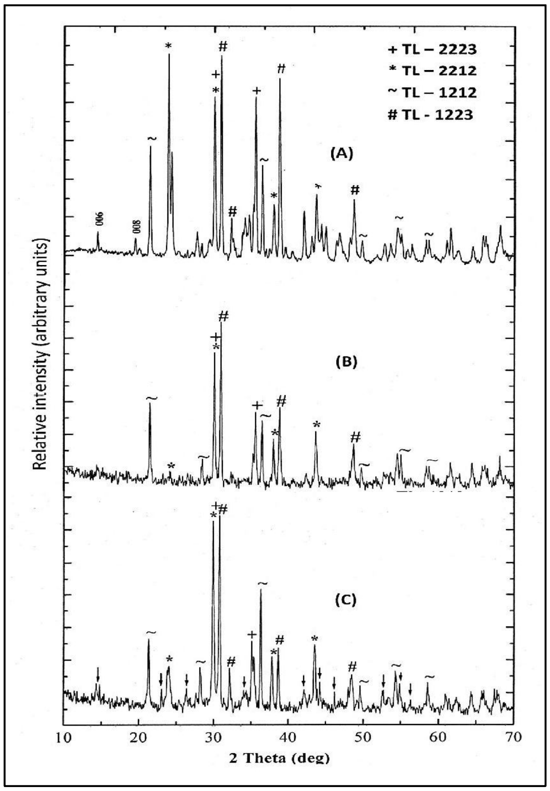
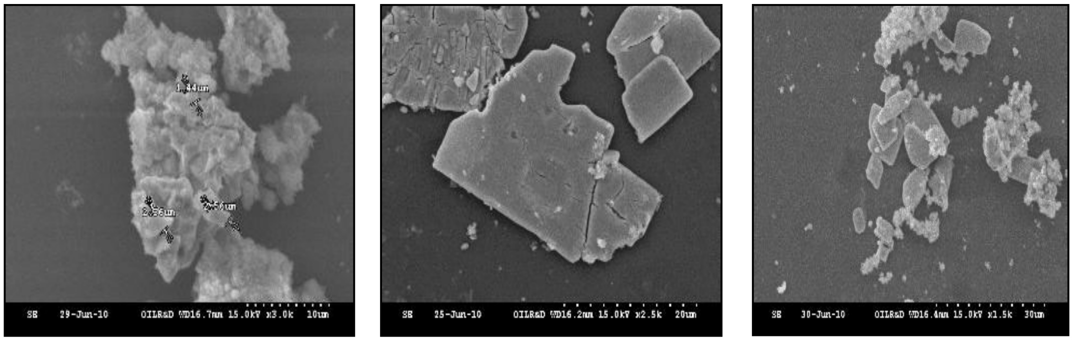


Figure1:X-raydiffractogram of (A) Tl-based undoped cuprate (B) Re-doped Tl- based bulk cuprate sample and (C) (Te, Re)-doped Tl-based cuprate compound



(a) (b) (c)

Figure 2: Scanning electron micrograph of (a)Tl-based undoped cuprate, (b) Re-doped Tl-based bulk cuprate sample and (c) (Te, Re)-doped Tl-based cuprate compound

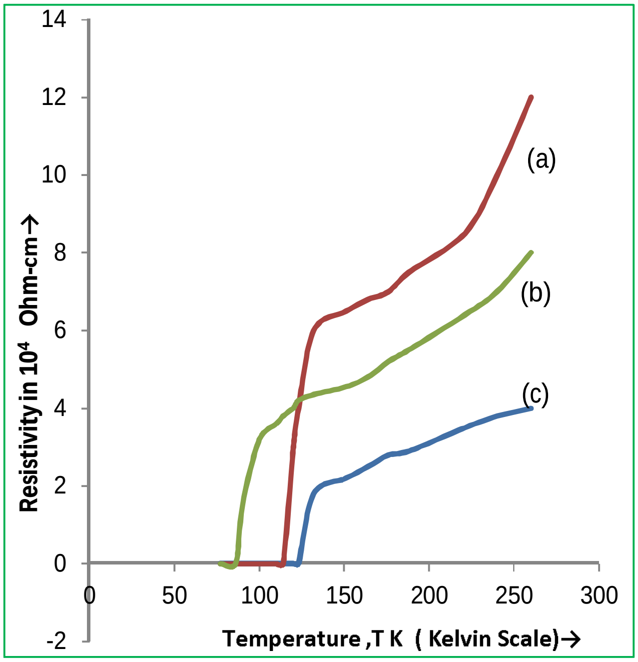


Figure 3: Temperature vs resistivity Curves of (a) Tl-based undoped cuprate, (b) Re- doped Tl-based bulk cuprate and (c) (Te, Re)-doped Tl-based cuprate compounds

The separation of polygon shaped plate like grains is much more complete in case of the (Te, Re)-doped compound (figure 2 (c)). More completed separation reduces lump formation and leads to much better grain homogeneity. The grain size of the compound $Tl_2Ba_2Ca_2Cu_3O_x$ ranges from 1-3 µm whereas the grain size varies from 8-10 µm in the case of $Tl_{1.88}Re_{0.12}Ba_2Ca_2Cu_3O_y$. 3-6 µm sized grain sizes were found for $Tl_{1.85}Re_{0.12}Te_{0.03}Ba_2Ca_2Cu_3O_z$ superconductors.

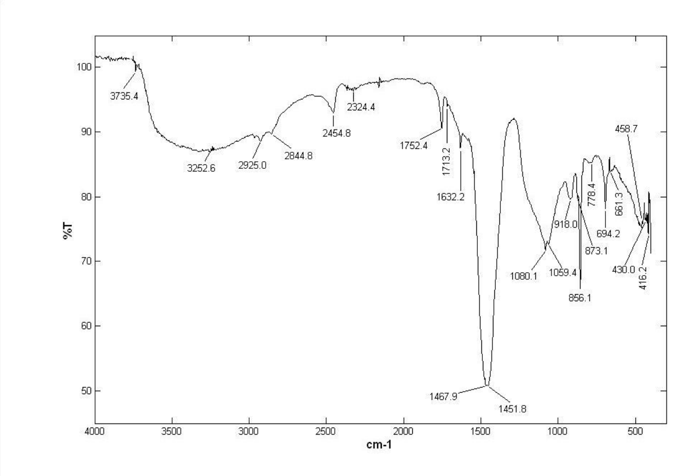


Figure 4(a): FTIR plot of Re-doped Tl-based bulk cuprate sample

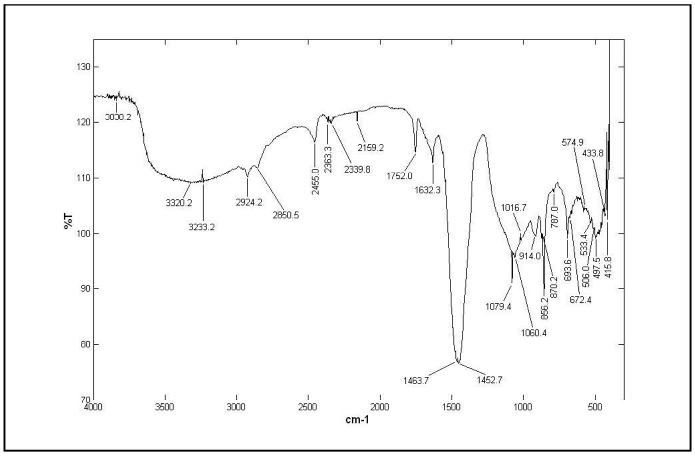


Figure 4(b): FTIR plot of (Te, Re)-doped Tl-based cuprate compound

Conclusion

In conclusion, $Tl_{1.88}Re_{0.12}Ba_2Ca_2Cu_3O_y$ and $Tl_{1.85}Re_{0.12}Te_{0.03}Ba_2Ca_2Cu_3O_z$ compounds are fabricated by doping Rhenium and Rhenium- tellurium in the Ba-Ca-Cu-O precursor. It is found that the presence of non volatile Re in the cuprate lowers the T_c value for disturbing the local oxygen distribution in the phases. The Re-Te doping induces the additional pairing mechanisms by forming charge reservoir layers with Tl (negative U-centers). This may lead to different types of phase formation and hence to enhance Tc(0) of the superconducting material. It can also be concluded that high temperature superconductors (HTS) for energy applications with different doping materials is an invaluable reference tool for anyone involved or interested in HTS materials and their applications. The use of it results substantial environmental improvements by energy saving.

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