International Journal of scientific research and management (IJSRM)
||Volume||2||Issue||5 ||Pages|| 850-854 ||2014||
Website: www.ijsrm.in ISSN (e): 2321-3418

SYNTHESIS, CHARACTERIZATION AND STUDY THE EFFECT OF *Te* ON CRITICAL TEMPERATURE OF *Tl-Re* BASED CUPRATES.

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Financial support :-Financial support through Major Research Project was received from University Grants Commission, Bahadur Shah Zafar Marg, New Delhi, India.

ABSTRACT

In the present work, we presented the synthesis, characterization and study the effect of Te on Critical Temperature Tl-Re based Cuprates: one was Re-doped and another one was simultaneously doped with Te and Re. All these materials were prepared by the usual solid-state reaction method. Ba-Ca-Cu-O precursors were made by taking $BaCO_3$, $CaCO_3$ and CuO in the ratio of 2:2:3. The Tl-Re based cuprate with the formal composition $Tl_{1.88}$ $Re_{0.12}Ba_2Ca_2Cu_3O_{10+\delta}$ was formed by adding stoichiometric amount of Tl_2O_3 and Re_2O_7 to the precursor and whole mixture was synthesized under closed condition at 950^oC. For Tellurium doping, Tl_2O_3, TeO_2, and Re_2O_7 were taken in the ratio of 1.85:0.03:0.12 and synthesized at the same condition .The samples are then palletized and annealed at oxygen environment. The Tellurium doping increased the critical temperature from 86K to 123K. X-ray diffraction reveals the presence of a number of more superconducting phases in Tl-Re-Te compounds. The gain homogeneity of the powered samples were excellent.

Keywords: thallinated, stoichiometry, calcined, cuprate

1. INTRODUCTION

Since the discovery of superconductivity, continuous and enormous efforts have been made by different group of researchers for searching new high T_c superconducting materials. In 1988 Sheng and Hermann reported Tl-based superconducting cuprates with high critical temperature (120K) [1]. The importance of bulk thallium based cuprates as practical high-temperature superconductors has already been established by the works of different researchers worldwide. Tl-1223 and Tl-2223 phases were important due to relatively high critical temperature shown by them. Tl- based copper oxides are thermally unstable phases, and rapid loss of thallium takes place above 875^{0}C. Hence it was 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 superconducting materials with well-connected grains by doping with rare earth oxides. The Tl-1212 crystallites are usually polygon shaped with dimensions in the order of 2-5μm [3]. Doping with Re makes the thallinated precursor highly resistant to air degradation and chemical stabilization of the sample [4]-[6]. and it has a significant influence on the magnetic, microwave and structural properties of the Tl-superconductor in thin film[6],[7]. Rhenium efficiently prevents the Ba carbonation of the $Ba_2Ca_2Cu_3O_x$ precursors [8]. Small amounts of a dopant like Te (less than 0.05) strongly favoured the formation of the Tl-1223 superconducting phases in bulk samples [9]. The motivation behind the work was to see the effect of doing of tellurium on the phase formation and critical temperature of Tl-Re based cuprates. X-ray powder diffraction reveals a multiphase mixture in the two samples.

2. MATERIAL AND METHODS

The Tl-Re superconducting oxide powders were synthesized by the conventional solid-state reaction route

[10]. For precursors, stoichiometric amounts of $BaCO_3$, $CaCO_3$ and CuO were mixed and finely grinded in an

agate mortar. The mixture was calcined in an open platinum crucible at 850^{o}C 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. For the preparation of Tl-Re (TR)- based cuprate superconductor, a mixture of Tl_2O_3 and Re_2O_7 at ratio of 1.88 **:** 0.12 was synthesized by sintering with one of the precursor samples in a platinum crucible with a lid at 950^{o}C for 8 hours. Intermittent grindings were carried out after every 2 hours. For that of Tl-Te-Re (TRT)-cuprate, the ratio of Tl_2O_3, TeO_2 and Re_2O_7 was maintained at 1.85 **:** 0.03 **:** 0.12. This mixture was added to another prepared precursor and sintered under the same experimental procedures and conditions as that of TR cuprate. In order to prevent severe thallium loss and maintaining the stoichiometry of the products to ensure the formation of the desired phase during the reaction, samples were wrapped by Ag foil.

The resulting powders were pelletized for resistivity measurements at a pressure of 0.0280tonne/mm^2 using polymer press (PF-M15). The pellets were annealed at 600^{0}C for 6 hours under oxygen flow.

Finally pellets were characterized by resistance vs. temperature (R-T) measurement using a standard dc four probe measuring technique. The phase compositions of the final powdered sample were analyzed by X-ray diffraction by using a Philips PW1710 with Cu Kα1/Kα2 radiation. Scanning electron microscopies (SEM) were performed using Hitachi field emission S-3600N.

3. RESULTS AND DISCUSSION

3.1 Results

The x-ray diffractograms of the compounds are shown in Fig. 1a & 1b. Some

peaks are common for both compounds at 2θ = 21.3°, 24.06°, 28.24°, 29.8°, 30.8°, 35.2°, 36.3°, 38.6°, 43.6°, 48.5°, 49.6°, 54.5°, and 58.5°. The (006) peak (at 2θ = 15°) and (110) peaks (at 2θ=32.2) are appeared for TRT-Cuprates. But, it vanished completely for TR-compounds. Again, one of the (101) peaks of the Tl-2212 phase (at 2θ=24.06) is prominent in TRT- compound whereas, it is much diminished in the TR-based cuprate.. Moreover, there are peaks at 26.4°, 32.05°, 34.08°, 44.3°, 46.3° and 52.7°, which were not present before Te doping. The disappearance of peaks at 42° and 46.8° in the diffractogram for the TR- compound is another important observation. On examining the XRD of the TRT-sample (Fig. 1b), numerous new peaks were found.

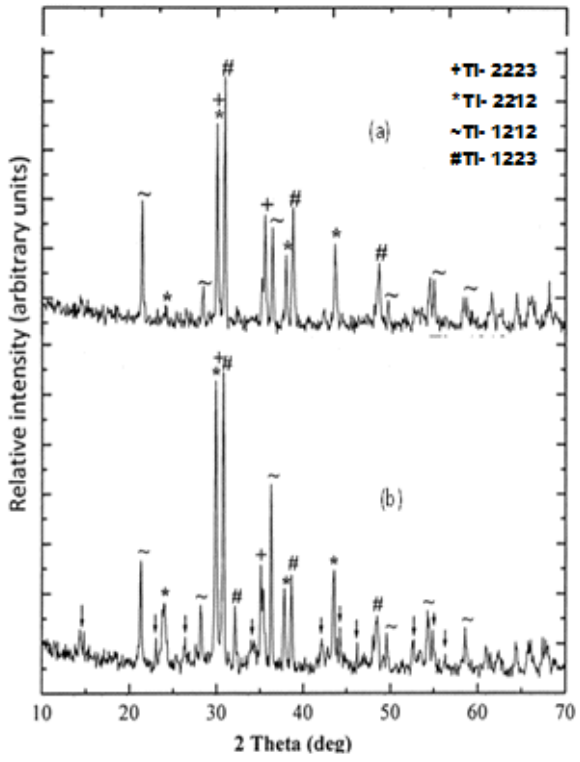

Figure 1: X-ray diffractogram of

(a) $Tl_{1.88}Re_{0.12}Ba_2Ca_2Cu_3O_x$ and

(b) $Tl_{1.85}Re_{0.12}Te_{0.03}Ba_2Ca_2Cu_3O_y$ sample

From scanning electron micrographs, the surface morphology of the compound of $Tl_{1.88}Re_{0.12}Ba_2Ca_2Cu_3O_x$ cuprate is seen partially separated and relatively large plate-like structures (Fig 2a). Polygon shaped plate like grains were seen in case of $Tl_{1.85}Re_{0.12}Te_{0.03}Ba_2Ca_2Cu_3O_y$ compound with better grain homogeneity (Fig 2b). The $T_{c(0)}$ value of TRT- cuprate was found to be 123 K and that of TR -cuprate was 86 K as indicated by Figs. 3a & 3b.

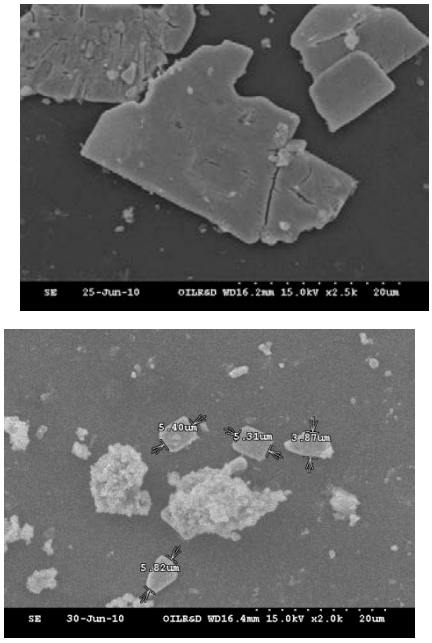

Figure 2: Scanning electron micrograph of

(a) $Tl_{1.88}Re_{0.12}Ba_2Ca_2Cu_3O_x$ and

(b) $Tl_{1.85}Re_{0.12}Te_{0.03}Ba_2Ca_2Cu_3O_y$ samples

Figure 3 : Temperature vs resistivity Curves of

(a) $Tl_{1.88}Re_{0.12}Ba_2Ca_2Cu_3O_x$ and

(b) $Tl_{1.85}Re_{0.12}Te_{0.03}Ba_2Ca_2Cu_3O_y$

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3.2 Discussion:

The new peaks obtained in the XRD of the TRT-cuprate sample can be explained from the fact that some additional possible unconventional pairing interactions may occur outside the CuO_2 layers in high-T_c superconductors due to fluctuations between two degenerate valence states. These indicated that simultaneous doping of rhenium and tellurium in Tl-based cuprate leaded to some new pairing mechanism of Tl with either of the dopants by forming charge reservoir layers (negative U-centers) and hence enhanced Tc [11]. The Re doping promoted the growth of Tl-1212 superconducting phases and suppressed other phases. On the other hand Re dopants may disturb the local oxygen distribution in both in the 2212 and 1212 lattice, which in turn lowers the T_c of the material [5]. The separation of polygon shaped plate like grains was much more complete in case of the TRT-doped $Tl_{1.85}Re_{0.12}Te_{0.03}Ba_2Ca_2Cu_3O_y$ compound (figure 2(b)). More completed separation reduces lump formation and leads to much better grain homogeneity. The grain size of $Tl_{1.88}Re_{0.12}Ba_2Ca_2Cu_3O_y$ varied from 8-10 μm and for that of $Tl_{1.85}Re_{0.12}Te_{0.03}Ba_2Ca_2Cu_3O_{y.}$ was 3-6 μm

4. CONCLUSIONS

In conclusion, $Tl_{1.88}Re_{0.12}Ba_2Ca_2Cu_3O_x$ and $Tl_{1.85}Re_{0.12}Te_{0.03}Ba_2Ca_2Cu_3O_y$ compounds are fabricated by doping Rhenium and Rhenium-tellurium in the Ba-Ca-Cu-O precursor at nominal starting compositions. It is found that the lowering of T_c value in the Re doped Tl-cuprates may be for disturbing the local oxygen distribution in the phases [5]. The Re-Te doping in Tl- Cuprates induces the additional pairing mechanisms by forming charge reservoir layers with Tl (negative U-centers). This may lead to different types of superconducting phase formation and hence to enhance *Tc(0)* of the compound.

ACKNOWLEDGEMENT Financial support through Major Research Project by University Grants Commission, India is gratefully acknowledged.

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