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## Influence of Tl-Containing Coprecipitation Derived Precursor Powder on Microstructure and Transport Properties of $Tl_{0.8}Bi_{0.2}Sr_2Ca_{0.8}Y_{0.2}Cu_2O_7$ Superconducting Dip-Coated Tapes

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**Abstract:** Ag clad dip-coated superconductor tapes were fabricated from Tl-1212 superconducting powders of  $Tl_{0.8}Bi_{0.2}Sr_2Ca_{0.8}Y_{0.2}Cu_2O_7$  starting composition derived via two different methods; the conventional Solid-State (SS) and coprecipitation (CP) methods. The effects of using superconducting powders derived from SS and CP methods on phase formation, microstructure and critical current density were investigated for tapes annealed at 850°C for 60 min in flowing  $O_2$ . XRD analysis of the tapes oxide cores indicated a lower volume percentage of 1212 phase for the CP tape (84%) compared to the SS tape (97%). However, the transport  $J_c$  of the CP tape (4670 A cm<sup>-2</sup>) was significantly higher than the SS tape (560 A cm<sup>-2</sup>). The SEM micrograph of the CP tape's oxide core revealed partially melted microstructure with fused grains in contrast to the microstructure of the SS tape's core which shows porous microstructure with irregularly shaped grains. The higher  $J_c$  for the CP tape is suggested to be due to improved inter-grain connectivity as a result of the partial melting.

**Key words:** Coprecipitation, dip-coated tapes, Tl1212 superconductor

### INTRODUCTION

Superconductivity in thallium-based (Tl-based) materials was first reported by Sheng and Hermann (1988) and since then there have been numerous effort to fabricate these materials into superconducting wires and tapes for applications in superconducting motors, transformers, transmission lines and magnets (Selvamanickam *et al.*, 1996; Goyal *et al.*, 1995). Amongst Tl-based superconductors, much interest have been focused on single Tl-O layered Tl1223 and Tl1212 materials as they have been suggested to show better in-field behavior compared to multiple Tl-O layers system (Jergel *et al.*, 1996). However, Tl1212 is expected to produce higher transport critical current density ( $J_c$ ) and better performance in magnetic fields compared to Tl1223 due to reduced anisotropy through its stronger interlayer coupling and less severe thermally activated flux motion (Lao *et al.*, 2000). Some research have been reported on Tl1212/Ag tapes using Powder-In-Tube (PIT) method (Hamid and Abdul Shukor, 2001) but no report was found for Tl1212 using Dip-Coating (DC) which is a faster and simpler method of tape fabrication. A key factor in processing Tl1212 tapes to achieve high  $J_c$  is to optimize not only the processing details of the tapes but also the quality of the superconductor powders used.

For Tl-based superconductor system, synthesis of single-phased samples is difficult due to the constrain imposed by  $Tl_2O_3$  that melts and evaporates at 717°C at atmospheric pressure whereas formation of the high- $T_c$  phase typically takes place at higher temperatures (Abdul Shukor, 1993). Conventional solid-state method using oxide powders has employed the two-step method, which used non Tl-containing precursor in the first stage before addition of Tl in the final sintering stage to reduce Tl-loss. On the other hand, chemical coprecipitation method offers unique advantages over conventional solid-state method such as lower temperature synthesis, precise composition control, high purity and chemical homogeneity due to the fact that in the process the mixing of components occurs at the atomic level (Segal, 1997; Rao *et al.*, 1993). Interestingly, single-step synthesis of bulk Tl1212 using Tl-containing coprecipitated precursors have been recently reported (Salleh *et al.*, 2005). However, the use of the coprecipitation derived powder in fabrication of dip-coated tapes has not been reported. In this paper, we report on Tl1212/Ag dip-coating tapes fabrication using powders prepared by SS and CP methods. Results of powder X-ray Diffraction (XRD) and Scanning Electron Microscope (SEM) investigations on the tapes are presented and analyzed.

**MATERIALS AND METHODS**

In the first stage, bulk  $Tl_{0.8}Bi_{0.2}Sr_2Ca_{0.8}Y_{0.2}Cu_2O_7$  superconductor was prepared via two different routes, i.e., the coprecipitation route and the conventional solid-state route. The bulk SS sample was prepared by a two-step solid-state reaction, as described in previous research (Yahya *et al.*, 2004). In the coprecipitation route, a similar synthesis process as described by Salleh *et al.* (2005) was used. Reagent-grade Tl, Bi, Sr, Ca, Y and Cu acetates with stoichiometric ratios of 0.8:0.2:2:0.8:0.2:2 were dissolved in glacial acetic acid and was reacted with excess oxalic acid at 50°C under continuous stirring, yielding precipitation of the mixed oxalates. The precipitate was filtered and dried overnight at 80°C and then was ground to powdery state. Calcination of the powder was done in air at 600°C, to avoid loss of Tl for a duration of 20 h with heating and cooling rates of 1°C min<sup>-1</sup> following which the powder was reground and sintered at 1000°C for 6 min in flowing O<sub>2</sub> followed by furnace cooling to room temperature. A slurry solution was then prepared by mixing the SS (and CP) superconductor powder with appropriate amounts of organic binder, dispersant and solvent (Sung *et al.*, 2000). Strips of silver substrate were dipped into the slurry and coated films of 0.25-0.40 mm in thickness were achieved and monitored. The coated silver strips were dried at 80°C for 2 days and were separately wrapped using silver foil and were heated at 450°C in air for 3 days, after which the tapes were deformed by cold rolling to 45-55% deformation in tapes thickness. The tapes were annealed in flowing O<sub>2</sub> at 850°C for 60 min annealing duration.

Electrical resistance (dc) measurements for both tape samples were carried out using the standard four-point-probe method with silver paint contacts in a Janis model CCS 350ST cryostat in conjunction with a closed cycle refrigerator from CTI Cryogenics model 22. The phase characterization was done by powder XRD using Bruker D8 Advanced diffractometer with Cu-K<sub>α</sub> source. Transport critical current densities ( $J_c$ ) were measured in zero magnetic fields using standard four-point-probe method and evaluated using the electric field criterion of 1 μV cm<sup>-1</sup>. SEM micrographs were recorded using JEOL model JSM-6360LA scanning electron microscope.

**RESULTS AND DISCUSSION**

XRD patterns for both fabricated tapes are shown in Fig. 1. The volume fractions of 1212 and 1201 phase and SrCO<sub>3</sub> impurity are estimated using the same technique employed by Matsushita *et al.* (1994). The normalized resistances versus temperature curves of the bulk and tape samples are shown in Fig. 2. All bulk and tape

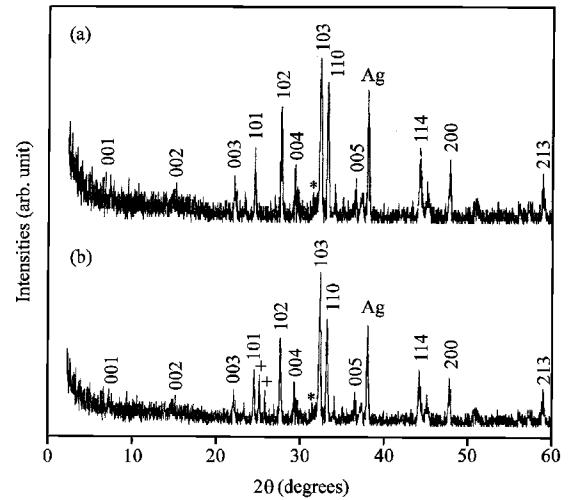


Fig. 1: Powder X-ray diffraction pattern for tape samples (a) SS and (b) CP. Peaks due to 1201 phase and SrCO<sub>3</sub> impurities are indicated by (\*) and (+), respectively

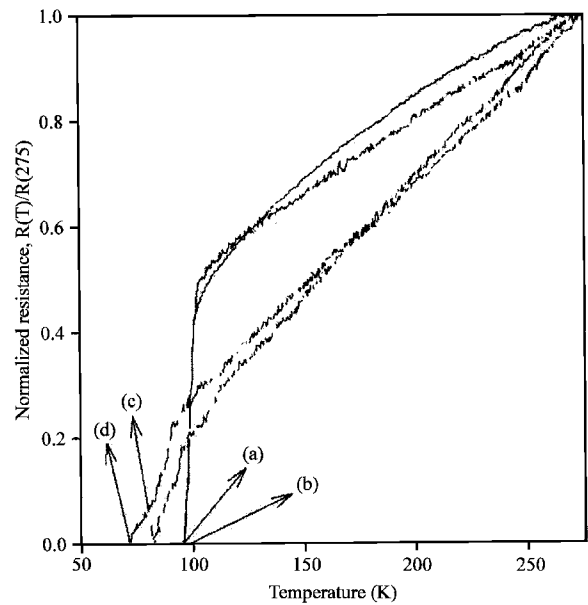


Fig. 2: Normalized resistance versus temperature curve for (a) SS bulk, (b) CP bulk, (c) CP tape and (d) SS tape samples

samples exhibit metallic behavior in its normal state with  $T_{c\ onset}$  of between 85-103 K. Based on the  $T_{c\ onset}$  values, it is suggested that 1212 phase is responsible for the observed superconductivity as superconductivity due to 1201 phase is reported to be below 50 K (Sheng *et al.*, 1991). Table 1 show sample, 1212:1201: SrCO<sub>3</sub> phase Vol. %, zero-resistance critical temperature ( $T_{c\ zero}$ ) of bulk

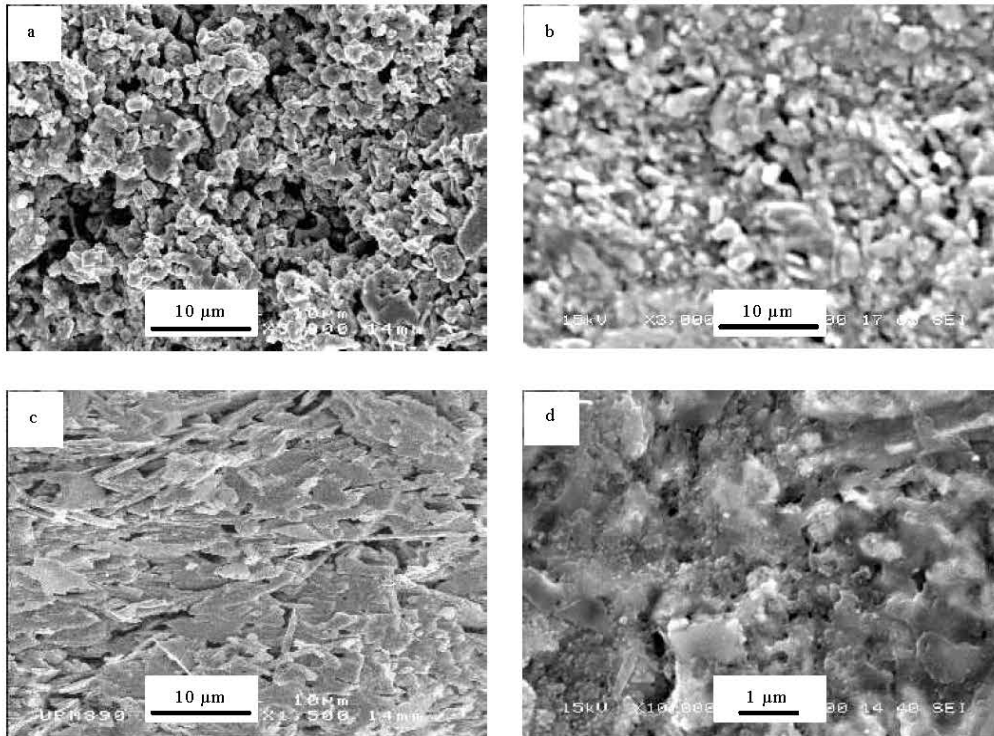


Fig. 3: Microstructures of bulk and tapes (a) SS bulk, (b) SS tape, (c) CP bulk and (d) CP tape

Table 1: Sample, 1212:1201:SrCO<sub>3</sub> phase Vol. % of tape samples, zero-resistance critical temperature ( $T_{c, zero}$ ) of bulk samples, zero-resistance critical temperature ( $T_{c, zero}$ ) of tape samples and transport critical current density ( $J_c$ ) at 40 K of tape samples

Samples	1212:1201: SrCO <sub>3</sub> of tape sample (Vol. %)	$T_{c, zero}$ of bulk sample (K)	$T_{c, zero}$ of tape sample (K)	$J_c$ at 40 K of tape sample (A cm <sup>-2</sup> )
SS	97:3:0	94	71	560
CP	84:3:13	94	82	4670

samples, zero-resistance critical temperature ( $T_{c, zero}$ ) of tape samples and transport critical current density ( $J_c$ ) at 40 K of tapes prepared in this study.

In this study, higher  $J_c$  has been obtained for the CP tape compared to the SS tape. In view of the fact that the 1212 volume fraction for CP tape was only 84%, it is possible that  $J_c$  can be further increased if the 1212 volume fraction is higher and the amount of impurity is lower. Work on addition of a sintering aid to the Tl-containing precursor that results in SrCO<sub>3</sub>-free superconducting sample has been carried out and will be reported elsewhere. Because the volume fraction of the 1212 phase in the CP tape was lower than that of the SS tape (Table 1), it is suggested that the higher magnitude of  $J_c$  for the CP tape is not due to 1212-phase volume fraction. In addition, differences in thermal and mechanical

treatments of the tapes may not be the reason as both tapes were subjected to the same thermomechanical processes during fabrication. The minor 1201 phase and the SrCO<sub>3</sub> impurity observed for the CP tape are also not known to introduce magnetic pinning effects in high-temperature superconductors. As such, the reason for the differences in  $J_c$  can be due to other reasons, such as, the microstructure of the tapes core.

Figure 3 shows SEM micrographs of fractured surface of the bulk and superconducting oxide core of the tape samples. The micrograph of CP bulk revealed elongated plate-like grains with sizes between 7-15 μm, which is remarkably different from the micrograph of SS bulk that revealed slightly rounded grains of 3-4 μm in average size. Interestingly, the micrograph of CP tape's core displayed partially melted and fused grains in contrast to micrograph of SS tape's core which revealed porous microstructure consisting of irregular shaped grains of 1-4 μm in average size. No particular texturing was observed from the SEM micrograph of the tapes core for both tapes (Fig. 3a-d).

From the microstructural observations, it is possible that the partially melted microstructure with fused grains and the accompanying reduction in porosity of the CP

tapes has lead to improved connectivity between grains which may be the reason for the higher  $J_c$  compared to the SS tape. While, reduction in sample porosity is known to cause increase in sample  $J_c$  (Jeong and Sohn, 1998), partially melted microstructure with fused grains have shown higher  $J_c$  values for Tl1212/Ag tapes fabricated via the powder-in-tube method (Hamid and Abdul Shukor, 2001).

### CONCLUSION

Tl<sub>0.8</sub>Bi<sub>0.2</sub>Sr<sub>2</sub>Ca<sub>0.8</sub>Y<sub>0.2</sub>Cu<sub>2</sub>O<sub>7</sub> tapes employing powders derived from the conventional Solid-State (SS) and Tl-containing coprecipitation (CP) methods were fabricated using dip-coating technique on silver substrate. The tape fabricated using Tl-containing coprecipitation derived oxide powder has superior transport  $J_c$  than the tape that used conventional solid-state derived powder. The enhanced  $J_c$  of the CP tape compared to the SS tape is attributed to improved inter-grains connectivity as a result of partial melting.

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