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# FABRICATION OF $Tl_{0.9}Bi_{0.1}Sr_{1.9}In_{0.1}Ca_{0.9}Y_{0.1}Cu_2O_7$ AND $YBa_2Cu_3O_{7-\delta}$ SUPERCONDUCTOR COATED TAPES

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## ABSTRACT

In this paper, preparation of Y123 and Tl1212 thick films on silver substrate using a simple coating procedure are reported. In this procedure $YBa_2Cu_3O_{7-\delta}$ and $Tl_{0.9}Bi_{0.1}Sr_{1.9}In_{0.1}Ca_{0.9}Y_{0.1}Cu_2O_7$ superconductors were synthesized using the conventional solid state method and transformed into slurry by mixing with appropriate amounts of organic binder, dispersant and solvent. The slurry was then transferred on silver substrates. The resulting thick film was annealed at low temperature for 2-3 hours to remove the organic material. It was then subjected to thermal treatment before electrical resistance measurements (dc) using the four-point-probe method. The Y123 and Tl1212 coated tapes showed metallic normal state behaviors with critical temperature $T_{c\ zero}$ of 62 K and 72 K, respectively. The critical current ($I_c$) of the Y123 and Tl1212 tapes measured at 40 K were 390.5 mA and 864.5 mA respectively.

## INTRODUCTION

Fabrication of superconducting tapes has been of interest to both scientist and engineers because of its high current carrying capacity and potential technological applications. The development of coated conductors with RE-Ba-Cu-O system superconductors has shown high critical current density ($J_c$) at the liquid nitrogen temperature in external magnetic fields [1]. In recent years, fabrication of Y123 coated tapes with different methods such as dip coating [2-4] and others were applied to fabricate coated conductors. Fabrication of coated Y123 tapes on crystallographically oriented substrates by dip-coating demonstrated remarkable success for example on polycrystalline $DyBa_2Sn_{5.5}$ [3] and $GdBa_2HfO_{5.5}$ [4] and gave critical temperature $T_{c\ zero}$ of 92 K.

However, although Y123 has excellent current carrying capacity, it suffers from weak-link problem [5,8,9]. IBAD technique [6] was developed to fabricate highly aligned Y123 coated tape with high critical current density in high magnetic fields. Meanwhile Tl1212 is one of the interesting compounds in Tl-based superconductor [7]. Single layer Tl1212 has shown better $J_c$ property in magnetic fields compared to other double layer Tl-based compounds [8]. This is suggested to be due to stronger coupling between conducting Cu-O planes separating single-layer insulating Tl-O plane. Quite a number of work has been reported on preparation of Tl1212 tape using powder in tube method [8-10] but report on Tl1212 coated tapes is limited. Moreover, there still remain difficulties to produce coated tapes with good superconductivity properties due to non uniform coatings and viscosity of slurry [11].

In this paper, fabrication of Y123 and Tl1212 tapes from $YBa_2Cu_3O_{7-\delta}$ and $Tl_{0.9}Bi_{0.1}Sr_{1.9}In_{0.1}Ca_{0.9}Y_{0.1}Cu_2O_7$ starting compositions, respectively using a simple coating method are reported. Preliminary results of electrical measurements (dc) and powder X-ray diffraction are included. We believe that this is the first successful fabrication locally and we are glad to report the results.

## EXPERIMENTAL PROCEDURE

### Preparation of bulk sample

**$YBa_2Cu_3O_{7-\delta}$**

Bulk Y123 was prepared by using conventional solid state reaction method. Chemicals of high purity (99.99%) $Y_2O_3$, $BaCO_3$ and $Cu_2O$ with nominal starting composition ratio of 1:2:3 were mixed and ground for 1 hour in an agate mortar. The resulting mixture was calcined at 900$^o$C for 24 hours with several intermittent grindings. The calcinated powder was reground and then pressed into pellets with 13 mm diameter under a load of 4-5 tons using a hydraulic press. After that, the pellets were sintered at 930$^o$C for 24 hrs in air and then slow cooled to room temperature at 40$^o$C/hr.

**$Tl_{0.9}Bi_{0.1}Sr_{1.9}In_{0.1}Ca_{0.9}Y_{0.1}Cu_2O_7$**

Precursor sample with nominal composition $Tl_{0.9}Bi_{0.1}Sr_{1.9}In_{0.1}Ca_{0.9}Y_{0.1}Cu_2O_7$ were prepared by mixing high purity (99.99%) $SrCO_3$, $Cu_2O$, CaO, $In_2O_3$ and $Y_2O_3$. The chemicals were ground in an agate mortar for an hour and then calcined in air at around 900$^o$C for 48 hours with several intermittent grindings. Appropriate amounts of $Tl_2O_3$ and $Bi_2O_3$ were added to the heated powder with nominal composition $Tl_{0.9}Bi_{0.1}Sr_{1.9}In_{0.1}Ca_{0.9}Y_{0.1}Cu_2O_7$. The powder was reground for another 1 to 2 hours to obtain homogenous powder and then pressed into pellets with 13 mm diameter under a load of 4-5 tons using a hydraulic press. The resulting pellets were placed into a ceramic boat for sintering. Sintering was done around 1005$^o$C for 7 minutes in flowing oxygen and furnace cooled to room temperature.

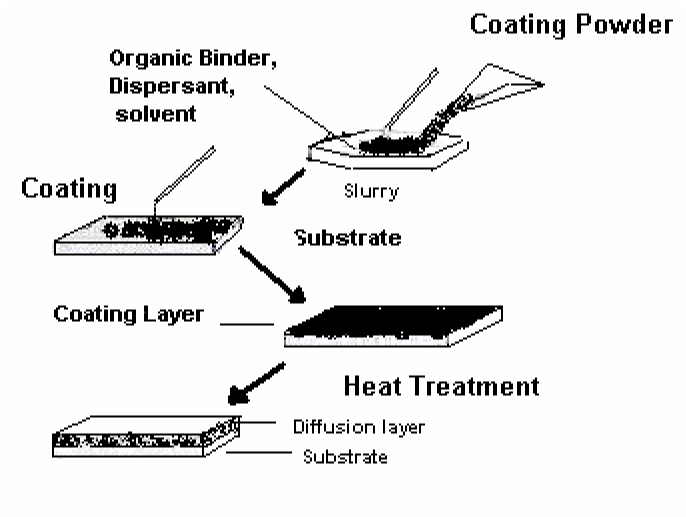


Figure 1: Slurry preparation and coating process for Y123 and Tl1212 tapes.

**Preparation of Y123 and Tl1212 coated tapes**

After electrical measurements, the superconducting pellets were reground to fine powder for coating. The slurry was made by mixing with appropriate amounts of solvent (trichloroethylene), superconducting powder, organic binder (polyvinyl butyral) and dispersant (sorbitan trioleate) with weight percent ratio of 68:22:6:4 and stirred for 5 hours to obtain a homogenously dispersed solution. The slurry was then transferred on silver substrates size 2.5 mm x 4 mm with thickness of 0.05 mm. The tape was annealed at 500$^{o}$C for 2-3 hours to remove the organic materials. The coated tapes were then wrapped with silver substrate to avoid any chemical losses during sintering at high temperature. The slurry preparation and coating process are shown in figure 1. The Y123 coated tape was then heated at 930$^{o}$C for 30 minutes in air and then slow cooled to room temperature at 40$^{o}$C/hr while the Tl1212 tape was heated at 910$^{o}$C for 60 minutes in flowing oxygen and then furnace cooled to room temperature. The final thickness of the Y123 tape is 0.19 mm and Tl1212 tape is 0.18mm

**Sample Characterization**

Powder X-ray diffraction analysis was carried out using a Bruker D8 Advance diffractometer with Cu-$K_\alpha$ radiation. For Y123 and Tl1212 bulk samples, the pellets were reground to fine powder and then XRD analysis were carried out. For Y123 and Tl1212 tape, the contents of the tape were scrapped and the powders were analyzed. However for Tl1212 tape, the amount of superconducting powder was too little and XRD do not show any meaningful peaks. The electrical resistance measurements between 16 K to 300 K were carried out using the 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 temperature sensor used was a TG-120P GaAlAs diode. The onset of superconductivity, $T_{c\ onset}$ was taken as the temperature at which the tangent of the resistance versus temperature curve intersects with the tangent of the part where resistance dropped abruptly. $T_{c\ zero}$ is defined as the temperature at which the electrical resistance readings reached zero.

## RESULTS AND DISCUSSION

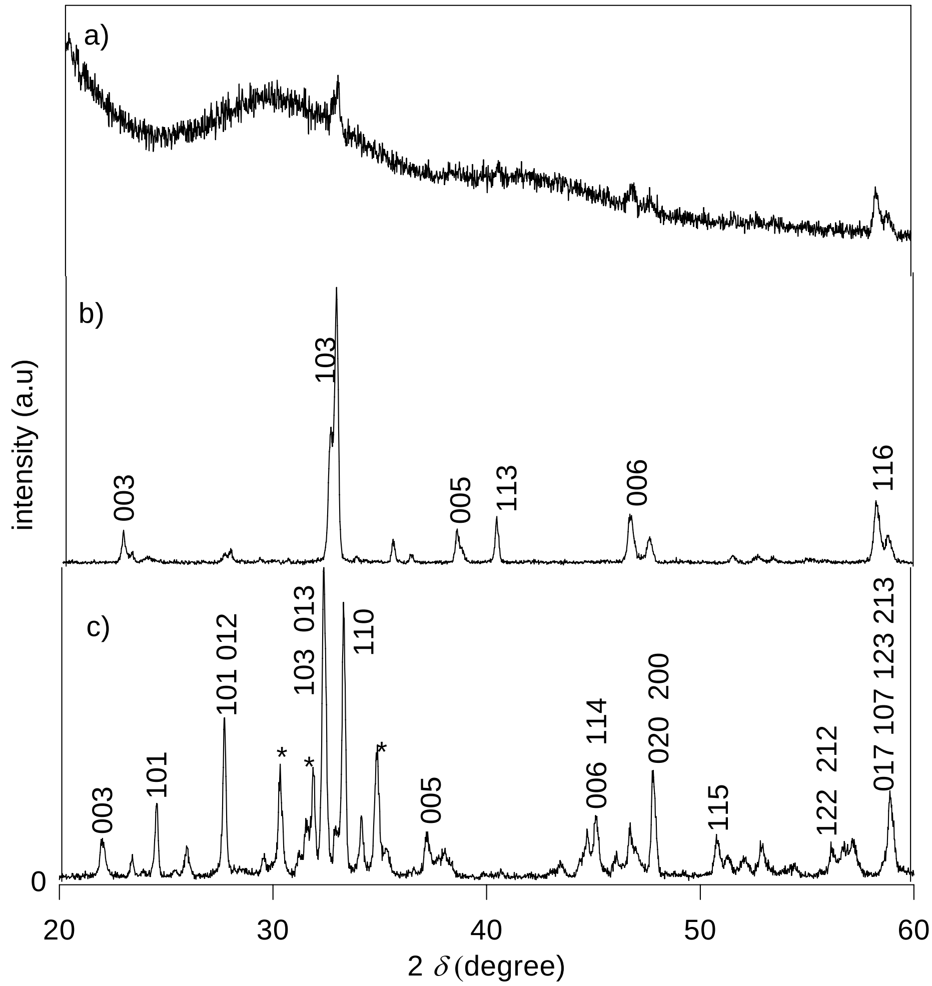


Figure 2: Powder X-ray diffraction pattern for a) Y123-tape b) Y123-bulk and c) Tl1212-bulk sample

Figure 2 a) and 2 b) shows the X-ray diffraction patterns for Y123-tape and Y123-bulk samples, respectively. The majority of the peaks are attributed to the $YBa_2Cu_3O_{7-\delta}$. For Y123 tape, several peaks identified with 123 phase can be observed. The background of X-ray diffraction is due to the glass sample holder. Meanwhile figure 2 c) shows the X-ray diffraction patterns for Tl1212 bulk sample consist of 1212 major peaks and 1201 minor peaks. The peaks due to the 1201 phase are indicated by (*) in the X-ray diffraction spectrum.

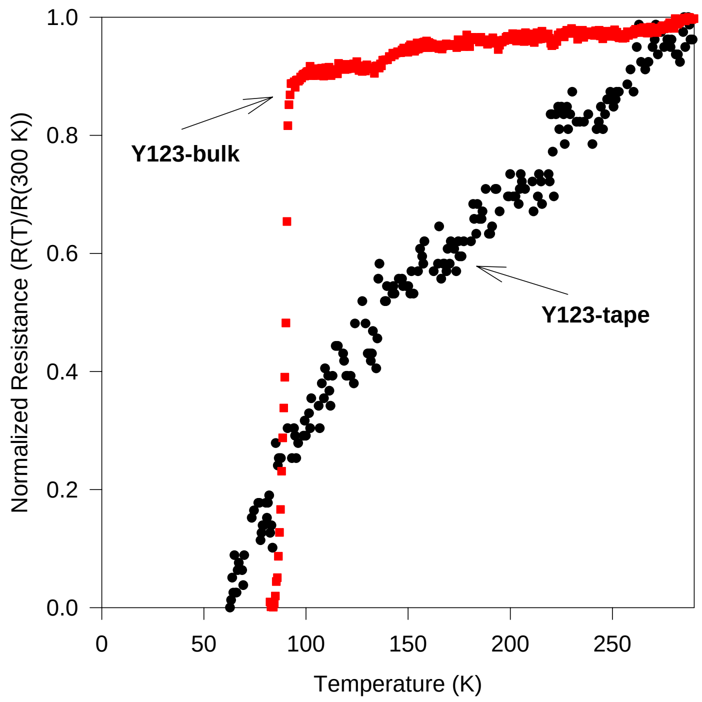


Figure 3: Normalized resistance versus temperature curve for Y123 bulk and Y123 tape sample

Figure 3 shows the normalized resistance versus temperature curves for Y123-bulk and Y123-tape samples. The Y123-bulk and Y123-tape samples showed metallic normal state behaviors with $T_{c\ zero}$ of 83 K and 62 K, respectively. Figure 4 shows the normalized resistance versus temperature curve for Tl1212-bulk and Tl1212-tape samples. Both samples showed the metallic normal state behavior with $T_{c\ zero}$ of 80 K and 72 K respectively. The critical currents ($I_c$) for Tl1212 and Y123 coated tapes at 40 K were 864.54 mA and 390.51 mA, respectively.

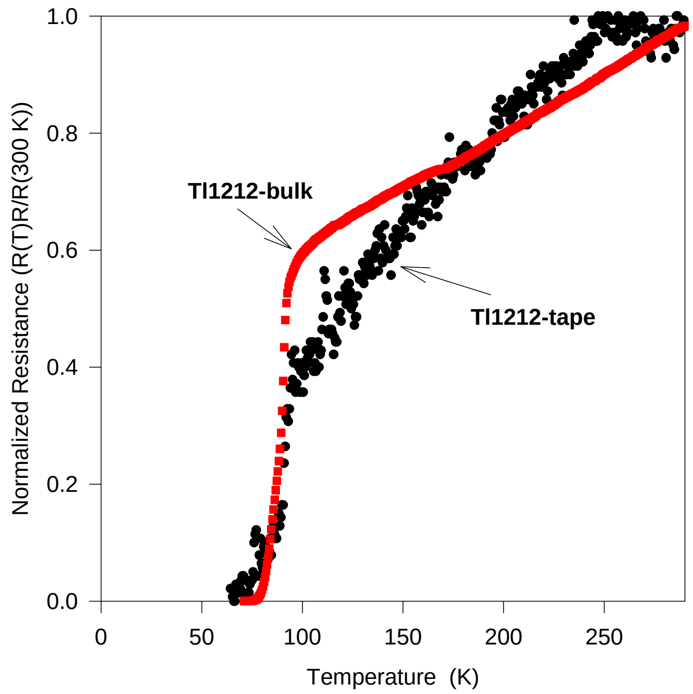


Figure 4: Normalized resistance versus temperature curve for Tl 1212 bulk sample and Tl1212 tape

Existence of organic impurity would have increased transition width ($\Delta T_c$) and lowered $T_{c\ zero}$ of the tapes. From our experience, preparation of the slurry is crucial in the fabrication process. During the preparation, only appropriate composition of binder, superconducting powder, solvent and dispersant can be applied for slurry formulation. Some of the formulation that we have tried resulted in the thick film to peel off during annealing. In forming the slurry, the powder preparation is also important because the particle size and distribution will affect the density and uniformity of the final film [5]. The choice of substrate is critical to minimize deleterious chemical interaction between the substrate and the film [5] and maintain substrate dimension during processing [11]. In this research, silver substrate has been used as it does not react to the superconducting layer material [5].

Our results show that we have successfully fabricated Y123 and Tl1212 high temperature superconductor coated tapes by the simple coating procedure. Electrical resistance measurements show that the superconducting properties of bulk and tape for both samples are maintained. Future work on the coated tapes, will concentrate on Tl1212. The effect of chemical doping and thermomechanical treatment on the Tl1212 coated will be studied. According to Alford *et al.* tape or film properties strongly depend on the microstructure. As such thermomechanical treatment can be used to vary the internal microstructure of the tapes. Future investigation on microstructure of the coated tape will be done later using scanning electron microscope (SEM) which can provide the information of cross sectional area, thus $J_c$ value can be calculated. According to T. Matsushita *et al.* one of the factors affecting performance of critical current density is thickness of superconducting tapes. For example critical current density is higher for a thinner taper, thus the thickness of coated tape should be monitored. This will also be considered in our future work.

## CONCLUSION

In conclusion, Y123/Ag ($T_{c\ zero}$ of 62 K) and Tl1212/Ag ($T_{c\ zero}$ of 72 K) coated tapes with $I_c$ of 390.5 mA and 864.5 mA, respectively, were successfully fabricated using a simple coating method. We observed that slurry preparation is crucial to produced coated tapes with good superconducting properties and does not peel off from the substrate or cracking during annealing and sintering. We also observed that superconducting properties of the tapes are comparable to that of bulk samples.

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