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Superconducting $YBa_2Cu_3O_{7-\delta}$ thick film ($T_c(0)$ = 92 K) on a newly developed perovskite ceramic substrate

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Abstract. A complex perovskite oxide, $YbBa_2NbO_6$, as a non-reacting substrate for $YBa_2Cu_3O_{7-\delta}$ superconducting film has been developed. The dielectric constant and loss factor values of the material are in the range suitable for its use as substrate for microwave applications. A $YBa_2Cu_3O_{7-\delta}$ superconducting thick film dip coated on $YbBa_2NbO_6$ substrate gave a T_c (0) of 92 K and current density of ~ $1{\cdot}3 \times 10^4$ A cm^{-2}.

Keywords. Ytterbium barium niobate; perovskite; $YBa_2Cu_3O_{7-\delta}$ superconducting film.

1. Introduction

Superconducting thick films have wide applications in transmission lines, integrated circuits and many other high frequency electronic devices (Khare *et al* 1991). Among the different copper oxide superconductors discovered, $YBa_2Cu_3O_{7-\delta}$ superconductor has gained considerable attention and a great deal of effort has been made for the production of high quality superconducting films. In the preparation of superconducting thick films, the selection of substrate is a key factor. The chemical non-reactivity between the substrate and the superconductor at the processing temperature is the most crucial factor for obtaining a high T_c superconducting (HTSC) film and the overriding importance of chemical compatibility over other substrate requirements in determining the suitability of a material as substrate for HTSC films has been emphasized in many articles (Humphereys *et al* 1990; Rowell 1991; Hollmann *et al* 1994; Koshy *et al* 1995; Phillips 1996). The high chemical reactivity of HTSC films with the substrate at the processing temperature imposes severe restrictions on the materials suitable as substrate for HTSC superconductors. MgO, the most commonly reported substrate material for $YBa_2Cu_3O_{7-\delta}$ (YBCO) does form an inter layer of barium salt at the interface if the processing temperature is above 700°C (Cheung and Ruekenstien 1989; Perng *et al* 1990; Hollmann *et al* 1994). It is also found that at higher temperature (≥ 900°C) the chemical interdiffusion of the cations across the film substrate boundary in YBCO–MgO is quite prominent. In the course of our work on the development of suitable substrate materials for HTSC (Koshy *et al* 1995; Kurien *et al* 1995), we identified a new perovskite ceramic substrate material, $YbBa_2NbO_6$, which is found to be chemically non-reacting with YBCO superconducting films even under extreme processing conditions. $YbBa_2NbO_6$ (YBNO) has moderately low dielectric constant and loss factor values making it suitable for microwave applications. YBNO has a complex cubic perovskite structure and did not show any phase transition in the temperature range 30–1300°C. Dip coated YBCO thick film on polycrystalline YBNO substrate gave a $T_c(0)$ of 92 K and J_c of ~ $1{\cdot}3 \times 10^4$ A cm^{-2}. Our results on the development and characterization of YBNO substrate and the fabrication of a superconducting YBCO thick film on it are reported in this paper.

2. Experimental

YBNO was synthesized following solid-state reaction method by thoroughly mixing high purity (99·9%) Yb_2O_3, $BaCO_3$ and Nb_2O_5 and calcining the mixture at 1100°C for 36 h with two intermediate grindings. The phase purity of the calcined mixture was checked by powder X-ray diffraction (XRD) technique and the finely ground powder was pelletized at a pressure of ~ 400 MPa in the form of circular discs having 13 mm diameter and thickness of about 1–2 mm. These discs were sintered at 1500°C for 10 h in air. The structure of the material was studied by using an X-ray diffractometer (Rigaku Dmax/2C Japan) with nickel filtered CuKα radiation (1·5406 Å). The dielectric constant (ε') and loss factor (tan δ) values of polycrystalline YBNO at room temperature and liquid nitrogen temperature were studied using an HP 4192A complex impedance analyser in the frequency range 30 Hz–10 MHz, with silver electrode on both sides of the sintered pellets. The differential thermal analysis of YBNO was carried out in the temperature range 30–1100°C.

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Pure YBCO was prepared by the solid state reaction method using high purity (99·9%) Y_2O_3, $BaCO_3$ and CuO. To study chemical reactivity between YBCO and YBNO, the two compounds were mixed in 1 : 4 vol.% ratio and pressed in the form of pellets. The pressed pellets were heated at 950°C for 15 h in air, followed by controlled slow cooling. The reactivity between YBNO and YBCO was studied by XRD technique.

The YBCO suspension for dip coating was prepared by mixing YBCO powder with isopropyl alcohol or *n*-butanol and the viscosity of the suspension was controlled by the addition of commercially available fish oil. The polished and cleaned YBNO substrate was dipped in the respective suspension till the required thickness was attained. The YBCO film was then dried in an oven and heated in a programmable furnace at the rate of 5°C/min in air up to 982°C and kept at this temperature for 2 min. The film was then cooled at the rate of 2°C/min up to 940°C and annealed at this temperature for 60 min. The film was then cooled to room temperature at the rate of 1°C/min. The structure of the dip coated YBCO thick film was examined by XRD technique and the superconductivity of the film was studied by temperature resistance measurements using four-probe technique. A Keithley current source model 181 was used for resistance measurements. The temperature of the sample was measured by a calibrated copper-constantan thermocouple with an accuracy of ± 0·2 K. The critical current density of the film was measured on thick films coated on rectangular (10×2 mm^2) substrate by standard four-probe method using the 1 μV cm^{-1} criterion.

3. Results and discussion

Figure 1 shows the XRD pattern taken on sintered YBNO sample. All peaks in the XRD pattern of the sintered YBNO sample have been indexed for a cubic perovskite structure ($A_2BB'O_6$). The *d* value is found to be in good agreement with the value reported in JCPDS file, in which doubling of the basic perovskite unit cell is observed. Doubling of the perovskite unit cell in YBNO is due to the ordering of Yb and Nb atoms in the octahedral sites (Zhang and Wang 1991). The presence of super structural lines in the XRD pattern (figure 1) indicates the ordering of the basic ABO_3 perovskite unit cell in YBNO material. The DTA analysis of YBNO showed that there was no phase transition up to 1100°C. The sintered density of YBNO measured by Archimedes method was ~ 98% of the calculated theoretical density and the room temperature resistivity of YBNO was ~ 10^{10} Ω-cm. The sintered YBNO samples were mechanically strong and could be sliced into thin pieces of 0·5 mm thickness by a diamond cutter. Good reflecting surfaces were obtained by mechanical polishing. The sintered discs were highly stable under atmospheric conditions and organic solvents such as alcohol, carbon tetrachloride, trichloroethylene etc could be used as effective cleansing agents.

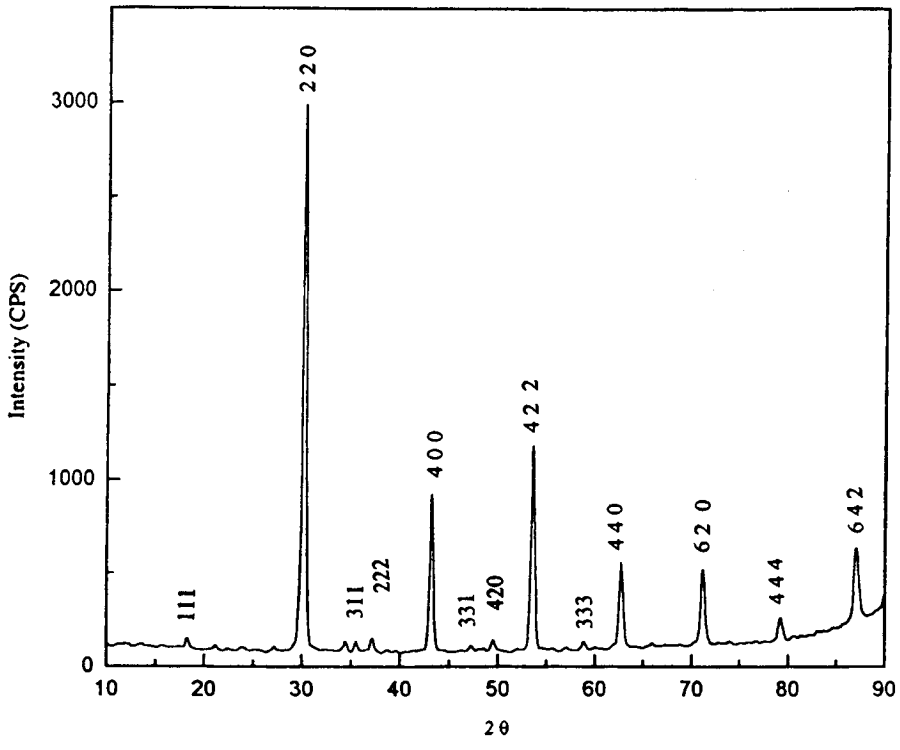

Figure 1. Powder X-ray diffraction pattern of sintered $YbBa_2NbO_6$.

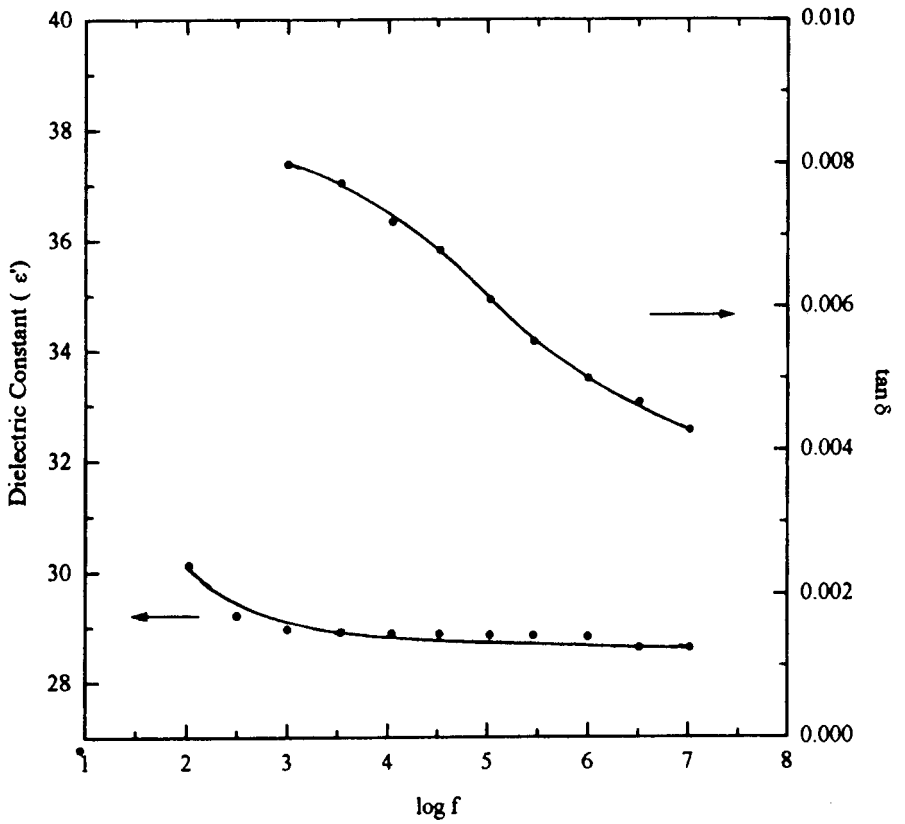

Figure 2. Variation of dielectric constant (ε') and loss factor (tan δ) for $YbBa_2NbO_6$ compounds.

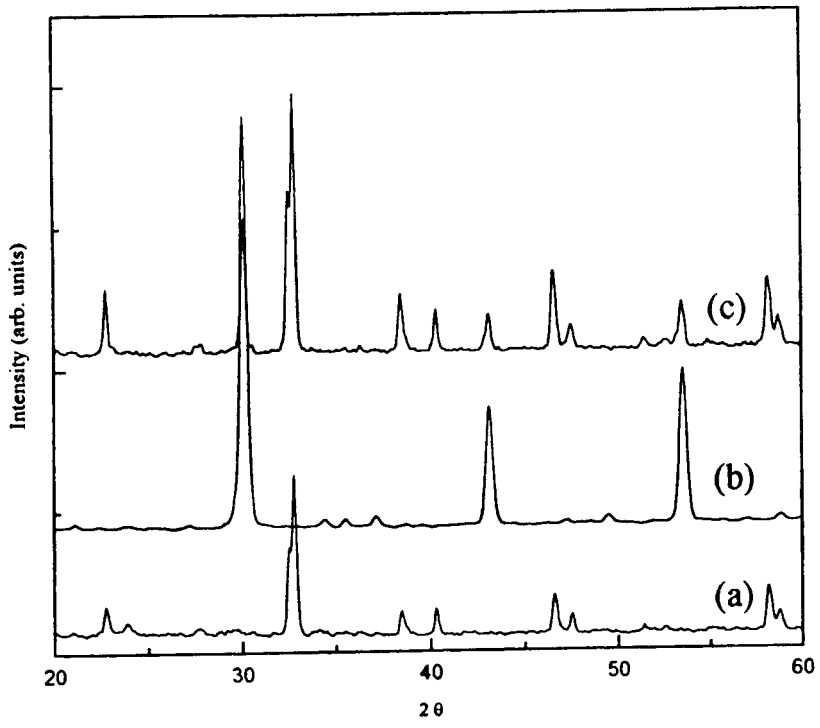

Figure 3. Powder X-ray diffraction pattern of (a) phase pure YBCO, (b) sintered $YbBa_2NbO_6$ and (c) 1 : 4 volume mixture of YBCO and $YbBa_2NbO_6$ annealed at 950°C for 15 h.

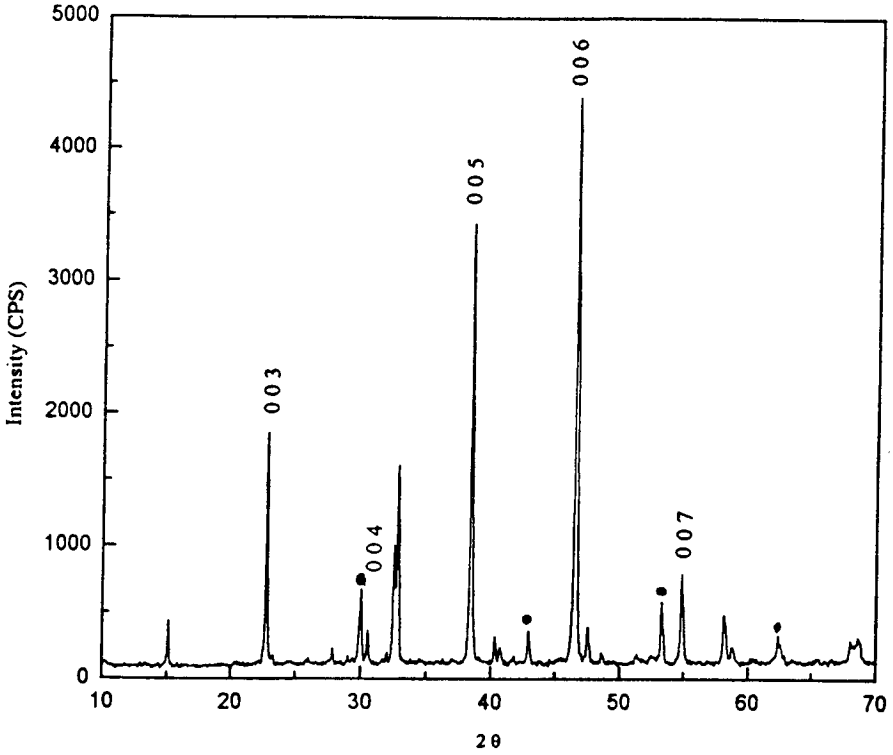

Figure 4. X-ray diffraction pattern of dip-coated $YBa_2Cu_3O_{7-\delta}$ thick film on $YbBa_2NbO_6$ substrate (substrate peaks are marked by '•').

The variation of ε' and tan δ with frequency of YBNO at liquid nitrogen temperature is shown in figure 2. The loss factor values for YBNO samples measured at liquid nitrogen temperature were found to be much less than the value obtained at room temperature. However, no substantial change was observed for dielectric constant (ε') at liquid nitrogen temperature. The ε' and tan δ values of YBNO were in a range suitable for microwave applications.

In order to see whether YBNO is chemically compatible with YBCO, its chemical reactivity with YBCO was studied at temperature up to 950°C. The XRD pattern of the annealed 1 : 4 volume ratio of YBCO and YBNO is shown in figure 3c and is compared with those of pure YBCO (figure 3a) and pure YBNO (figure 3b) samples. From figure 3 it is clear that there is no additional phase besides YBCO and YBNO composites, indicating that there is no detectable chemical reaction taking place between YBNO and YBCO even at the extreme processing conditions (within the precision of XRD technique). This indicates that YBNO is chemically compatible with YBCO superconductor even under severe heat treatment conditions.

The suitability of YBNO as a substrate for YBCO superconductor was confirmed by dip coating thick film of YBCO on polycrystalline YBNO substrate. The XRD pattern of YBCO thick film on YBNO is as shown in figure 4. Except for the characteristic peaks of YBNO all other XRD peaks in it are due to orthorhombic superconducting YBCO.

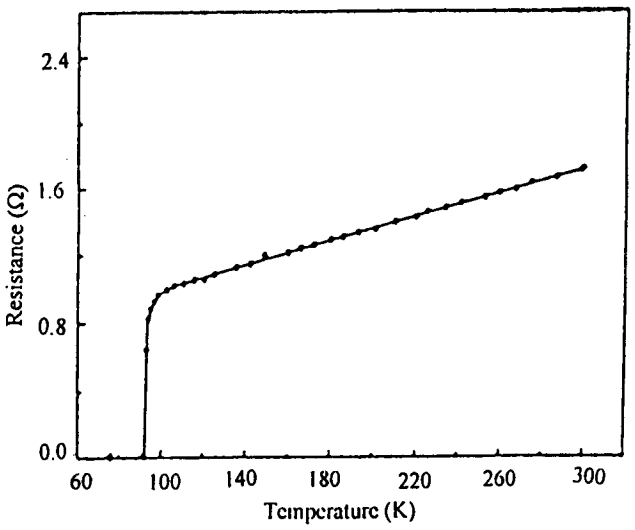

Figure 5. Temperature vs resistance curve of dip-coated YBCO thick film on $YbBa_2NbO_6$ substrate.

The superconductivity of YBCO thick film on polycrystalline YBNO substrate was studied by temperature-resistance measurement, using the standard four-probe technique. Figure 5 shows the temperature vs resistance curve of YBCO thick film developed on YBNO sub-

strate. The film shows a metallic behaviour in the normal state and gave a zero resistance transition at 92 K. The critical current density of YBCO thick film on YBNO measured at 77 K using 1 $\mu V\ cm^{-1}$ criterion under zero applied magnetic field was found to be $1{\cdot}3 \times 10^4$ A cm^{-2}. The dip coated YBCO thick film had excellent adhesion with the YBNO substrate.

4. Conclusions

$YbBa_2NbO_6$ has been synthesized, characterized and sintered as single-phase material by solid-state reaction method. It has an ordered cubic perovskite structure ($A_2BB'O_6$) with lattice constant, $a = 8{\cdot}39$ Å. The dielectric constant (28) and loss factor value (4×10^{-3}) of sintered YBNO were in a range suitable for its use as a substrate for microwave applications. The DTA studies reveal that no phase transition is occurring in YBNO in the temperature range 30–1100°C. It was found that YBNO does not react with YBCO superconductor even at severe heat treatment conditions. Superconducting YBCO thick film prepared by dip coating in polycrystalline YBNO gave $T_c(0)$ of 92 K. The critical current density of YBCO thick film developed on YBNO was $1{\cdot}3 \times 10^4$ A cm^{-2} at 77 K and zero magnetic field.

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