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Ba_2HoNbO_6: A new perovskite ceramic substrate for superconducting $YBa_2Cu_3O_{7-\delta}$ thick film ($T_c(0) = 92$ K)

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A complex perovskite oxide Ba_2HoNbO_6 has been developed as a non-reacting substrate for $YBa_2Cu_3O_{7-\delta}$ superconducting film with lattice constant $a = 8.3905$ Å. The dielectric constant (30) and loss factor value (5×10^{-3}) 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 Ba_2HoNbO_6 substrate gave a $T_c(0)$ of 92 K and current density of $\sim 1.2 \times 10^4$ A cm^{-2}. © *2003 Kluwer Academic Publishers*

1. Introduction

The discovery of superconductivity above liquid nitrogen temperature has evoked widespread interest in the field of superconductivity. The immediate application of high-T_c superconductors is likely to be in the form of thick and thin films in magnetic shielding devices, flux transformers, bolometers microwave devices etc. [1–3]. 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 of this superconductor for suitable electronic applications. In the preparation of superconducting thick films, substrate plays a vital role. 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) films [4–7]. 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 [8–10] and reduces superconducting transition temperature of the film drastically. It is also found that at higher temperatures (≥900°C) the chemical interdiffusion of the cations across the film substrate boundary in YBCO-MgO is quite prominent. In the course of our research work on the development of suitable substrate materials for HTSC [11–14], we identified a new perovskite ceramic substrate material Ba_2HoNbO_6 which is found to be chemically non-reacting with YBCO superconducting films even under extreme processing conditions. Ba_2HoNbO_6 (BHNO) has moderately low dielectric constant and loss factor values making it suitable for microwave applications. BHNO 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 BHNO substrate gave a $T_c(0)$ of 92 K and J_c of $\sim 1.2 \times 10^4$ A cm^{-2}. Our results on the development and characterisation of BHNO substrate and the fabrication of a superconducting YBCO thick film on it are reported in this paper.

2. Experimental details

BHNO was synthesized following solid-state reaction method by thoroughly mixing high purity (99.9%) $BaCO_3$, Ho_2O_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 is pelletised at a pressure of ~400 MPa in the form of circular discs having 13 mm diameter and thickness of about 1 to 2 mm. These discs were sintered at 1510°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 Cu K_α radiation (1.5406 Å). The dielectric constant (ε') and loss factor (tanδ) values of polycrystalline BHNO at room temperature and liquid nitrogen temperature were studied using an HP 4192A complex impedance analyser in the frequency range 30 Hz to 10 MHz, with silver electrode on both sides of the sintered pellets. The differential thermal analysis of BHNO was carried out in the temperature range 30–1100°C.

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 BHNO, 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

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controlled slow cooling. The reactivity between BHNO and YBCO were 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 BHNO 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 Keithly 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

Fig. 1 shows the XRD pattern taken on sintered BHNO sample. All peaks in the XRD pattern of the sintered BHNO 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 BHNO is due to the ordering of Ho and Nb atoms in the octahedral sites [15]. The presence of super structural lines in the XRD pattern (Fig. 1) indicates the ordering of the basic ABO_3 perovskite unit cell in BHNO material. The DTA analysis of BHNO showed that there was no phase transition up to 1100°C. The sintered density of

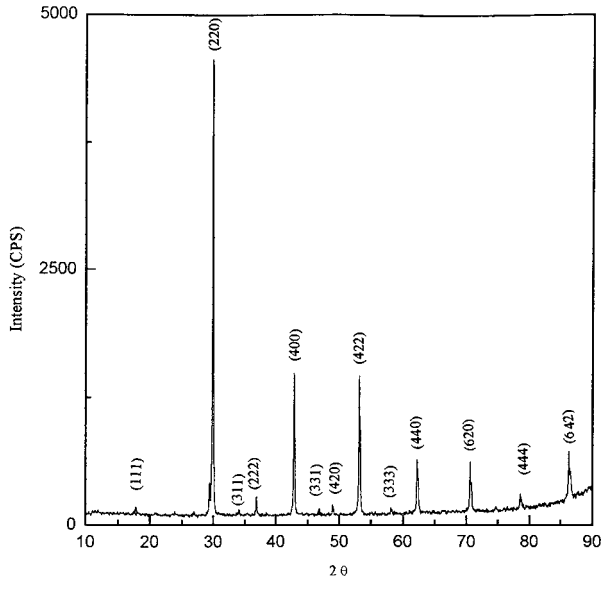

Figure 1 Powder X-ray diffraction pattern of sintered Ba_2HoNbO_6.

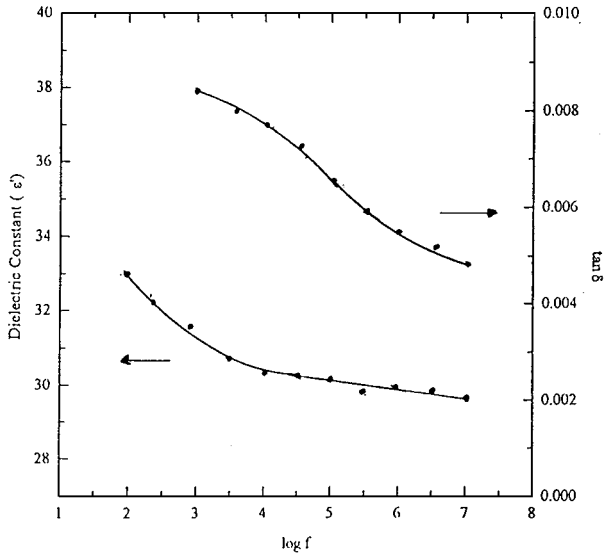

Figure 2 Variation of dielectric constant (ε') and loss factor (tanδ) for Ba_2HoNbO_6 compounds.

BHNO measured by Archimedes method was ~98% of the calculated theoretical density and the room temperature resistivity of BHNO was $\sim 10^{10}$ Ω-cm. The sintered BHNO 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.

The variation of (ε') and tanδ with frequency of BHNO at liquid nitrogen temperature is as shown in Fig. 2. The loss factor value for BHNO 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 values ε' and tanδ of BHNO were in a range suitable for microwave applications.

In order to see whether BHNO 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 BHNO is shown in Fig. 3c and is compared with those of pure YBCO Fig. 3a and pure BHNO Fig. 3b samples. From Fig. 3 it is clear that there is no additional phase besides YBCO and BHNO composites, indicating that there is no detectable chemical reaction taking place between BHNO and YBCO even at the extreme processing conditions (within the precision of XRD technique). This indicates that BHNO is chemically compatible with YBCO superconductor even under severe heat treatment conditions.

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

The superconductivity of YBCO thick film on polycrystalline BHNO substrate was studied by

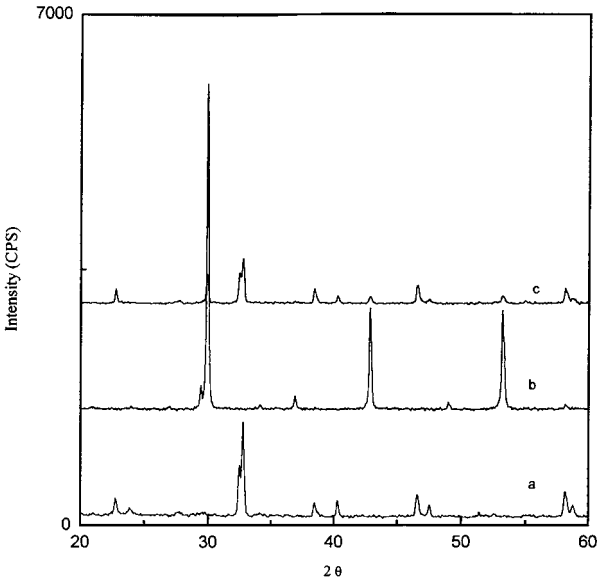

Figure 3 Powder X-ray diffraction pattern of (a) phase pure YBCO (b) sintered Ba_2HoNbO_6 (c) 1 : 4 volume mixture of YBCO and Ba_2HoNbO_6 annealed at 950°C for 15 h.

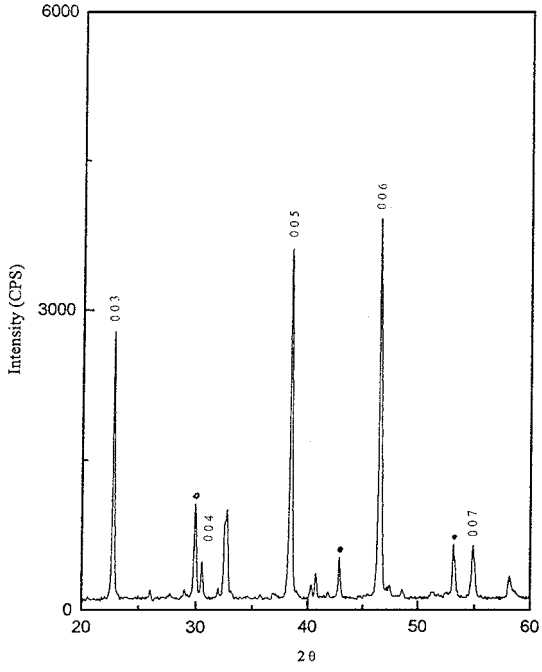

Figure 4 X-ray diffraction pattern of dip-coated $YBa_2Cu_3O_{7-\delta}$ thick film on Ba_2HoNbO_6 substrate (Substrate peaks are marked by 'o').

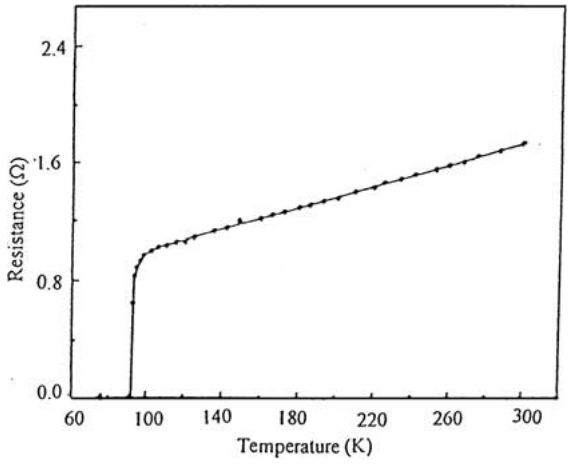

Figure 5 Temperature verses resistance curve of dip-coated YBCO thick film on Ba_2HoNbO_6 substrate.

temperature-resistance measurement, using the standard four-probe technique. Fig. 5 show the temperature vs. resistance curve of YBCO thick film developed on BHNO substrate. 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 BHNO measured at 77 K using 1 μ Vcm^{-1} criterion under zero applied magnetic field was found to be 1.2×10^4 A cm^{-2}. The dip coated YBCO thick film had excellent adhesion with the BHNO substrate.

4. Conclusion

Ba_2HoNbO_6 have 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 constants $a = 8.3905$ Å. The dielectric constant (30) and loss factor value (5×10^{-3}) of sintered BHNO 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 BHNO in the temperature range 30–1100°C. It was found that BHNO does not react with YBCO super conductor even at severe heat treatment conditions. Superconducting YBCO thick film prepared by dip coating in polycrystalline BHNO gave $T_c(0)$ of 92 K. The critical current density of YBCO thick film developed on BHNO was 1.2×10^4 A cm^{-2} at 77 K and zero magnetic field.

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