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$YBa_2SnO_{5.5}$, a novel ceramic substrate for YBCO and BiSCCO superconductors

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Abstract. $YBa_2SnO_{5.5}$ has been synthesized and sintered as single phase material for its use as substrate for both YBCO and BiSCCO superconductors. $YBa_2SnO_{5.5}$ has a complex cubic perovskite ($A_2BB'O_6$) structure with the lattice constant $a = 8{\cdot}430$ Å. The dielectric constant and loss factor of $YBa_2SnO_{5.5}$ are in a range suitable for its use as substrate for microwave applications. $YBa_2SnO_{5.5}$ is found to be chemically compatible with both YBCO and BiSCCO superconductors. The thick film of YBCO screen printed on polycrystalline $YBa_2SnO_{5.5}$ substrate gave a $T_{c(0)}$ of 92 K and a critical current density (J_c) of 4×10^4 A/cm^2 at 77 K. A screen printed BiSCCO thick film on $YBa_2SnO_{5.5}$ substrate gave $T_{c(0)} = 110$ K and current density 3×10^3 A/cm^2 at 77 K.

Keywords. Ceramic substrate; $YBa_2SnO_{5.5}$.

1. Introduction

Since the discovery of high T_c superconductors, there has been widespread interest in producing these materials in thick and thin film forms for device applications. Substrates play a vital role in the preparation of superconductor films. The chemical non-reactivity between the substrate and superconductor at the processing temperature is the most crucial factor for obtaining a high T_c superconducting film (Koinuma *et al* 1988; Rowell 1991). In addition, for the microwave applications of the film, the substrates should have low dielectric constant and loss factor. Many commonly available substrates such as Si, SiO_2, Ge, Al_2O_3 etc react with YBCO and BiSCCO superconductors at the processing temperature thereby reducing the superconducting transition temperature of the film drastically (Dou *et al* 1989; Stonstrop and Enguell 1990). MgO is the only substrate material reported to be suitable for both YBCO and BiSCCO films for microwave applications. However, MgO does form an interlayer of Ba salt at the YBCO–MgO interface if the processing temperature is above 700°C, reducing the superconducting transition temperature of YBCO considerably (Cheung and Ruckenstein 1989; Preng *et al* 1990). Recently we have reported a new ceramic material $YBa_2SnO_{5.5}$ (YBSO) which was found to be chemically compatible with YBCO even under severe heat treatment (Koshy *et al* 1993). Our further studies revealed that YBSO is chemically compatible with BiSCCO superconductors also. We have now successfully screen printed both YBCO and BiSCCO thick films on YBSO substrate. The YBCO thick film screen printed on YBSO substrate gave a zero resistivity superconducting transition $T_{c(0)} = 92$ K and a critical current density $J_c = 4 \times 10^4$ A/cm^2 at 77 K. A screen printed Bi(2223) thick film on YBSO substrate gave a $T_{c(0)} = 110$ K with a critical current density

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$J_c = 3 \times 10^3$ A/cm^2 at 77 K. In the present paper, we describe the development of YBSO as a polycrystalline substrate material for microwave applications, its chemical non-reactivity with BiSCCO superconductors and the fabrication of YBCO and BiSCCO superconducting thick films on this substrate.

2. Experimental

$YBa_2SnO_{5\cdot5}$ was synthesized following the conventional solid state reaction method. High purity (99·9%) Y_2O_3, $BaCO_3$ and SnO_2 were wet mixed in acetone medium using an agate mortar. The mixed powder was pressed in the form of pellets and was calcined in air at 1150°C for 72 h with three intermediate grindings. Prolonged calcination with multiple grindings was essential to get a single phase YBSO. The calcined material was then finely ground and pelletized at a pressure of 6 ton/cm^2 in the form of circular discs having 11 mm dia and 2 mm thickness. These discs were sintered at 1400°C for 12 h in air. The structure of the compound was examined by X-ray diffraction (XRD) method using a computerized Rigaku X-ray Diffractomer with nickel filtered CuKα radiation. The dielectric constant and loss factor of YBSO were measured using an HP 4192 A Complex Impedance Analyser in the frequency range 30 Hz to 13 MHz at room temperature with silver electrodes on both sides of the pellet.

Thick film paste of YBCO for screen printing was made by mixing superconducting YBCO powder (prepared by standard solid state reaction) with isopropyl alcohol. The viscosity of the thick film paste was controlled by the addition of commercially available fish oil. This paste was screen printed on the polished YBSO substrate using a screen of 325 mesh size. The printed film was heated in a programmable furnace in air at a rate of 2°C/min to 980°C. The film was cooled down slowly at 0·2°C/min to 940°C and was kept at this temperature for 30 min and then cooled to room temperature at a rate 1°C/min. Heating up to 980°C was necessary to get good adhesion and slow cooling at 1°C/min from 940°C to room temperature was essential for oxygenation.

For screen printing BiSCCO films, a thick film paste was prepared by mixing pure Bi(2223) powder prepared from the nominal composition $Bi_{1\cdot5}Pb_{0\cdot5}Sr_2Ca_2Cu_3O_x$ (BiSCCO) (see Koshy *et al* 1994 for details) and this paste was screen printed on a polished YBSO substrate using a screen of 325 mesh size. The printed film was heated in a programmable furnace in air at a rate of 5°C/min up to 880°C. The film was kept at this temperature for 2 min and then cooled at a rate of 0·1°C/min to 850°C and the film kept at this temperature for 12 h. The film was then furnace cooled to room temperature.

The structure of the screen printed YBCO and BiSCCO films on YBSO substrates was examined by X-ray diffraction. The superconductivity of the films was studied by temperature-resistance measurements using standard four-probe technique. A Keithley current source model 220 and a Keithley nanovoltmeter model 181 were used for resistance measurements. The temperature of the films were measured by a calibrated copper constantan thermocouple with an accuracy of ±0·2 K.

3. Results and discussion

The XRD pattern of a sintered YBSO for 2Θ values between 5° and 90° is shown in figure 1. The XRD peaks including the minor peaks of $YBa_2SnO_{5\cdot5}$ are indexed for a complex cubic perovskite ($A_2BB'O_6$) structure with the lattice constant $a = 8{\cdot}430$ Å and the computerized XRD data of $YBa_2SnO_{5\cdot5}$ is given in table 1. The DTA experiment carried out using a Shimadzu DTA-50 H machine showed that there is no phase transition in YBSO material up to a temperature of 1350°C. The sintered density of YBSO as measured by Archimedes method was 96% of the theoretical density (6·12 g/cm^3) and the room temperature resistivity of YBSO was 10^{10} Ω cm. The electrical resistivity and density of the humidity treated YBSO samples were same as that of the sintered sample indicating that YBSO is highly stable under atmospheric conditions. The YBSO samples were mechanically strong and could be sliced into thin pieces of 0·5 mm thickness by diamond cutter. Good reflecting surfaces were obtained by mechanical polishing and organic solvents such as alcohol, acetone, carbon tetrachloride could be used as effective cleaning agents.

The dielectric constant (ε′) and loss factor (tan δ) of YBSO were studied in the frequency range 10 Hz to 13 MHz at room temperature and the variation of ε′ and

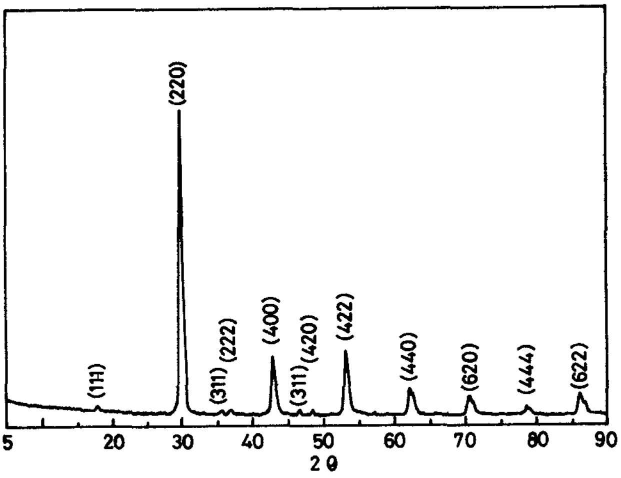

Figure 1. X-ray diffraction of phase pure sintered $YBa_2SnO_{5\cdot5}$.

Table 1. Computerized X-ray diffraction data of $YBa_2SnO_{5\cdot5}$.

2-Θ	Int.	Width	*d*	I/I_0	hkl
18·200	145	0·345	4·870	6	111
29·930	2652	0·780	2·983	100	220
35·720	156	0·495	2·512	6	311
37·080	207	0·780	2·423	8	222
42·820	588	0·510	2·110	22	400
46·710	191	0·465	1·943	7	331
48·560	208	0·435	1·873	8	420
53·640	509	0·510	1·707	19	422
62·090	302	0·510	1·494	11	440
70·560	248	0·855	1·334	9	620
78·340	120	0·330	1·220	5	444
86·200	252	0·810	1·127	10	642

tan δ are shown in figure 2. The dielectric constant and loss factor values of YBSO are comparable to those of MgO at these frequencies. Based on the doubling of simple perovskite unit cell, the lattice constant of YBSO is also comparable to that of MgO. The melting experiments conducted on the YBSO samples showed that this material melts congruently and could be grown as a single crystal from the melt.

One of the most important criteria for the selection of YBSO as a substrate for YBCO and BiSCCO superconductors is the chemical non-reactivity between YBSO and the superconductor films at the processing temperatures. The chemical non-reactivity of YBSO with YBCO has already been discussed in our earlier publication. The chemical compatibility between YBSO with Bi(2223) was studied by mixing 20 vol% of YBSO in Bi(2223) and annealing the pressed pellet at 850°C for 20 h and figure 3 (curve C) shows the XRD pattern of the annealed YBSO-Bi(2223) sample. The XRD pattern of the two phases (figure 3, curve C) in the annealed sample is compared with those of pure Bi(2223) (figure 3, curve A) and pure YBCO (figure 3, curve B) samples. Figure 3 (curve C) shows that there is no additional phase formed other than those of Bi(2223) and YBSO in the annealed YBSO-Bi(2223) composite (within the precision of XRD). These results indicate that YBSO is chemically compatible with Bi(2223) superconductor also.

The suitability of YBSO as substrate for YBCO and BiSCCO superconductors was confirmed by screen printing YBCO and Bi(2223) thick films on YBSO polycrystalline substrate. Figures 4a and b shows the XRD patterns of YBCO and BiSCCO thick films developed on YBSO substrates. Except for the characteristic peaks of YBSO, all the other peaks in figure 4a are those of orthorhombic superconducting YBCO. However in the case of BiSCCO (figure 4b) the screen printed film contained both Bi(2223) (~ 80 vol%) and Bi(2212) (~ 20 vol%) phases.

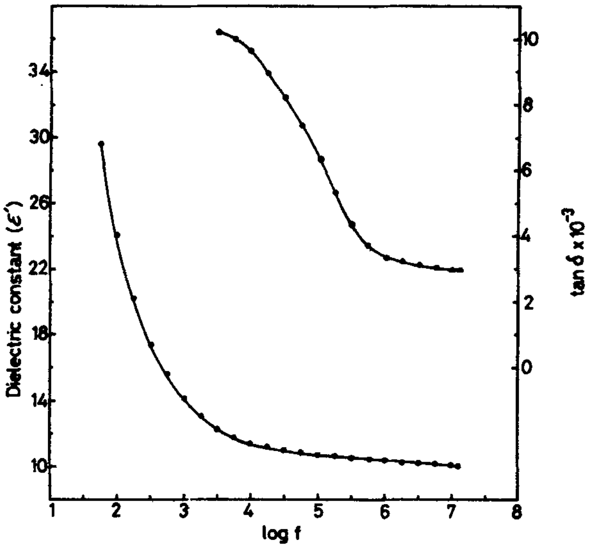

Figure 2. Variation of dielectric constant (ε′) and loss factor (tan δ) with frequency for $YBa_2SnO_{5.5}$.

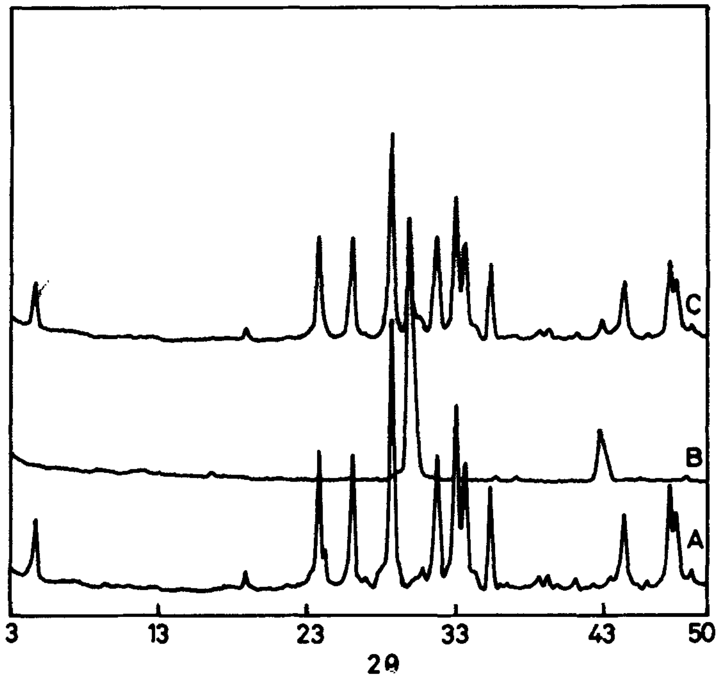

Figure 3. X-ray diffraction pattern for (A) Bi(2223) superconductor, (B) pure $YBa_2SnO_{5.5}$ and (C) 4 : 1 volume mixture of Bi(2223) and $YBa_2SnO_{5.5}$ annealed at 850°C for 20 h.

The formation of Bi(2212) phase may be due to the partial melting of the film during the thick film processing at 880°C. The volume fractions of Bi(2223) and Bi(2212) phase in the BiSCCO thick film were determined from the computerized XRD data of the thick film using the empirical relation (Chen *et al* 1992)

$$V = (Y - 29{\cdot}07)/(28{\cdot}81 - 29{\cdot}07)$$
$$28{\cdot}81 < Y < 29{\cdot}07,$$

where V is the volume fraction of Bi(2223) and $Y = 2\Theta$ for the peak appearing between 28·81 and 29·07, caused by the interplay of reflection of the crystal planes 0010 of Bi(2223) phase and 0012 of Bi(2223) phase. Figures 5a and b show the resistance-temperature curves for screen printed YBCO and BiSCCO thick films, respectively, developed on YBSO substrates. The films showed metallic behaviour in the normal state and gave a zero resistivity transition temperature of 92 K for YBCO and 110 K for BiSCCO thick films. The critical current density of the YBCO films at 77 K with 1 μV criterion was 4×10^4 A/cm^2 and for BiSCCO film, the critical current density was 3×10^3 A/cm^2 at 77 K. The screen printed YBCO and BiSCCO films had excellent adhesion to the YBSO substrates.

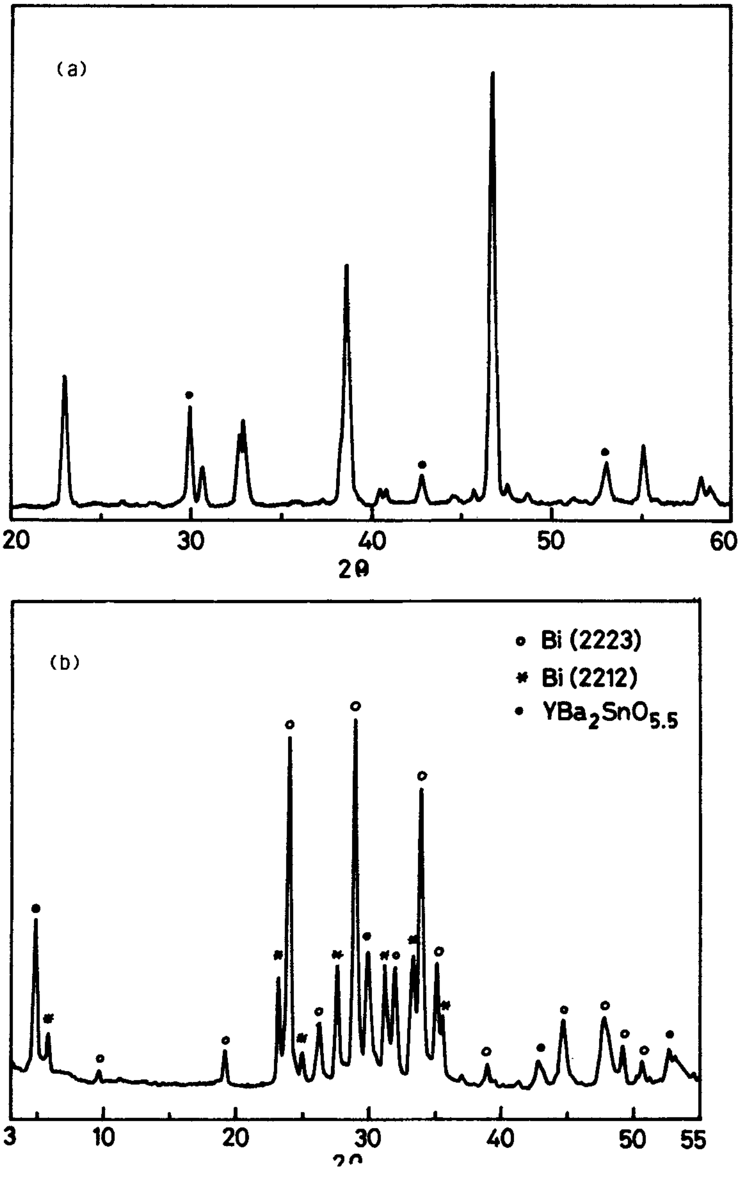

Figure 4. X-ray diffraction patterns of screen-printed (a) $YBa_2Cu_3O_{7-\delta}$ thick film and (b) BiSCCO thick film on $YBa_2SnO_{5.5}$ substrates. Substrate peaks are marked by '●'.

4. Conclusions

$YBa_2SnO_{5.5}$ has been synthesized and sintered as single phase material for its use as substrate for YBCO and BiSCCO superconductors for microwave applications. This material has a complex cubic perovskite ($A_2BB'O_6$) structure with the lattice constant $a = 8{\cdot}430$ Å. The dielectric constant and loss factor of the $YBa_2SnO_{5.5}$ are

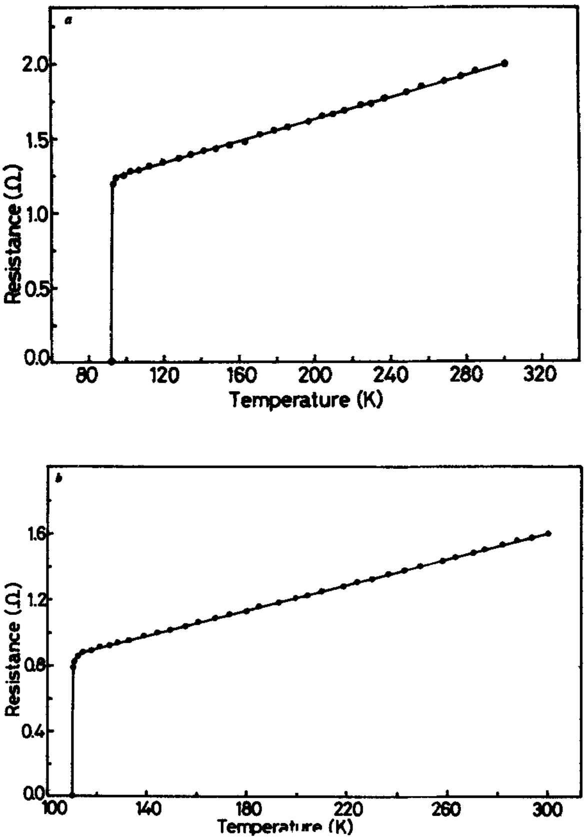

Figure 5. Temperature-resistance curve of screen-printed (a) $YBa_2Cu_3O_{7-\delta}$ thick film and (b) BiSCCO thick film on $YBa_2SnO_{5.5}$ substrates.

in a range suitable for microwave applications. $YBa_2SnO_{5.5}$ is found to be chemically compatible with YBCO and BiSCCO superconductors. The use of $YBa_2SnO_{5.5}$ as substrate for YBCO and BiSCCO superconductors was confirmed by screen printing superconducting YBCO and BiSCCO thick films on polycrystalline $YBa_2SnO_{5.5}$ substrate which gave $T_{c(0)}$ of 92 K and 110 K respectively.

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