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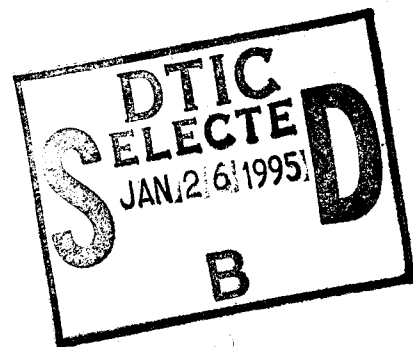


Electrical Characteristics of Barium Strontium Titanate-Oxide Composite Films

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13. ABSTRACT (Maximum 200 words) In a previous work, composites of BSTO combined with other nonelectrically active oxide ceramics have been formulated and have demonstrated adjustable electronic properties which can be tailored for use in various electronic devices. ¹ One application has been for use in phased array antennas and insertion has been accomplished into several working antenna systems. ² To further accommodate the frequencies required by these phased array antennas, thin films of the composites have been fabricated using a Krypton Fluoride excimer laser as an ablation source. The electrical properties, including the dielectric constant and the tunability (change in the dielectric constant with applied voltage) have been measured. The results have been compared to those obtained for the bulk ceramic composites and other BSTO/oxide composite thin film structures. ³				
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INTRODUCTION

Phased array antennas are currently constructed using ferrite phase shifting elements. These antennas can steer transmitted or received signals either linearly or in two dimensions without mechanically oscillating the antenna. Due to the circuit requirements necessary to operate these present day antennas, however, these are costly, large and heavy. Therefore, the use of these antennas has been limited primarily to military applications which are strategically dependent on such capabilities. In order to make these devices available for many other commercial and military uses, the basic concept of the antenna must be improved. Towards this goal, a ceramic Barium Strontium Titanate, $Ba_{1-x}Sr_xTiO_3$, (BSTO), phase shifter using a planar microstrip construction has been demonstrated.¹ However, to meet the required performance specifications (e.g., maximum phase shifting ability), the BSTO electronic properties must be optimized. As part of this optimization process, various composites of BSTO and non-ferroelectric oxides have been formulated and proven successful.

However, in order to obtain higher operating frequencies (30 GHz and beyond) and to decrease the voltage requirements, thin film fabrication of the above candidate materials is necessary. This paper outlines the work on the characterization of the thin films of undoped and modified BSTO deposited by the pulsed laser deposition (PLD) method. The electronic properties of the films were measured using an HP 4194A impedance analyzer. The results of these measurements will be discussed.

EXPERIMENTAL

The metallized films used for the electrical measurements were: (1) Sapphire / RuO_2 / BSTO / Pt and (2) Sapphire / RuO_2 / BSTO with 1 wt.% oxide II / Pt. Prior to PLD, the sapphire substrates utilized underwent a cleaning cycle which included an ultrasonic cycle of TCE followed by two methanol ultrasonic cycles. The samples were then rinsed with methanol and air dried. The lattice parameters and dielectric constants of the sapphire substrates used in this experiment are 4.76 Å and 11 (at 300 K) respectively.

Prior to the thin film deposition, a ground plane electrode of Ruthenium oxide (RuO_2) was sputtered onto the substrates at a substrate temperature of 200 °C and a O_2/Ar ratio of 1:4 with a total pressure of 10 mT. The Ruthenium oxide films were 3000 Å thick. The resistivity of the as-deposited films were in the order of 160 μohms-cm. They were annealed at 600 °C for 30 minutes to lower the resistivity and were cooled by furnace quenching. The resistivity of the annealed films were measured to be 110 μohms-cm.

The targets chosen for this work were $Ba_{0.6}Sr_{0.4}TiO_3$ (BSTO) and BSTO with 1 wt.% of an additive oxide, referred to hereafter as oxide II. The PLD of the ferroelectric thin films was accomplished using a Questek 2000 krypton-fluoride excimer laser with a wavelength of 248 nm and a repetition rate of 10 Hz. The substrate was held parallel to the target and their separation distance was maintained at 55 mm. The average pulse energy was 300 mJ with a 20 ns pulse width. The oxygen partial pressure in the chamber was 100 mT and the substrate temperature was 500 °C, which was monitored by a thermocouple clamped between the heater and the substrate. The powder pressed ceramic targets were prepared according to a description

published previously.² A Dektak-200 profilometer was used to measure the films thicknesses. It was measured to be 6000 Å for both the films. The compositions of the films were confirmed by Glancing Angle X-ray-diffraction (GAXRD).

After the deposition of the thin films, the top Pt electrodes were deposited by electron beam evaporation. The thicknesses of the top electrodes were measured to be approximately 3000 Å using a Dektak-200 profilometer.

The dielectric constant (ϵ') and % tunability were determined for both thin film/substrate combinations. The % tunability of a material is determined using the following equation:

$$\% \text{ tunability} = \{ \epsilon'(0) - \epsilon'(V_{\text{app}}) \} / \{ \epsilon'(0) \} \quad (1)$$

The tunability measurements were taken with an applied electric field which ranged from 0 to +/- 3.3 V/micron (μm). The electronic properties were measured at two frequencies, 30 KHz and 0.5 MHz. Capacitance versus voltage (C-V) measurements for the films were taken using an HP4194 impedance / phase gain analyzer. The voltage, applied internally through the HP 4194A, was varied from -2.0 V to +2.0 V.

RESULTS AND DISCUSSION

Electronic Measurements

Fig. 1 shows the capacitance versus voltage characteristics at 30 KHz for the BSTO (undoped) film deposited on RuO₂/sapphire. The curve shows a symmetric capacitance-voltage relationship which is characteristic of *paraelectric* films. The dielectric constant at zero bias was calculated to be 1380 and the tunability is 48% at a field of 3.3 V/ μm . The bulk undoped material has a dielectric constant of 3300 and a tunability of 20% at 0.73 V/ μm .¹ It has been previously shown that the dielectric constant of ferroelectric films are inherently less than the bulk ceramic values due to oxygen defects at the electrode/film interface.⁴ Also any porosity and/or leakage current in the films will tend to decrease the dielectric constants obtained.

The C-V curve at 30 KHz for the BSTO/1 wt% oxide II film deposited on RuO₂/sapphire is shown in Fig. 2. The curve shows a typical paraelectric behavior (i.e., a symmetric capacitance) with positive and negative bias applied. The dielectric constant at zero voltage calculated from this curve is 600. The tunability obtained up to 3.3 V/ μm was 34%. The value for the dielectric constant found in the bulk ceramic target of BSTO/1 wt% oxide II was 2700 and a tunability of 46% at 2.5V/ μm [1]. Table I summarizes the values of the dielectric constants and tunability of the various BSTO thin films measured at 30 KHz and their ceramic counterparts.

Similar measurements of the same thin films were also carried out at 0.5 MHz. For the undoped BSTO thin film, a dielectric constant of 940 and a tunability of 55% ($V_{\text{applied}} = 3.3 \text{ V}/\mu\text{m}$) were obtained. For the BSTO/oxide II thin film, a dielectric constant of 310 and a tunability of 39% ($V_{\text{applied}} = 2.0 \text{ V}/\mu\text{m}$) were obtained.

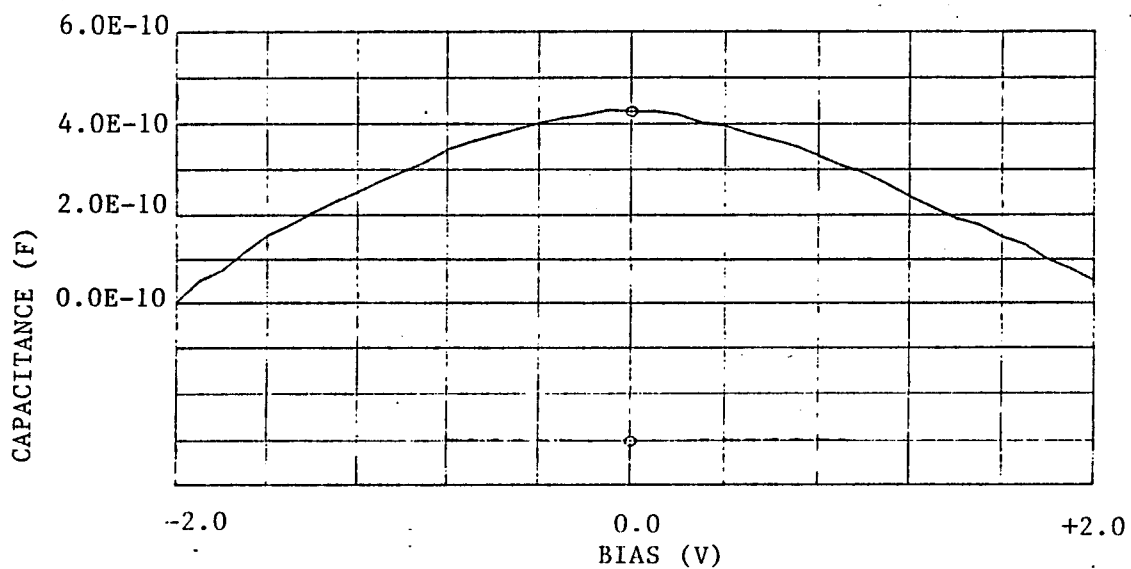


Fig. 1. Capacitance versus voltage for BSTO (undoped) deposited on RuO₂/Sapphire with Pt top electrode.

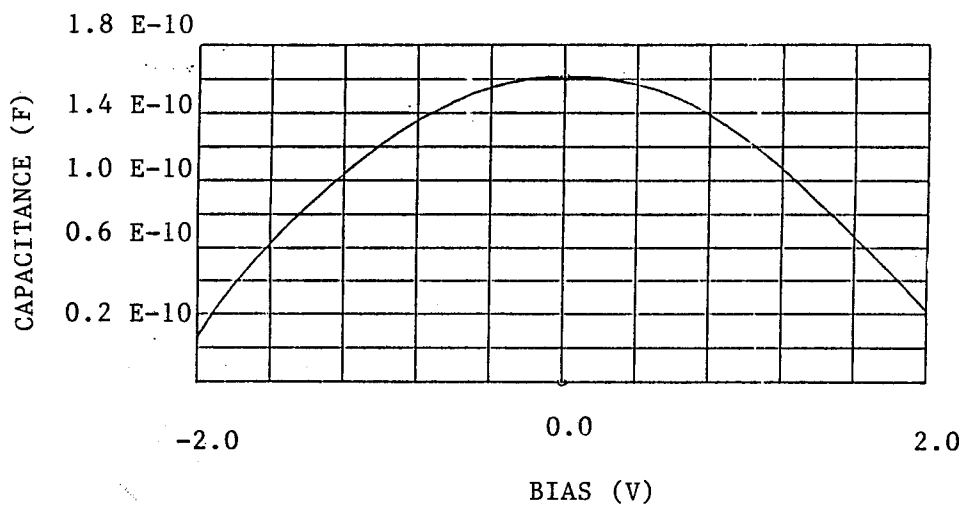


Fig. 2. Capacitance versus voltage for BSTO/1 wt% Oxide II deposited on RuO₂/Sapphire with Pt top electrode.

TABLE I. Electronic Properties of BSTO (Ba = .6) and BSTO/Oxide Composite Thin Films.

<u>Material</u>	<u>Applied Field (V/μm)</u>		<u>ϵ'(V=0)</u>		<u>% Tunability</u>	
	<u>Film</u>	<u>Bulk</u>	<u>Film</u>	<u>Bulk</u>	<u>Film</u>	<u>Bulk</u>
BSTO	3.3	0.73	1380	3300	48	20
BSTO / OXIDE II	3.3	2.5	600	2700	34	46
BSTO / OXIDE III (Ref. 3)	2.0	2.3	398	1276	79	16

CONCLUSION

Thin films of both undoped and oxide modified BSTO have been deposited by PLD onto RuO₂/Sapphire substrates. The electronic properties of the undoped and oxide modified BSTO thin films exhibited similar trends relative to the bulk materials as shown in Table I.

We have shown that the tailoring of the electronic properties of BSTO thin films in the low frequency region is possible through the incorporation of metal oxides. The lowering of the dielectric constants, alongwith a high tunability, plays an important role in the impedance matching of these films into the electronic circuits. Further investigation of such tailoring in the microwave region through the incorporation of such oxides is underway.

ACKNOWLEDGMENTS

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REFERENCES

1. L.C. Sengupta, S. Stowell, E. Ngo, M.E. O'Day and R. Lancto, *J. of Integrated Ferroelectrics*, (1994), in press.
2. R. Babbitt, T. Koscica, W. Drach, and D. Didomenico, *J. of Integrated Ferroelectrics*, (1994), in press.
3. S. Sengupta, L.C. Sengupta, S. Stowell, W.E. Kosik, E. Ngo, D.K. Vijay, and S.B. Desu, IEEE Transactions of the Proceedings of ISAF '94, Penn State, University Park, PA, (in press).
4. C.J. Brennan, *Integrated Ferroelectrics* **2**, 73 (1992).

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