

Fabrication and characterisation of barium strontium titanate thick film device structures for microwave applications

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Abstract

Tunable capacitor elements for microwave phase shifter applications have been fabricated by screen printing barium strontium titanate (BST) films on alumina substrates. A $\text{Ba}_{0.70}\text{Sr}_{0.30}\text{TiO}_3$ composition was chosen for the initial devices as it has been shown to exhibit high tunability at room temperature. A vertical capacitor test structure involving a Pt lower electrode, BST film and Ag top electrode has been used throughout the work. The tunability and figure of merit (phase shift/dB of insertion loss) at 2–3 GHz were found to be strongly dependent on the sintering temperature of the BST layers, with properties improving as the sintering temperature was increased. However, for sintering temperatures $>1280^\circ\text{C}$, the device properties could not be measured, possibly indicating a problem with the lower Pt electrodes. In order to reduce the sintering temperature required for densification, test structures have also been fabricated using other BST compositions to which sintering aids have been added. Finally, a reflection-type phase shifter (RTPS) based on the capacitor test structure and optimised processing conditions is presented with microwave measurements results.

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1. Introduction

Phase shifters are important components in tunable communication systems such as steerable antennas. Barium strontium titanate, $\text{Ba}_x\text{Sr}_{1-x}\text{TiO}_3$ [(BST), $0 \leq x \leq 1$], has attracted most attention in the development of tunable microwave devices as it exhibits a large change in dielectric constant with an applied dc electric field.^{1–3} Most of the work reported to date in this area focuses on BST thin films.^{4–6} Recently BST thick films have been investigated for such applications as a cost effective route using alumina substrates, which because of their low relative permittivity and low cost, are commonly used in microwave devices applications.^{7–9} Compared to bulk materials, the thick films often show inferior properties due to much more complex materials systems, for example, the introduction of interfaces between the thick films, substrate, and electrodes. As a deleterious reaction between BST thick films and alumina substrates

occurs at sintering temperatures $>1200^\circ\text{C}$, sintering temperatures for BST films should be limited to $<1300^\circ\text{C}$.¹⁰ For BST films without sintering aids this leads to films with a porous microstructure possessing poor adhesion to the substrate.

In this paper, efforts towards solving the problems of porosity and poor adhesion of BST thick films are reported. A vertical capacitor test configuration has been employed in this study in order to understand the impact of composition and processing conditions on the device performance, and has been transferred to a phase shifter design to test the microwave properties.

2. Experimental

$\text{Ba}_x\text{Sr}_{1-x}\text{TiO}_3$ thick films with composition of Ba/Sr = 50/50, 60/40 and 70/30 ($x = 0.50, 0.60$ and 0.70 , respectively) were fabricated on 99.6% alumina substrates using a conventional screen printing method. The ink was prepared by mixing BST powders produced via a solid state route with a commercial vehicle (Blythe 6321 Medium) at a solids loading of 40 vol.% via three-roll milling. Platinum bottom electrodes (between the substrate and BST film) were applied and sintered at 1400°C for

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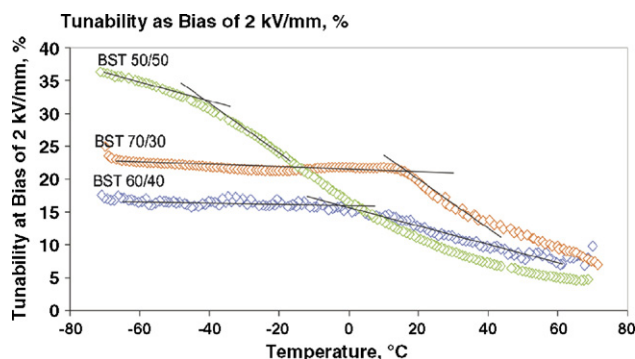


Fig. 1. Tunability of $\text{Ba}_x\text{Sr}_{1-x}\text{TiO}_3$ thick films with different compositions over temperature range of -70 to $+70$ °C.

2 h prior to BST film deposition. Silver top electrodes (applied on the top surface of the sintered BST film) were also screen printed, and sintered at 850 °C for 10 min. Glass frit (Johnson Matthey Glass) which contains 43 wt.% PbO was used as the sintering aid.

The dielectric properties of the films were characterised using an impedance analyser (HP 4914A) with a metal-ferroelectric-metal (MFM) capacitor configuration at frequencies from 100 Hz to 10 MHz and over the temperature range from -70 to 70 °C. The tunability of the films was measured at 10 kHz with an internal bias up to 40 V and calculated from $1 - \varepsilon(V)/\varepsilon(0)$, where $\varepsilon(V)$ and $\varepsilon(0)$ is the relative permittivity with bias V and without bias respectively. However, in the capacitor test structure, the tunability was calculated by using $C(V)$ and $C(0)$, which is the capacitance with bias V and without bias, respectively. The performance of the phase shifter was measured using a HP8722ES Network analyzer with the packaged devices mounted in an Anritsu Model 8680V test fixture.

3. Results and discussion

The tunabilities of BST thick films with different compositions are shown in Fig. 1. The Curie temperature of the films changes with different composition, and generally, shifts to lower temperature with the decrease of the barium content. The films all show two stage tunability curves. In the high tem-

perature paraelectric (PE) region, tunability increases as the temperature approaches the Curie point but becomes stable in the ferroelectric (FE) region. Higher tunability and good temperature stability are exhibited in the ferroelectric region for all compositions. BST thick films with composition of $x = 0.70$ show higher tunability in the paraelectric region than other two compositions. The diffuse transitions exhibited in all the compositions were caused by porous microstructures and inhomogeneous compositions due to the low sintering temperature.⁸

BST70/30 thick films capacitor test structures sintered at two different temperatures are shown in Fig. 2. In the test structures, a platinum film underneath the BST thick film acts as the bottom electrode. The top electrode on the BST thick film and the connection to the bottom electrode are both silver thick films. Notably, the area of the top electrode varies in size. Other structural parameters in the capacitor test structure also vary, such as the working gap which is the perspective distance between the top silver electrode and the bottom platinum electrode.

The BST films sintered at 1200 °C have smooth surfaces and clear edges, as shown in Fig. 2(a), but exhibit a porous structure, as evidenced by the mesh marks in the top electrode layer, indicating the suction of the ink solvent by the porous BST films. In comparison, the films sintered at 1260 °C show much denser microstructures, with no observable mesh marks in the top electrode, as shown in Fig. 2(b). SEM micrographs of the film surfaces are shown in Fig. 3. Higher sintering temperature of 60 °C promotes greatly the development of the grain size and the densification of the BST thick film. However, the edges of the BST film in contact with the alumina substrate in Fig. 2(b) show much brighter surface, indicating an interfacial reaction between the BST film and the alumina substrate at the elevated sintering temperature. For test samples sintered ≥ 1280 °C, the device properties could not be measured, possibly indicating the loss of the lower Pt electrodes due to the interactions between the alumina substrate, BST and Pt.

Another example shown in Fig. 3 (c) is the BST55/45 film with 10 wt.% glass frit sintered at 900 °C. Although the microstructure still shows porosity and non-uniformity, lower porosity was obtained compared to BST70/30 thick film in Fig. 3(a), indicating the effectiveness of glass frit as a sintering aid. In fact, the sintering temperature has been effectively

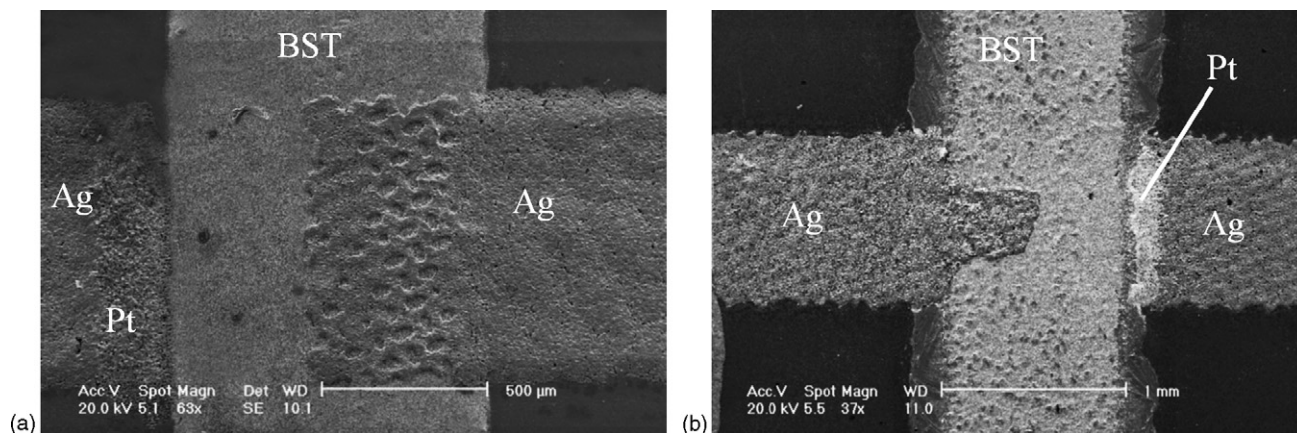


Fig. 2. SEM photos of the BST70/30 thick films capacitor structures. (a) Sintered at 1200 °C; (b) sintered at 1260 °C.

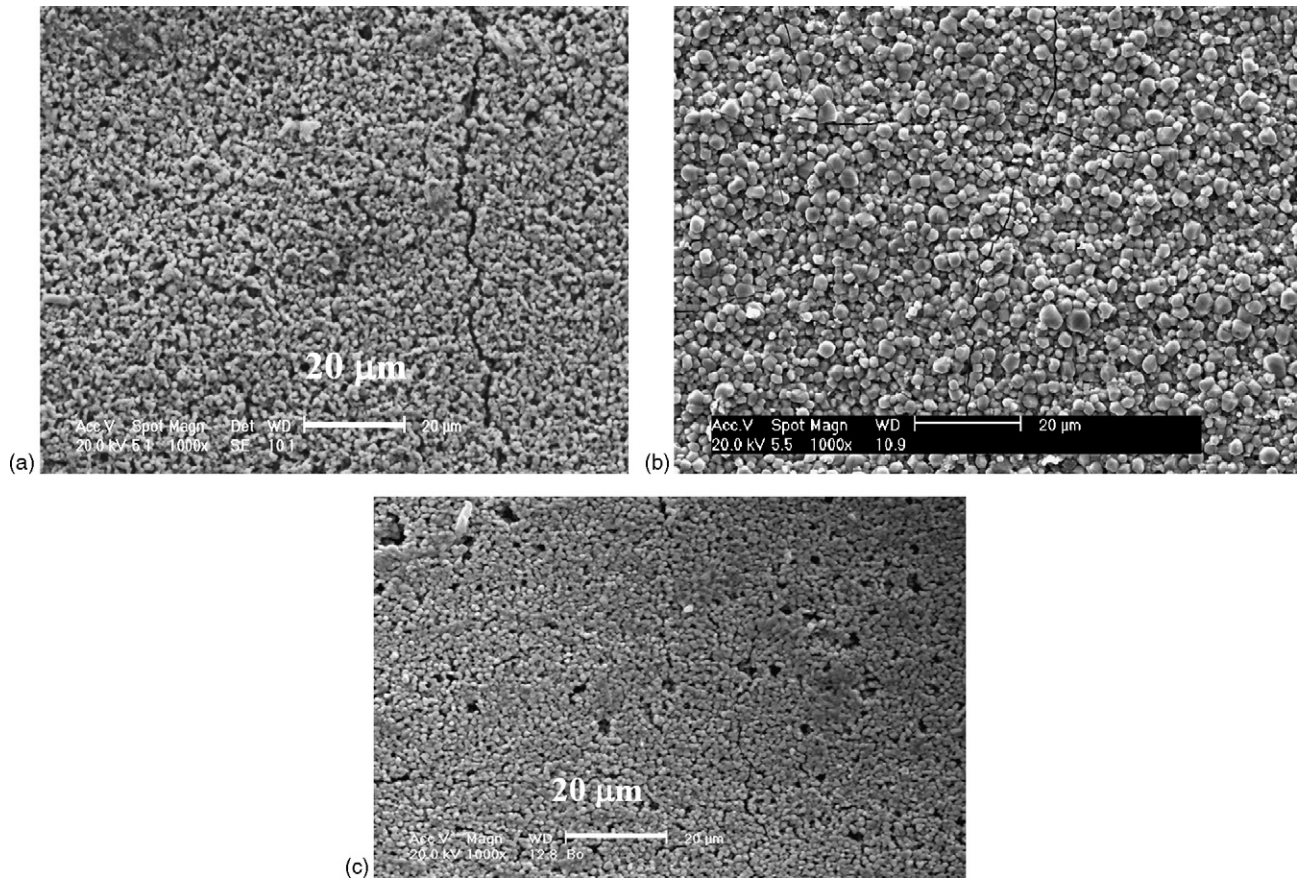


Fig. 3. SEM photos of the microstructures of BST thick films capacitor structures. (a) BST70/30 sintered at 1200 °C; (b) BST70/30 sintered at 1260 °C; (c) BST55/45 + 10 wt.% glass frit sintered at 900 °C.

reduced by 300 °C by introducing the glass frits as the sintering aid.

The dielectric properties of BST thick film test structures are shown in Fig. 4. BST70/30 film sintered at 1260 °C shows much higher tunability than the film sintered at 1200 °C. BST55/45 film with 10 wt.% glass frit exhibits a lower tunability, compared to BST70/30 film sintered at 1260 °C. Moreover, one factor that has to be taken into account is the film thickness. While the thickness of BST70/30 films reduces with the increase of sintering temperature, the bias field applied is constant at 40 V, effectively

increasing the applied electric field and the measured tunability.

Table 1 summarizes the microwave properties of the BST film capacitor structures. For BST70/30 films sintered at the temperature ≤ 1260 °C, the performance improves with the increase of sintering temperature to a figure of merit (FOM) of 35.6°/dB for the film sintered at 1260 °C. For BST55/45 film with 10 wt.% glass frit, a similar FOM value of 35.9°/dB was measured, but with a decrease of sintering temperature of 360 °C, from 1260 to 900 °C.

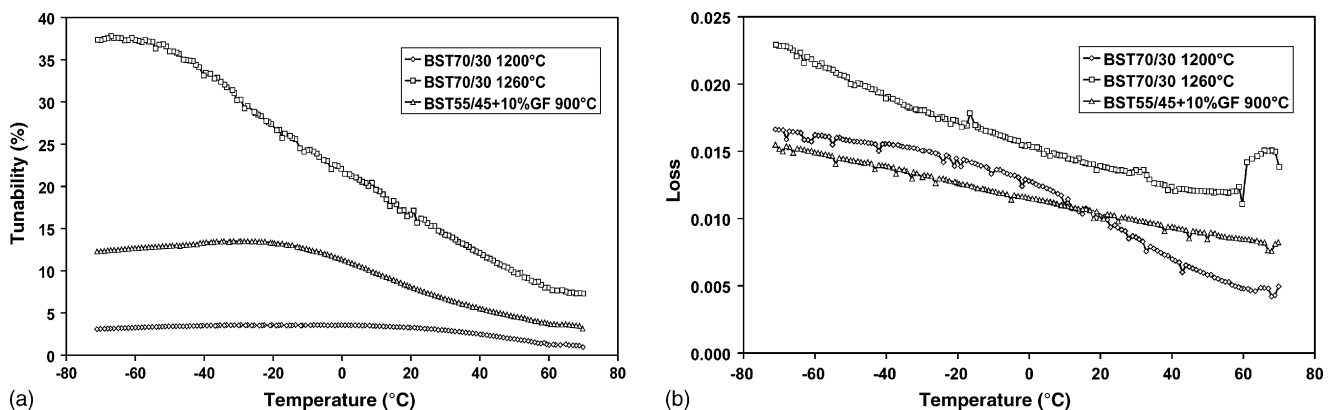


Fig. 4. Dielectric properties of BST thick films with the same capacitor test structure measured at 10 MHz and under 40 V bias.

Table 1
Microwave properties of BST films

Sintering temperature (°C)	Insertion loss (dB)	Phase shift (°) (at 100 V)	F_0 (GHz)	FOM (°/dB)
BST70/30				
1200	−4.30	38.5	2.5	11.2
1220	−2.40	80.0	1.6	26.6
1260	−1.38	65.5	1.5	35.6
BST55/45 + 10%GF				
900	−1.90	62.0	2.2	35.9

All results based on same test structure. Figure of merit (FOM) has been normalized at frequency of 2 GHz.

Obviously, the sintering temperature plays a very important role in optimizing the device performance. On the one hand, elevated sintering temperature can promote the densification of the film and its adhesion on the substrate. On the other hand, too high a sintering temperature will result in strong interfacial reaction which can degrade the properties of the devices. Therefore, care has to be taken in finding an optimum sintering temperature to satisfy both these criteria. These preliminary results show that BST films containing glass frits have a similar performance compared to conventional BST samples, but the sintering temperatures can be reduced by about 360 °C. This enlarges the processing window greatly and also provides the possibility of integrating with low temperature co-fired ceramic (LTCC) systems which are also processed around 900 °C.¹¹

Fig. 5 shows the photograph of a reflection-type phase shifter (RTPS). Capacitor test structures were incorporated into the RTPS, as indicated by the circles. For this device, BST 70/30 was utilized and sintered at 1250 °C. Silver thick film was employed as the main transmission line. The two lower electrodes of silver lines (shown in the left part of the device) were grounded by the silver paste prior to the microwave tests. With 100 V dc bias, the phase shifter circuit shows a 49° phase shift and insertion loss about −2.4 dB at 2.5 GHz, as shown in Fig. 6, resulting in a FOM of ~20°/dB. Compared to the FOM values of the capacitor test structures in Table 1, the decrease of FOM of the whole RTPS circuit is probably a result of additional conductor and reflection losses.

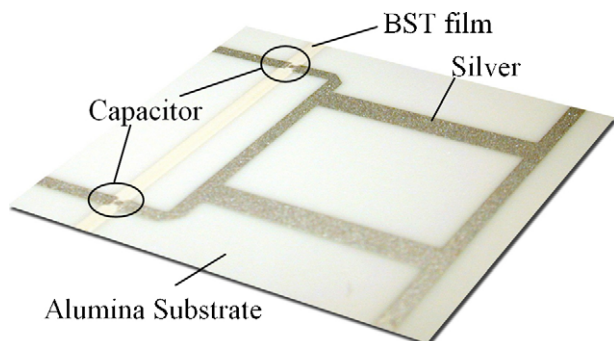


Fig. 5. Optical photograph of a thick film phase shifter on an alumina substrate.

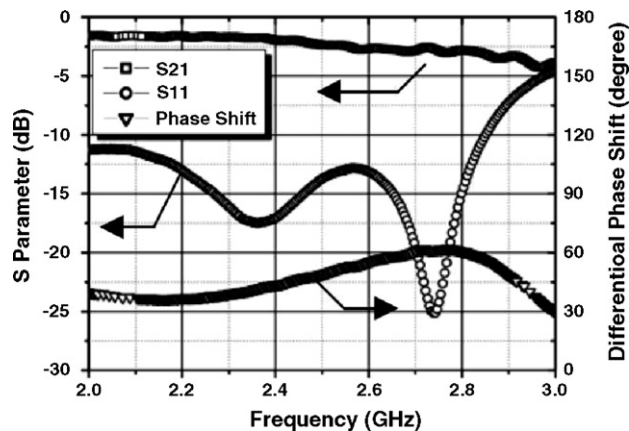


Fig. 6. Microwave measurement result of the phase shifter under 100 V bias.

4. Conclusion

A composition of $\text{Ba}_{0.70}\text{Sr}_{0.30}\text{TiO}_3$ was chosen for the fabrication of BST thick film capacitor test structures due to its high tunability at room temperature. The tunability and figure of merit at 2–3 GHz were found to be strongly dependent on the sintering temperature of the BST films. Capacitor test structure with BST70/30 films sintered at 1260 °C show the best performance with FOM = 36°/dB, however, slight increase of sintering temperature above 1260 °C leads to the failure of the device. A glass frit was shown to be useful for BST55/45 composition and films of this composition sintered at 900 °C produced similar performance to BST70/30 sintered at 1260 °C. A reflection-type phase shifter based on the capacitor test structure with BST70/30 film sintered at 1250 °C showed a FOM of ~20°/dB.

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References

- Davis, J. L. and Rubin, L. G., Some dielectric properties of barium–strontium titanate ceramics at 3000 megacycles. *J. Appl. Phys.*, 1953, **24**, 1194–1197.
- Galt, D., Price, J. C., Beall, J. A. and Ono, R. H., Characterization of a tunable thin-film microwave $\text{YBa}_2\text{Cu}_3\text{O}_{7-x}/\text{SrTiO}_3$ coplanar capacitor. *Appl. Phys. Lett.*, 1993, **63**, 3078–3080.
- Outzourhit, A., Trefny, J. U., Kito, T., Yasar, B., Nazirpour, A. and Hermann, A. M., Fabrication and characterization of $\text{Ba}_{1-x}\text{Sr}_x\text{TiO}_3$ tunable thin-film capacitors. *Thin Solid Films*, 1995, **259**, 218–224.
- Ott, R. and Wordenweber, R., Improved designs of tunable ferroelectric capacities for microwave applications. *Appl. Phys. Lett.*, 2002, **80**, 2150–2152.
- Cole, M. W., Nothwang, W. D., Hubbard, C., Ngo, E. and Ervin, M., Low dielectric loss and enhanced tunability of $\text{Ba}_{0.6}\text{Sr}_{0.4}\text{TiO}_3$ based thin films via material compositional design and optimized film processing methods. *J. Appl. Phys.*, 2003, **93**, 9218–9225.
- Moon, S. E., Kim, E. K., Kwak, M. H., Ryu, H. C., Kim, Y. T., Kang, K. Y. et al., Orientation dependent microwave dielectric properties of fer-

- roelectric $\text{Ba}_{1-x}\text{Sr}_x\text{TiO}_3$ thin films. *Appl. Phys. Lett.*, 2003, **83**, 2166–2168.
7. Ngo, E., Joshi, P. C., Cole, M. W. and Hubbard, C. W., Electrophoretic deposition of pure and MgO-modified $\text{Ba}_{0.6}\text{Sr}_{0.4}\text{TiO}_3$ thick films for tunable microwave devices. *Appl. Phys. Lett.*, 2001, **79**, 248–250.
 8. Su, B., Holmes, J. E., Meggs, C. and Button, T. W., Dielectric and microwave properties of barium strontium titanate (BST) thick films on alumina substrates. *J. Eur. Ceram. Soc.*, 2003, **23**, 2699–2703.
 9. Zimmermann, F., Voigts, M., Menesklou, W. and Ivers-Tiffée, E., $\text{Ba}_{0.6}\text{Sr}_{0.4}\text{TiO}_3$ and $\text{BaZr}_{0.3}\text{Ti}_{0.7}\text{O}_3$ thick films as tunable microwave dielectrics. *J. Eur. Ceram. Soc.*, 2004, **24**, 1729–1733.
 10. Su, B. and Button, T. W., Interactions between barium strontium titanate (BST) thick films and alumina substrates. *J. Eur. Ceram. Soc.*, 2001, **21**, 2777–2781.
 11. Gongora-Rubio, M. R., Espinoza-Vallejos, P., Sola-Laguna, L. and Santiago-Aviles, J. J., Overview of low temperature co-fired ceramics tape technology for meso-system technology (MsST). *Sens. Actuator A: Phys.*, 2001, **89**, 222–241.