

Investigation of barium strontium titanate thick films for tunable phase shifters

F. Zimmermann^{a,*}, M. Voigts^a, C. Weil^b, R. Jakoby^b, P. Wang^c,
W. Menesklou^a, E. Ivers-Tiffée^a

^a*Institut für Werkstoffe der Elektrotechnik (IWE), Universität Karlsruhe (TH), Kaiserstraße 12, D-76128 Karlsruhe, Germany*

^b*Institut für Hochfrequenztechnik, Universität Darmstadt (TU), Merckstraße 25, 64283 Darmstadt, Germany*

^c*DaimlerChrysler AG, Forschung und Technologie, Ulm, Germany*

Received 4 September 2000; received in revised form 2 November 2000; accepted 5 December 2000

Abstract

The influence of the microstructure of $\text{Ba}_{0.6}\text{Sr}_{0.4}\text{TiO}_3$ (BST) bulk ceramics and thick films on the dielectric properties have been studied. Thick films have been prepared by screen printing technique on Al_2O_3 substrates. The powder has been prepared by using the common mixed oxide technique. In comparison to dense bulk ceramics, the permittivity of thick films is approximately 10 times less. The effect of temperature on the permittivity and the tunability (change of the dielectric constant with applied voltage) has also been investigated at low frequency (1 kHz). At microwave frequencies, BST thick films have been characterized by measuring coplanar waveguides (CPW) at room temperature and utilizing an enhanced quasi-static CPW model for multilayer dielectric substrates. © 2001 Elsevier Science Ltd. All rights reserved.

Keywords: BaTiO_3 and titanates; Dielectric properties; Grain size; Phase shifter; Porosity

1. Introduction

Phased array antennas with steerable electronic beam will gain increasing importance in future applications in radar, satellite and communication systems.¹ The use of ferroelectric ceramics, like barium strontium titanate (BST), for phase shifters could be a cheap alternative to the commonly used components in such applications.^{2,3} For an application in a beam steering device, the required properties for the ferroelectric material are: low dielectric constants $\epsilon_r \leq 50$, low losses ($\tan \delta$), high tunability ($\Delta \epsilon_r / \epsilon_r$) and a low temperature dependence.

It has been shown that the permittivity of barium titanate can be reduced by increasing the porosity⁴ or by admixing different materials⁵ and that the temperature dependence of the permittivity can be decreased by reducing the grain size.^{6,7}

This paper describes the investigation of $\text{Ba}_{0.6}\text{Sr}_{0.4}\text{TiO}_3$ bulk ceramics with different porosity and grain sizes, as well as of $\text{Ba}_{0.6}\text{Sr}_{0.4}\text{TiO}_3$ thick films with different porosity printed on alumina (99%) substrate. The dielectric properties (ϵ_r , $\tan \delta$) of thick films have been measured at low frequency (1 kHz) as a function of temperature ($70 < T/K < 320$), applied electrical field and compared to those of bulk ceramics. Furthermore, results are presented for a ferroelectric transmission line phase shifter in coplanar technology at 24 GHz, suitable for an integration in planar phased array antennas.

2. Material preparation and experimental methods

Disk-shaped dense and porous $\text{Ba}_{0.6}\text{Sr}_{0.4}\text{TiO}_3$ bulk ceramics were prepared by the mixed oxide method. BaCO_3 , SrCO_3 and TiO_2 was weighed out in stoichiometric proportions, calcined at 1050°C, milled to a median grain size of $d_{50} < 0.5 \mu\text{m}$. The porous samples were hand-pressed and sintered at 1280°C. Dense samples were cold isostatically pressed at 200 MPa before being sintered at 1300°C. The ceramics were then cut to disks

* Corresponding author. Tel.: +49-721-608-7769; fax: +49-721-608-7492.

E-mail address: frederic.zimmerma@etec.uni-karlsruhe.de (F. Zimmermann).

of 8 mm diameter and of 1.4 mm thickness. Thin gold electrodes were sputtered on both faces of the disk to build a simple capacitor so that the permittivity and the losses could easily be calculated from the complex capacity.

In order to build thick film capacitors, a 3 μm platinum bottom electrode was screen printed on an Al_2O_3 -substrate and fired at 1340°C. Then a layer of BST was printed on the top of the platinum electrode and sintered at 1250°C. The average diameter of the grains and the layer thickness after sintering was determined by scanning electron microscopy. For the top electrode, a gold layer of 1.5 μm thickness was printed on the BST and fired at 900°C. A summary of the examined BST-ceramics is given in Table 1.

A cryostat was used to cover the temperature range from 70 to 340 K. The complex permittivity was measured by a HP4274A LCR-meter (discrete frequencies from 100 Hz to 100 kHz) which was separated from the applied high voltage up to 1 kV by low loss capacitors.

Tunable microwave transmission line phase shifters were realized on BST thick films. The uniplanar concept of the coplanar waveguide (CPW) was chosen here, since the electrical field of the CPW is mainly concentrated between the line slots, i.e. in the ferroelectric film. The dielectric constant of the BST film can be controlled by simply applying a dc voltage V_{dc} between the inner conductor and the ground metallizations of the CPW, perpendicular to the direction of propagation of the microwave signal.

Coplanar waveguides have been fabricated on $\text{Ba}_{0.6}\text{Sr}_{0.4}\text{TiO}_3$ thick films of approximately 5 μm thickness on top of 0.635 mm thick alumina substrates using standard photolithography and thin film technology. A gold metallization has been electroplated up to a thickness of $t = 3 \mu\text{m}$. The strip width w and the slot width s of the CPW were chosen to be $w = 18 \mu\text{m}$ and $s = 16 \mu\text{m}$, respectively. The scattering parameters of CPW's on various BST thick films were measured at room temperature up to 26.5 GHz using a HP 8510B network analyser and an on-wafer probe station. Since maximum internal dc voltage of the network analyser was limited to only $V_{\text{dc}} = 40 \text{ V}$, a maximum electric field strength of $E_{\text{max}} = 2.5 \text{ kV/mm}$ was achieved in the narrow CPW slots.

The differential phase shift, $\Delta\Phi$, of the ferroelectric CPW-phase shifter at a certain frequency, f_0 , can be defined as:

$$\Delta\Phi = \Phi_{\text{max}} - \Phi_{\text{min}}$$

$$= \frac{2\pi}{c_0} f_0 \left(\sqrt{\varepsilon_{\text{r,eff}}(0)} - \sqrt{\varepsilon_{\text{r,eff}}(E_{\text{max}})} \right) L, \quad (1)$$

where $\varepsilon_{\text{r,eff}}(0)$ and $\varepsilon_{\text{r,eff}}(E_{\text{max}})$ denote the effective dielectric constants for zero and maximum dc bias, respectively.

Table 1
 $\text{Ba}_{0.6}\text{Sr}_{0.4}\text{TiO}_3$ ceramics examined at low frequencies (1 kHz)

	System	Porosity (%)	Grain size (μm)	Thickness
BST60	Bulk	< 1	≤ 25	1.4 mm
RB20	Bulk	30–40	0.7	1.4 mm
S3	Thick film	30–40	0.7	7 μm
S2	Thick film	> S3	0.7	8.5 μm

Furthermore, c_0 is the velocity of light in free space and L is the physical length of the transmission line.

According to Eq. (1), the measurement of the phase shift, Φ , as a function of frequency makes it possible to derive the effective dielectric constant, $\varepsilon_{\text{r,eff}}$, of the thick film-BST CPW and its tunability. A quasi-static CPW model for multilayer dielectric substrates was concatenated and extended, taking into account finite metallization thickness, dispersion as well as ohmic conductor losses and dielectric losses, in order to estimate the relative permittivity, $\varepsilon_{\text{r,BST}}$, the loss tangent, $\tan\delta_{\text{BST}}$, and the tunability of the BST thick film.⁸ This simple and fast analytical CPW model has shown quite reasonable agreement with a numerical full wave analysis.

The phase shifter device can be characterized by its differential phase shift, $\Delta\Phi$, according to Eq. (1) and the insertion loss. Excluding the return loss, the intrinsic loss of a phase shifter is

$$\alpha = -10 \log(|S_{11}|^2 + |S_{21}|^2) (\text{dB}), \quad (2)$$

where S_{11} and S_{21} are the measured reflection and transmission coefficients, respectively. Since a high phase shift and low losses are aimed for a phase shifter, the figure-of-merit (FoM) of a phase shifter can be defined as the quotient of the differential phase shift, $\Delta\Phi$, and the insertion loss, α :

$$FoM = \frac{\Delta\Phi}{\alpha} (\text{deg/dB}). \quad (3)$$

3. Results and discussion

Fig. 1 shows the temperature dependence of the permittivity and the loss tangent of the dense and the porous BST bulk ceramics. The dense bulk ceramic clearly shows the three well-known⁹ phase transitions from cubic to tetragonal (275 K), tetragonal to orthorhombic (220 K) and orthorhombic to rhombohedral (175 K), and a strong increase of losses with decreasing temperature. In contrast, the permittivity curve of the porous bulk ceramic shows only one broad peak and a strongly reduced permittivity compared to the dense ceramic. The sharp peak at the Curie point disappears, and the losses below 280 K show an almost linear behavior and are smaller than those of the dense ceramic.

The porosity seems to be the main reason for the reduction of the permittivity,⁴ and the small grain size causes the Curie-point to be smeared out⁷

Fig. 2 compares the dielectric properties of the porous ceramic with the properties of the thick film. The thick film (S3) which has a similar microstructure as the porous ceramic (RB20) shows significant smaller permittivity which further decreases by increasing porosity. The temperature dependence decreases also with the increase of the porosity, and the more porous sample (S2) has the lowest temperature dependence and the smallest permittivity of all the shown samples. The maximum of the temperature curve moves to lower temperatures. This can result from a shift of the lower two transition points to higher temperature with a concurrent decrease of the Curie temperature T_c .¹⁰

The more porous thick film S2 has the lowest losses ($\tan\delta_{S2} = 0.007$) of all the three samples at room temperature at the frequency $f = 1$ kHz.

The influence of microstructural changes in BST ceramic on the tunability is of particular interest. In Fig. 3, the tunability t_u

$$t_u = \frac{\Delta\epsilon_r}{\epsilon_r(0)} = \frac{\epsilon_r(0) - \epsilon_r(E_{\max})}{\epsilon_r(0)} \quad (4)$$

is shown for all the four examined samples. In this formula, $\epsilon_r(0)$ is the permittivity of the ceramic without applied external field, and $\epsilon_r(E_{\max})$ is the permittivity of the ceramic with the maximum applied field. Since the thick films have a thickness of $\approx 8 \mu\text{m}$ in comparison to ≈ 1 mm of the bulks, much higher fields can be applied with the same voltage so that tunability of thick films and bulks in Fig. 3 should not be compared directly. In the ferroelectric region, the tunability of the porous ceramic is much lower than the tunability of the dense ceramic. Nevertheless, at room temperature the porous ceramic shows a tunability of 8% which is almost half of the tunability of the dense bulk. The thick films, however, are nearly temperature-independent and show a very high tunability of 70–80% at the maximum and about 45–70% at room temperature with an applied electrical field close to breakdown.

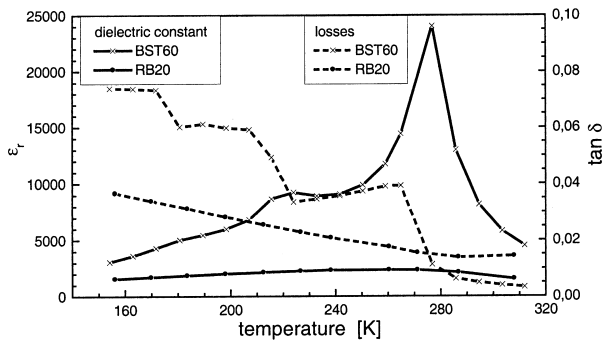


Fig. 1. Dielectric constant, ϵ_r , and losses, $\tan\delta$, versus temperature of dense (BST60) and porous (RB20) $\text{Ba}_{0.6}\text{Sr}_{0.4}\text{TiO}_3$ bulk ceramic measured at 1 kHz.

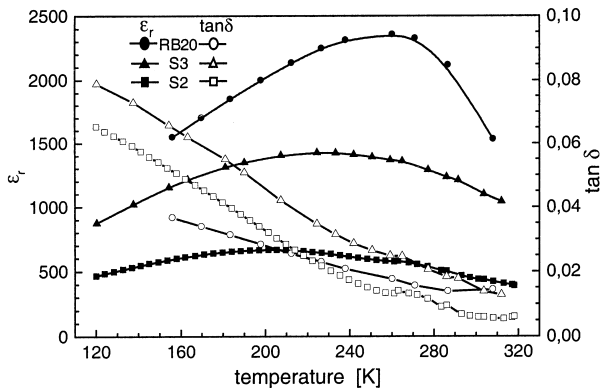


Fig. 2. Dielectric constant and loss tangent versus temperature of $\text{Ba}_{0.6}\text{Sr}_{0.4}\text{TiO}_3$ porous bulk ceramic (RB20), a microstructural comparable thick film (S3), and a thick film with increased porosity (S2) measured at 1 kHz.

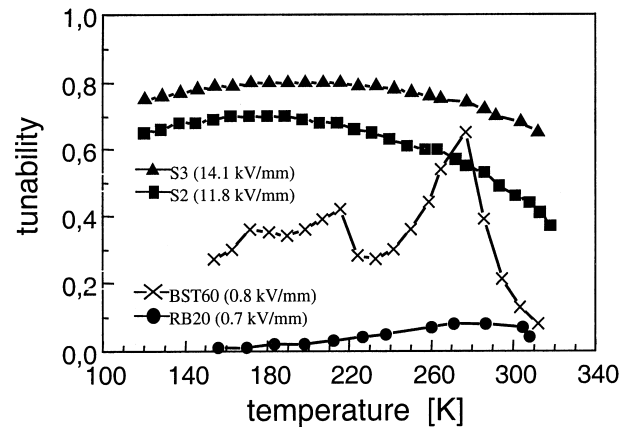


Fig. 3. Tunability versus temperature of a dense (BST60) and a porous (RB20) $\text{Ba}_{0.6}\text{Sr}_{0.4}\text{TiO}_3$ bulk ceramic and the thick films S2 and S3 at 1 kHz.

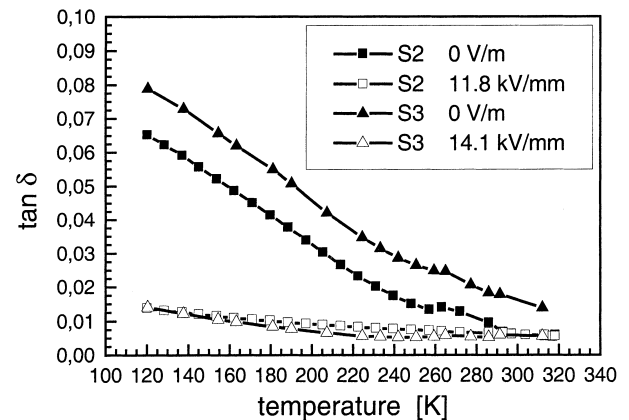


Fig. 4. Loss tangent versus temperature of $\text{Ba}_{0.6}\text{Sr}_{0.4}\text{TiO}_3$ thick films S2 and S3 as a function of the applied electrical field (1 kHz).

Table 2

Dielectric material properties at K-band frequencies and room temperature and performances of a CPW-phase shifter at 24 GHz for a $\text{Ba}_{0.6}\text{Sr}_{0.4}\text{TiO}_3$ thick film ceramic

Material concept	BST material data at K-band			CPW phase shifter performance @ 24 GHz		
	$\varepsilon_{r,\text{bst}}$ at 0 kV/mm	$\tan\delta_{\text{BST}}$	Tunability (%) at 2.5 kV/mm	$\Delta\Phi$ (deg/mm) at 2.5 kV/mm	α (dB/mm)	FoM (deg/dB) at 2.5 kV/mm
$\text{Ba}_{0.6}\text{Sr}_{0.4}\text{TiO}_3$ thick film	321	0.090	9.8	10.26	2.12	4.84

Fig. 4 shows the $\tan\delta$ of the thick films S2 and S3 as a function of temperature with and without an electrical field. At zero field, both thick films have a strong temperature dependence of $\tan\delta$ which disappears with an applied electrical field. A possible explanation for this circumstance might be a fixing of the polarization due to the strong electric field.

Table 2 shows the model-aided estimations of the dielectric properties of a $5\text{ }\mu\text{m}$ $\text{Ba}_{0.6}\text{Sr}_{0.4}\text{TiO}_3$ thick film. The results were gained from CPW measurements at K-band frequencies and at room temperature. In addition, the measured performance of a CPW phase shifter at 24 GHz is given for this BST-material concept. In comparison with interdigital capacitor measurements at 1 MHz, only a slight degradation in $\varepsilon_{r,\text{BST}}$ and tunability of the BST thick film is observed at K-band frequencies. A tunability of nearly 10% was achieved based upon an applied electrical field strength of 2.5 kV/mm. However, the BST loss tangent has increased crucially by a factor of about 10.⁸ These high losses are assumed to be due to BST-film porosity, imperfections, surface stress, and interdiffusions at the boundary layer. Due to the dominating losses and the relatively low electrical field strength, the phase shifter FoM was only about 5 deg/dB.

For microwave-phase shifter applications, where high tunability and low losses, i.e. a high figure-of-merit according to Eq. (3), are desirable, one has to find the BST-material composition, offering the best trade-off of both parameters.

4. Conclusions

Dense and porous bulk ceramics with the composition $\text{Ba}_{0.6}\text{Sr}_{0.4}\text{TiO}_3$ were prepared to show the influence of porosity and grain size on the dielectric behavior. The desired effect of reduction of permittivity and of temperature dependence by means of increasing the porosity has been achieved.

$\text{Ba}_{0.6}\text{Sr}_{0.4}\text{TiO}_3$ thick films prepared by the screen printing method have similar dielectric properties, low temperature dependence and low permittivity. The tunability of the more porous thick film S2 at room temperature is $t_{u,\text{S2}} \approx 45\%$ and the losses $\tan\delta_{\text{S2}} \approx 0.007$ with an applied electric field of $E_{\text{max},\text{S2}} = 11.8\text{ kV/mm}$ at 1

kHz. Since the losses of the thick films with an applied bias field are almost independent of the temperature and the porosity, an application with the operating point near breakdown would be quite conceivable. Room temperature measurements of a thick film CPW phase shifters at K-band frequencies indicated that the tunability values are transferable from the kHz to the GHz region, but the dielectric loss tangent of BST increases by an order of magnitude. A material optimization regarding losses and applying higher electrical field strengths should allow a distinctly improved figure-of-merit of the phase shifter device at room temperature.

Acknowledgements

The authors thank Silvia Schöllhammer for her help in preparing the bulk ceramics and the thick films and Jin Xu for support with the measurement. The authors also thank Professor Karl Heinz Hårdtl for helpful discussions. This work was supported by the BMBF, NMT No. 03 N 10574.

References

1. Varadan, V. K. and Varadan, V. V., Smart electronics and sensors for IVHS and automobile collision warning antenna Systems. In *Proceedings of SPIE*, No. 2448, 1995, pp. 35–40.
2. Babbitt, R., Kosica, T., Drach, W. and Didomenico, Ferroelectric phase shifters and their performance in microwave phased array antennas. *Integrated Ferroelectrics*, 1995, **8**, 65–76.
3. De Flaviis, F. and Alexopoulos, N. G. *et al.*, Planar microwave integrated phase-shifter design with high purity ferroelectric material. *IEEE Transactions on Instrumentation and Measurement*, 1997, **45**, 963–969.
4. Delfrate, M. A., Leoni, M., Nanni, L., Melioli, E., Watts, B. E. and Leccabue, F., Electrical characterization of BaTiO_3 made by hydrothermal methods. *Journal of Materials Science — Materials in Electronics*, 1994, **5**, 153–156.
5. Sengupta, L. C., Stowell, S., Ngo, E., O'Day, M. E. and Lancto, R., Barium strontium titanate and non-ferroelectric oxide ceramic composites for use in phased array antennas. *Integrated Ferroelectrics*, 1995, **8**, 77–88.
6. Bell, A. J. and Moulson, A. J., The effect of grain size on the dielectric properties of barium titanate ceramic. *British Ceramic Proceedings*, 1985, **36**, 57–66.
7. Arlt, G., Hennings, D. and de With, D., Dielectric properties of fine-grained barium titanate ceramics. *Journal of Applied Physics*, 1985, **58**(4), 371–373.

8. Weil, C., Wang, P., Downar, P., Wenger, J. and Jakoby, R., Ferroelectric thick film ceramics for tuneable microwave coplanar phase shifters. *Frequenz*, 11–12/2000, **54**, 250–256.
9. Landolt-Börnstein, Numerical data and functional relationships in science and technology. In *New Series, Group III: Crystal and Solid State Physics*. Springer Verlag, Berlin/Heidelberg, 1981, p. 416.
10. Kinoshita, K. and Yamaji, A., Grain-size effects on dielectric properties in barium titanate ceramics. *Journal of Applied Physics*, 1976, **47**(1), 371–373.