Available online at www.sciencedirect.com

SCIENCE DIRECT

Journal of the European Ceramic Society 26 (2006) 1873–1877

www.elsevier.com/locate/jeurceramsoc

Thickness effects on microwave properties of $(Ba,Sr)TiO_3$ films for frequency agile technologies

K. Kageyama *, A. Sakurai, A. Ando, Y. Sakabe

Murata Manufacturing Co., Ltd., Nagaokakyo-shi 617-8555, Japan

Available online 1 December 2005

Abstract

We fabricated $(Ba_{0.6}Sr_{0.4})TiO_3$ (BST) thin films of various thicknesses on sapphire (−1 1 2 0) substrates using metal-organic decomposition method. These films showed grain growth from 160 to 650 nm with an increase in the thickness from 90 to 1050 nm. At microwave frequencies, the measured capacitances of the planar capacitors decreased with the film thickness because the electro-magnetic field propagates across high permittivity BST films to the low permittivity sapphire substrate. However, we found that the BST-thin film permittivity remained large up to 90 nm thick, based on electro-magnetic field analysis using the finite element method. On the other hand, the BST thin film tunability decreased with the film thickness.

Keywords: Precursors-organic; Dielectric properties; Perovskite

1. Introduction

Frequency agile technologies are promising for next-generation mobile telecommunication systems. There is a lot of research on tunable devices, such as antennas, filters, and phase shifters. Many materials are available that can achieve frequency tunability, such as PIN/Schottky diodes, GaAs/FET-s, GaAs/varactors, MEMS, and ferroelectrics. We can select the most suitable material for potential applications using frequency agile technology.

Ferroelectric materials have a large dielectric loss, due to the domain wall movement, and they show a large dielectric dispersion at microwave frequencies. Above Curie temperatures (T_c), ferroelectric material enters a para-electric state, and the ensuing dielectric loss drastically decreases while the materials retain a large permittivity. These materials are thus very effective for use in microwave-frequency tunable devices.

Of all the ferroelectric materials $(Ba,Sr)TiO_3$ (BST) is considered the most promising for microwave frequency agile applications.[1] For BST systems, T_c can be easily adjusted by substituting barium ions for the strontium ions. The resulting BST has a large permittivity and low loss ($\tan\delta < 0.01$) at microwave frequencies. The effect of thickness on dielectric properties has been reported for these systems.[2] A lot of research has focused on this phenomenological field. The dielectric constant of BST decreases as it is thinned, and the dielectric tunability decreases with the thickness. These effects are caused by the interlayer between the electrodes and BST. This is true for metal–insulator–metal/parallel-plate capacitor structures.[3]

On the other hand, such thickness effects are not clear in planar capacitor structures,[3] although the formation of interdigitated electrode-array (IDEA)/planar-type structures is common for actual microwave devices. Moreover, planar-type devices have a productivity advantage because of their simple structure. They are more reliable and offer a choice of electrode metals compared to MIM/parallel-plate capacitor structures, in which the electrode metal on the bottom sometimes degrades reliability and quality factors. For these reasons, we must obtain more information through experiments on the thickness dependence of planar-type components.[3] We fabricated BST films of various thicknesses on sapphire substrates and used a planar/IDEA structure to investigate the thickness dependence of the microwave-tunable properties.

2. Experimental

As shown in Fig. 1, BST thin films were fabricated on sapphire (−1 1 2 0) using metal-organic decomposition (MOD). Reagent-grade strontium, barium and titanium alcohol oxides were mixed to form the precursor solution. We chose $(Ba_{0.6}Sr_{0.4})TiO_3$ for our investigation. We dropped a fixed amount of the solution on the sapphire substrate (0.366-mm

* Corresponding author.

0955-2219/$ – see front matter
doi:10.1016/j.jeurceramsoc.2005.09.090

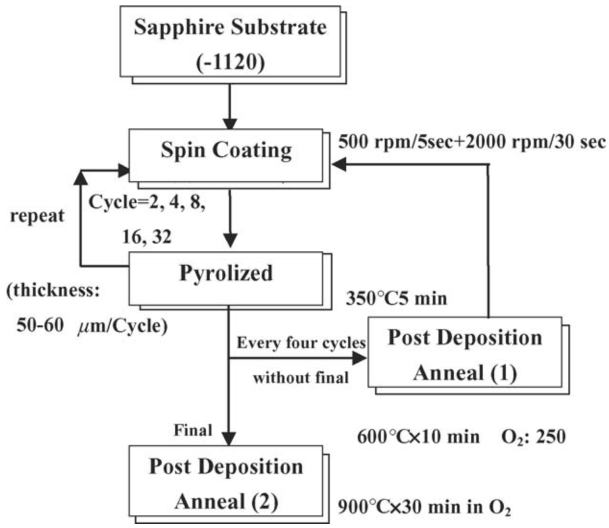

Fig. 1. Fabrication of (Ba,Sr)TiO_3 thin films using metal-organic decomposition.

thick) and coated it, using a spin coater with preset times for each sample. After they were coated with the MOD solution, the samples were annealed at 900 °C for 1 h.

The microstructures were examined using field-emission scanning electron microscopy (FESEM) and atomic force microscopy (AFM). The crystalline phases were identified using X-ray diffraction analysis (XRD). To measure the microwave dielectric properties, we patterned interdigitated electrode arrays (Fig. 2) on top of the BST films using a photolithographic lift-off process. Two-micron thick Cu-electrodes formed using e-beam evaporation.

We measured the microwave dielectric properties and tunability using a vector network analyzer (HP8510C) and applying a DC bias voltage thru bias-T, at a frequency below 10 GHz. We measured permittivity using the conformal matching method,[4,5] assisted by electro-magnetic field analysis using the finite element method (FEM). Permittivity measurements at microwave frequencies were confirmed using the perturbation method.[6] The

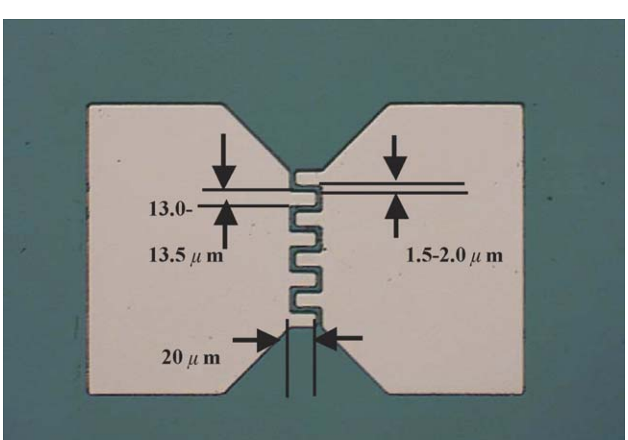

Fig. 2. Interdigitated electrode array.

Fig. 3. Cross-sectional FESEM photograph of $(Ba_{0.6}Sr_{0.4})TiO_3$ thin film.

tunability of each sample was calculated using:

$$\text{Tunability}\,(\%) = \frac{C_0 - C_{\text{bia}}}{C_0 \times 100} \tag{1}$$

where C_0 and C_{bia} are the capacitances with no bias and applied bias voltage, respectively.

3. Results and discussion

Fig. 3 shows an FESEM photograph of a cross-section of the BST film. Surface images of the films analyzed using AFM are shown in Fig. 4. The BST deposited on the sapphire/R-plane (−1 1 0 2) had a dense film microstructure. In the AFM images, we observed an accelerated grain growth from 160 to 650 nm, with increases in the film thickness from 90 to 1050 nm. The grain size compared to the film thickness is shown in Fig. 5. When the thickness was below 300 nm, grain growth was constrained. This could have been caused by the interaction with the substrate. The results of the XRD analysis are illustrated in Fig. 6. They reveal a random growth orientation.

Fig. 7 shows the results of the FEM simulation for the interdigitated-electrode array structure shown in Fig. 2. We clearly observed a decrease in the capacitance when the BST film was thinned. As shown in Fig. 7(a), the electro-magnetic field between the interdigitated electrode arrays propagated across the BST thin films with a high dielectric constant (>300) to the sapphire substrate with the low dielectric constant (<10) and became strong enough to propagate to the sapphire substrate, as the film thickness decreased. Consequently, we observed a decrease in the capacitance with the film thickness, when the dielectric constant of the BST thin film was held constant as the thickness decreased. In general, planar-capacitor permittivity is the effective permittivity because the actual permittivity is smaller than that of capacitor dielectrics. Because the electro-magnetic field propagates to the substrate, and to the air, it decreases the actual planar-capacitor permittivity. The results shown in Fig. 7 demonstrate that the effective permittivity of the BST-deposited substrate decreases with the film thickness.

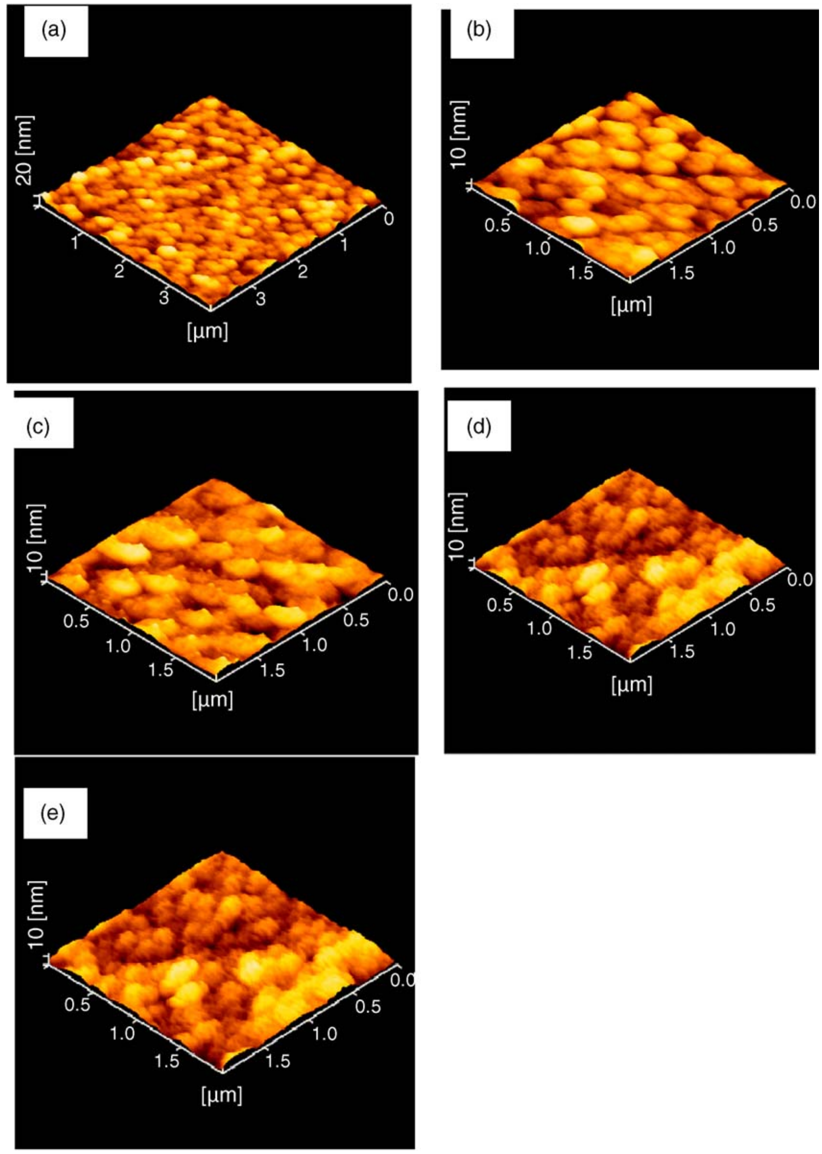

Fig. 4. Surface images of BST thin films analyzed using AFM: (a) 90 nm; (b) 150 nm; (c) 240 nm; (d) 510 nm; (e) 1050 nm.

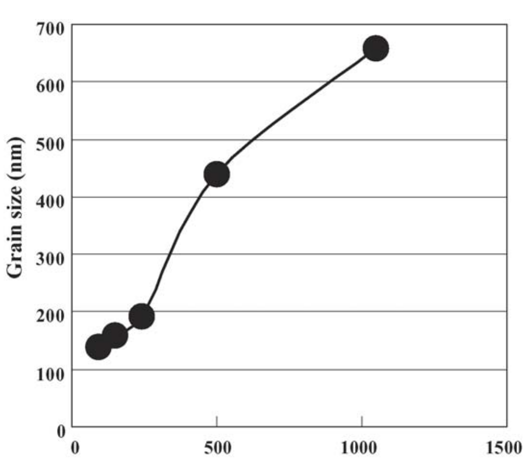

Fig. 5. Grain growth of BST thin film.

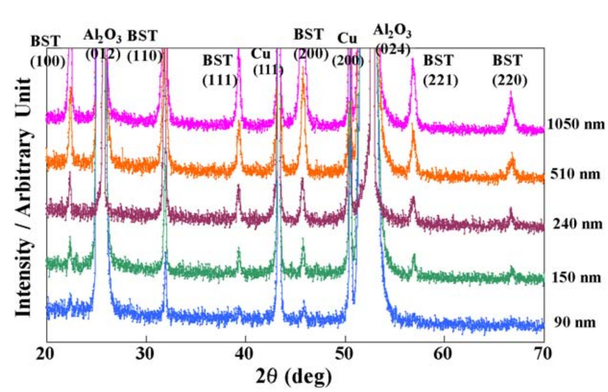

Fig. 6. XRD patterns of BST thin films.

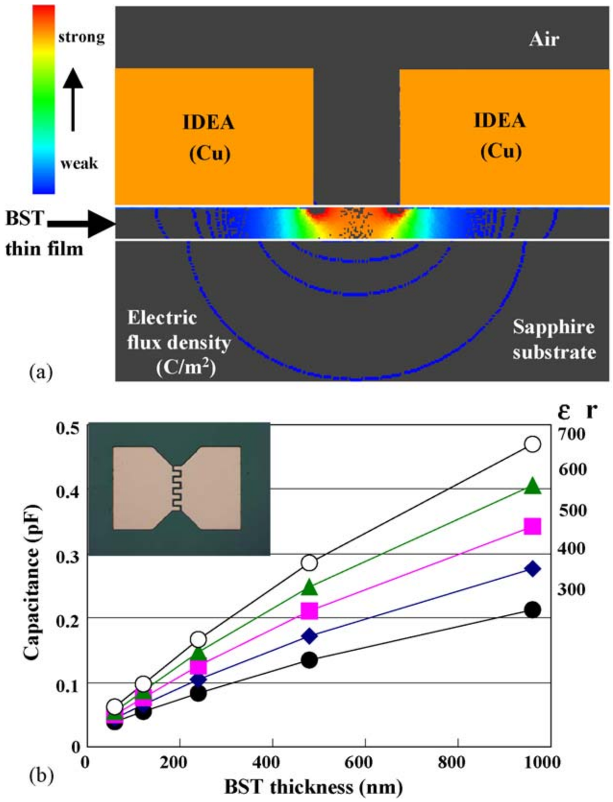

Fig. 7. Results of FEM simulations for capacitances of interdigitated electrode array compared to various $(Ba_{0.6}Sr_{0.4})TiO_3$ film thickness. (a) Results of electro-magnetic field analysis. (b) Summary of simulated capacitances compared to BST film thickness for various film permittivities.

The measured dielectric constants for various film thicknesses are summarized in Fig. 8. They remained large, even when the thickness decreased to below 200 nm. The measured dielectric tunabilities obtained under various film thickness conditions are summarized in Fig. 9. We observed that the tunability decreased with the thickness. Higher permittivity causes higher tunability.[3] Bellotti et al.[7] measured a significant decrease in the tunability as the film thickness dropped below 180 nm, even in planar structures. However, the thickness dependence of permittivity was not presented in their work. We still do not know whether the thickness dependence of permittivity in planar capacitors is the same as in parallel-plate capacitors.

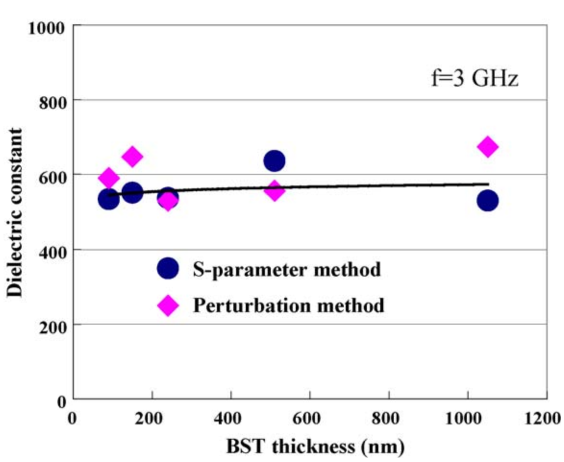

Fig. 8. Thickness dependence of dielectric constant for BST thin films.

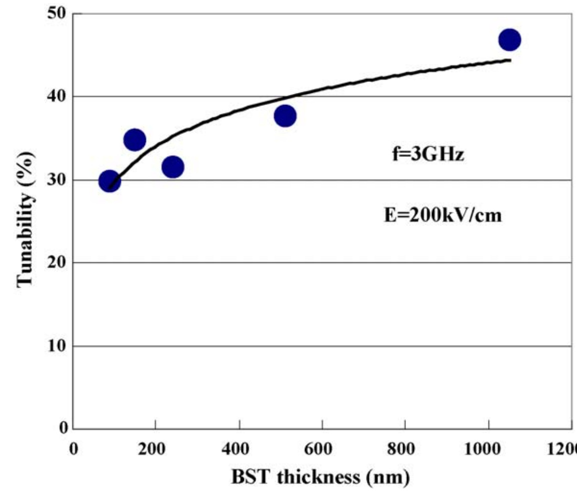

Fig. 9. Thickness dependence of dielectric tunability for BST thin films.

In this work, we found that permittivity remains large, even at thicknesses below 200 nm. On the other hand, tunability decreased with the thickness in the planar structures. Therefore, in planar capacitors, permittivity does not significantly depend on the BST film thickness, whereas tunability does. The dielectric tunability of BST films in planar capacitors increased with the capacitance or effective permittivity. This indicates that the decrease in the tunability of planar capacitors (caused by decreasing the BST thickness), is accompanied by a decrease in the effective permittivity of the BST deposited substrate.

4. Summary

We fabricated $(Ba_{0.6}Sr_{0.4})TiO_3$ films of various thicknesses using the metal-organic decomposition. These films have a dense, randomly oriented, polycrystalline microstructure. They showed a grain growth from 160 to 650 nm, and a corresponding film thickness increase from 60 to 960 nm. The measured capacitances for the interdigitated electrode-array structures at microwave frequencies decreased with the film thickness because the electro-magnetic field propagated across the film to the sapphire substrate. This caused a decrease in the measured capacitance. However, permittivity remained large, even when the thickness decreased to below 200 nm. Tunability decreased with the thickness. This indicates that the decrease in planar-capacitor tunability caused by thinning the film is accompanied by a decrease in the effective permittivity of the BST-deposited substrate, while maintaining a large permittivity. Therefore, planar-capacitors have markedly different behaviors in their thickness dependence on the dielectric properties from those of parallel-plate capacitors.

References

1. Streiffer, S., An introduction to ferroelectrics for tunable microwave devices. In *IEEE MTT-S 2003 Microwave Symposium Workshop Notes, Ferroelectric Materials and Devices for Microwave Applications*, 2003.

2. Streiffer, S., Basceri, C., Parker, C. B., Lash, S. E. and Kingon, A. I., Ferroelectricity in thin films: The dielectric response of fiber-textured $(Ba_xSr_{1-x})Ti_{1+y}O_{3+z}$ thin films grown by chemical vapor deposition. *J. Appl. Phys.*, 1999, **8**, 4565–4575.
3. Tagantsev, A. K., Sherman, V. O., Astafiev, K. F., Venkatesh, J. and Setter, N., Ferroelectric materials for microwave tunable applications. *J. Electroceramics*, 2003, **11**, 5–66.
4. Bedair, S. S. and Wolff, I., Fast, accurate, and simple approximate analytic formulas for calculating the parameters of supported coplanar waveguides for (M)MIC's. *IEEE Trans. Microwave Theory Technol.*, 1992, **MTT-40**, 41–48.
5. Gillick, M., Robertson, I. D. and Joshi, J. S., An analytical method for direct calculation of E & H-field patterns of conductor-backed coplanar waveguides. *IEEE Trans. Microwave Theory Technol.*, 1993, **MTT-43**, 1606–1610.
6. Nishikawa, T., Wakino, K., Tanaka, H. and Ishikawa, Y., Precise measurment method for complex permittivity of microwave dielectric substrate. *CPEM '88 Digest*, 1988, 155–156.
7. Bellotti, J., Akdogan, E. K., Safari, A., Chang, W. and Kirchoefer, S., Tunable dielectric properties of BST thin films for RF/MW passive components. *Integr. Ferroelectr.*, 2002, **49**, 113.