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Ceramics International 41 (2015) S567–S571

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# Composite thin films consisting of fine-grained barium strontium titanate for tunable microwave devices

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Received 26 October 2014; accepted 19 March 2015 Available online 2 April 2015

#### Abstract

It has been reported that grain size has significant influence on the dielectric properties of barium strontium titanate (BST) thin films. In this paper we have proposed a multilayer structure in which BST layers are separated by thin MgO layers which are expected to function as barrier to atomic diffusion so that the grain size of BST could be confined. By simulation we have found that for a device with a coplanar/composite film/ substrate configuration, the introduction of MgO can effectively change the dielectric constant of the composite while the tunability is not significantly affected, provided the amount of MgO is smaller than 25%. A prototype device was prepared to demonstrate the feasibility of this idea. By X-ray diffraction measurements it is revealed that BST and MgO coexist in the composite. The dielectric property of the composite film was determined and it was found that the film has a tunability of up to  $\sim 7\%$  at 500 MHz and under an electric field of 13.3 kV/mm. © 2015 Elsevier Ltd and Techna Group S.r.l. All rights reserved.

Keywords: Barium strontium titanate; Composite thin films; Tunable; Coplanar capacitor

### 1. Introduction

Thin films of barium strontium titanate  $(Ba_{1-x}Sr_xTiO_3)$  have been widely studied for applications in phase shifters, flexoelectric devices, tunable filter, voltage controlled oscillators, delay lines, and so on [1–3]. One of the major problems of this material is that the dielectric loss is relatively large at frequency of GHz. In literature, several ways have been developed aiming to lowering the loss. For example, BST has been combined with low loss oxides (typically MgO or MgTiO<sub>3</sub>) to form a 0–3 type ceramic composite (where BST is a continuous phase with MgO particles randomly distributed in it). The introduction of a second phase leads to a suppression of the dielectric loss at a cost of significant reduction of dielectric constant and tunability

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http://dx.doi.org/10.1016/j.ceramint.2015.03.263

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[4–7]. Recently it has also been reported that, by controlling the grain size of BST, it is possible to make thin films with both a large dielectric tunability and small loss tangent. As reported by Riehl et al [8], BST prepared by a sophisticatedly modified PLD process could have a very small average grain size of  $\sim 20$  nm in diameter, which is much smaller than that of BST prepared by a conventional pulsed laser deposition (PLD) process (typically  $\sim 50$  nm in diameter). Microwave measurements on such small-grained films indicated that a good compromise between dielectric tunability and loss tangent was achieved. The problem is that modification to the PLD system is needed and the processing window is rather narrow.

In light of the technical problems, we propose to develop a composite thin film structure that consists of several BST and MgO layers which are alternatively laminated and grown on conventional single crystal substrates like  $(LaAlO_3)_{0.3}(Sr_2Al-TaO_6)_{0.7}$  (LSAT). What makes our work different from the literature is that the purpose of introducing MgO in our samples

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is mainly for constraining the growth of BST and therefore: (1) the amount of MgO needed to meet this purpose is significantly smaller than that in conventional BST–MgO composites and (2) sophisticated processing technique is not necessary and conventional PLD (or sputtering) can be employed for making the composites. Because of the fine grain size of BST and small concentration of MgO, the composites are expected to exhibit a property of large dielectric tunability and small loss tangent.

In this paper, we report the simulation work that was conducted aiming to establish a relationship between the sample structure and performance as well as preliminary experimental results on sample fabrication and characterization.

#### 2. Simulation and experiment work

Fig. 1 shows the structure of the composite thin films to be developed, which is rather complicated. One possible way to establish the relationship between the device capacitance and structure is by conducting simulation using commercial software. In this work, however, we managed to simply the calculation by: (i) counting only the top two layers (i.e., 1 layer of BST and 1 layer of MgO) and neglecting the contribution of capacitance from the other layers and the substrate, (ii) adopting the so-called partial-capacitance model to divide a multilayer capacitor into several single-layer capacitors, and (iii) estimating the capacitance of a single-layer coplanar capacitor by adopting the capacitance formula for parallel plate capacitors (this is feasible because the dielectric constant of both BST and MgO are much larger than that of the air). The simplification can certainly lead to much lowered accuracy; nevertheless, the calculation results can still provide useful information that can be of help for the design and fabrication of such devices, as to be demonstrated later in the paper.

To verify the feasibility of the above idea, we have conducted experiments to grow BST/MgO composite thin films on LSAT (0 0 1) single crystal substrates by pulsed laser ablation (PLD) using a krypton fluoride (KrF) excimer laser (Lambda PhysikCOMPex 205) with a wavelength of 248 nm,

energy of 250 mJ and a repetition rate of 10 Hz. Ceramic targets composed of Ba<sub>0.6</sub>Sr<sub>0.4</sub>TiO<sub>3</sub> and MgO respectively were prepared via conventional ceramic processing. The distance between the target and substrate was fixed at 5 cm. The substrate temperature was maintained at 650 °C during the deposition. The oxygen partial pressure was kept at 27 Pa during the laser ablation process. The structure of the thin films was examined by X-ray diffractometer (XRD) (Bruker D8 Discover), scanning electron microscope (SEM) and atomic force microscopy (AFM). The dielectric parameters of the thin films were characterized along an in-plane direction on interdigital electrode capacitors with a configuration of electrode/BST+MgO/LSAT. The top interdigital electrodes were prepared by the deposition of a 200-nm-thick Au layer on the film surface followed by patterning using standard photolithography and wet chemical etching techniques [8]. The IDE has a total of 21 fingers with the finger length of 925 µm and finger width of 5  $\mu$ m. The finger gap spacing is 3  $\mu$ m. The dielectric tunability is defined as the following equation: Tunability=  $(C_0 - C_E)/C_0 \times 100\%$ , where  $C_0$  and  $C_E$  denote the capacitance without and with dc bias field, respectively. The dielectric properties and tunable properties were measured using HP 4291B RF impedance/material analyzer.

## 3. Results and discussion

The influence of the amount of MgO on the overall properties of the composite was studied by simulation. As shown in Fig. 1(b), the total capacitance equals to the sum of the capacitances of two capacitors—the MgO-based capacitor and the "virtual" capacitor. The thickness of the MgO-based capacitor equals to  $(t_{BST} + t_{MgO})$ and the thickness of the virtual dielectric layer equals to  $t_{BST}$ , according to the model. As the dielectric constant of BST and MgO are both much larger than the dielectric constant of air (=1), it is reasonable to assume that the capacitance of each interdigital capacitor is proportional to the thickness of the dielectric film and inversely proportional to the distance between two top electrodes. With this estimation, one can easily find out that the apparent



Fig. 1. (a) Schematic structure of BST–MgO composite thin films; (b) Partial capacitance model. The second capacitor has a virtual dielectric layer of which the dielectric constant equals to  $(\varepsilon_{BST}-\varepsilon_{MgO})$ .

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