

Structure and Dielectric Properties of Co-Sputtered (Ba_{0.5}Sr_{0.5})TiO₃ Thin FilmsChen Xiaofeng¹, Zhu Weiguang¹, and Lim Su Yin²¹ Microelectronics Centre, School of Electrical and Electronic Engineering, Nanyang Technological University, Nanyang Avenue, Singapore 639798

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We demonstrate the structural evolution and dielectric properties of co-sputtered (Ba_{0.5}Sr_{0.5})TiO₃ thin films deposited at two sputtering pressures, 3.2×10⁻³ and 8.3×10⁻³ mbar, respectively. It is revealed that the sputtering pressure impacts on the growth of BST thin films significantly. Low sputtering pressure would lead to the off-stoichiometric composition and poor crystallinity in the sputtered BST thin films. In particular, the presence of TiO₂ rutile structure could be observed at the pressure of 3.2×10⁻³ mbar. The off-stoichiometric effect could be slightly eliminated in the co-sputtered BST thin films by increasing sputtering pressure. The structural evolution and dielectric properties of these sputtered BST thin films are discussed by correlating with the sputtering pressure and substrate temperature. The film with optimized figure-of-merit (FOM) achieved in this work shows low zero-bias permittivity of 80.5, low tangent loss of 0.007 and FOM of 44.1, which is potential for frequency-agile microwave device applications.

Key words: sputtering, (Ba,Sr)TiO₃ thin film, tunability, microwave application

1. INTRODUCTION

Owing to the electric-field-dependent non-linear dielectric property close to the Curie temperature T_C , ferroelectric materials offer an enticing prospect for introducing tuning into microwave and millimeter circuits, which cannot be achieved with existing ferrite and semiconductor tuning technologies. Examples of such passive frequency-agile components are dielectric resonators, phase shifters, phased array antennas, transmission lines, bandpass filters, and coplanar waveguides. Since the Curie temperature T_C of (Ba,Sr)TiO₃ (BST) solid solution could be adjusted to room temperature by tailoring the Ba-to-Sr ratio, BST-based ceramic thin films are considered as the most promising candidate material for room temperature frequency-agile microwave device applications. BST films have been deposited by chemical vapor deposition (CVD) [1], pulsed laser deposition (PLD) [2], sputtering [3], and sol-gel process [4]. Conventional parallel plate RF sputtering of BST thin films is very susceptible to back sputtering the growing film by negative oxygen ions. It may lead to off-stoichiometric films having inferior dielectric properties. These adverse effects can be minimized by using composition compensated target [5], high sputtering gas pressure [3, 6], or unconventional off-axis growth geometry [7, 8]. In this work, we reported results on the structural evolution and the dielectric properties of co-sputtered BST thin films deposited in two sputtering pressures, 3.2×10⁻³ and 8.3×10⁻³ mbar, and correlated to the influence of sputtering pressure and deposition temperature.

2. EXPERIMENTAL

BST50/50 thin films were fabricated on Pt(150 nm)/TiW(30 nm)/SiO₂(500 nm) silicon wafers using Leybold UNIVEX450B confocal multi-target RF magnetron sputtering system. The deposition was

carried out by co-sputtering from 3-inch stoichiometric BaTiO₃ and SrTiO₃ ceramic targets in two Ar-20%O₂ gas mixtures with a sputtering pressure of 3.2×10⁻³ and 8.3×10⁻³ mbar respectively. The composition of the films was controlled via varying the RF powers applied to BaTiO₃ and SrTiO₃ targets, according to the pre-calibrated sputtering rates. The films were grown at a substrate temperature range between 500°C and 620°C. Typical deposition parameters are summarized in Table I.

TABLE I Deposition Parameters for BST Thin Films.

	Condition 1	Condition 2
Target size	3 inch	
Target	BaTiO ₃ and SrTiO ₃	
Base pressure (mbar)	5×10 ⁻⁷	
Gas flow (sccm)	16 for Ar, 4 for O ₂	64 for Ar, 16 for O ₂
Sputtering pressure (mbar)	3.2×10 ⁻³	8.3×10 ⁻³
RF power (W)	BaTiO ₃ 60, SrTiO ₃ 220	BaTiO ₃ 76, SrTiO ₃ 220
Substrate temperature (°C)	500 - 620	
Thickness (nm)	200-300	

The crystal structure of the films was investigated by a Siemens D5005 X-ray diffractometer with grazing angle thin film attachment. Cu K_α was used as the radiation source. For dielectric characterization, 180-nm-thick Pt top electrodes with a dimension of 0.3×0.3 mm² were deposited on BST films by RF sputtering using a lift-off technique. Dielectric measurements of Pt/BST/Pt capacitors were performed

on a Mitutoyo vibration-proof microprobe station using HP 4155B precision semiconductor parameter analyzer and HP 4284 LCR meter.

3. RESULTS

3.1 Structural evolution

Figure 1 illustrates the X-ray diffraction spectra of the BST films deposited at a low sputtering pressure of 3.2×10^{-3} mbar with the substrate temperature range from 500 to 620°C. It is seen that the films deposited at 500°C exhibits a complicated diffraction spectrum with the existence of TiO_2 rutile (110) at 27.5°. It is believed that such complicated XRD pattern might originate in the off-stoichiometric composition of the BST thin film with excess Ti while deposited at a low pressure [3]. With temperature increasing, the excess Ti could be dissolved into the perovskite structure. As a result, the pure perovskite structure with a broaden peak at 31.5°, corresponding to BST (110), could be observed in a temperature range between 540°C and 580°C. It is shown clearly that these BST films exhibit a poor crystallizability with nano-sized crystallite due to their off-stoichiometric composition. At 620°C, the TiO_2 rutile structure comes again, probably related to the bottom electrode stability during the high-temperature deposition process [9]. It has been known that the out-diffusion of Ti at high process temperature may result in the degradation of bottom electrode.

In contrast, the films deposited at a higher sputtering pressure of 8.3×10^{-3} mbar show typical polycrystalline perovskite diffraction spectra as illustrated in Fig. 2. These films exhibit an improved crystallizability owing to their minimized off-stoichiometric composition. The film grown at 500°C exhibits a broaden peak at 31.5°, corresponding to smaller grain size. With the increase of deposition temperature, the peak is sharpened and intensity is increased, indicating the improved crystallinity in the BST films. However, further increasing deposition temperature would lead to poor adhesion of BST due to the degradation of bottom electrode.

3.2 Leakage current

Figure 3 illustrates the leakage current of co-sputtered BST thin films deposited at 3.2×10^{-3} mbar. The leakage current of Pt/BST/Pt thin film capacitors is generally dominated by Schottky-barrier emission, which is sensitive to interface trap states. As expected, the deposition temperature affects the leakage current of BST films significantly. The film deposited at 500°C shows an electrical leaky behavior, exhibiting high leakage current and low breakdown voltage. It is consistent with our XRD finding, of which the underdeveloped microstructure with the presence of TiO_2 rutile structure is shown correspondingly. With the increase of deposition temperature, the leakage current is lowered and the breakdown voltage is increased. However, the breakdown voltage tremendously drops down while grown at 620°C, probably originated in the degradation of bottom electrode. Similarly, the leakage current of BST thin films grown at 8.3×10^{-3} mbar are also demonstrated in Fig. 4. Generally, these films have a higher leakage current and lower breakdown voltage than those grown at 3.2×10^{-3} mbar correspondingly. It

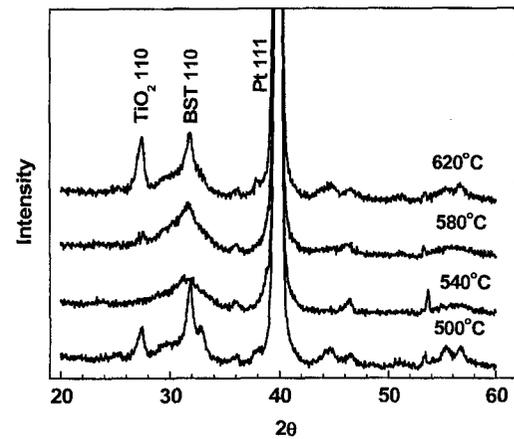


Fig. 1 Dependence of deposition temperature on the X-ray diffraction spectra for the BST thin films sputtered at 3.2×10^{-3} mbar.

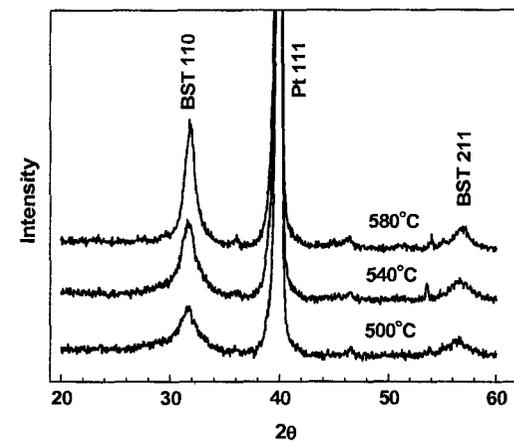


Fig. 2 Dependence of deposition temperature on the X-ray diffraction spectra for the BST thin films sputtered at 8.3×10^{-3} mbar.

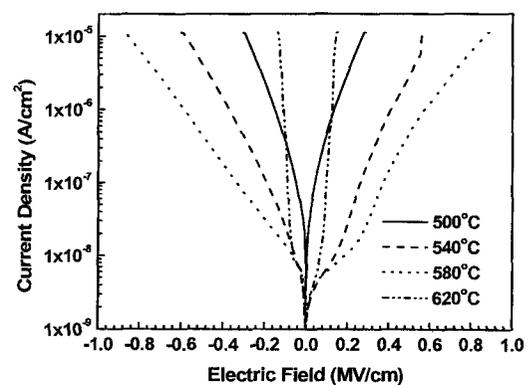


Fig. 3 Leakage currents of the BST thin films sputtered at different temperatures in an Ar-20% O_2 ambient of 3.2×10^{-3} mbar.

has been understood that the off-stoichiometric composition of the sputtered BST thin films is the probable reason [3].

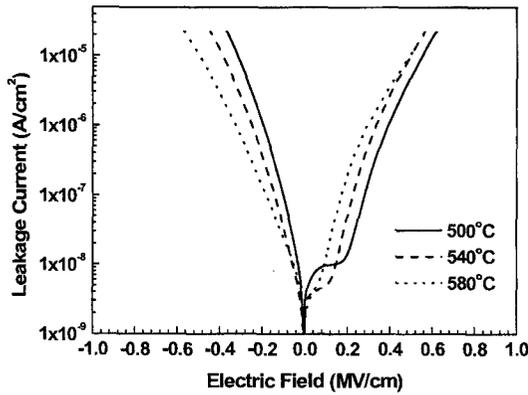


Fig. 4 Leakage currents of the BST thin films sputtered at different temperatures in an Ar-20%O₂ ambient of 8.3×10^{-3} mbar.

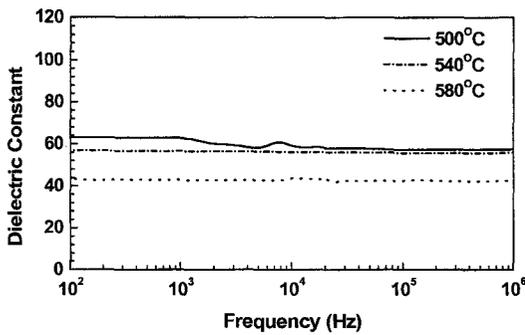


Fig. 5 Frequency dependence of the BST thin films sputtered at 3.2×10^{-3} mbar, from 500 to 580°C.

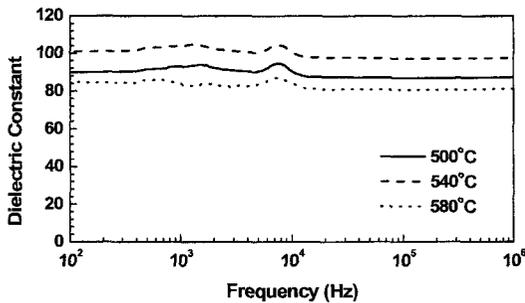


Fig. 6 Frequency dependence of the BST thin films sputtered at 8.3×10^{-3} mbar, from 500 to 580°C.

3.3 Dielectric properties

Frequency dependence of the co-sputtered BST films was investigated at room temperature with an oscillating level of 100 mV, from 100 Hz to 1 MHz. Figure 5 illustrates the frequency dependence of permittivity of the BST films deposited at 3.2×10^{-3} mbar, from 500 to 580°C. It is revealed that there is no remarkable dispersion measured in the testing frequency range. But their permittivity values are very low indeed, around 57.3, 55.6, and 42.8 at 100 kHz for the films deposited at 500, 540 and 580°C, respectively. Correspondingly, their tangent loss is 0.009, 0.010, and 0.011, respectively. The decrease of permittivity is probably related to their off-stoichiometric composition [1, 3]. Similarly, the frequency dependence of the films deposited at 8.3×10^{-3}

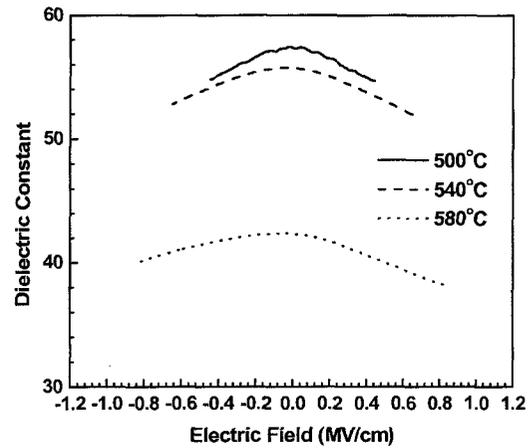


Fig. 7 Dielectric tunability of the BST thin films sputtered at 3.2×10^{-3} mbar, characterized at 100 kHz.

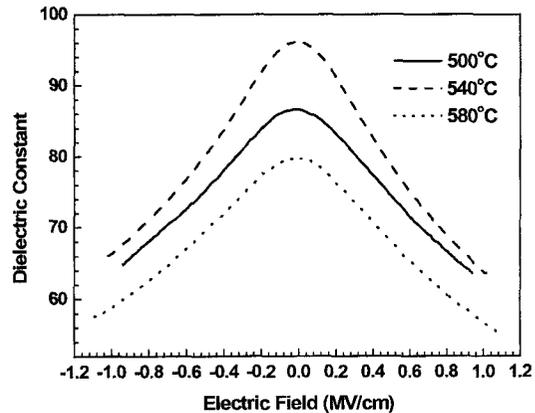


Fig. 8 Dielectric tunability of the BST thin films sputtered at 8.3×10^{-3} mbar, characterized at 100 kHz.

mbar is demonstrated in Fig. 6. Because of slightly eliminating the off-stoichiometric composition, these films exhibit the improved permittivity, of which the value of 86.4, 97.2 and 80.5 is measured at 100 kHz corresponding to the substrate temperature of 500, 540 and 580°C, respectively. The reduction of permittivity for the film deposited at 580°C is believed to relate to the degradation of bottom electrode. The tangent loss values are acceptable, around 0.009, 0.009, and 0.007, respectively. Also no remarkable dispersion is found in the testing frequency range.

The electric-field dependent dielectric properties of the co-sputtered BST thin films was investigated at room temperature with an oscillating source of 100 mV at 100 kHz and a maximum dc sweep range from -40 V to +40 V. Because of the low zero-bias permittivity, the BST films deposited at 3.2×10^{-3} mbar display a very poor tunability, as shown in Fig. 7. To evaluate the benefits of incorporating the BST films as tunable elements in microwave circuits, the figure-of-merit (FOM), defined by

$$\text{FOM} = \frac{\epsilon_{\max} - \epsilon_{\min}}{\epsilon_{\max}} \cdot \frac{1}{\tan \delta}$$

is usually used to give a correlation between the permittivity and tangent loss. The calculated value is 5.2, 6.7, and 8.7 for the film deposited at 500, 540 and 580°C, respectively. In contrast, the films deposited at 8.3×10^{-3} mbar exhibit the improved tunability because of slightly eliminating the off-stoichiometric composition in the films, as shown in Fig. 8. The FOM value is estimated to be 29.5, 37.6, and 44.1 for the film deposited at 500, 540 and 580°C, respectively.

4. CONCLUSIONS

We have compared the structural evolution and dielectric properties of co-sputtered $(\text{Ba}_{0.5}\text{Sr}_{0.5})\text{TiO}_3$ thin films deposited at 3.2×10^{-3} and 8.3×10^{-3} mbar respectively. The low sputtering pressure results in the off-stoichiometric composition in the BST thin films with the presence of TiO_2 rutile structure and poor crystallinity. Increasing sputtering pressure would slightly eliminate this off-stoichiometric effect, showing the improved crystallinity. Meanwhile, the microstructure evolution of the films may be promoted by increasing substrate temperature. However, the stability issues of Pt/TiW bottom electrode should be considered in the deposition process. The off-stoichiometric BST thin films exhibit poor dielectric properties with the low zero-bias permittivity value and the poor tunability. Whereas, increasing the sputtering pressure would result in the improved dielectric properties. The film with optimized FOM achieved in this work exhibits low zero-bias permittivity of 80.5, low tangent loss of 0.007 and FOM of 44.1.

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