Characterization of BST Thin Films using MgO(100) Buffer Layer for Tunable Device

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In this paper, we have investigated the structure and dielectric properties of the $(Ba_{0.6}Sr_{0.4})TiO_3$ (BST) thin films fabricated on MgO(100)/Si substrate by an alkoxide-based sol-gel method. Both the structure and morphology of those films were analyzed by x-ray diffraction (XRD) and atomic force microscope (AFM). For the MgO(100)/Si substrate, the BST thin films exhibited highly (100) orientation. The highly (100)-oriented BST thin films showed high dielectric constant, tunability, and figure of merit (FOM). The dielectric constant, dielectric loss and tunability of the BST thin films annealed at 700 °C deposited on the MgO(100)/Si substrate measured at 10 kHz were 515.9, 0.0082, and 54.3 %, respectively.

Keywords : BST, Sol-gel, Tunability, Dielectric properties, MgO

1. INTRODUCTION

$(Ba,Sr)TiO_3$ (BST) is a promising dielectric material for a wide variety of tunable microwave devices such as electrically tunable mixer, delay line, filter, capacitor, oscillator, resonator, and phase shifter[1,2]. However, for a successful application of BST in these fields, there is a set of properties to be satisfied: moderate-to-low dielectric constant at microwave frequencies, low dielectric loss factor, a large-scale variation of the dielectric constant by direct current (DC) biasing condition and low leakage current density[3]. It is well known that the dielectric properties of BST films depend on the dielectric material-electrode interface, Ba/Sr molar ratio, microstructure, the stress state of the film, surface morphology, dopant, and texture[4-6]. Although many researchers have studied the relations between electrical and structural properties for the dielectric thin films[7,8], the effects of interface or buffer layer between BST and substrate are not explored enough. At the same time, the problem of interface seems to be a key point to obtain optimum electrical characteristics and to adjust them through an obtaining of preferred orientation of BST thin films. The main purpose of our work was to investigate the role of MgO buffer layer aimed to offer the benefits of better-preferred orientation of BST thin films.

In this article, we report about highly (100)-oriented MgO thin films used as a buffer layer between BST and Si substrate as well as about dielectric BST thin films fabricated by sol-gel method. The effects of orientation on the crystallinity, microstructure and dielectric properties of BST thin films were investigated.

2. EXPERIMENTAL

The BST films were fabricated on the MgO/Si substrate. First, the MgO thin films were prepared by the MOD method. The precursor material was magnesium acetate tetrahydrate ($(CH_3CO_2)_2Mg{\cdot}4H_2O$). Acetic acid and de-ionized water were used as solvents. Initially, the magnesium acetate tetrahydrate was dissolved in acetic acid and water at room temperature and under a constant stirring for 12 h. Total concentration of the synthesized solution was 0.4 M. The final solution became transparent. The MgO solutions were spun to a Si (100) substrate using the spin-coating method[4000 rpm, 30 s], and then pre-baked on a hot plate at 350 °C for 10 min. The pre-baked films were annealed at various temperatures in the range 550–700 °C for 1 h in the

oxygen atmosphere to obtain crystallization. The total thickness of the MgO films was about 120 nm.

The precursor solutions for $Ba_{0.6}Sr_{0.4}TiO_3$ were prepared by the sol-gel method using barium acetate, strontium acetate, and Ti-isopropoxide as the starting materials. The solid-state barium acetate, strontium, and acetate were initially dissolved in acetic acid and then mixed to obtain a (Ba, Sr) stock solution. Titanium isopropoxide was dissolved in 2-methoxyethanol under N_2 atmosphere. Finally, both starting solutions were mixed to prepare the stoichometric, clear, transparent, and stable BST precursor. For the fabrication of BST thin films, the BST precursor solution was syringed through a 0.2 μm syringe filter on the MgO annealed at 650 °C. The films were deposited by the spin-coating technique at 4000 rpm for 30 s. After the spin-coating procedure the films were kept on hot plate at 400 °C for 10 min to remove the organic contamination. Then, the pre-baked films were annealed at 700 °C for 1 h in oxygen atmosphere for crystallization. The final thickness of BST thin films was about 200 nm. For the electrical measurements, Au interdigital electrodes (IDEs) were deposited on the BST films using E-beam evaporator and lift-off photolithography. The top electrodes were about 0.11 μm thick. The widths of finger lines and the gap between lines were 10-20 μm for the IDE capacitor.

The crystalline structures of the MgO and BST thin films were analyzed using X-ray diffraction (XRD) with a Rigaku-D/MAX diffractometer with CuK_α emission. Surface morphology of BST films was analyzed and quantified by a atomic force microscope (Digital Instrument NanoScope IIIa). Capacitance-voltage characteristics as well as dielectric constant and dielectric loss were measured using HP 4192 impedance analyzer at 20 mV oscillation level.

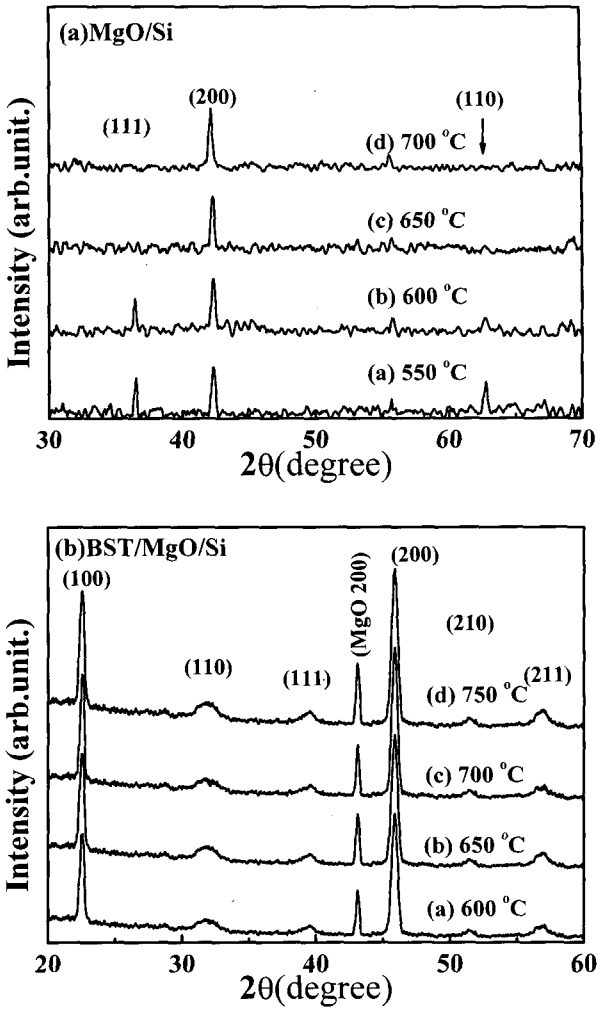

Fig. 1. X-ray diffraction patterns of (a) MgO thin films annealed at different temperatures and (b) BST/MgO/Si structure annealed at different temperatures.

3. RESULTS AND DISCUSSION

Figure 1(a) shows the XRD patterns of MgO thin films prepared under various conditions. As the annealing temperature increases to 550 °C, the MgO thin film begins to crystallize. The XRD pattern of the MgO film annealed at 550 °C shows the presence of such peaks as (111), (200) and (110). This fact indicates that the annealing temperature of 550 °C gives a poly-crystalline MgO film. However, when the annealing temperature increases above 550 °C, the ratio of the intensity of the (200) peak increases while the intensities for (111) and (110) peaks decrease. It is well known that the degree of preferred orientation may be estimated using the ratio of relative intensity as follows: $\alpha_{(hkl)} = I_{(200)}/(I_{(111)} + I_{(200)})$, where I is peak intensity[9]. Considering data of Fig. 1(a), corresponding α is 0.53, 0.62, 0.95 and 0.96 for the annealing temperatures of 550, 550, 600, and 650, respectively. This result suggests that the annealing temperature of 650 °C is enough to obtain (100)-oriented MgO film on Si substrate.

Figure 1(b) shows XRD patterns of the BST films deposited on MgO buffer layer annealed at 700 °C. In this figure, we can see that the BST thin films annealed above 600 °C show the perovskite structure and the absence of the pyrochlore phase. The α values derived from XRD are found to be above 0.95 for all the BST samples. Accordingly, we can conclude that the BST films deposited on MgO(100)/Si substrate are crystallized with (100) preferred orientation. Therefore our data confirms the fact that the crystallization and growth of the BST thin films are influenced by the substrate. The XRD data shows also that the increase in annealing temperature leads to the increase of the intensity

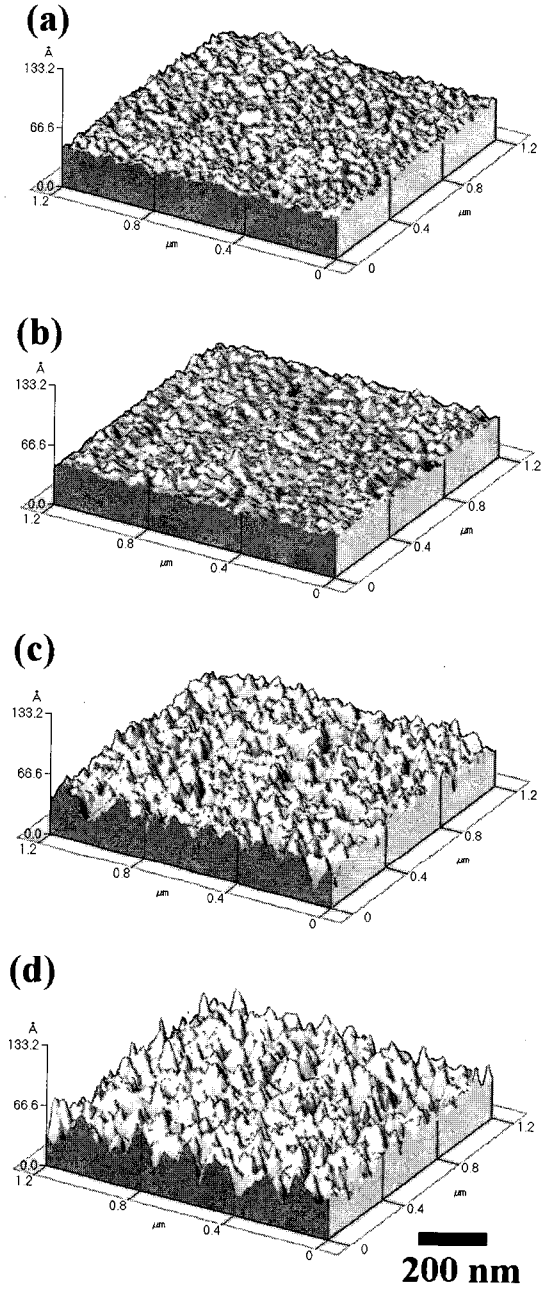

Fig. 2. Surface morphology of BST/MgO/Si structure annealed at (a) 600 °C, (b) 650 °C, (c) 700 °C, and (d) 750 °C using atomic force microscope.

of diffraction peak. However, the full widths at half maximum (FWHM) were found to be lowered. In our opinion, this effect may be explained by the increase of the grain size with the annealing temperature[10].

The average grain size and the surface roughness of the BST thin films were investigated using AFM. Figure 2 shows the surface morphology of the BST films on the MgO film. It can be seen that all AFM images show small surface roughness (1.96~3.5 nm), the grain size in

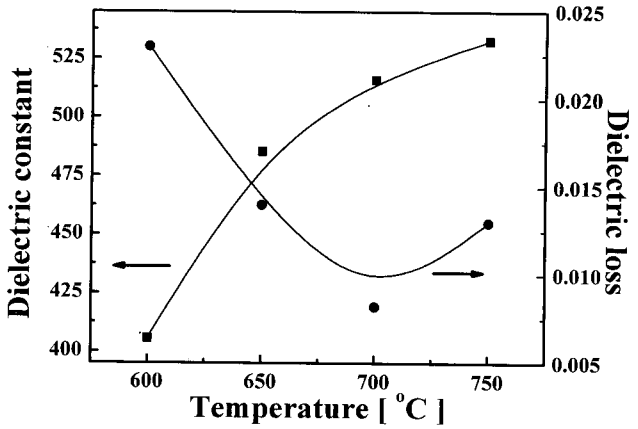

Fig. 3. Dielectric constant and dielectric loss of BST/MgO/Si structure as a function of the annealing temperature.

the range of 25-60 nm and the absence of both cracks and pin-holes. However we have found that the increase of annealing temperature causes the noticeable increase of the surface roughness. This effect seems to be reasonable and may be related to the increase of the grain size of BST thin films. This conclusion is in a good agreement with the XRD measurements, which indicate the increased peak sharpness with increasing annealing temperature.

In one of previous works, Chivukula et al. suggests the negligible frequency dispersion in the capacitance up to 10 GHz. Based on this data, we measured dielectric constant and tunability of BST films at 10 kHz assuming good correlation with the behavior of these parameters at microwave frequencies[11]. Although the dielectric loss usually shows higher value at microwave frequencies, the quantitative correlation between the results of low-frequency measurements and high-frequency measurements is also present. In other words, the films showed lower dielectric loss at low (10 kHz–1 MHz) frequencies, have lower dielectric loss at microwave frequencies.

Therefore we can assume that low-frequency measurements provide a convenient method to characterize electrical properties without the complicated technique required for microwave frequencies. To calculate the dielectric constant of the BST film with the IDTs, an analytical model suggested by Farnell et al. was adopted[12].

As the result, we have found increasing annealing temperature of BST thin films causes an increase in dielectric constant, which reaches a maximum of (551) for the films annealed at 750 °C. At the same time, Fig. 3 shows that the dielectric loss decreases with increasing annealing temperature. However, when the annealing temperature exceeds 750 °C, relative dielectric loss begins to increase. It is well known that the dielectric

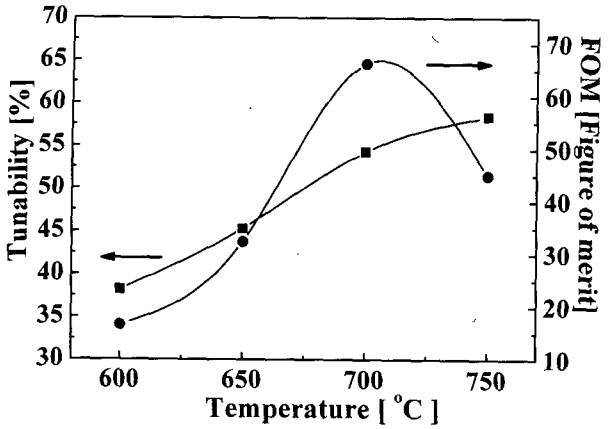

Fig. 4. Tunability and FOM of BST/MgO/Si structure as a function of the annealing temperature.

loss in ferroelectric and dielectric materials is affected by various factors such as space charge polarization, crystallinity, domain wall pinning, secondary phase, and interfacial diffusion. Therefore, highest dielectric loss obtained for the films annealed at 750 °C may be explained by interfacial diffusion.

Figure 4 shows the figure of merit (FOM) and tunability of BST/MgO thin films as a function of annealing temperature. The tunability was determined as $(\varepsilon_{max}-\varepsilon_{min})/\varepsilon_{max}$, where ε_{max} and ε_{min} are the maximum and minimum values of permittivity measured at the zero electric field and 500 kV/cm electric field, respectively. The FOM is a frequently used parameter to characterize correlations between tunability and dielectric loss. This parameter is defined as FOM =[(%) tunability / tanδ (%)], where dielectric loss is given on a percentage scale[13]. The FOM reflects that a tunable microwave circuit cannot take full advantage of high tunability if the loss factor is too high. Ideally, the FOM value should be as high as possible. From this figure we obtained the increase of both tunability and FOM with increasing annealing temperature. The tunability and FOM of the BST/MgO annealed at 700 °C were 54.3 % and 66.2, respectively.

4. CONCLUSION

In this work, we have shown that BST films with high tunability, low dielectric loss and high FOM can be prepared onto the MgO/Si substrate by sol-gel method. For the MgO/Si substrate, the BST thin films exhibited highly (100) orientation. The highly (100)-oriented BST thin films showed high dielectric constant, tunability, and figure of merit (FOM). The dielectric constants, dielectric loss and tunability of the BST thin films annealed at 700 °C deposited on the MgO/Si substrate measured at 10 kHz were 515.9, 0.0082, and 54.3 %, respectively.

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