Phase-shifters 응용을 위한 MgO 박막위에 성장된 BST(100) 박막의 유전적 특성
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Dielectric properties of (100)-oriented Ba₀.₆Sr₀.₄TiO₃ Thin Films grown on MgO (100) thin films for phase-shifters
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Abstract

In this paper, we have investigated the structure and dielectric properties of the (Ba₀.₆Sr₀.₄)TiO₃ (BST) thin films film fabricated on MgO(100)/Si substrate by an alkoxide-based sol-gel method. Both the structure and morphology of films were analyzed by x-ray diffraction (XRD) and atomic force microscope (AFM). For the MgO(100)/Si substrates, 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(100)/Si substrates measured at 10 kHz were 515.9, 0.0082, and 54.3 %, respectively.

Key Words: Dielectric properties, Sol-gel, Tunable,

1. 서 론

(Ba,Sr)TiO₃ (BST) is a promising dielectric material for a variety of tunable microwave devices such as electrically tunable mixers, delay lines, filters, capacitors, oscillators, resonators, and phase shifters [1, 2]. However, for a successful application of BST in this field, there is set of properties to be satisfied: moderate-to-low dielectric constant at microwave frequencies, low dielectric loss factor, large-scale variations of the dielectric constant by direct current (DC) biasing fields 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 relationships between electrical and structural properties for the dielectric thin films [7-8], the effects of interface or buffer layers between BST and substrate are not explored enough. At the same time, the problem of interface seems to be a key problem to obtain optimal 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
(001)-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. 실험

The BST films were fabricated on the MgO/Si substrate. First, the MgO thin films were prepared by the MOD method. The precursor materials were magnesium acetate tetrahydrate ((CH₃CO₂)₂Mg·4H₂O). 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 0.4M. The final solution became transparent. The MgO solutions were spun on 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°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 Ba0.6Sr0.4TiO3 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 a N₂ atmosphere. Finally, both starting solutions were mixed to prepare the stoichiometric, clear, transparent, and stable BST precursor. For the fabrication of BST thin films, the BST precursor solution was syringed through a 0.2 m 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 contaminations. Then, the pre-baked films were annealed at 700°C for 1 hr in oxygen atmosphere for crystallization. The final thickness of BST thin film 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 m thick. The widths of finger lines and the gap between lines are 10-20 m for the IDE capacitor.

3. 결과 및 고찰

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 polycrystalline 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 intensities as follows: \( I_{hkl} = \frac{I_{(200)}/I_{(111)} + I_{(200)}}{I_{(111)}} \), where \( I \) is peak intensity [9]. Considering data of Fig. 1(a), corresponding is 0.53, 0.62, 0.95 and 0.96 for the annealing temperatures of 550, 550, 600, and 650°C, respectively. This result suggests that the annealing temperature of 650°C is enough to obtain (100)-oriented MgO film on Si substrates.

Fig. 2(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 values derived from XRD were 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 were crystallized with (100) preferred orientation. Therefore our data confirm the fact that the crystallization and growth of the BST thin films are influenced by the substrate. The XRD data show also that the increase in annealing temperature leads to the increase of the intensity of diffraction peaks. However, the full widths at half maximum (FWHM) were found to be lower. 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 film were investigated using AFM. Figure 2 shows the surface morphology of the BST films on the MgO films. It can be seen that all AFM images show small surface roughness (1.96~3.5 nm), the
grain size in the range of 25–60 nm as well as the absence of both cracks and pin-holes. However we have found that the increase in 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, the dielectric loss was found to be independent on the annealing temperature. This parameter was below 0.0023 for all BST films.

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 (max−min)/max, where max and min are the maximum and minimum values of permittivity, respectively, measured at the zero electric field and 500 kV/cm electric field. The FOM is a frequently used parameter to characterize correlations between tunability and dielectric loss. This parameter is defined as FOM = (% tunability / tan(%)), where dielectric loss is given on a percentage scale [13]. The FOM reflectsthat 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. In Fig. 4, 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. 결론

In this work, we have shown that BST films with high tunabilities, low losses and high FOM can be prepared onto the MgO/Si substrate by sol-gel method. For the MgO/Si substrates, 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 films annealed at 700 C deposited on the MgO/Si substrates measured at 10 kHz were 515.9, 0.0082, and 54.3%, respectively.

참고 문헌

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

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

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

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