

Tunable 소자 응용을 위한 Sol-gel 법으로 제작된 BST 박막의 Bi 첨가에 따른 구조적, 유전적 특성

김태형, 김경태, 김창일
여주대학, 중앙대학교

Structure and Dielectric properties of BST Thin Films prepared by Sol-gel method for Tunable element application

Tae-Hyung Kim, Kyoung-Tae Kim, and Chang-Il Kim
Yejoo Institute, Chungang Uni.

Abstract

An alkoxide-based sol-gel method was used to fabricate $Ba_{0.6}Sr_{0.4}TiO_3$ thin films doped by Bi from 5 to 20 mol% on a Pt/Ti/SiO₂/Si substrate. The structural and dielectric properties of BST thin films were investigated as a function of Bi dopant concentration. The dielectric properties of the Bi doped BST films were strongly dependent on the Bi contents. The dielectric constant and dielectric loss of the films decreased with increasing Bi content. However, the leakage current density of the 10 mol% Bi doped $Ba_{0.6}Sr_{0.4}TiO_3$ thin film showed the lowest value of 5.13×10^{-7} at 5 V. The figure of merit (FOM) reached a maximum value of 32.42 at a 10 mol% Bi doped $Ba_{0.6}Sr_{0.4}TiO_3$ thin films. The dielectric constant, loss factor, and tunability of the 10 mol% Bi doped $Ba_{0.6}Sr_{0.4}TiO_3$ thin films were 333, 0.0095, and 31.1%, respectively.

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

1. 서론

Recently, there has been a great deal of interest in the application of ferroelectric thin films for tunable microwave devices, such as electrically tunable mixers, delay lines, filters, capacitors, oscillators, resonators and phase shifters [1-2]. Tunable devices using $(Ba,Sr)TiO_3$ (BST), which has high tunability [$(C_{max}C_{min})/C_{max}$], low loss tangent, and high power handling capability, are very promising as a replacement for traditional ferrite and semiconductor device [3].

The optimal electronic parameters of a dielectric material for tunable microwave device applications are: (i) a moderate-to-low dielectric constant at microwave frequencies; (ii) a low dielectric loss factor; and (iii) a large variation in the dielectric constant in an applied DC bias field. In addition, the thin film must also possess low leakage current characteristics [4].

According to the results of previous investigations, the electrical properties such as dielectric constant, dielectric loss, and leakage current of BST thin films depend upon the deposition method, composition, dopant, electrode,

microstructure, interfacial quality, and thickness of the films [5-6]. Many researches have studied the undoped BST thin films, which offer tunability upward of 50% at bias voltage of less 10 V. However, high tunability of undoped BST thin films are high loss tangents, which are much larger than that of 0.02. It is well known that dopants can significantly modify the dielectrical and electrical properties of ferroelectric materials such as BST. In most cases, it has been researched that a donor for the B site of the perovskite structure, which have been known to lower leakage current and dielectric loss [7-8]. In this paper, we report the influence of the cationic substitution of Bi in A site such as Ba or Sr site of the ABO perovskite structure on structure and dielectric properties of BST thin films.

2. 실험

Thin films of Ba_{0.6}Sr_{0.4}TiO₃ were prepared using the sol-gel method from barium acetate, strontium acetate, bismuth acetate (the dopant precursor, in concentrations ranging from 5 to 20 mol%), each initially dissolved solutions in acetic acid were mixed together to obtain Bi doped (Ba, Sr) stock solution. Then titanium isopropoxide was added under N₂ atmosphere into 2-methoxyethanol. After refluxing for several hours at room temperature, the Bi doped (Ba, Sr) stock solution was added to the Ti stock solution. The undoped and Bi doped BST precursor solutions were spin-coated onto a Pt (120 nm) / Ti (30 nm) / SiO₂ / Si substrate using a spinner operated at 4000 rpm for 30s, and then pre-baked on a hot plate at 400 C for 10 min to remove the organic materials. The pre-baked films were then annealed at 700 C for 1 h under an oxygen atmosphere to allow crystallization to take place. The final thickness of the BST thin film was about 200 nm.

Dielectric measurements were carried out using the metalinsulatormetal (MIM) capacitor

configuration. Pt electrodes having a diameter of 200 m were then dc sputter-deposited on the BST film to form the top electrode. X-ray diffraction (XRD) profiles were obtained using a CuK radiation source on a Rigaku-D/MAX diffractometer to determine the phase formation in, and the sample crystallinity, and the orientation of the thin films. Auger electron spectroscopy (AES) was employed to assess the elemental distribution within the film and across film-Pt interface. The surface morphology of the film was analyzed by atomic force microscope (AFM). Capacitance-voltage, dielectric constant, and loss measurements were carried out using an HP 4192 impedance analyzer. The leakage current was measured by a HP 4155A semiconductor parameter analyzer at 0-5 V.

3. 결과 및 고찰

Figure 1 shows the XRD patterns of the Bi doped (020 mol%) BST thin films deposited on Pt bottom electrodes annealed at 700 C for 1 h. The (100), (110), (111), (200), (210), and (211) peaks corresponding to the BST perovskite structure are obtained in all films. All films possessed a non-textured polycrystalline structure with no evidence of secondary phase formation. Each film exhibited similar XRD patterns, so there was no change in peak intensity as a result of the Bi doping. However, the full width at half maximum (FWHM) of the peaks increased with increasing Bi content. This peak broadening effects may be connected with decreasing on grain size with increasing of Bi content [2]. The RMS roughness of the undoped, 5 mol%, 10 mol%, 15 mol%, and 20 mol% doped BST thin films were 2.95 nm, 2.86 nm, 2.48 nm, 2.21 nm, 2.09 nm, respectively.

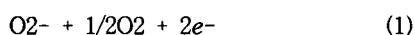
Figure 2 shows the dielectric constant and the dielectric loss of the BST thin films as a function of the Bi content at a frequency of 100 kHz. The values represented in Fig. 4 that Bi doping BST films had a dependence on the

material properties of the BST films. The dielectric constant and dielectric loss factor decreased with increasing Bi content in the BST thin films. The decreasing dielectric constant of the sample with a small grain size is attributed to the increasing amounts of low dielectric constant grain boundaries. Many researchers have been reported that was the grain size effect on dielectric properties in BST films [10]. The dielectric constant and the dielectric loss factor of the 10 mol% Bi doped BST thin films were 333 and 0.0095, respectively.

Figure 3 shows the tunability and the FOM of the undoped and Bi doped BST thin films as a function of the Bi content. The tunability is defined as $(\max - \min) / \max$, where max and min are the maximum and minimum permittivity measured at 0 and 10V, respectively. In addition, an FOM is frequently used for the observed correlation between the tunability and loss, and is defined as $\text{FOM} = (\%) \text{ tunability} / \tan (\%)$, where dielectric loss is given as a percentage [11]. The FOM value reflects the fact that a tunable microwave circuit cannot take full advantage of high tunability if the loss factor is high; the FOM needs to be as high as possible. The tunability decreased with increasing Bi content of the BST thin films. The highest FOM value found for BST thin films with Bi concentration of 10 mol% was 32.42. The tunability and the FOM of the 10 mol% Bi doped BST film were 31.1% and 32.42, respectively.

Figure 4 shows the leakage current-voltage characteristics, measured for the undoped and doped BST film as a function of the Bi content at room temperature with a positive voltage applied to the top electrode. The value of the leakage current densities at applied voltage shows a dependence of Bi contents. As a Bi content increases from 0 to 10 mol%, the value of leakage current densities for the BST thin films decreased. At 10 mol% Bi doped BST film, we obtained the maximum leakage current densities. Further addition of Bi content, the leakage

current densities increased as shown in Fig.6. The leakage current densities of the BST films doped by less than 10 mol% Bi decreased and that of the specimen doped with more than 10 mol% increased. The decreased leakage current density could be explained as follows. Aliovalent, A site dopants such as La^{3+} , Ce^{3+} , and Bi^{3+} are known to lead n-type semiconductivity in BaTiO_3 [12] and Bi^{3+} with charge 3+ behaves similarly to La^{3+} [13]. The charge balance compensation mechanism when Ba^{2+} is replaced by Bi^{3+} as following defect reaction equation.



This result can be easily explained by the Bi acting as a donor in the BST structure suppresses the inherent oxygen vacancy concentration when formed at the top electrode/BST interface, leading to prohibit a formation of charge carriers. The decreased leakage current also was dependent on grain size of the film. The undoped BST films with large grain size have short conduction paths along the highly resistive grain boundary, which causes an increase higher leakage current than that of Bi doped BST film with smaller grain size. The leakage current density of the 10 mol% Bi doped thin film is less than $5.13 \times 10^{-7} \text{ A/cm}^2$ at 5 V.

4. 결론

We have shown that Bi doped BST films having variable Bi contents to form high tunabilities, low losses, and high FOMs, can be prepared on a Pt (120 nm)/ Ti (30 nm)/ SiO_2/Si substrate using the sol-gel method. The dielectric constant and dielectric loss of the films decreased with increasing Bi content, while leakage current decreased with increasing Bi content of the BST thin films, reaching a minimum leakage current at 10 mol%, and then

increased with further addition. The dielectric constant, loss factor, and tunability of the 10 mol% Bi doped BST thin films were 333, 0.0095, and 31.1%, respectively. These results suggest that the improved dielectric properties such as dielectric loss and leakage current densities of Bi doped BST films are suitable for tunable microwave device.

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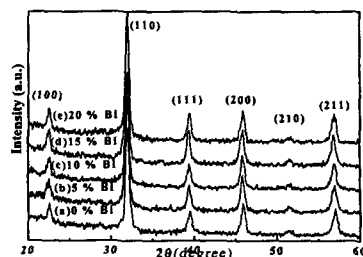


Figure 1. XRD patterns of the undoped and Bi doped BST thin films annealed at 700 C.

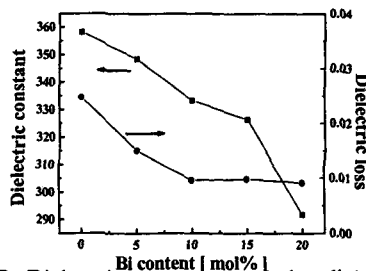


Figure 2. Dielectric constant and the dielectric loss of the BST thin films as a function of the Bi content at a frequency of 100 kHz.

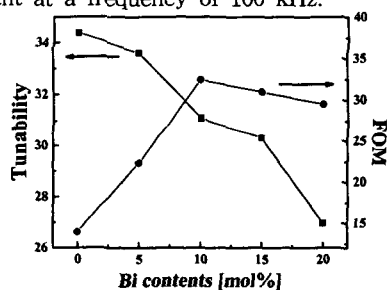


Figure 3. Tunability and the FOM of the undoped and Bi doped BST thin films as a function of the Bi content.

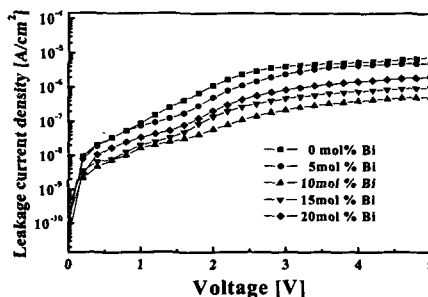


Figure 4. Leakage current-voltage characteristics of the undoped and doped BST film as a function of the Bi content.