# 초고주파 응용을 위한 MgO 버퍼층을 이용한 PST(100) 박막의 유전적 특성

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# Dielectric properties of highly (100) oriented (Pb0.5, Sr0.5)TiO3thin films grown on Si with MgO buffer layer

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**Abstract**

Pb0.5,Sr0.5TiO3(PST) thin films were deposited on Si with MgO (100) buffer layer by the alkoxide-based sol-gel method. Structural and dielectric properties of PST thin films for the tunable microwave device applications were investigated. For the MgO/Si buffer layer, the PST thin films exhibited highly (100) orientation. The MgO buffer layer affects the stress state of the (100)-oriented PST thin films. The dielectric constant, tunability, and FOM of the highly (100)-oriented PST thin film increased with increasing annealing temperature due to the decrease in lattice distortion. The differences in dielectric properties may be attributed to the change in the film stress. The dielectric constants, dielectric loss and tunability of the PST thin films deposited on the MgO/Si substrates measured at 10 kHz were 822, 0.025, and 80.1%, respectively.

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

## 1. 서 론

Due to their unique properties, dielectric/ferroelectric thin films are very attractive for applications such as capacitors in dynamic random access memories (DRAMs), piezo micro-actuators, pyroelectric infrared detectors, tunable devices, and non-linear optical devices .1-2 Among the various ferroelectrics, Pb(Zr,Ti)O3 thin films have been intensively investigated while PbTiO3 thin films have receivedless attention due to their high coercive field and large tetragonal distortion c/a of 1.064.3-4 The addition of La or Sr decreasesthe coercive field and tetragonality.5 A perovskite solid solution of PbTiO3 and SrTiO3 (PST) has the merits of the high dielectric constant of PbTiO3 and the structural stability of SrTiO3. The properties of the Pb1-xSrxTiO3 bulk for x less than 0.7 are known to be ferroelectric with a tetragonal structure at room temperature.6 To date, there have been a number of published papers dealing with the preparation and the dielectric properties of (Pb,Sr)TiO3 bulk ceramics.7

The dielectric properties of dielectric materials depend on the structural properties of the films such as phase structures developed during an annealing process, orientations of crystalline phase and the interface structure between an electrode and a film. Many researchers have studied the relationships between electrical and structural properties for the dielectric thin films19, the effects of interface or buffer layers between dielectric thin films and substrate are not explored enough. At the same time, the problem of interface layers seems to be a key problem to obtain optimal electrical characteristics and to adjust them through an obtaining of preferred orientation of PST 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 PST thin films.

## 2. 실 험

The PST films were fabricated on the MgO/Si substrate. First, the MgO thin films were prepared by the MOD method. PST thin films were deposited on the MgO(100)/Si buffer layer by a sol-gel method. Thin films of (PbxSr1-x)O3 (x=0.5) with excess Pb-acetate of 10 mol % were prepared using lead acetate trihydrate, acetate and titanium iso-propoxide as the starting materials. Acetic acid and 2-methoxyethanol were used as the solvent forthe sol-gel method. PST solutions were spin-coated on the MgO(100)/Si substrates, and then pre-baked on a hot plate at 400C for 10 min to remove the organic materials. The pre-baked films were annealed at various temperatures in the range 500650C for 1 h in the oxygen atmosphere to obtain crystallization. Thefinal thickness of PST 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 filmsprepared under various conditions. As the annealing temperature increases to 550C, the MgO thin film begins to crystallize. The XRD pattern of the MgO film annealed at 550C 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: *(hkl)* = I*(200)*/(*I*(*111*) +*I*(*200*)), where I is peak intensity.20 Considering data of Fig. 1(a), corresponding is 0.53, 0.62, 0.95 and 0.96 for the annealing temperatures of 550, 600, 650, and 700C, respectively. This result suggests that the annealing temperature of 650C is enough to obtain (100)-oriented MgO film on Si substrates.

Fig. 1(b) XRD patterns of the PST thin films deposited on MgO buffer layer annealed at 700 C. All the films show a well-developed perovskite structure with no evidence of secondary phase formation. The XRD pattern of the PST film annealed at 500C shows the presence of such peaks as (100), (110), and, (200). This fact indicates that the onset of crystallization into the perovskitephase is close to 500C. It is noted that highly (100)-oriented PST thin films were obtained when annealed over 550C. Considering data of Fig. 1(b), the value increased with increasing annealing temperature. The values of the PST thin films deposited on MgO buffer layer were 0.75, 0.86, 0.95, and 0.98 for the annealing temperatures of 500, 550, 600, and 650C, respectively. The values derived from XRD suggest that the PST films deposited on MgO/Si buffer layer were crystallized with (100) preferred orientation. The results mentioned above indicate that the crystallization and growth of the PST thin films are influenced by the substrate used. The XRD data show also that the increase in annealing temperature leads to the increase of the intensity of diffraction peaks. However, the full width at half maximum (FWHM)of the peaks decreased with increasing annealing temperature; these can be assumed that the grain size was increasing with annealing temperature.21

Figure 2 shows a cross-section TEM micrographs of the PST/MgO/Si structure annealed at 650C. The thickness of the PST and MgO layer is estimated to be 200 nm and 120 nm, respectively. The cross-sectional TEM image revealed a dense microstructure with a uniform thickness, and no significant reaction between the PST film and the MgO layer. Additionally, the TEM image shows the amorphous SiO2 layer, which was not grown severely. In general, the thickness of the native oxide is in the range of 3-4 nm.

Figure 3 illustratesthe lattice parameters along the surface normal, in-plane, and lattice distortion of the PST/MgO/Si structure measured from the XRD patterns for (100)-orientated PST films. The in-plane lattice parameters of the PST thin films were larger than surface normal lattice parameters. This result indicates that the possibility of the in-plane lattice distortion, although the corresponding bulk structure is a cubic. Additionally, these lattice parameters were elongated along the in-plane direction in the PST films. As it can be seen in Fig. 3, the variation of the lattice distortion, which is the lattice parameter ratio of in-plane direction to surface normal, decreased with increasing annealing temperature. Generally speaking, there are three stresses such as intrinsic stress, extrinsic stress and thermal stress in a thin film. The intrinsic stress is caused by an incomplete structural order dependence on the film deposition process and parameters. The thermal stress is induced by the difference in thermal expansion coefficients between the film and substrate.22 The extrinsic stress comes from the lattice parameter changes owing to a phase change in the lattice structure. As shownin Fig. 3, the lattice distortions decrease with increasing annealing temperature. Therefore, we suggest that the extrinsic stress diminished by the decreased lattice distortion is gradually relaxed during a high annealing temperature.

In one of previous works, Chivukula et al. reported about the negligible frequency dispersion with the capacitance up to 10 GHz.23Based on this data, we carried out the measurements of the dielectric constant and tunability of the PST films at 10 kHz assuming the same behavior of these parameters at the microwave frequencies. Although the dielectric loss is usually higher at the microwave frequencies, there is the qualitative correlation between the results of low-frequency and high-frequency measurements. In other words, the film showing, for example, lower dielectric loss in the frequency range of 10 kHz 1 MHz, also has lower dielectric loss at microwave frequencies. Therefore, low-frequency measurements provide a convenient method to characterize and to compare qualitatively the electrical properties without the complicated technique required for microwave frequencies.

In other words, the films that showed lower dielectric loss at low (10 kHz 1 MHz) frequencies, also 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 PST film with the IDTs, an analytical model suggested by Farnell et al. was adopted.24 Fig. 4 shows the voltage dependent dielectric properties such as dielectric constant of the PST thin film on the MgO buffer layer annealed at 650 C. We have found increasing annealing temperature of PST thin films causes an increase in dielectric constant, which reaches a maximum of (822) measured at zero field. At the same time, the dielectric loss was found to be independent of the annealing temperature. The dielectric loss of the PST thin films deposited on MgO buffer layer were 0.029, 0.026, 0.03, and for the annealing temperatures of 500, 550, 600, and 650C, respectively. The increase of the dielectric constant may be explained by the increase of the grain size as well as by the reduced lattice distortion.

Figure 5 shows the figure of merit (FOM) and tunability of PST/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.11 The FOM reflects that a tunable microwave circuit cannot take full advantage of high tunabilityif the loss factor is too high. Ideally, the FOM value should be as high as possible. In Fig. 5, we obtained the increase of both tunability and FOM with increasing annealing temperature. The increase of the tunability may be explained by the reduced lattice distortion, whereby small stresses in the PST film should lead to enhance a tunability. The tunability and FOM of the PST/MgO annealed at 650C were 80.1% and 32, respectively.

## 4. 결 론

In this work, we have shown that PST 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 buffer layer, the PST thin films exhibited highly (100) orientation. The dielectric constant, tunability, and FOM of the highly (100)-oriented PST thin film increased with increasing annealing temperature due to the decrease in lattice distortion. The differences in dielectric properties may be attributedto the change in the film stress. The dielectric constants, dielectric loss and tunability of the PST thin films deposited on the MgO/Si substrates measured at 10 kHz were 822, 0.025, and 80.1%, respectively.

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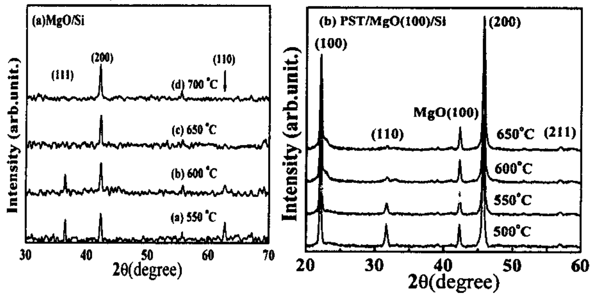


Figure 1. XRD patterns of (a) Mgo/Si and (b) PST/MgO/Si for various annealing temperatures.

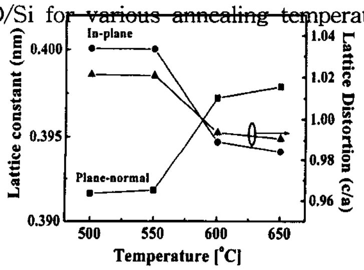


Figure 2. The lattice constant along in-plane and surface-normal directions, and lattice distortion of PST/MgO/Si structure as a function of annealing temperature.

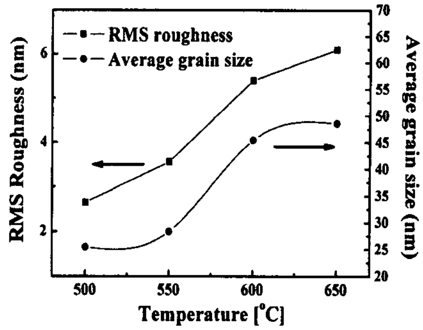


Figure 3. Grain size and RMS roughness of PST/MgO/Si structure as a function of annealing temperature.

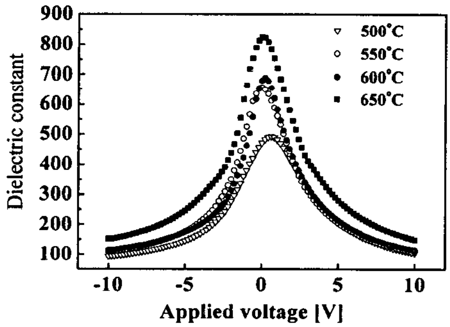


Figure 4. Dielectric constant voltage curves of the PST/MgO/Si as a function of annealing temperature.

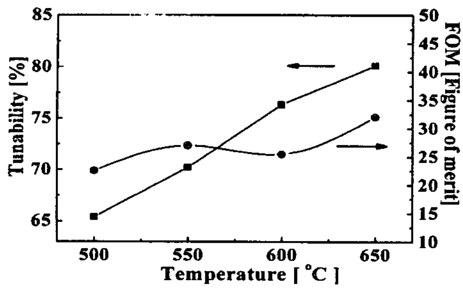


Figure 5. Tunability and FOM of the PST/MgO/Si as a function of annealing temperature.