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PST (20/80)/ PST(80/20) 이종층 박막의 유전특성

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Dielectric properties of PST (20/80)/ PST(80/20) heterolayered thin films

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Abstract - Dielectric PST (20/80) / PST (80/20) heterolayered thin films structures were created by a consequent deposition of the PST (20/80) and PST (80/20) thin films on the Pt/Ti/SiO_2/Si substrate using alkoxide-based sol-gel method. Both structural and dielectric properties of heterolayered PST thin films were investigated for the tunable microwave device applications. As the number of coating increases, the lattice distortion decreased. It can be assumed that the lower PST layer affects a nucleation site or a seeding layer for the formation of the upper PST layer. The dielectric constant, dielectric loss and tunability of the PST-6 heterolayered structure measured at 100 kHz were 399, 0.022 and 57.9%, respectively. All these parameters showed an increase with increasing number of coatings due to the decrease in lattice distortion.

1. 서론

During the past decade, the investigations of ferroelectric materials have been quite active because of promising expectations on their applications in such fields as dynamic random access memories (DRAMs), piezo micro-actuators, pyroelectric infrared detectors, tunable devices, and non-linear optical devices [1-3]. Particularly, (Ba,Sr)TiO_3 (BST), Pb(Zr,Ti)O_3 (PZT), (Pb_x,Sr_{1-x})TiO_3 (PST), (Pb,Ca)TiO_3 (PCT) and some similar titanates characterized by close values of Curie temperature (T_c) and room temperature were studied for tunable microwave applications [4]. From these works, it can be understood that optimal working parameters of the dielectric materials for tunable microwave device applications are moderate to low dielectric constant at microwave frequencies, low dielectric loss and leakage current and a large-scale variation of the dielectric constant by direct current (DC) biasing fields applied [5]. It was found also that, for the thin film - based dielectric structures, all these properties depend on the structural properties of the films such as phase structures developed during an annealing process, orientations of crystalline phase as well as the interface structure between an electrode and a film [6]. However, the effects of interface or buffer layers between dielectric thin films and substrate are not explored enough, and the problem of interface layers seems to be a key problem to obtain optimal electrical characteristics for tunable devices.Among ferroelectrics materials listed above, a dominant attention was attracted to the Pb(Zr,Ti)O_3 thin films while the $PbTiO_3$ thin films have received less attention due to their high coercive field and large tetragonal distortion c/a of 1.064. Later, it was found that the addition of La or Sr decreases the coercive field and tetragonality. For example, Normura et al. investigated polycrystalline samples of (Pb_x,Sr_{1-x})TiO_3 ceramic system. They established a complete series of solid solution from $PbTiO_3$ (PTO) (T_c = 485 C) to $SrTiO_3$ (STO) (T_c = -237 C)and found that the lattice constant and Curie temperature depend on the Pb/Sr ratio [7]. Also, in our previous work, we reported that both lattice constant and dielectric properties of PST thin films depend on Pb/Sr ratio [8]. The main purpose of present work was to investigate structural and dielectric properties for the heterolayered PST thin film structure prepared by a consequent deposition of PST (20/80) and PST (80/20) thin films on the Pt/Ti/SiO_2/Si substrate using the sol-gel method.

2. 실험

(Pb_xSr_{1-x})O_3(x=0.2, 0.8) thin films with 10 mol % excess of Pb-acetate were prepared using lead acetate trihydrate, acetate and titanium iso-propoxide as the starting materials. Acetic acid and 2-methoxyethol were used as the solventfor the sol-gel method. The PST (20/80) precursor solution was spin-coated on the substrates using a spinner operated at 4000 rev/min for 30 s to form the first layer. These PST (20/80) films were dried at 400 C for 30 min to remove organic materials and sintered at 650 C for 1 h to crystallize them into a perovskite structure. Then, the PST (80/20) precursor solution was spin-coated and dried/sintered on the PST (20/80) films to form the second layer under the same conditions. Such procedure was repeated six times and the PST-n (n is the number of coatings) heterolayered thin films structures were fabricated. The final thickness of the heterolayered structure was about 260 nm.Measurements of dielectric properties were carried out in the metal-insulator-metal (MIM) capacitor cell. For these purpose, the top-side Pt electrodes with a diameter of 300 m were deposited on the PST heterolayered films by dc sputtering method. Capacitance-voltage characteristics as well as dielectric constant and dielectric loss were measured using HP 4192 impedance analyzer. To investigate the crystallinity, phase formation parameters and orientation of the PST thin films, X-ray diffraction (XRD) profiles were obtained using CuK radiation source (Rigaku-D/MAX). The XRD carried out to examine the lattice distortion along the surface normal and in-plane directions of the PST heterolayered thin films. The surface morphologies of the PST heterolayered thin films were examined using the JEOL 6330F field emission scanning electron microscope (FE-SEM). The cross-sectional microstructures were examined using the JEOL 6330F field emission scanning electron microscope (FE-SEM). The compositional depth profile between the PST film and the Pt electrode was investigated using the CAMECA ims-4f Secondary Ion Mass Spectrometry (SIMS). Also, the SIMS depth profile using an oxygen ion beam (5.5 keV and 15 nA) was obtained.

3. 본론

Figure 1(a) shows the X-ray diffraction patterns for the heterolayered PST thin films. The figure shows typical XRD patterns of perovskite polycrystalline structure while both pyrochlore phase and preferred orientation are not observed. From these data, it can be assumed that the lower PST layers play an important role of nucleation site or seeding layer for the formation of the upper PST layers. With a furthermore increase in the number of coatings, all the XRD diffraction peaks become sharper while the full width at a half maximum (FWHM) decreases. These results indicate that the grain size increases with increasing the number of coating. Figure 1(b) illustrates the lattice parameters along the surface normal, in-plane, as well as the lattice distortion extracted from the XRD patterns for the heterolayered structures with various numbers of coatings. It can be seen that normal lattice parameters are larger than in-plane ones. This result indicates the possibility of the in-plane tetragonal lattice distortion, although the corresponding bulk structure has a cubic shape. Additionally, these lattice parameters are elongated along the in-plane direction in the PST films, and the variation of the lattice distortion (which is the lattice parameter ratio of surface normal to in-plane direction) decreases with increasing the number of coating. Generally speaking, there are two stresses such as intrinsic and extrinsic stress in a thin film. The intrinsic stress appears in a bulk as the response for the defect such as dislocation in the film. The extrinsic stress comes from the contact layer between film and substrate. The possibility of chemical reactions. As shown in Fig. 1(b), the lattice distortion decreases with an increase in number of coatings.Such effect confirms the assumption that the lower PST layer acts as nucleation site or seeding layer resulting in improvement of the structural characteristics for upper PST layer. We suggest also that the thermal stress developed by the difference in the thermal expansion coefficient is gradually relaxed during a number of coatings. This result indicated that the tetragonal lattice is strained. It can be assumed that heterolayeredPST lattice is strained due to a tensile stress. This tensile stress of the films affects a dielectric property [9].

Figure 2 shows the surface SEM micrographs of heterolayered PST-n films with various numbers of coatings (n). The PST-5 structure with the cubic PST (20/80) thin film as a top layer shows a plated-like grain structure. The PST-6 structure with a tetragonal PST (80/20) thin film as a top layer shows an enlarged grain structure and void-free grain structure. In our opinion, the crystal growth of the upper PST layers is influenced by the lower PST layers. Therefore, the selection of the initial PST layer or a seeding layer gives a way to control the microstructure behavior of the resulted film.

Figure 3 shows dielectric constant and dielectric loss for heterolayered PST thin films measured at 100 kHz as functions of number of coatings. It can be seen that an increase in both number of coatings and film thicknessleads to increasing dielectric constant, but to decreasing dielectric loss. Both effects can be easily attributed to an increase in the grain size as well as to reduced lattice distortion. Absolute values of dielectric constant and dielectric loss for heterolayered PST-6 structure were 399 and 0.022, respectively.

Figure 4(a) illustrates the behavior of the voltage dependent dielectric properties such as dielectric constant measured at 100 kHz for the heterolayered PST thin film with the various numbers of coatings. We assume that the changes of dielectric constantmay be attributed to the changes in film stress as well as in the in-plane oriented polar axis. This result is in a good agreement with the data reported by P. Padmini et al. [10]. Particularly, they suggested that BST film was subjected to tensile stress, and a contraction occurred along the caxis resulting in the enhancement of the in-plane oriented polar axis.

Figure 4(b) shows the figure of merit (FOM) and tunability of heterolayered PST thin films as a function of coatings number. 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 fieldand at 190 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 both terms in the RHS are given on a percentage scale. 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 should be as high as possible. In Fig. 5(b), we obtain an increase in both tunability and FOM with increasing number of coatings. The increase of the tunability may be explained by reduced lattice distortion, whereby small stresses in the heterolayered PST thin film should enhance a tunability. The tunability and FOM for the heterolayered PST-6 structure were 57.9% and 15.8, respectively.

4. 결 론

In this work, we have shown that PST heterolayered thinfilms with high tunabilities, low losses and high FOM can be prepared onto the Pt/Ti/SiO_2/Si substrate by sol-gel method. As the number of coating increases, the lattice distortion decreased. It can be assumed that the lower PST layer affects a nucleation site or a seeding layer for the formation of the upper PST layer. The dielectric constant, dielectric loss and tunability of the PST-6 heterolayered structure measured at 100 kHz were 399, 0.022 and 57.9%, respectively.

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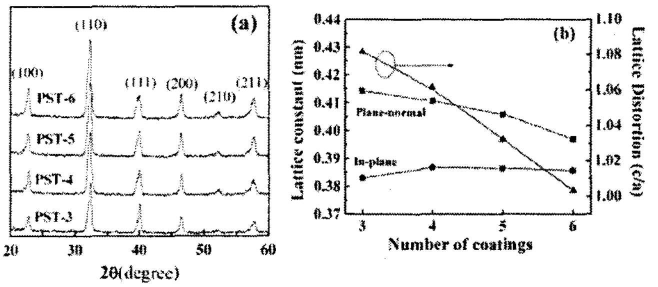
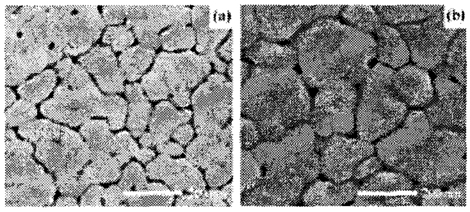
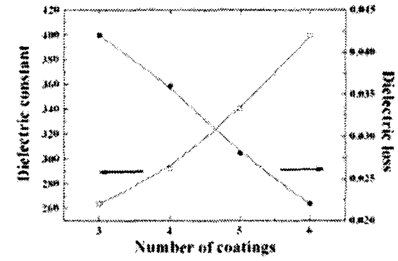


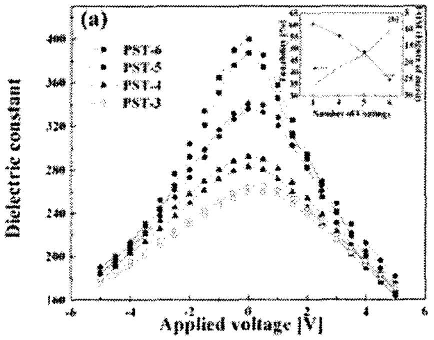
<Figure 1> (a) XRD patterns and (b) lattice constant along in-plane and surface-normal directions, and lattice distortion of PST heterolayered thin films as a function of the number of the coatings.



<Figure 2> SEM micrographs of PST heterolayered films: surface morphologies of (a) PST-5 film and (b) PST-6.



<Figure 3> Dielectric constant and dielectric loss at 100kHz of PST heterolayered films as a function of the number of the coatings.



<Figure 4> (a) Dielectric constant-voltage characteristics and (b) Tunability and FOM of PST heterolayered films.