Phase shifters 응용을 위한 Sol-gel 법으로 제작된 PST 박막의 Pb/Sr 비에 따른 구조적, 유전적 특성

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Structure and Dielectric properties of PST Thin Films with Pb/Sr ratio prepared by Sol-gel method for Phase shifter

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Abstract

The object of investigation is represented by PbxSr1-xTiO3(PST) thin films, which were fabricated by the alkoxide-based sol-gel method on Pt/Ti/SiO2/Si substrate. We have investigated both structural and dielectric properties of PST thin films aimed to tunable microwave device applications as a function of Pb/Sr ratio. PST thin films showedtypical polycrystalline structure with a dense microstructure without secondary phase formation. Dielectric properties of PST films were found as strongly dependent on Pb/Sr composition ratio. Increasing of Pb content leads to simultaneous increasing of both dielectric constant and dielectric loss characteristics of PST films. The figure of merit (FOM) parameter (FOM = (%) tunability / tan (%)) reached a maximal value of 27.5 corresponding to Pb/Sr ratio of 40/60. The tunability increased with increasing Pb content. The dielectric constants, dielectric loss and tunability of the PST thin films at Pb/Sr ratio of 40/60 measured at 100 kHz were 335, 0.0174 and 47.89 %, respectively.

Key Words : Dielectric properties; Sol-gel, Tunable

1. 서 론

Recently, there is a great interest in the application of ferroelectric and paraelectric thin *films for tunable microwave device such as* electrically tunable mixers, delay line, filters, capacitor, oscillators, resonators and phase shifters [1,2,3]. Ferroelectric materials are characterized by sufficient advantages including the possibility of dielectric constant adjustment by the electric field applied.Ferroelectric materials such as (Ba,Sr)TiO3 (BST) [4], PST [5] and (Pb,Ca)TiO3 (PCT) [6] and similar titanates, which are characterized by close values of Curie temperature and room temperature are well studied for tunable microwave applications. The optimal working parameters of the dielectric materials for tunable microwave device applications may be specified as follows:

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 [7].

Normura and Sawada investigated polycrystalline samples of (PbxSr1-x)TiO3 ceramic system and established a complete series of solid solution from PbTiO3 (PTO) (*Tc* = 485 C, *Tc* Curie temperature) to SrTiO3 (STO) (*Tc* = -237 C)[8]. The lattice constant and Curie temperature were found to be dependent on Pb/Sr composition ratio in the PST ceramics. STO films have a cubic phase at room temperature and characterized by lower crystallization temperature in comparison with BST films.On the other hand, the dielectric constant is lower than that of BST films. In contrary, tetragonal PTO films showedhigher dielectric constant at room temperature. H. J. Chung et al. proposed that addition of lead oxide into STO resulted not only in higher dielectric constant but in lower deposition temperature too [5]. The main peculiarity of PST films is that they have Curie temperature below room temperature level. Therefore, it is possible to expect that PST films will show the properties of paraelectric materials with a high dielectric constant. This fact makes PST films as a very perspective material for tunable devices applications.

The sol-gel and MOD techniques are widely used for ferroelectric thin films fabrication due to several advantages in comparison with dry methods. The most important advantages are the simple equipment, the ability of accurate control of stoichiometry over large area, high homogeneity and relatively low process temperature [9]. Since the PST films for tunable microwave device are not widely studied,the main aim of our work includes the investigations of structural and electrical properties of PST thin films prepared using sol-gel method as a function of Pb/Sr compositions.

2. 실 험

Thin films of (PbxSr1-x)O3(x=0.2 ~ 0.8) 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-methoxyethol were used as the solvent forthe sol-gel method. PST precusor solutions were spin-coated on the Pt (120 nm) / Ti (30 nm) / SiO2 / Si substrate, and then pre-baked on a hot plate at 400 C for 10 minto remove the organic materials. The pre-baked films were annealed at 650 C temperatures for 1 hr in oxygen atmosphere for crystallization. The final thickness of PST thin film was about 200 nm. Measurements of dielectric properties were carried out in the metal-insulator-metal (MIM) capacitor cell. For these purposes, top-side Pt electrodes with diameter of 300 m were deposited on the PST 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). Surface morphology of PST films was analyzed and quantified by atomic force microscope (AFM). The cross-sectional microstructures of PST films were examined using a JEOL 6330F field emission scanning electron microscope (FE-SEM). The chemical compositions of PST films were determined using an electron probe microanalysis (EPMA). Compositional depth profiles of both PST films and substrate were investigated by auger electron spectroscopy (AES).

3. 결과 및 고찰

Figure 1 shows XRD patterns of the PST thin films annealed at 650 C as a function of Pb/Sr compositions. It is evidently clear from Fig.1 that all tested PST films at any Pb/Sr ratios are characterized by non-textured polycrystalline structure with no evidence of the secondary phases formation. The XRD patterns analysis of PST films with Pb/Sr ratios of (20/80), (30/70) and (40/60) showed a domination of cubic structure while the films with Pb/Sr ratios of (50/50), (60/40), (70/30) and (80/20) are characterized by tetragonal structure. Experiments showed that phase transitions from the tetragonal structure of PTO to the cubic structure of STO occurred at stoicheometric composition of x ~ 0.5.

Figure 2 illustrates the lattice parameters of the PST films as a function of Pb/Sr compositions. Lattice parameters were calculated on the base of the XRD patterns corresponded to room temperature. Fig.2 shows an abrupt changes of lattice parameters of PST thin films, which appear at the PST composition corresponding to Pb/Sr ratio of (50/50). Therefore, it is possible to assume that this ratio corresponds to transition composition. The c/a value of the PST films with ratios of (40/60) and(50/50) thin film calculated from the XRD pattern are 0.9982 (a=0.3907 nm, b=0.391 nm) and 1.014 (a=0.3925 nm, b=0.395 nm), respectively.

Figure 3 shows the experimental data concerning dielectric constant and dielectric loss of PST thin films as a function of Pb/Sr compositions at frequency of 100 kHz. Data of Fig.6 show that increasing of Pb content in PST film leads to increasing of dielectric constantbut to decreasing of dielectric loss. This fact is a rather predictable and may be easily explained by improvement of ferroelectric properties [5]. Absolute values of dielectric constant and dielectric loss of PST (40/60) thin films were 335, 0.0174, respectively.

Figure 4 represents figure of merit (FOM) and tunability parameters of the PST thin films as a function of Pb/Sr compositions. The tunability was defined as (max-min)/ max, where max and min are the maximum and minimum values of permittivity, which were measured at zero electric field and 250 kV/cm electric field, respectively. 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 in percentage scale [10]. FOM value reflects the fact that a tunable microwave circuit cannot take full advantage of high tunability if the loss factor is high too. Ideally, FOM value is desired to as high as possible. In our case, tunability of PST films increase with increasing of Pb content. The dependence of FOM on Pb ratio shows an extreme behavior with a maximum at 40% of Pb. The maximum value of FOM corresponded to this point is about 27.5. The tunability and FOM of the PST (40/60) film were obtained on the level of 47.89 %, 27.52, respectively.

Figure 5 shows the voltage dependent dielectric properties such as dielectric constant measured at 100 kHz of the PST (80/20) and PST (40/60) capacitors. Capacitor with PST (40/60)film showed the absence of splitting in the capacitance curve for decreasing and increasing voltage in the directions of various polarities. This fact indicates that PST thin film with Pb/Sr ratio of (40/60) is actually corresponds to paraelectric state but not to ferroelectric state. In contrary, PST (80/20) capacitor showed a typical ferrolectric butterfly shape.

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 Pt (120 nm) / Ti (30 nm) / SiO2/ Si substrate by sol-gel method as a function of Pb/Sr compositions.The phase transition from the tetragonal phase of PTO to the cubic phase of STO occurred at x~0.5. We inferred that the transition composition were near the PST (50/50) thin film. The c/a value of the

PST (40/60) and PST (50/50) thin film calculated from the XRD pattern are 0.9982 (a=0.3907 nm, b=0.391 nm) and 1.014 (a=0.3925 nm, b=0.395 nm), respectively. The tunability and FOM of the PST (40/60) film were 47.89%, 27.5, respectively.

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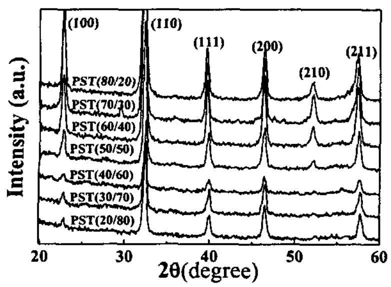

Figure 1. XRD patterns the PST thin films annealed at 650 Cas a function of Pb/Sr compositions.

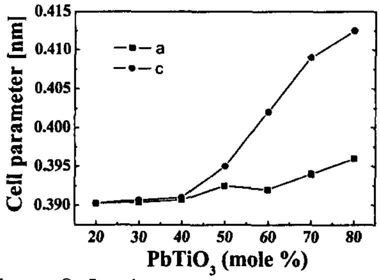

Figure 2. Lattice parameters at room temperature as a function of Pb/Sr compositions.

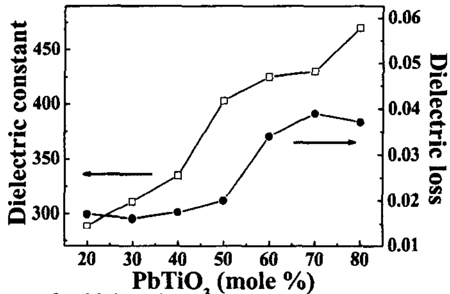

Figure 3. Dielectric constant and dielectric loss of PST thin films as a function of Pb/Sr compositions at frequency of 100 kHz .

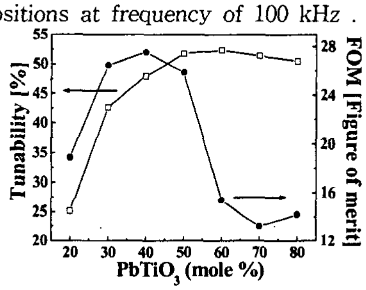

Figure 4. Tunability and FOM of the PST thin films as a function of Pb/Sr compositions

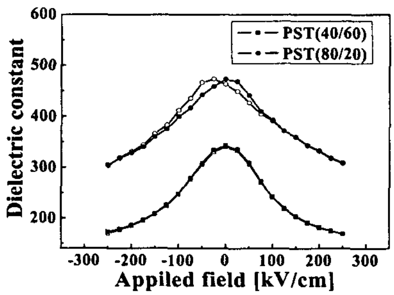

Figure 5. Dielectric constant voltage curves of the PST thin film at frequency of 100 kHz.