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Journal of the European Ceramic Society 26 (2006) 2169–2173
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Effects of composition and stacking sequence on dielectric properties of the multilayered (Pb,Sr)TiO_3 thin films for tunable device application

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Available online 3 November 2005

Abstract

Multilayered ($Pb_{1-x}Sr_x$)TiO_3 (PST(x)) thin films consisted of uniform, PST(x) and heterostructure, PST(x)–PST80 were synthesized by coating the solutions with different Sr contents ($50 \leq x$, Sr(mol%) $\leq$80), respectively. Their structural and dielectric properties were investigated in terms of composition and stacking sequence of each film. Among uniform PST(x) thin films, the PST60 films showed the highest dielectric constant and tunability, while so lower figure of merit which is an important parameter for microwave tunable device application was obtained due to relatively higher dielectric loss. In an effort to bring down the dielectric loss, the PST(x) thin films were alternately coated with PST80 thin layer. Dielectric properties of the heterostructured PST(x)–PST80 films were found to be dependent on the intrinsic dielectric values of each film composition and corresponding phase transition temperature shift effect. Furthermore, surface roughness became smoother by inserting PST80 thin layer, resulting in decrease in dielectric loss. In case of the PST60–PST80 heterostructured film, despite of slight decrease in tunability, the figure of merit on account of lowered dielectric loss was effectively improved (>40%), compared to that of the uniform PST60 film.

Keywords: Films; Sol–gel process; Dielectric properties; Perovskite; Tunable device

1. Introduction

Thin films based on ($Ba_{(1-x)}Sr_x$)TiO_3 (BST) have recently received considerable attention as promising candidates for applications in tunable microwave devices. These tunable devices are based on the large variation of permittivity with electric field, which results in a change in phase velocity in the microwave device, allowing it to be tuned in real time for a particular application.[1] In the case of the ($Pb_{(1-x)}Sr_x$)TiO_3 (PST), systems, Sr addition to $PbTiO_3$ offers good control over many of the desired room temperature dielectric and piezoelectric properties as reported in the studies based on bulk ceramics[2] and thin films.[3,4]Especially Somiya et al.[5] have suggested these PST ceramics for promising tunable device application. While in the case of PST based thin film, there are few studies regarding dielectric tunable characteristics. Considering that PST thin films with high dielectric constant also have high dielectric loss, there is a trade-off between dielectric tunability and material loss tangent, practically for microwave tunable device application. Tunability means dielectric constant of nonlinear response to an applied dc electric field at room temperature. Also this is defined by $\{\varepsilon r(\max) - \varepsilon r(\min)\}/\varepsilon(\max)$.[1] Hence, it is needed to engineer some reliable material, which can reduce the dielectric constant and loss tangent, by keeping the tunability in the necessary range to get optimum figure of merit (FOM, tunabilty/loss tangent). For BST thin film systems, various experimental methods, such as use of dopants,[6] control of annealing process[1] and formation of $(BT/ST)_n$[7] or (Ba_xSr_{1-x})$Ti_{1+y}O_{3+z}$ layer,[8] have been tried to improve its FOM.

In this study, the structural and dielectric properties of ($Pb_{1-x}Sr_x$)TiO_3 ($0.5(PST50) \leq x(\mathrm{mol}) \leq 0.8(PST80)$) films were investigated. And PST(x)–PST80 heterostructured thin films, by sequential deposition of PST(x) and PST80 layer, were synthesized with the aim to achieve composite PST films and with the desired dielectric properties, where the PST80 film composition, containing relatively higher Sr content, was chosen primarily because of its low dielectric constant and low loss tangent. And it also might influence the surface morphology

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doi:10.1016/j.jeurceramsoc.2005.09.087

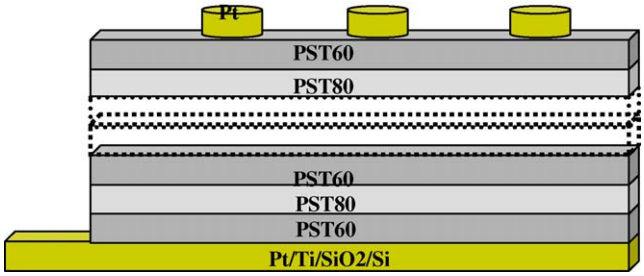

Fig. 1. Cutting plain of the heterostructure film.

of thin film. As a result, the dielectric properties and tunable device characteristics, especially FOM, of the heterostructured thin films were discussed in terms of film composition and surface roughness.

2. Experimental

A precise procedure for preparing the PST precursor solution was presented in previous report.[3] Uniform PST(x) thin films were consisted of same composition layer containing x mol% Sr. While the heterostructured films, PST(x)–PST80 were synthesized by depositing the PST(x) and PST80 layer alternately. The coated films were dried at 100°C for 1 min and then layer-by-layer heat-treated at 700°C for 1 min, respectively, for each layer deposition. Such procedures were repeated (nine times) until a desired thickness was obtained. Finally, well-crystallized PST thin films were obtained at 700°C for 10 min by directly inserting the coated thin films into the preheated Rapid Thermal Annealer (Mila-3000-P-N, Sinku-Riko), resulting in film of about 300 nm in thickness and each layer thickness is about 320–340 nm. Fig. 1 shows the cutting plain of heterostructured film. X-ray Diffractometer (D/MAX 2C, Bruker) and Atomic Force Microscopy (SPM-9500J3, Shimadzu) were used to determine crystal structure, lattice parameter and surface morphology of the films. The dielectric properties were measured by employing an impedance analyzer (4294A, HP) at 100 kHz with an electric field of 200 kV/cm and Precision Pro (Radiant Tech), and then tunability and FOM values were calculated.

3. Results and discussion

Fig. 2 shows the XRD patterns of uniform (($Pb_{1-x}Sr_x$)TiO_3) and heterostructured (($Pb_{1-x}Sr_x$)TiO_3–($Pb_{0.2}Sr_{0.8}$)TiO_3) thin films. All the films exhibited a polycrystalline perovskite. With increasing Sr (x) content, the peaks slightly shifted towards higher angles, which implies the incorporation of Sr into the crystal structure by considering relatively smaller ionic size of Sr^{2+}(0.144 nm) compared to Pb^{2+}(0.149 nm).[9–11] It was confirmed by measuring the lattice parameters for both thin film series as plotted in Fig. 3. In the uniform PST films (Fig. 3A), c/a value decreases with adding $SrTiO_3$ with cubic structure and for $x \geq 70$, convergences about 1. These results further indicated that the composition range of tetragonal/cubic phase boundary at room temperature corresponds to $x = \sim 0.7$ (mol) for the PST thin film system, where this composition seems to be similar

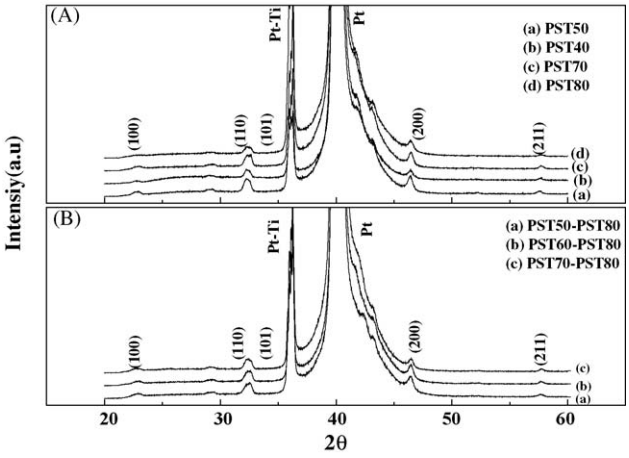

Fig. 2. XRD patterns of the uniform PST(x) thin films and heterostructure PST(x)–PST80 thin films.

to the reported boundary composition in the Sr-modified PZT study and our previous report of PST thin film.[2,3,12] The effect of Sr content on tetragonality of PST thin film was well observed in the case of the heterostructured films which were coated alternately with PST80 thin film (Fig. 3B). For all PST(x)–PST80 heterostructured film, lower tetragonality value compared to that of uniform PST(x) film was obtained due to the low tetragonality of PST80 film, implying the shift of phase transition boundary of uniform PST(x) film to lower temperature by PST80 film.

Figs. 4 and 5 display the microstructure and surface roughness imaging of PST thin films, which were measured with an AFM. For both uniform and heterostructured films, a crack free microstructure with similar grain size (0.08–0.1 μm) was exhibited. While the film surface seems to be smoother with higher Sr content. Also Fig. 5 shows that the surface roughness of PST(x) film was much decreased by forming the heterostructured films, PST(x)–PST80. These variations of surface state were confirmed by analyzing the root-mean-square (rms) roughness of uniform and heterostructured films as shown in Fig. 6. Such improvement in surface roughness of the heterostructure thin film indicates the effectiveness of PST80 layer with relatively smoother surface.

Fig. 7 shows that various electrical characteristics of uniform PST(x) thin films, where the PST60 films showed the

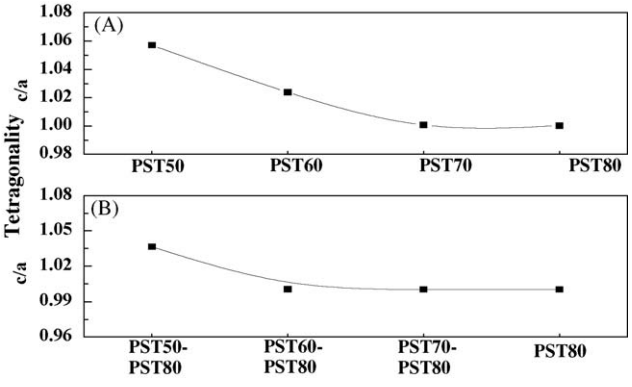

Fig. 3. Tetragonality (c/a) variation for the uniform PST(x) thin films and heterostructure PST(x)–PST80 thin films.