Tunability and Calculation of the Dielectric Constant of Capacitor Structures with Interdigital Electrodes

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Abstract. The voltage dependence of the dielectric constant of ferroelectric materials makes them attractive for use as tuning elements in microwave circuits. In this study, capacitance tuning and loss measurements were performed on ferroelectric materials prepared by the sol-gel process and RF magnetron sputtering. We find that Pb(Zr,Ti)O$_3$ (PZT) thin films with interdigital electrodes can be fabricated with reasonably low loss to make them useful for room temperature tuning applications. In addition, it is found that high temperature post-deposition annealing of both sputtered SrTiO$_3$ (ST) and sol-gel derived BaTiO$_3$ (BT) films markedly improves their tuning and loss factor characteristics. By annealing the samples in the range of 1000–1100°C, the tunability was increased by as much as a factor of seven, while the dissipation factors were decreased to values of 0.3–0.5%. In addition, it is shown that the permittivity of the films in these interdigitated capacitor structures can be calculated using an analytical model previously described by Farnell et al. [1].

Introduction

Ferroelectric thin films are currently being considered for a variety of electronic applications. Some of these applications take advantage of the voltage dependence of the dielectric constant. This feature can be used to produce voltage-tunable microwave devices such as resonators, filters, and phase shifters [2,3]. For these applications, it is desirable to produce thin films which have maximum tunability (change in capacitance under applied dc bias) with a minimum loss factor. Since the tunability of a ferroelectric material is maximized near its Curie point, different materials are preferred for different application temperatures. Thus, materials such as SrTiO$_3$ are useful for tuning applications at cryogenic temperatures (the Curie point of SrTiO$_3$ is 65 K), whereas materials such as BaTiO$_3$ and Pb(Zr,Ti)O$_3$ are more suitable for room-temperature tuning applications.

A useful figure of merit for a tunable capacitor is given by:

$$\text{FOM} = \frac{\left(\%\text{tunability} \equiv \dfrac{C_{\max} - C_{\min}}{C_{\max}}\right)}{\tan\delta(\%)} \quad (1)$$

where $C_{\max}$ is the maximum capacitance and $C_{\min}$ is the minimum capacitance over the investigated voltage range, and the dissipation factor ($\tan\delta$) is given as a percentage. This figure of merit reflects the fact that a tunable microwave circuit cannot take full advantage of a high tunability if the loss factor is high. While this figure of merit should be as high as possible, previously reported values have typically been less than 50.

Previous work has been mainly focused on two ferroelectric materials, SrTiO$_3$ and Ba$_{1-x}$Sr$_x$TiO$_3$ [4–6]. In the present study, SrTiO$_3$ (ST), BaTiO$_3$ (BT), and Pb(Zr,Ti)O$_3$ (PZT) thin film capacitors are

evaluated. The effect of post-deposition annealing on the tuning and loss factor of selected samples is discussed. In addition, it is shown that the permittivity of these ferroelectric thin films with interdigital electrodes can be calculated based on an analytical technique described by Farnell et al. [1].

Experiment

The PZT and BT films were deposited using the sol-gel process while the ST films were deposited using RF magnetron sputter-deposition. The details of PZT precursor solution preparation have been previously described [7]. Briefly, the PZT process utilizes lead acetate, $Pb(CH_3COO)_4$, Zr-butoxide, $Zr(OC_4H_9)_4$, and Ti isopropoxide, $Ti(OC_3H_7)_4$, as precursors and acetic acid and methanol as solvents. The BT process utilizes barium acetate, $Ba(OOCCH_3)_2$, and Ti isopropoxide $Ti(OC_3H_7)_4$, as starting precursors and acetic acid and methanol as solvents [8].

The BT and PZT films were grown from the precursor solutions on Al_2O_3 and $LaAlO_3$ single crystal substrates using a multilayer spin-casting approach. Each layer was spin-coated using 3000 rpm for 30 s and then pyrolyzed on a hot plate at 300°C. After the desired number of layers have been deposited, the PZT films were crystallized at 650°C and the BT films were crystallized at 750°C. The ST films were deposited using RF magnetron sputtering at about 800°C from a bulk $SrTiO_3$ target. Selected ST and BT samples were subjected to post-deposition annealing at 1000–1100°C for up to 16 h.

Pt interdigital electrodes (IDEs) were then deposited on all films using a combination of magnetron sputtering and lift-off photolithography. The top Pt electrodes were about 0.1 μm thick. The IDE patterns consisted of 100 or 200 finger pairs with identical finger width and spacing (12 or 24 μm); the fingers were 2 mm long. Figure 1 shows a schematic of the cross section of the finished capacitor structure and defines the relevant variables used in the dielectric constant calculation. The capacitance and loss factor measurements were done on these films using an HP 4194A impedance analyzer and a 20 mV oscillation level. For the tunability measurements, a dc bias was also applied across the IDE pattern. The dielectric properties of the films and their tunability were measured at temperatures ranging from 77 K to 393 K.
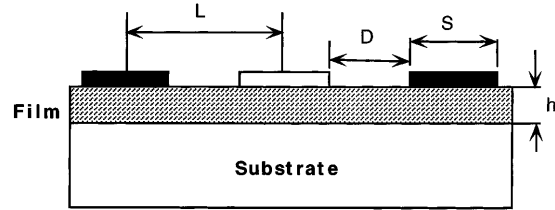
The dielectric measurements were mostly carried

Fig. 1. A schematic of a cross section of the capacitor structure used in the tuning experiments. Pt interdigital electrodes were used in all capacitors.

out at 10 kHz. However, it should be pointed out that we have observed, and others have reported [9], negligible frequency dispersion in the capacitance up to 10 GHz. Thus, film capacitance and tunability measured at 10 kHz are actually a good indicator of film properties at microwave frequencies. However, the dissipation factor ($\tan \delta$) did increase at microwave frequencies. Nonetheless, the films that had lower losses at low frequencies (10 kHz-1 MHz) also displayed lower losses at microwave frequencies. Thus, low frequency measurements provide a convenient method for determining the suitability of thin films for tuning applications without the difficulties associated with subtracting electrode losses at microwave frequencies. Finally, the microstructure and grain morphology of selected samples were examined using field-emission scanning electron microscopy (FESEM).

To calculate the dielectric constant of the thin film capacitors with the interdigital electrode configuration shown in Fig. 1, an analytical model previously derived by Farnell et al. was employed [1]. Based on Farnell's analysis, it can be shown that the dielectric constant of a thin film having the configuration shown in Fig. 1 can be calculated using the following expression:

$$\varepsilon_f = \varepsilon_s + \left\{ \frac{C - [K(1 + \varepsilon_s)]}{K\left[1 - \exp\left(\frac{-4.6h}{L}\right)\right]} \right\} \quad (2)$$

where ε_f and ε_s are the film and substrate dielectric constants, respectively; h is the film thickness, K is a constant which has units of pF, and C is the measured capacitance per unit finger length per electrode section of width L, and is given by:

$$C = \frac{C_m}{FL \times N} \quad (3)$$

where C_m is the experimentally measured capacitance

in (pF), FL is the IDE finger length in meters, and N is the number of sections of width L (L is half the IDE pattern period or $\lambda/2$). The number of sections of width L in an IDE pattern can be calculated from the following formula:

$$N = 2P - 1 \tag{4}$$

where P is the number of finger pairs in the IDE pattern. The constant K is given by [1]:

$$K = 6.5\left(\frac{D}{L}\right)^2 + 1.08\frac{D}{L} + 2.37 \tag{5}$$

For an IDE pattern with equal finger width and spacing, L is equal to 2D, where D is the IDE finger width, and $K = 4.53$ pF. The IDE patterns used in these experiments all had equal finger width and spacing. This means that the value of K used throughout these calculations is 4.53 pF.

Similarly, for the case where the dielectric thin film is grown on a bottom electrode (ground plane), the dielectric constant following Farnell's analysis can be calculated from:

$$\varepsilon_f = \frac{(C - K\varepsilon_s)}{K + \left\{0.29\left(\frac{h}{L}\right)^{-1.5} - 1\right\}} \tag{6}$$

where all the variables and constants have the same definition as described above. This expression holds for the case when the alternating fingers in the IDE pattern have opposite polarity and the ground plane is floating [1].

Results and Discussion

A. Pb(Zr,Ti)O₃ Capacitors

Pb(Zr,Ti)O$_3$ films have received less consideration than ST and BST films for tuning applications, presumably due to the lower microwave losses reported for ST and similar materials [10]. However, our evaluation of the tuning and loss characteristics of several PZT thin film compositions indicate that PZT films with interdigital electrodes have reasonably low loss, which makes them potentially useful for room temperature tuning applications. Figure 2a shows the voltage dependence of the capacitance and loss factor of a Nb-doped PZT 4/50/50 film (0.5 μm thick) grown on a LaAlO$_3$ single crystal. Niobium is a donor dopant
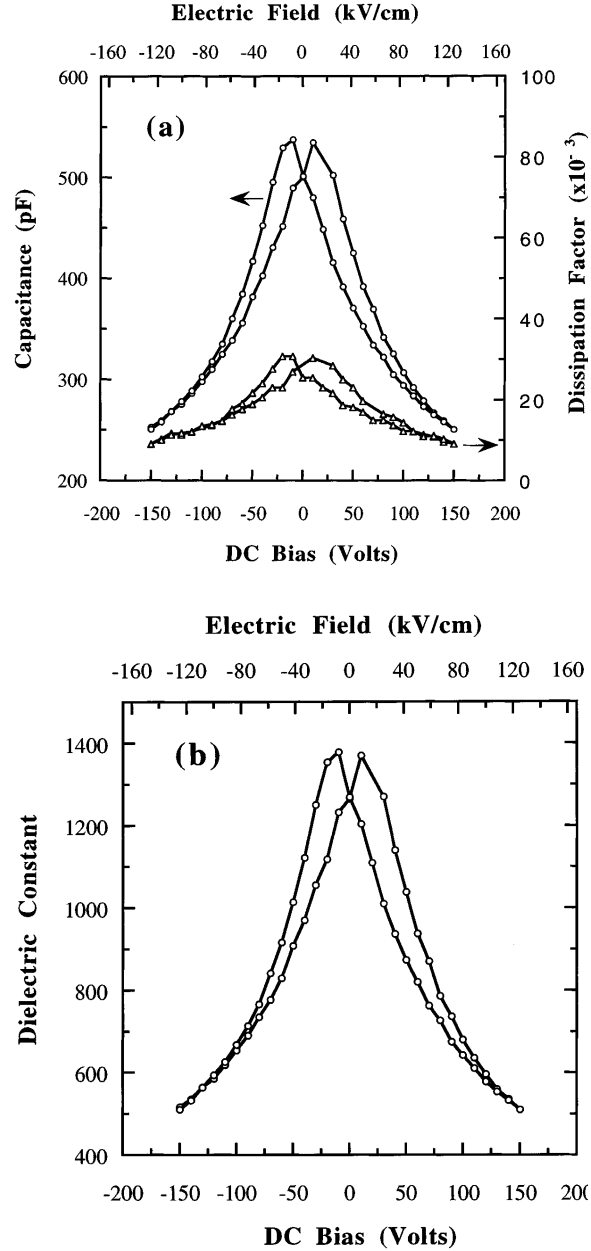

Fig. 2. (a) Capacitance and loss factor and (b) dielectric constant of a PNZT 4/50/50 film as a function of dc bias. The PNZT film was deposited on a LaAlO$_3$ single crystal substrate. The measurements were performed at 10 kHz and room temperature.

that has previously been shown to improve the leakage current and lifetime of PZT thin film capacitors [11]. As Fig. 2 shows, the PNZT 4/50/50 capacitor exhibits 54% tuning over an applied bias of ±150 V (±125 kV/cm). This is the largest tuning value obtained for any of the samples tested in this

study. The loss factor for this sample ranged from 1% at 150V to about 2% at zero bias.

Figure 2b shows the voltage dependence of the dielectric constant of the same PNZT 4/50/50 film. The %tunability calculated from the change in the dielectric constant shown in Fig. 2(b) is 68% compared to 54% calculated from the change in the capacitance for the composite structure (Fig. 2a). This difference is due to the fact that the capacitance is the sum total of the contribution from the high-permittivity film, which exhibits tuning, and the substrate, which has a constant permittivity. This result implies that this film thickness (0.5μm) is not optimized to provide maximum tuning of the capacitance. It further points out that reporting the change of the dielectric constant rather than of the capacitance, as is typically done, is preferable for understanding the materials properties.

Table 1 shows that an identical PNZT 4/50/50 thin film grown on a C-cut Al_2O_3 single crystal showed lower tuning (47%) and a 30% smaller dielectric constant (1260 vs. 850) at zero bias as calculated from Eq. (2). The difference in magnitude of the tuning and in-plane permittivity of the two PNZT 4/50/50 films grown on $LaAlO_3$ and Al_2O_3 can be attributed to differences in film microstructure. Specifically, SEM analysis of the PNZT films grown on C-cut Al_2O_3 revealed that the near-surface region of the films contains some ($\sim$25%) of the non-ferroelectric, low-permittivity fluorite phase, that is typically observed

in our films. The identity of this phase has been previously confirmed by electron diffraction analysis. In contrast, the PNZT 4/50/50 film grown on $LaAlO_3$ appeared to consist entirely of the perovskite phase with no evidence of a near-surface fluorite phase.

The in-plane dielectric constant values of the PNZT 4/50/50 films calculated from Eq. (2) are comparable to, although slightly lower than, the values that are usually obtained using a parallel-plate capacitor geometry. For instance, the in-plane dielectric constant value of the PNZT 4/50/50 thin film grown on $LaAlO_3$ substrate is 1260 at zero bias (Table 1), whereas the permittivity determined from a parallel-plate geometry is typically 1400–1500. Since the capacitance values calculated from Farnell's analysis and Eq. (2) are only supposed to be accurate to within 10%, the agreement between the in-plane and out-of-plane agreement is good, especially considering that the films for the out-of-plane measurements are grown on a base electrode (Pt) rather than directly on $LaAlO_3$.

Tuning experiments were also performed on undoped PZT films. Figure 3 shows the voltage dependence of the dielectric constant and loss factor of a 0.4μm thick PZT 53/47 thin film grown on a C-cut sapphire substrate. As indicated in Table I, this film exhibits 43% capacitance tuning over a bias of ± 150 V (± 125 kV/cm). Similar to the PNZT films discussed earlier, the in-plane loss factor of this film is relatively low ($\sim$2% at zero bias). These results

Table 1. Summary of the dielectric properties of various capacitor stacks with interdigital electrodes (@10 kHz and zero bias). All values were measured at room temperature except for $SrTiO_3$ which was measured at 77 K

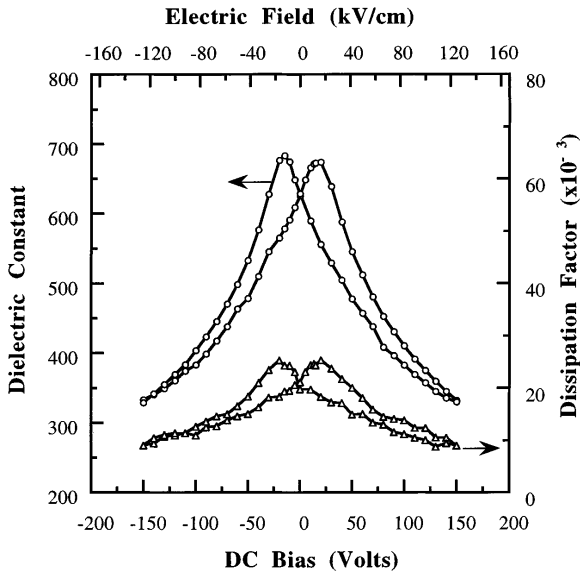| Sample	Capacitance, pF	Permittivity	$\tan \delta \times 10^{-3}$	%Tunability
PNZT 4/50/50 on $LaAlO_3$	500	1260	18	54% ($\pm$125 kV/cm)
PNZT 4/50/50 on C-Al_2O_3	305	850	14	47% ($\pm$125 kV/cm)
PZT 53/47 on C-Al_2O_3	190	620	16	43% ($\pm$125 kV/cm)
PZT 4/30/70 on Pt/SiO_2/Si	23 000	574	80	—
$BaTiO_3$ (750°C)	55	280	38	6% ($\pm$83 kV/cm)
$BaTiO_3$ (1100°C)	86	1200	5	21% ($\pm$83 kV/cm)
$SrTiO_3$ (800°C)	80	320	5	3.5% ($\pm$83 kV/cm)
$SrTiO_3$ (1000°C)	112	720	3–4	21% ($\pm$83 kV/cm)

Fig. 3. Dielectric constant and loss factor of a PZT 53/47 film as a function of dc bias. The PZT film was deposited on an Al_2O_3 single crystal. The measurements were performed at 10 kHz and room temperature.

indicate that PZT thin film compositions with 43–54% tunability and relatively low loss can be fabricated using interdigital electrodes. Consequently, these films are potentially useful for capacitance tuning applications. However, additional measurements especially at higher frequencies ($\omega > 1$ GHz), are needed to evaluate film properties for microwave applications.

Another capacitor geometry that was investigated using PZT thin film capacitors included a ground plane between the ferroelectric thin film and the single crystal substrate. In this case, the PZT films were grown on Pt electrodes that were sputter-deposited on SiO_2/Si substrates using RF magnetron sputtering. The dielectric constant of such a capacitor structure with interdigital top electrodes was calculated using Eq. (6). As Table 1 shows, a dielectric constant value of about 574 was obtained for a La-doped PZT thin film (PLZT 4/30/70). This value is somewhat smaller than obtained by parallel-plate capacitor geometry, where a dielectric constant of 650–750 has been reported [12].

B. BaTiO_3 Capacitors

Tuning experiments were performed on $BaTiO_3$ films at room temperature and at 120° C. The $BaTiO_3$ films

were typically crystallized at 750°C for 1/2 h in air. Under these conditions, the films exhibited granular morphology with a grain size of $\sim$50 nm as shown in Fig. 4. The tuning performance of these films at room temperature is illustrated in Fig. 5, which shows both the capacitance and dielectric constant change with applied bias for two BT films. As Fig. 5a illustrates, the BT films crystallized at 750° exhibit only about 6% capacitance tunability over $\pm$83 kV/cm applied field. In addition, Table 1 indicates that the in-plane dielectric constant and loss factor of these films are about 280 and 4% at zero bias, respectively. These values are comparable to those obtained using BT thin films in parallel-plate capacitor geometry.

However, we find that the BT film properties can be substantially improved by annealing the films at higher temperatures. Figure 5a also shows the capacitance tuning behavior of a $BaTiO_3$ film that has been processed at 1100°C for 16 h. It is clear from Fig. 5 that the film post-annealed at 1100°C is superior to the film crystallized at 750°C. The film annealed at 1100°C exhibits 21% capacitance tunability over $\pm$83 kV/cm applied field and a loss factor of 0.5% at zero bias. This is compared to 6% capacitance tunability and 4% loss factor for the film crystallized at 750°C. Furthermore, the BT film annealed at 1100°C exhibits significant hysteresis relative to the film crystallized at 750°C. The superior performance of the film annealed at 1100°C is also reflected in the change in the dielectric constant with bias as shown in Fig. 5b. As discussed in the previous section, the tunability values calculated from the change in the dielectric constant of the films with applied bias are larger than those calculated from the capacitance values of the film/substrate structure. This difference, as discussed earlier, has to do with field penetration into the substrate and the resultant contribution of the substrate to the measured capacitance.

In addition to being fired at higher temperature, the BT film also had 2.5 mol% excess Ti, since Ti is known to enhance grain growth in BT ceramics. In contrast, the film crystallized at 750°C was stoichiometric. Figure 6 shows an SEM micrograph of the BT film annealed at 1100°C. The figure reveals that the grain size of this film is relatively uniform and is on the order of 0.2 μm, which is approximately four times larger than that of the film crystallized at 750°C. The combination of excess Ti and high annealing temperature is believed to be the reason for the increased grain size in the case of the 1100°C process.
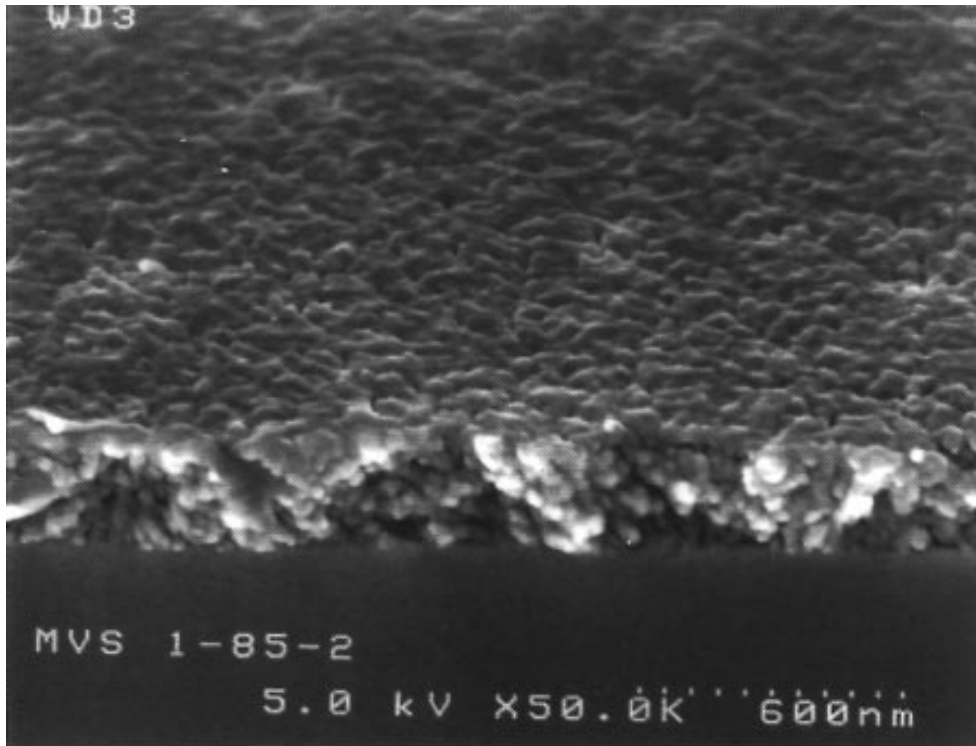
Fig. 4. Scanning electron micrograph of a BaTiO$_3$ film deposited on a LaAlO$_3$ substrate and cystallized at 750°C. The film has a grain size on the order of 50 nm.
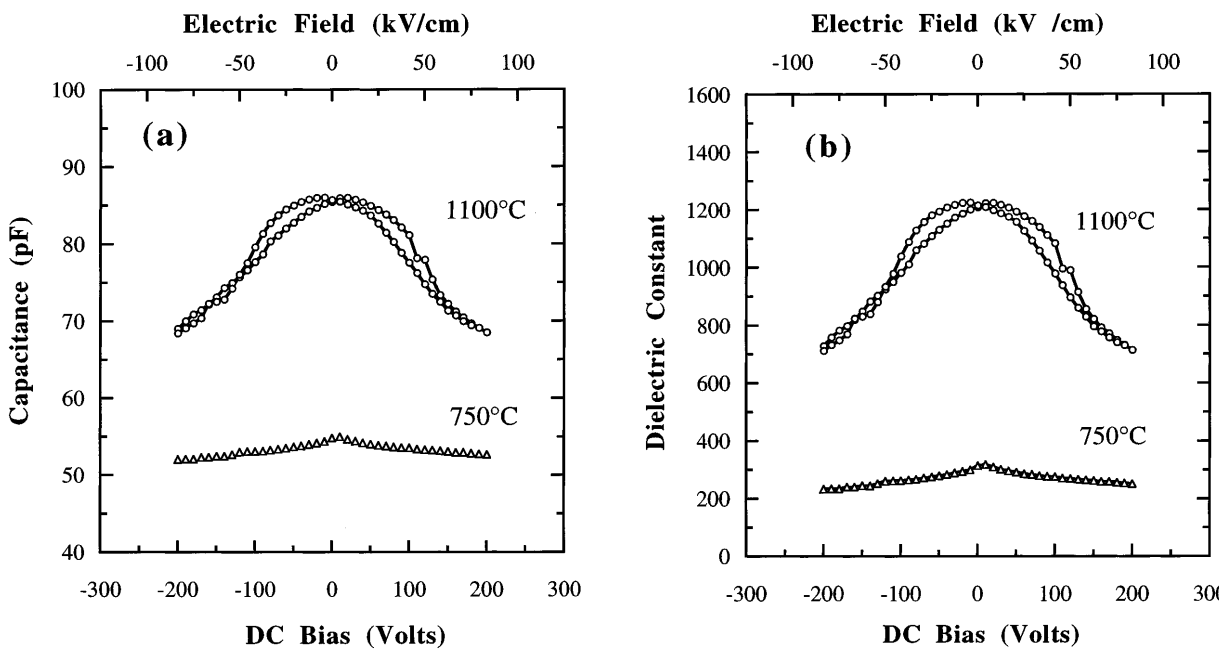

Fig. 5. (a) Capacitance and (b) dielectric constant of BaTiO$_3$ films as a function of dc bias. The films were deposited on LaAlO$_3$ substrates and annealed at 750°C and 1100°C. The measurements were performed at 10 kHz and room temperature.
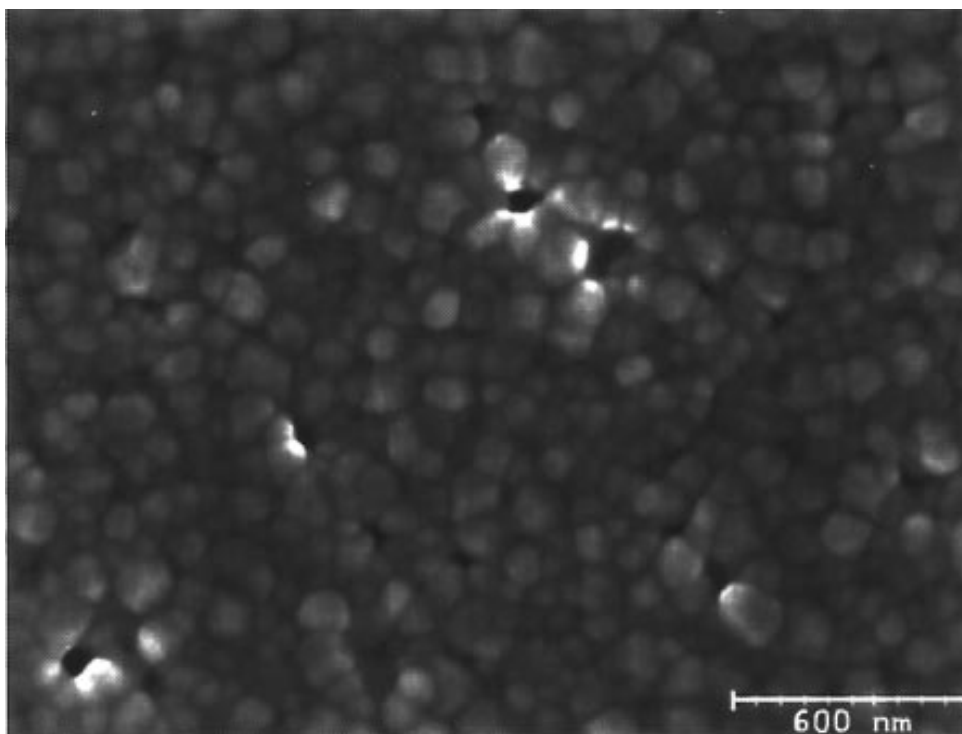
Fig. 6. Scanning electron micrograph of a BaTiO$_3$ film deposited on a LaAiO$_3$ substrate and annealed at 1100°C. The film has a grain size on the order of 0.2 μm.

The dielectric constant of both BT films showed little change with temperature and exhibited a diffuse peak in the dielectric constant near the transition temperature ($\sim$127°C for these films). As a result, the tuning properties of the BT films near the Curie point (at 120°C) were very similar to their tuning behavior at room temperature. In fact, the BT film fired at 1100°C showed 22% capacitance tunability at 120°C compared to 21% capacitance tunability measured at room temperature under otherwise identical testing conditions. This indicates that these BT films can be processed with good tunability and a minimal temperature coefficient, which is a desirable characteristic.

Figure 5b also indicated that the in-plane permittivity of the BaTiO$_3$ film annealed at 1100°C is around 1200 at zero bias. This is substantially higher than the dielectric constant values measured for our sol-gel BT films crystallized at 750°C, which exhibit values comparable to what is typically reported for BaTiO$_3$ thin films (200–300). The improved permittivity values as a result of high temperature annealing are believed to be due to the larger grain size, based on the well-known effect that grain size has on the permittivity of BaTiO$_3$ [13–15].

C. SrTiO$_3$ Capacitors

The SrTiO$_3$ thin films were deposited on LaAlO$_3$ substrates using RF magnetron sputter deposition at about 800°C [16]. The as-deposited films were 0.69 μm thick and had a typical grain size of less than a 100 nm. The typical tuning performance of these films at 77 K is illustrated in Fig. 7, which shows both the capacitance and dielectric constant change with applied bias for two ST films. As Fig. 7a illustrates, the ST films deposited at 800°C exhibit only about 3.5% capacitance tunability over $\pm$83 kV/cm applied field. The loss factor for the same sample was about 0.5% at 77 K and zero bias.

In order to determine if high temperature post-deposition annealing improves the tunability of ST films, selected ST samples were subjected to 1000°C, 5 h post-deposition annealing treatment. As Fig. 7 also shows, the tunability of the ST films improves dramatically following post-deposition annealing at
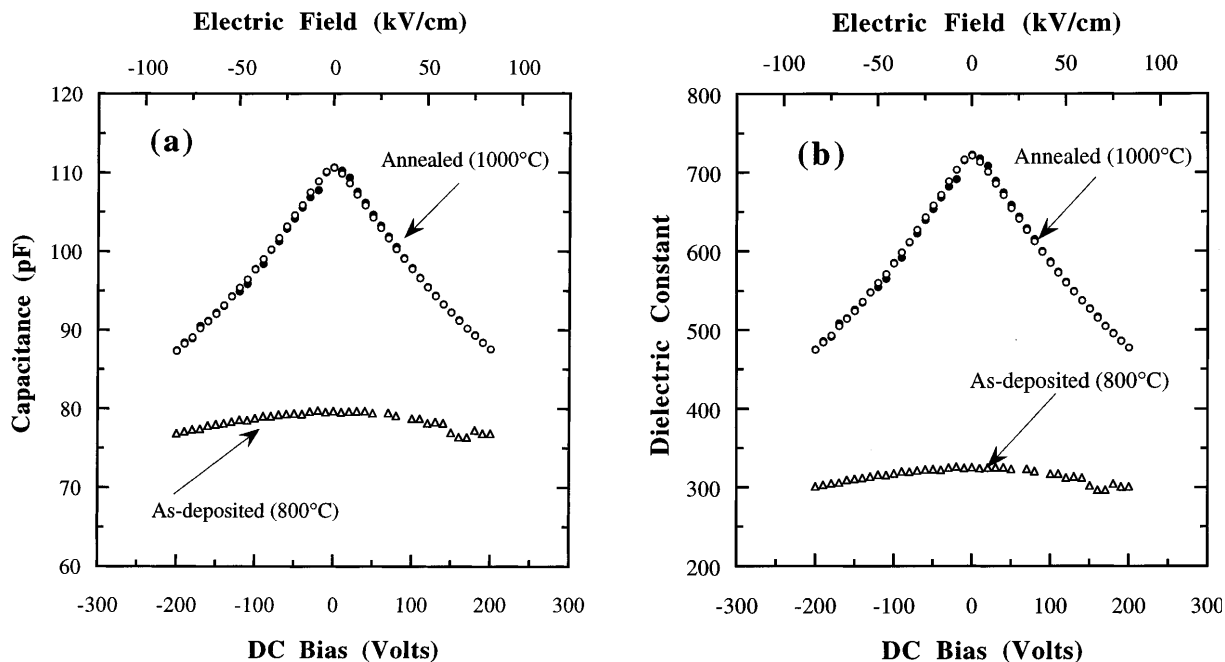
Fig. 7. (a) Capacitance and (b) dielectric constant of SrTiO$_3$ films as a function of dc bias. The films were deposited on LaAlO$_3$ substrates at 800°C with one film subsequently annealed at 1100°C for 5 h. The measurements were performed at 10 kHz and 77 K.

1000°C. Specifically, the % tunability of the ST films annealed at 1000°C was 21% compared to 3.5% for the as-deposited samples. The loss factor of the ST films annealed at 1000°C was 0.3–0.4% compared to 0.5% for the as-deposited film. Furthermore, the tuning curve of the annealed ST film shows smaller hysteresis than has been observed for ST films deposited by other techniques [17]. In addition, the in-plane permittivity of the ST film subjected to post-deposition annealing (1000°C) showed a dramatic increase as shown in Fig. 7b. Specifically, the in-plane dielectric constant of the ST films increased from a typical value of roughly 300 to a value of 730 (zero bias) after annealing; this is similar to the change exhibited by the annealed BaTiO$_3$ films. The exact reason for this abnormally large dielectric constant value has not yet been determined. However, there are at least two possible reasons for the increase in permittivity and improved tuning after post-deposition annealing: 1) an increase in grain size, and 2) a reduction in film stress.

AFM analysis revealed that the grain size of the post-deposition annealed ST film was 300–500 nm, which is 3–5 times larger than the grain size of the as deposited ST film [16]. It is well known that a larger grain size can improve the dielectric constant values

of perovskite materials [13–15], which can explain the increase in dielectric constant values of the ST films after post-deposition annealing. Furthermore, X-ray analysis revealed that the strain estimated from the shift in film lattice parameter from the bulk value is different for the two films. Specifically, the as-deposited ST films showed a 0.67% strain while the films annealed at 1000°C showed a much smaller value of 0.26%. This result indicates a difference in film stress which may have contributed to the difference in tuning behavior.

Finally, it should be noted that tuning values as high as 48% have been reported for SrTiO$_3$ films deposited by laser ablation [18]. However, those films had a larger loss factor than the sputter-deposited films reported in this study, 2.4% vs. 0.4%, respectively. Thus, considering the figure of merit (ratio of %tunability to loss factor), our sputtered SrTiO$_3$ films actually have a larger figure of merit (50 vs. 20).

Summary

Capacitance tuning of several ferroelectric thin films has been evaluated. It is found that PZT and PNZT

exhibit high tunability with loss factors of 1–3%. The high tunability may make these materials appropriate for certain varactor applications, but probably makes them inappropriate for microwave devices. In the case of $BaTiO_3$ and $SrTiO_3$ films, post-deposition annealing at elevated temperature (1000–1100°C) is beneficial for both capacitance tuning and loss factor, so that the figure of merit is substantially improved. The annealing treatment lead to increased grain size and reduced film stress, which are presumably responsible for the improved dielectric properties. It has also been shown that the dielectric constant of these ferroelectric materials can be calculated based on an analytical model that has been previously developed. This expression permits the tunability of the dielectric constant to be reported, rather than just the capacitance as has typically been done before.

Acknowledgments

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