

# Dielectric and tunable properties of K-doped $\text{Ba}_{0.6}\text{Sr}_{0.4}\text{TiO}_3$ thin films fabricated by sol-gel method

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$\text{Ba}_{0.6}\text{Sr}_{0.4}\text{TiO}_3$  (BST) thin films doped by K (BSTK) from 1 to 20 mol % were fabricated by sol-gel method on a Pt/TiO<sub>2</sub>/SiO<sub>2</sub>/Si substrate. Thermal evolutionary process of the  $\text{Ba}_{0.6}\text{Sr}_{0.4}\text{TiO}_3$  and  $(\text{Ba}_{0.6}\text{Sr}_{0.4})_{0.95}\text{K}_{0.05}\text{TiO}_3$  dry gel was carried out by thermogravimetry and differential thermal analysis system. The structure and surface morphology of BST thin films were investigated as functions of K concentration by x-ray diffraction and atomic force microscopy. The dielectric measurements were conducted on metal-insulator-metal capacitors at the frequency from 100 Hz to 1 MHz. The K concentration in BST thin films has a strong influence on the material properties including surface morphology and dielectric and tunable properties. The grain size, surface root-mean-square roughness, dielectric constant, dissipation factor, and tunability all increased with increasing K content up to 7.5 mol % and then decreased with increasing K content from 7.5 to 20 mol % in the BSTK thin films at 1 MHz. The effects of K doping on the microstructure and dielectric and tunable properties of  $\text{Ba}_{0.6}\text{Sr}_{0.4}\text{TiO}_3$  thin films were analyzed. The  $(\text{Ba}_{0.6}\text{Sr}_{0.4})_{0.925}\text{K}_{0.075}\text{TiO}_3$  thin film exhibited the highest dielectric constant of 1040 and the largest tunability of 73.6%. The dielectric constant, dielectric loss, and tunability of K-doped BST thin films with the optimal K content of 5 mol % were about 971, 0.023, and 69.96%, respectively. In addition, its figure of merit showed a maximum value of approximately 28.52. © 2006 American Institute of Physics. [DOI: 10.1063/1.2189976]

## I. INTRODUCTION

Barium-strontium titanate,  $\text{Ba}_{1-x}\text{Sr}_x\text{TiO}_3$  (BST), has been investigated with considerable interest as a dielectric material for future-generation dynamic random access memories (DRAM),<sup>1,2</sup> infrared detectors based on uncooled focal plane arrays (UFPA),<sup>3,4</sup> and electrically tunable microwave devices, such as voltage tunable phase shifters, oscillators, delay lines, and parametric amplifiers.<sup>5-7</sup> In the tunable microwave devices, it is desirable to have a high dielectric tunability under a certain electric field range and a low dielectric loss. To improve the material performance for the use of tunable devices many efforts have been tried in the doping. Some dopants including  $\text{Mg}^{2+}$ ,  $\text{Ni}^{2+}$ ,  $\text{Fe}^{2+}$ ,  $\text{Fe}^{3+}$ ,  $\text{Mn}^{2+}$ ,  $\text{Mn}^{3+}$ ,  $\text{Co}^{2+}$ ,  $\text{Co}^{3+}$ ,  $\text{Al}^{3+}$ ,  $\text{Cr}^{3+}$ , and  $\text{Bi}^{3+}$ , which can occupy the B sites of the  $\text{ABO}_3$  perovskite structure and behave as electron acceptors, have been known to lower dielectric loss.<sup>8-17</sup> However, few papers reported the effect of dopants which can occupy the A sites of perovskite structure on the properties of BST, except some rare-earth elements, such as  $\text{La}^{3+}$  and  $\text{Er}^{3+}$ .<sup>18,19</sup> In fact,  $\text{K}^+$  that nearly has the same radius as the  $\text{Ba}^{2+}$  can substitute  $\text{Ba}^{2+}$  or  $\text{Sr}^{2+}$  and can act as

acceptor in the A sites of BST perovskite structure. Nevertheless, the effect of K doping on the material properties of BST thin films has not been reported yet.

At present, BST thin films have been prepared by a variety of techniques involving pulsed laser deposition (PLD),<sup>11,14-16</sup> rf-magnetron sputtering,<sup>20</sup> metal-organic solution deposition (MOSD),<sup>8-10,19</sup> and sol-gel process.<sup>13,17,18,21</sup> Among these techniques, the sol-gel technique offers significant advantages over others in such as purity, better homogeneity, precision stoichiometry control, substrates with non-planar shapes, large area deposition, and ease of doping. Especially, its lower processing temperature (not more than 750 °C) avoids the problems of volatilization of K element by ordinary sintering. So it is desirable to prepare K-doped BST thin films.

In the present work,  $(\text{Ba}_{0.6}\text{Sr}_{0.4})_{1-x}\text{K}_x\text{TiO}_3$  thin films were prepared by sol-gel method on Pt/TiO<sub>2</sub>/SiO<sub>2</sub>/Si in order to investigate the influence of K doping (A site of the  $\text{ABO}_3$  perovskite structure) on the microstructure, surface morphology, and dielectric properties of BST thin films.

## II. EXPERIMENT

Thin films of  $(\text{Ba}_{0.6}\text{Sr}_{0.4})_{1-x}\text{K}_x\text{TiO}_3$  (BSTK<sub>x</sub>) ( $x = 0-20$ ) were prepared using the sol-gel method. BSTK precursor solutions were synthesized using strontium acetate semihydrate [ $\text{Sr}(\text{CH}_3\text{COO})_2 \cdot 1/2\text{H}_2\text{O}$ ], barium acetate [ $\text{Ba}(\text{CH}_3\text{COO})_2$ ], kalium acetate, and titanium tetra-

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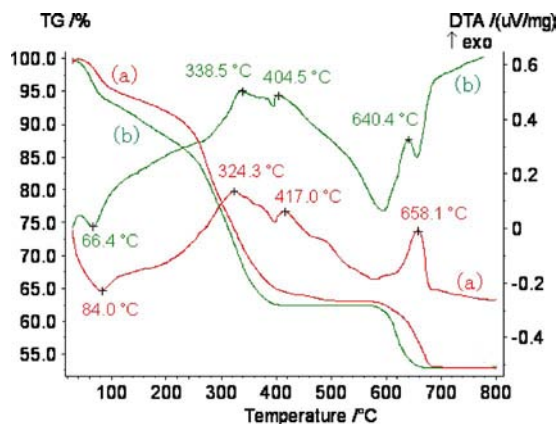


FIG. 1. (Color online) TG and DTA curves of (a)  $\text{Ba}_{0.6}\text{Sr}_{0.4}\text{TiO}_3$  and (b)  $(\text{Ba}_{0.6}\text{Sr}_{0.4})_{0.95}\text{K}_{0.05}\text{TiO}_3$  dry gels.

*n*-butoxide  $[\text{Ti}(\text{OC}_4\text{H}_9)_4]$  as source materials. Glacial acetic acid and 2-methoxyethanol were selected as solvents. Strontium acetate, barium acetate, and kalium acetate were dissolved into a hot acetic acid, and titanium tetra-*n*-butoxide mixed with 2-methoxyethanol was added into the above solution with constant stirring. Then ethylene glycol was added into the mixture to avoid film cracking. *pH* value was adjusted using glacial acetic acid to remain about 4. The solutions were mixed and stirred at a temperature of about 65 °C for 0.5 h to obtain clear solution. The concentration of the final solution was adjusted to 0.5M. The precursor solution was coated on the Pt/TiO<sub>2</sub>/SiO<sub>2</sub>/Si substrates. Spin coating was performed using a spinner rotated at a rate of 4000 rpm for 25 s. The as-deposited BST thin films were heated by a three-step heating procedure: first heated at a low temperature of 100 °C for 15 min to dry the gel, then pyrolyzed at 400 °C in air for 60 min, and crystallized at about 750 °C in O<sub>2</sub> atmosphere for 60 min.

Thermogravimetry (TG) and differential thermal analysis (DTA) (TG/DTA NETZSCH STA449C) analyses were carried out in air with a heating rate of 5 °C/min by a NETZSCH STA449C thermal analysis system. The structural and dielectric properties of BSTK thin films were characterized by various techniques. X-ray diffraction (XRD) profiles were obtained using a BSX3200 diffractometer with Cu *K*α radiation to determine the phase formation, the crystallinity, and the orientation of the films. The surface morphologies of the films were analyzed by atomic force microscope (AFM) (SPM-9500J3). Dielectric measurements were carried out using the metal-insulator-metal (MIM) capacitor configuration. Cu top electrode with 0.3 mm diameter was deposited on the film by direct current sputtering. Dielectric constant, tunability, and loss were measured using an Agilent 4294A precision impedance analyzer.

### III. RESULTS AND DISCUSSION

The thermal analysis results of the  $\text{Ba}_{0.6}\text{Sr}_{0.4}\text{TiO}_3$  and  $(\text{Ba}_{0.6}\text{Sr}_{0.4})_{0.95}\text{K}_{0.05}\text{TiO}_3$  dry gels at a heating rate of 5 °C/min are shown in Fig. 1. For the TG and DTA curves of the  $\text{Ba}_{0.6}\text{Sr}_{0.4}\text{TiO}_3$  dry gel, there is an endothermic peak at near 84 °C and there are three exothermic peaks at 324.3, 417.0, and 658.1 °C in the DTA curve, respectively. The

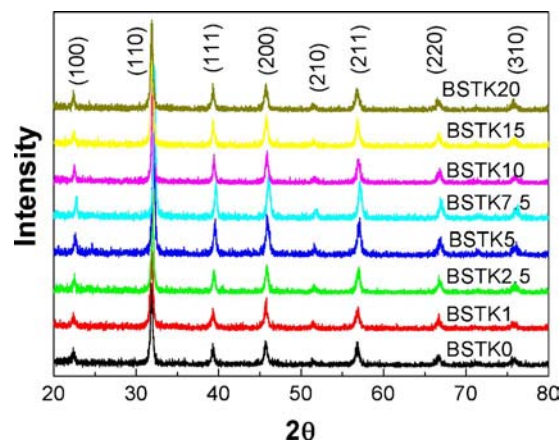


FIG. 2. (Color online) X-ray diffraction patterns of undoped and K-doped BST powders calcined at 750 °C.

endothermic peak at 84 °C in the DTA curve mostly results from the rapid volatilization of water. The exothermic peaks at 324.3 and 417.0 °C are related to the decomposition of carboxylate-alkoxide precursors. The end exothermic peak at 658.1 °C is associated with the formation of perovskite BST phase. At the same time, a significant weight loss is observed below 400 °C due to the evaporation of water and decomposition of organic compound in the TGA curve. With increasing temperature, a flat region is then followed in up to 600 °C. With further increasing temperature, a step is found in the TGA curve in a temperature range of 600–680 °C, accompanied by the exothermic peak of 658.1 °C in the DTA curve. For the  $(\text{Ba}_{0.6}\text{Sr}_{0.4})_{0.95}\text{K}_{0.05}\text{TiO}_3$  dry gel, its thermal analysis figures present a similar tendency to that of the  $\text{Ba}_{0.6}\text{Sr}_{0.4}\text{TiO}_3$  dry gel. However, its step in the TGA curve moves toward lower temperature and the end exothermic peak associated with the formation of perovskite phase lies at 640.4 °C which is about 20 °C lower than that of the  $\text{Ba}_{0.6}\text{Sr}_{0.4}\text{TiO}_3$  dry gel. It demonstrates that K-doped BST can reduce the formative temperature of BST perovskite phase and promote the crystallization of BST.

Figure 2 shows XRD patterns of the  $(\text{Ba}_{0.6}\text{Sr}_{0.4})_{1-x}\text{K}_x\text{TiO}_3$  (BSTK $x$ ) ( $x=0-20$ ) powders calcined at 750 °C. It indicates that the crystallization structures are cubic phase and no second phase appears for all the samples even with a 20 mol % K in the BST powder. It is implied that the dopants have entered the unit cell maintaining the perovskite structure of the solid solution. The increased K-doping ratio from 0% up to 7.5% leads to the increase in intensity of diffraction peaks and slightly shifts the diffraction peak position toward higher diffraction angles, which demonstrates that K doping promotes crystallization of BST and increases the film grain size with increasing K content up to 7.5 mol % as determined by the Scherrer formula.<sup>22</sup>

The microstructure, such as grain size and surface roughness, is one of the key parameters determining the dielectric properties. The average grain size and the surface root-mean-square (rms) roughness as functions of K content are shown in Fig. 3. The grain sizes are estimated from the AFM micrographs using the linear intercept method. The AFM images (Fig. 3) show all films to be well crystallized,

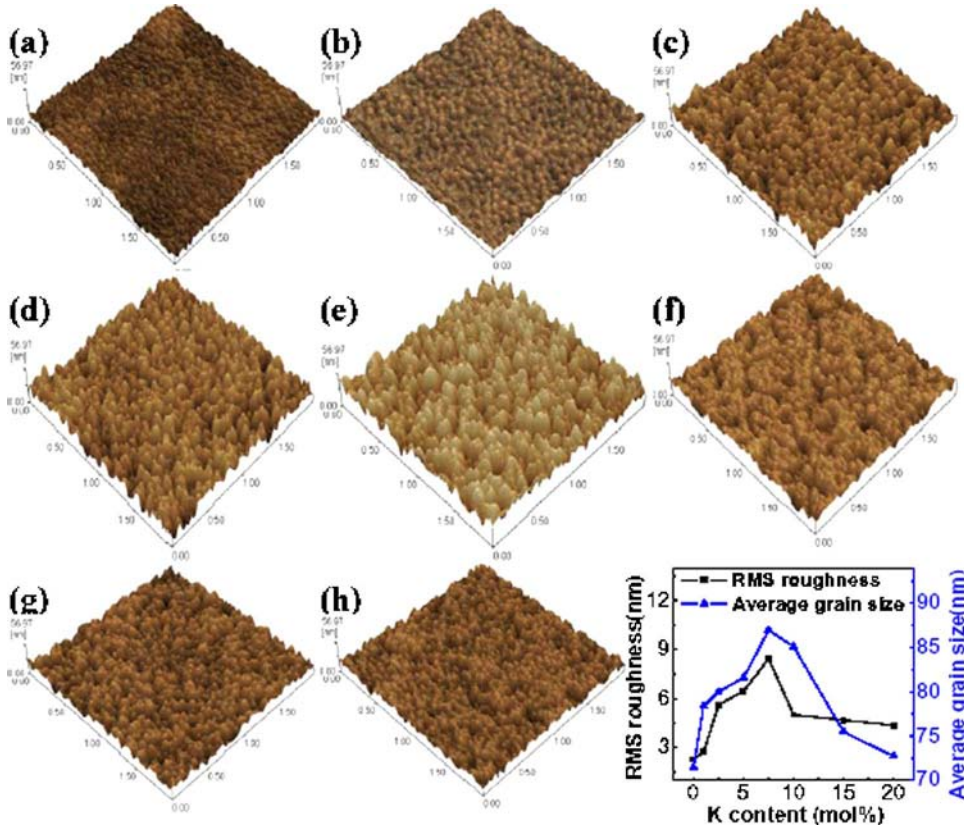


FIG. 3. (Color online) AFM micrographs of the 750 °C annealed (a) undoped, (b) 1 mol %, (c) 2.5 mol %, (d) 5 mol %, (e) 7.5 mol %, (f) 10 mol %, (g) 15 mol %, and (h) 20 mol % K-doped BST thin films and the rms roughness and average grain size of BST thin films as functions of K content.

crack-free, and with a dense microstructure. It was found that the grain size and the surface rms roughness of BSTK thin films were strongly influenced by the K content. The grain size and rms roughness both increased with increasing K content up to 7.5 mol % and then decreased with increasing K content from 7.5% to 20% in the BSTK thin films. This result is in a good agreement with the XRD measurements, as shown in Fig. 2. The higher surface roughness of BST films is attributed to better crystallization and larger grain size.<sup>23</sup> The doping of K ions leads to different grain sizes and surface roughnesses, which is likely to result in a different dielectric behavior. This concept can be further confirmed by the correlative dielectric measurement of thin films.

The room temperature dielectric permittivity and dielectric loss of the  $(\text{Ba}_{0.6}\text{Sr}_{0.4})_{1-x}\text{K}_x\text{TiO}_3$  as functions of frequency for different  $x$  (K content) are given in Fig. 4. These results were obtained using an oscillation voltage of 0.5 V and a frequency range of 100 Hz–1 MHz. As shown in the

figure, the dielectric constant rather slowly decreases with frequency up to 1 MHz. On the other hand, the dielectric loss of the films shows small dispersion at higher frequency ranges and the degree of dispersion is directly related to the surface roughness of the thin films as shown in Fig. 3. The  $(\text{Ba}_{0.6}\text{Sr}_{0.4})_{0.925}\text{K}_{0.075}\text{TiO}_3$  thin film has the highest surface roughness and also has the largest degree of dielectric dispersion. The phenomenon of dispersion of dielectric loss may be explained by the presence of interfacial layers, such as surface pyrochlore phases or electrode/film interface in the thin films.<sup>24–26</sup> At 1 MHz, the dielectric constant and dielectric loss of BSTK thin films as functions of K content are as shown in Fig. 5. The dielectric constant and dielectric loss increased with increasing K content from 1% to 7.5% and then decreased with increasing K content from 7.5% to 20% in the BST thin films. Actually, as the K content increases up to 7.5 mol %, the K-doped BST thin film reaches the highest dielectric constant of 1040. At the same time, the dielectric

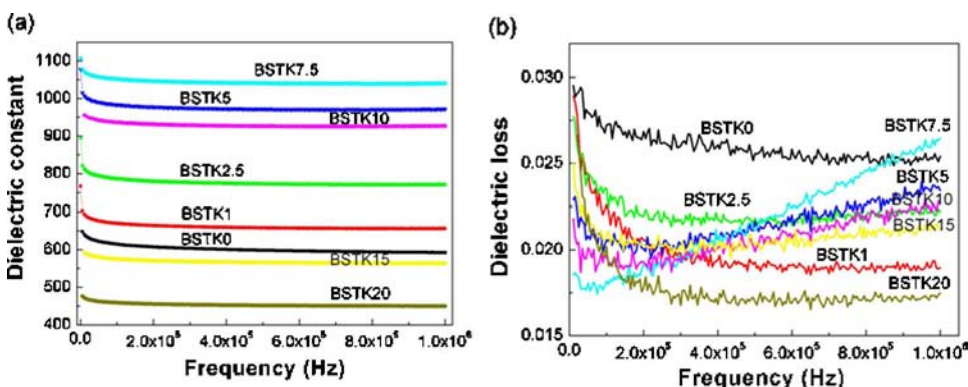


FIG. 4. (Color online) Dielectric constant and dielectric loss of the  $(\text{Ba}_{0.6}\text{Sr}_{0.4})_{1-x}\text{K}_x\text{TiO}_3$  ( $x=0-20$ ) thin films as functions of frequency at room temperature.



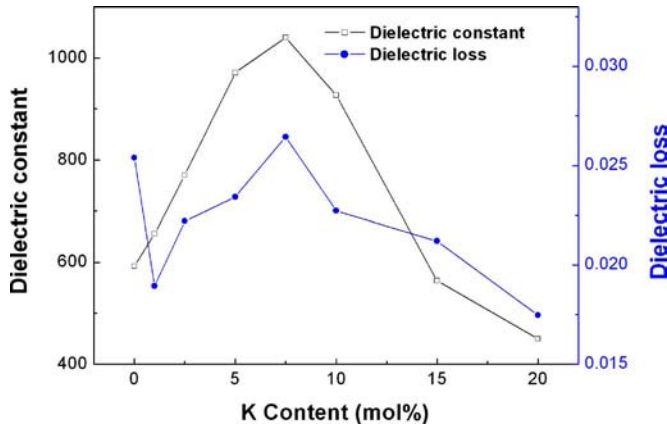


FIG. 5. (Color online) Dielectric constant and dielectric loss of BSTK thin films as functions of K content at 1 MHz.

loss of all K-doped BST thin films was lower than that of the undoped BST thin film except for the doped BST thin film with K content of 7.5 mol %, and the smallest dielectric loss is 0.0174 for the  $(\text{Ba}_{0.6}\text{Sr}_{0.4})_{0.99}\text{K}_{0.01}\text{TiO}_3$  thin film.

Such improvement in dielectric constant of BSTK thin films with increasing K content from 0 to 7.5 mol % is believed to be due to the effect of increased grain size. It is well known that the value of the dielectric constant of ferroelectric thin film is strongly affected by microstructure, grain structure, and the grain size. As for the last factor, the larger grain size usually results in larger polarization and therefore a higher value of dielectric constant.<sup>17</sup> After that, the decrease in dielectric constant of BSTK thin films with increasing K content from 10 to 20 mol % is attributed to the decrease in grain size.

At the same time, the dielectric loss has the similar change tendency to the dielectric constant with the increasing of K content, which may be the result of the corporate effect of acceptor doping and surface roughness. On one hand, acceptor-type dopants can prevent the reduction of  $\text{Ti}^{4+}$  to  $\text{Ti}^{3+}$  by neutralizing the donor action of the oxygen vacancies. Because the electrons resulting from the generation of oxygen vacancy can hop between different titanium ions and provide a mechanism for dielectric losses, the compensation for oxygen vacancy with the correct amount of acceptor dopant should, in theory, help to lower the loss tangent.<sup>27</sup> On the other hand, the surface roughness of the

thin film rapidly increases with increasing K content up to 7.5 mol % and an interface layer may appear between the top electrode and this rough surface of the thin film. Cole *et al.* pointed out that the dispersion of dielectric loss may have resulted from the interface layer.<sup>25,26</sup> As shown in Fig. 4, the degree of dispersion increased with increasing surface roughness of the thin films. So the rough surface of the thin film may result in a higher dielectric loss. It was found that the effect of surface roughness on the dielectric loss may be stronger than that of acceptor doping while the surface is very rough. Therefore, the BSTK thin film doped with 7.5 mol % K has the highest dielectric loss among all samples for its largest surface roughness. However, for the  $(\text{Ba}_{0.6}\text{Sr}_{0.4})_{0.99}\text{K}_{0.01}\text{TiO}_3$  thin film, its surface is relatively flat and the effect of acceptor doping on the dielectric loss plays a primary role. So its dielectric loss is lower than that of the undoped BST thin film.

The potential of the BST thin film to be used in voltage tunable devices depends on the ability to change the dielectric constant by means of an applied electric field. Figure 6 shows the dielectric constant ( $\epsilon$ ) and dielectric loss ( $\tan \delta$ ) of  $(\text{Ba}_{0.6}\text{Sr}_{0.4})_{1-x}\text{K}_x\text{TiO}_3$  films as functions of electrical field with different  $x$ . The curves were measured at room temperature and 1 MHz. As shown in Fig. 6, the relative dielectric constant and dielectric loss of the BSTK thin films nonlinearly decrease with increasing applied dc field. The nonlinearity of permittivity with electric field at the paraelectric phase of the material results from anharmonic interaction of titanium ions in perovskite structure.<sup>28</sup> The anharmonic interaction can be visualized as an inelastic feature of the covalent bond constructed by  $\text{Ti}^{4+}$  and  $\text{O}^{2-}$ . Such a nonlinearity could be explained by the phenomenological theory of Devonshire.<sup>30</sup> Based on the framework, two phenomenological equations were proposed by Johnson<sup>29</sup> to describe the dielectric constant and the intrinsic loss under the dc field which can be represented as

$$\epsilon_r/\epsilon_{r0} = 1/(1 + \alpha\epsilon_{r0}^3 E^2)^{1/3},$$

$$\tan \delta/\tan \delta_0 = 1/(1 + \alpha \tan \delta_0^3 E^2)^{1/3},$$

where  $\epsilon_{r0}$  and  $\epsilon_r$  are the permittivities under zero electric field and under electric field  $E$ , respectively.  $\alpha$  is the anharmonic coefficient, and  $\tan \delta_0$  and  $\tan \delta$  are the dielectric losses under zero electric field and under electric field  $E$ .

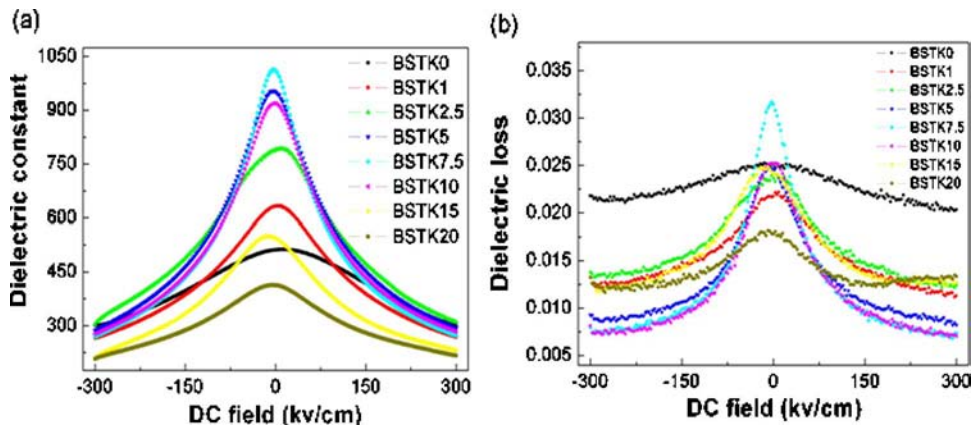


FIG. 6. (Color online) Variation of the dielectric constant and loss of BSTK thin films with applied dc electric field at 1 MHz.

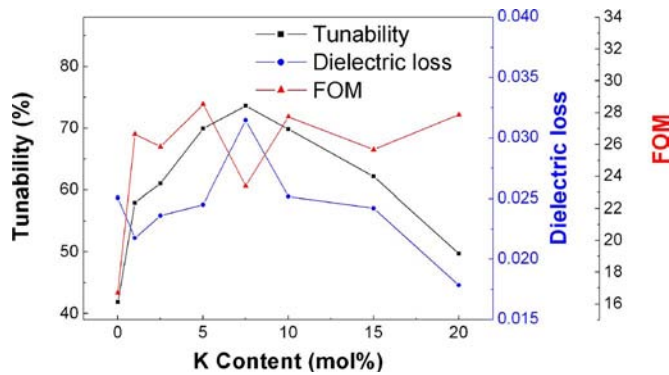


FIG. 7. (Color online) Tunability, dielectric loss, and the figure of merit (FOM) of BSTK thin films as functions of K content at 1 MHz.

Johnson's model shows clearly that the permittivity and dielectric loss of the thin films decrease with the increasing electric field.

The tunability, loss tangent ( $\tan \delta$ ), and figure of merit (FOM) of BSTK films as functions of K-dopant concentration are shown in Fig. 7. The tunability is defined as  $(\epsilon_{\max} - \epsilon_{\min}) / \epsilon_{\max}$ , where  $\epsilon_{\max}$  and  $\epsilon_{\min}$  are the maximum and minimum values of permittivity, respectively, measured at the zero electric field and 300 kV/cm electric field. The tunability of the BSTK thin films increases with increasing K concentration to 7.5% and then decreases after that. The dielectric loss of the BSTK thin films shows similar tendency as the behavior of the tunability and showed a minimum of 0.017 at the K concentration of 20 mol %, which may be related to its relatively flat surface and the effect of K acceptor doping. The highest tunability value found for BSTK films with K concentration of 7.5 mol % was approximately 73.6%. The increase in tunability is attributed to the increase in grain size.

It is noticed that the tunability of K-doped BST thin films obviously increased at K content no more than 7.5 mol % and the tunability is not less than that of the undoped BST thin film even at K content reaching to 20 mol %. At this point, it is different with the effect of other acceptors which occupy the B sites of the  $ABO_3$  perovskite structure on tunability. In early reports, the Curie temperature ( $T_C$ ) and tunabilities of BST thin films doped by  $Mg^{2+}$ ,<sup>8,31,32</sup>  $Co^{3+}$ ,<sup>33</sup> and  $Mn^{2+}$  (Refs. 34 and 35) are all decreased. The authors of Refs. 33–35 pointed that these acceptor ions occupying the  $Ti^{4+}$  ion sites result in oxygen vacancies and lead to a “break” of the cooperative vibration of the Ti–O chains and therefore shift the doped BST system to lower  $T_C$ . At the same time, it is known that the dielectric tunability increases near the  $T_C$ .<sup>36</sup> So the decreased tunability of BST thin films doped by these acceptor ions at room temperature was explained by the shifting of  $T_C$  to lower temperatures. For K-doped BST thin films,  $K^+$  ions occupying the A sites do not break the cooperative vibration of the Ti–O chains and do not reduce the anharmonic interaction of titanium ions in perovskite structure, which may be the reason of different tunabilities of K and other acceptors occupying the B sites of the  $ABO_3$  perovskite structure doped BST.

A tunable microwave circuit should take advantage of a high tunability together with a low loss factor ( $\tan \delta$ ). Figure

of merit is a frequently used parameter to characterize correlations between tunability and dielectric loss ( $\tan \delta$ ) and is defined as  $FOM = \text{tunability} / \tan \delta$ , which is desired to be as high as possible. The FOM value reflects the fact that a tunable microwave circuit cannot take full advantage of high tunability if the loss factor is too high. As shown in Fig. 7, the FOM values of undoped, 1%, 2.5%, 5%, 7.5%, 10%, 15%, and 20% K-doped BST films are 16.69, 26.64, 25.85, 28.52, 23.39, 27.74, 25.66, and 27.86, respectively. In the case of the 5% K-doped BST film, the tunability and loss are 69.96% and 0.024, respectively. Although the tunability becomes lower than that (73.6%) of the 7.5% K-doped BST film, the loss is lowered even more. As a result, the FOM has been improved. Considering the trade-offs between dielectric loss and tunability, the optimal doping content of K is 5%.

#### IV. CONCLUSION

In this study,  $(Ba_{0.6}Sr_{0.4})_{1-x}K_xTiO_3$  thin films have been fabricated on a Pt/ $TiO_2$ / $SiO_2$ /Si substrate by sol-gel method. The structural, surface morphological, and dielectric properties of BST thin films were investigated as functions of K concentration. It is found that the K concentration in BST thin films has a strong influence on the material properties including surface morphology and dielectric and tunable properties. The grain size, rms roughness, dielectric constant, dissipation factor, and tunability all increased with increasing K content up to 7.5 mol % and then decreased with increasing K content from 10 to 20 mol % in the BSTK thin films. Though the  $(Ba_{0.6}Sr_{0.4})_{0.925}K_{0.075}TiO_3$  thin film show the highest dielectric constant and tunability such as 1040 and 73.6%, respectively, the  $(Ba_{0.6}Sr_{0.4})_{0.95}K_{0.05}TiO_3$  thin film is the best choice for tunable device applications for its highest FOM value of 28.52.

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