Ceramics International xxx (xxxx) xxx–xxx

Contents lists available at ScienceDirect

Ceramics International

journal homepage: www.elsevier.com/locate/ceramint

# Effect of sputtering pressure on structural and dielectric tunable properties of $BaSn_{0.15}Ti_{0.85}O_3$ thin films grown by magnetron sputtering

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ARTICLE INFO

*Keywords:*
Ceramics
Tunability
Thin films
Dielectric

ABSTRACT

Lead−free ferroelectric $BaSn_{0.15}Ti_{0.85}O_3$ (BTS) thin films are grown on Pt-coated Si substrates by magnetron sputtering at 650 °C, the effect of sputtering pressure on the microstructural, surface morphological, dielectric properties and leakage characteristic is systematically investigated. XRD analysis shows the crystallinity of BTS thin films with perovskite structure can be improved by appropriate control of the sputtering pressure. The surface morphology analyses reveal that grain size and roughness can be affected by sputtering pressure. The BTS thin films prepared at sputtering pressure of 3.0 Pa exhibit a low dispersion parameter of 0.006, a medium dielectric constant of ~357, a high dielectric tunability of 65.7%@ 400 kV/cm and a low loss tangent of 0.0084 @ 400 kV/cm. Calculation of figure of merit (FOM) displays a high value of 84.1, and the measurement of leak current shows a very low value of $4.39 \times 10^{-7}$ A/cm$^2$ at 400 kV/cm. The results indicate that BTS thin film deposited sputtering pressure of 3.0 Pa is an excellent candidate for electrically steerable applications

## 1. Introduction

Ferroelectric oxide tunable thin films have been investigated for their applications in tunable capacitors, phase shifters, resonators, antennas, tunable filters, electrical controlled oscillators, and so on, owing to their excellent nonlinear dielectric response [1–4]. Currently, lead free barium zirconate titanate ($BaZr_xTi_{1-x}O_3$, BZT) [5,6] and barium strontium titanate ($Ba_xSr_{1-x}TiO_3$, BST) thin films [7,8] are being widely studied due to their large dielectric tunability (> 50%), high dielectric constants (> 300), and ease of wide−area deposition by simple techniques. However, the high dielectric loss (~0.02) strongly inhibits their further practical application in tunable devices [9,10]. Alternative tunable thin films are therefore of great scientific and technological interest.

Barium tin titanium ($BaSn_{0.15}Ti_{0.85}O_3$, BTS) is a kind of lead−free ferroelectric material with perovskite structure [11,12]. Due to the fact that the BTS thin films exhibit the large tunability and relatively low loss tangent, it has attracted the researchers' attention recently [12–14]. Magnetron sputtering [12], sol−gel [13,14] and pulsed laser deposition [15,16] technologies have been utilized for growing BTS thin films. Among these techniques, the BTS thin films grown by pulsed laser deposition show the highest figure of merit, and the lowest loss tangent of ~0.01 @ 100 kHz under an applied bias field of 400 kV/cm and at room temperature [16]. However, the pulsed laser deposition technique cannot be used for the deposition of large−area thin film, and the spatial nonuniformity of plasma results in the nonuniformity in the thickness of thin films [17]. This has been hampered pulsed laser deposition technique further developments and applications in the industry. The magnetron sputtering is a kind of technique for the deposition of large−area thin film, and it is also the most common deposition technique for the growth of thin films in industry. Also, the surface of the thin films deposited by magnetron sputtering is very smooth, which is advantageous for preparing the tunable devices [18,19].

Magnetron sputtering is a gaseous plasma−based coating process where a magnetically confined gaseous plasma is created near the surface of target. The surface of the target is eroded by high−energy Ar ions, and the liberated atoms travel through the vacuum environment and deposit onto a substrate to form a thin film [20,21]. Therefore, the investigation of the sputtering pressure dependence on dielectric properties of the BTS thin films is very decisive for the performance optimization. In this paper, the $BaSn_{0.15}Ti_{0.85}O_3$ (BTS) thin films with thickness of 260 nm were grown on $Pt/Ti/SiO_2/Si$ (Pt−Si) substrates at various sputtering pressures via using radio frequency (RF) magnetron sputtering. The BTS thin films grown at sputtering pressure of 3.0 Pa exhibit the largest figure of merit value (84.1), while the tunability and loss tangent are respectively 65.7% and 0.0084 @ 400 kV/cm. The influences of sputtering pressure on the microstructure and dielectric

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https://doi.org/10.1016/j.ceramint.2018.03.026
Received 26 January 2018; Received in revised form 28 February 2018; Accepted 4 March 2018

properties as well as leakage current density have been systematically investigated.

## 2. Experimental procedures

High purity $BaCO_3$ (99.99%), $SnO_2$ (99.99%) and $TiO_2$ (99.99%) were utilized as raw powders for the preparation of BTS target. The target was sintered in air at 1300 °C for 10 h. The diameter, thickness and density of the obtained BTS ceramic target were 51 mm, 2.9 mm and 95.3%, respectively. Before the deposition of BTS thin films, the distance from target to substrate was fixed at 8.0 cm and the base pressure of deposition chamber was evacuated to $3 \times 0^{-4}$ Pa. The thin films were deposited on Pt − Si substrates as bottom electrodes by RF magnetron sputtering, the power is 80 W, the substrate temperature is 650 °C. The $O_2$ (15%) and Ar (85%) mixture gas was used as the sputtering gas. The sputtering pressures are 0.3, 1.0, 3.0, and 10 Pa, respectively. The thickness of BTS thin films was fixed at 260 nm and estimated by control of deposition rate and time. At last, all the BTS thin films were in − situ annealed at 650 °C for 20 min in oxygen pressure of 1000 Pa.

The density of the BTS ceramic target was measured by density meter (Alfamirage SD − 200L). The thickness of BTS thin films was determined by Alpha–Step D–100 profilometer (KLA–Tencor, California, USA). X − ray diffraction (XRD) measurements of BTS thin films were carried out by a Bruker diffractometer (D8–Focus; Bruker AXS GmbH, Karlsruhe, German) with CuKα radiation. The surface morphologies and roughness were characterized by the atomic force microscopy (AFM, Bruker, Santa Barbara, CA, USA) using a Nanoscope Multimode 8. The cross-sectional photography was characterized by scanning electronic microscopy (SEM, JSM-6701F). For the electrical measurement, the Au top with a thickness of 200 nm and a diameter of 200 μm was deposited by thermal evaporation using a shadow metal mask. The loss tangent and dielectric constant were measured by an Agilent 4294 A LCR meter (Santa Clara, California, USA), the sample's temperature was varied by means of a high and low test chamber. The leakage current density was measured by ferroelectric test system (Radiant Technologies, Inc. of Albuquerque, New Mexico, USA.).

## 3. Results and discussion

Fig. 1a provides the XRD patterns of BTS thin films grown at various sputtering pressures. There are four growth directions that are (1 1 0), (1 1 1), (2 1 0) and (2 1 1) phases for all the BTS films, indicating the films exhibit single perovskite phase. As the increase in sputtering pressure, the intensity of main (1 1 0) peak in XRD patterns increases at first, reaches the maximum value at sputtering pressure of 3.0 Pa, and then decreases. This indicates that the BTS films grown at 3.0 Pa have the best crystal quality. The full − width at half − maximum (FWHM) values of (1 1 0) peak for the BTS films grown at different sputtering pressures are shown in Fig. 1b. The FWHM is 0.43, 0.32, 0.29 and 0.34 for the BTS films grown at sputtering pressures of 0.3, 1.0, 3.0 and 10.0 Pa, respectively. It follows the same trend as the peak intensity with increasing the sputtering pressure. The BTS thin films deposited at 3.0 Pa have the minimum FWHM value, which is generally related to an improvement of the crystallinity and an enlargement of the crystallite size. Since dielectric properties of thin films may be influenced by crystallite size [22,23], we can calculate the crystallite size by using Scherrer's formula as follow:

$$D = \frac{0.9\lambda}{\beta cos\theta} \quad (1)$$

where, $D$ denotes the average crystallite size, $\beta$ denotes the (1 1 0) peak width, $\theta$ is the Bragg diffraction angle, and λ denotes the wave length of Cu Kα radiation (1.5418 Å). According to the Eq. (1), the crystallite sizes are calculated are19.01, 25.56, 28.21 and 24.06 nm for the BTS films grown at sputtering pressures of 0.3, 1.0, 3.0 and 10.0 Pa,

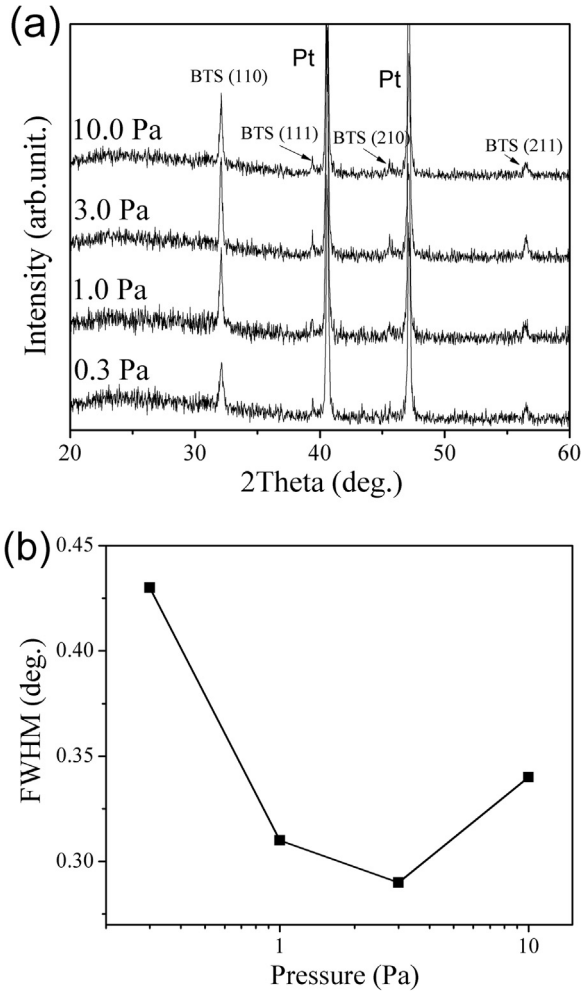


**Fig. 1.** (a) XRD patterns of BTS thin films with various sputtering pressure, and (b) sputtering pressure dependence of FWHM values of (1 1 0) peak for BTS thin films.

respectively. The change of the average crystallite size with sputtering pressure can be explained on the basis of Zener pinning effect (induced defects created in the system) [24]. The outward movement of grain boundaries is restricted by the retarding force generated by the defects like vacancies and dislocations. The BTS films grown at 0.3 Pa exhibit the poor crystal quality, and a large number of defects present in the films, which suppress the grain growth owing to the Zener pinning effect. With the sputtering pressure increases to 3.0 Pa, the defects in BTS thin films greatly decrease due to the improvement of crystallinity. Therefore, the restriction of growth of the crystallites is greatly weakened, and the crystallite size increases. With further increasing the sputtering pressure to 10.0 Pa, the crystallinity deteriorates again, the crystallite size decreases due to the Zener pinning effect caused by the increase of defects. In addition, the dislocation density can be used to estimate the amount of defects in a crystal, and it can be calculated as follow [25]:

$$\delta = 1/D^2 \quad (2)$$

where D denotes the crystallite size and δ defines the length of dislocation. As the dislocation density means the length of dislocation lines per unit volume of the crystal. Thus, the dislocation densities can be calculated by Eq. (2) are $2.77 \times 10^{15}$, $1.55 \times 10^{15}$, $1.26 \times 10^{15}$ and $1.73 \times 10^{15}$ lin/m$^2$ for the BTS films grown at sputtering pressures of 0.3, 1.0, 3.0 and 10.0 Pa, respectively. The BTS films grown at 3.0 Pa have the minimum dislocation density among all the BTS films, which indicates appropriate control of sputtering pressure can prevent defects generated in the BTS lattice.