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Aymen Selmi, O. Khaldi, Manuel Mascot, F. Jomni, Jean-Claude Carru. High-performance screen-printed Au/Ba<sub>0.85</sub>Sr<sub>0.15</sub>TiO<sub>3</sub>/Pt capacitors for tunable devices. *Journal of Alloys and Compounds*, 2021, 878, pp.160340. 10.1016/j.jallcom.2021.160340 . hal-04460074

**HAL Id: hal-04460074**

**<https://ulco.hal.science/hal-04460074>**

Submitted on 22 Jul 2024

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## High-performance screen-printed Au/Ba<sub>0.85</sub>Sr<sub>0.15</sub>TiO<sub>3</sub>/Pt capacitors for tunable devices

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### Abstract

Metal-Ferroelectric-Metal capacitor structure, employing the lead free ferroelectric Ba<sub>0.85</sub>Sr<sub>0.15</sub>TiO<sub>3</sub> thick films **are** successfully prepared by screen printing process on silicon platinized Pt/Ti/SiO<sub>2</sub>/Si substrates. The surface morphology and microstructure **are** analyzed by scanning electron microscope and X-ray diffraction. The thick film showed a well adhesion to the Pt/Ti/SiO<sub>2</sub>/Si substrate and uniform thickness of about ~20 μm. The frequency **dependence of** dielectric characteristics of BST thick films **are** systematically discussed and compared in a large temperature range. Ferroelectric characterizations, tunability and leakage current in the Au/BST(thick film)/Pt capacitor were also studied at room temperature by polarization cycles (P-V) , Capacitance (C-V) and current (I-V) measurements. The high tunability (81%) combined with the low dielectric loss **tangent** and low leakage current makes Ba<sub>0.85</sub>Sr<sub>0.15</sub>TiO<sub>3</sub> thick films one of the encouraging candidates for various modern **electronic** tunable devices.

**Key Words:** Lead-free BST thick film; Screen printing; Dielectric properties; ac conductivity; Tunability, leakage current

## 1. Introduction

The demand **for** tunable ferroelectric components in microelectronic field increased during the last years. Tunable microwave systems such as tunable varactors diodes, tunable oscillators, filters, FeRAM or tunable ferroelectric capacitors are becoming more and more important [1-7]. For this type of application the ferroelectric oxide materials are the most promising candidates. In terms of tunability, Barium Strontium Titanate ( $\text{Ba}_x\text{Sr}_{1-x}\text{TiO}_3$  or BST) is considered to be the archetype of electrically tunable materials [2, 8]. BST materials are known for their exceptional properties. So, aside from their high tunability and their environment friendly nature, ferroelectric BST can be prepared with **high** dielectric permittivity, small leakage-current and low dielectric losses [9, 10]. Thanks to all these unique **characteristics**, BST shows great promise for the conception of microwave tunable devices and are the more **most extensively** studied **material** to date because of the demand for lead-free ferroelectric materials [11]. For integration in microelectronic devices, BST has been investigated in various forms, including thin films with a thickness typically **less than**  $1\mu\text{m}$ , thick films with a few micrometers thick and ceramics with millimeter size. During these last years, the majority of the studies on BST **were** oriented towards the study of thin films and to a lesser extent towards ceramics but **there have been few studies published on** BST thick. Currently, the thick films of BST have received **some** attention; several **recent** research works **have** mentioned the importance of these materials in various applications of microelectronics [2, 12]. BST Thick films with thickness **ranging from**  $1\mu\text{m}$  **to**  $100\mu\text{m}$  can significantly replace BST ceramics in **case of** miniaturization **and can be** useful to fulfill the demands for multilayer assembles and circuit complexity and also can bridge the technological gap between BST ceramics and BST thin films [2]. To prepare BST thick films with a few micrometers thick**ess**, the preferred technique is the screen printing; this technique has **received** a lot of attention **due to its** exceptional characteristics including the good quality of the deposited films, the potential benefits of lower fabrication costs compared to other technique **and** the possibility to use different types of substrates whatever their geometry and morphology [13, 14]. This process **depends on** two essential parameters, the preparation of BST ink and heat treatments at high temperatures. The major frequent difficulty in depositing BST thick films by screen printing technology is the **quality of the** adhesion of the **film** to the substrate; this phase requires good ink quality, well studied thermal treatments and a good choice of substrate. **Presently**, Alumina  $\text{Al}_2\text{O}_3$  is **the** commonly used substrate in the deposition of BST thick films using screen printing process. However, some studies showed that deposited BST films are very reactive with  $\text{Al}_2\text{O}_3$  substrates during heat treatment at high

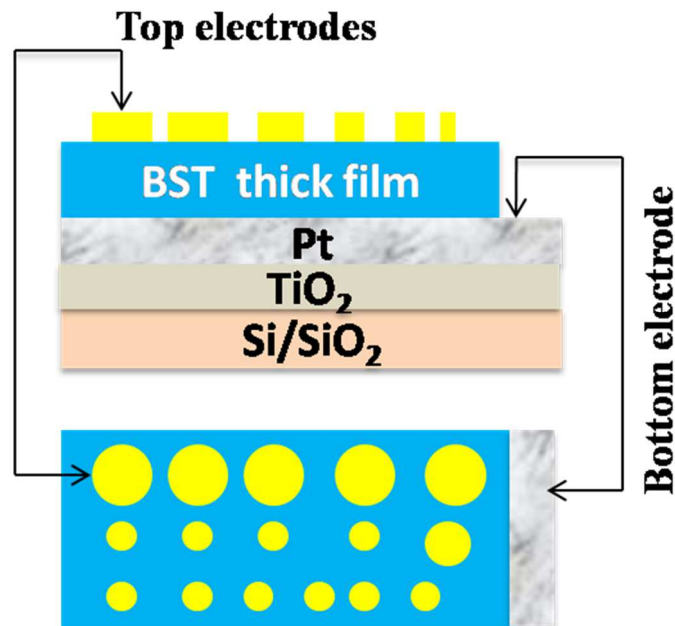
temperature [15]; this phenomenon strongly influences the performance of **the** elaborated thick films [16]. To the best of our knowledge, to date the deposition of BST in thick films form by **performed** screen printing process on Pt/Ti/SiO<sub>2</sub>/Si substrates has not yet been studied despite the technological importance of this type of substrate. **The usage of Pt/Ti/SiO<sub>2</sub>/Si substrates is known as for a high temperature stable metallization which withstands high temperatures (up to 750-800°C). In order to ensure good adhesion and low resistivity suitable for depositing ferroelectric oxide thin films, we have used Titanium as an adhesion layer. Then In consequence, the electrical performance of BST thick films on these platinized silicon substrates can could be higher than the ones deposited on alumina substrates.** In this view, we propose a detailed study on elaborations and characterizations of Ba<sub>0.85</sub>Sr<sub>0.15</sub>TiO<sub>3</sub> thick films on Pt/Ti/SiO<sub>2</sub>/Si substrates. **The composition BST85/15 is a good trade-off between having very good ferroelectric and dielectric properties for applications at temperatures close to room temperature.** In this work, high-quality Ba<sub>0.85</sub>Sr<sub>0.15</sub>TiO<sub>3</sub> thick films **of** 20 μm thick are successfully fabricated by screen-printing technique on Pt/Ti/SiO<sub>2</sub>/Si. Dielectric permittivity, dielectric loss and AC conductivity of Ba<sub>0.85</sub>Sr<sub>0.15</sub>TiO<sub>3</sub> thick films **are** investigated as a function of temperature, frequency and applied voltage. Ferroelectric properties and tunability were also analyzed using Polarization-Voltage (P-V) and Capacitance-Voltage (C-V) measurements. Moreover, the conduction mechanisms in Au/Ba<sub>0.85</sub>Sr<sub>0.15</sub>TiO<sub>3</sub>/Pt thick capacitors **are investigated** using current-voltage (J-V) measurements. It is shown that our ferroelectric Ba<sub>0.85</sub>Sr<sub>0.15</sub>TiO<sub>3</sub> thick films present high performance and are therefore encouraging candidates in the areas of thick film technology.

## 2. Experimental process

### 2.1 The different **steps** of thick films **préparation**

Ba<sub>0.85</sub>Sr<sub>0.15</sub>TiO<sub>3</sub> thick layer **is** prepared by employ a screen printing **device** (TIFLEX - HL570S). The powders of BST **are synthesized** by the solid-state procedure from Strontium oxide (SrO), Barium oxide (BaO), and Titanium oxide (TiO<sub>2</sub>) **from Alfa Aesar(99.5% purity)**. The ink for screen printing **is synthesized** by blending BST powders with organic vehicles. The organic vehicles **is** composed of {alpha-terpineol (C<sub>10</sub>H<sub>18</sub>O) **from Across Organics (97% purity)**, polyvinyl butyral (C<sub>8</sub>H<sub>14</sub>O<sub>2</sub>)<sub>n</sub>, **from Across Organics(98% purity)**, 2-(2-butoxyethoxy)-ethyl acetate (C<sub>10</sub>H<sub>20</sub>O<sub>4</sub>) **from Sigma Aldrich (99% purity)** and polyethylene glycol (C<sub>2</sub>H<sub>4</sub>O) **from Across Organics (99% purity)**. The ink **is** mixed and

milled by a roller to obtain a perfect uniform homogeneity. Initially polyvinyl butyral is dissolved in  $\alpha$ -terpineol at 60°C with vigorous stirring for 30 minutes. The  $\text{Ba}_{0.85}\text{Sr}_{0.15}\text{TiO}_3$  powder (previously dissolved in 2-(2-Butoxyethoxy) ethyl acetate) is added, along with polyethylene glycol to stabilize the mixture. One hour after, the ink of BST is heated at 100 °C for 1 hour to evaporate the solvent. Finally the ink is submitted to ultrasound and passed between the three-cylinder rollers in order to break the agglomerates. The different BST85/15 ink preparation procedures are highly detailed in our earlier published work [2]. The elaborated BST ink is deposited by screen printing on Pt/Ti/SiO<sub>2</sub>/Si substrates, the Pt (200nm thick) film serves as the bottom electrode for our capacitor which are then sintered for 1 hour at 100 °C. This operation was repeated for two times. Thereafter, the BST thick films were sintered in air for 2 h at 1100 °C to vaporize the organic compounds from the ink. Finally, to take electric measurements on the thick films, circular gold (top electrodes) with various diameters (0.1mm ~ 1mm) are deposited by thermal evaporation to obtain Metal-Ferroelectric-Metal (MFM) Au/BST/Pt capacitor structure. Fig. 1 shows the final structure of this kind of capacitor.



**Fig. 1.**

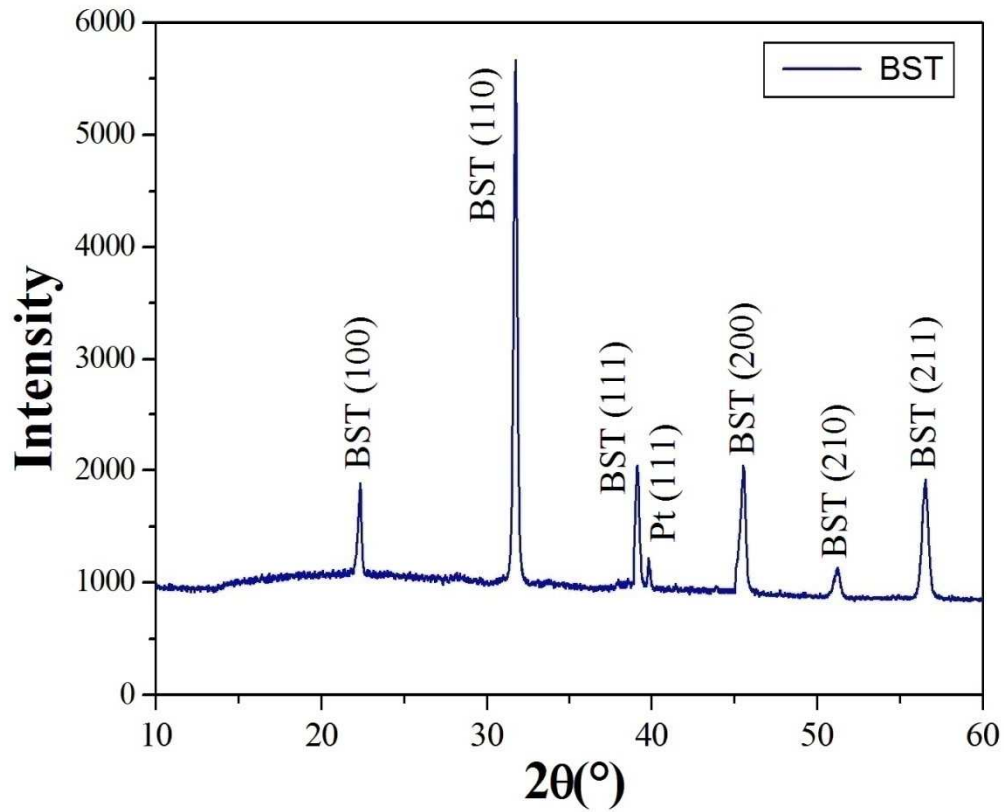
## 2.2 Characterizations techniques

The microstructure and the morphology of the prepared thick films were analysed by X-ray diffraction **with a Philips X-ray diffractometer (model PW1710) using CuK $\alpha$ 1 radiation ( $\lambda = 1,5406 \text{ \AA}$ )** and scanning Electron Microscopy **with a JEOLSEM (model JSM-5400)**. The dielectric measurements **are** carried out over a temperature range from room temperature (RT =25°C) up to 300°C and over a large frequency range ( $10^{-1}$  to  $10^6$  Hz) using a temperature regulator (Linkam TMS94) and an impedance analyzer (Solartron 1260 coupled to a dielectric interface 1296). Capacitance-Voltage (C-V) measurements on the Au/BST(thick film)/Pt capacitor **are** studied using an Agilent LCR meter E4980A. The hysteresis characteristics **are** analyzed by a modified Sawyer Tower circuit at 10 kHz. The leakage currents characterizations (J-V) **are** recorded using a Keithley electrometer 6517A **in order to** identify the conduction mechanisms involved in these structures.

## 3. Results and discussion

### 3.1 Structural and morphological characterization

**Fig. 2 presents** the XRD patterns for the BST thick film. It can be observed from the graph the existence of different diffraction peaks of BST in the ( $10^\circ$ - $60^\circ$ )  $2\theta$  range they are characteristics of BST (JCPDS File no. 39–1395) in this  $2\theta$  range. In addition, the calculated a-axis and c-axis lattice values are  $3.9942 \text{ \AA}$  and  $4.0171 \text{ \AA}$ , respectively. This suggests that the film is crystallized with the tetragonal structure phase at room temperature. The small Pt (111) phase detected in  $2\theta \approx 40^\circ$  is associated with the platinum bottom electrode.



**Fig. 2.**

Fig. 3a and 3b exhibit the surface and the cross-section vision of the BST thick film, respectively. It is clear that a no cracks on the BST thick film are observed. The cross-section images show a generally uniform thickness of about  $\sim 20\mu\text{m}$  and an excellent adhesion to the Pt/Ti/SiO<sub>2</sub>/Si substrate. The average grain size was measured to be  $0.4\ \mu\text{m}$ .

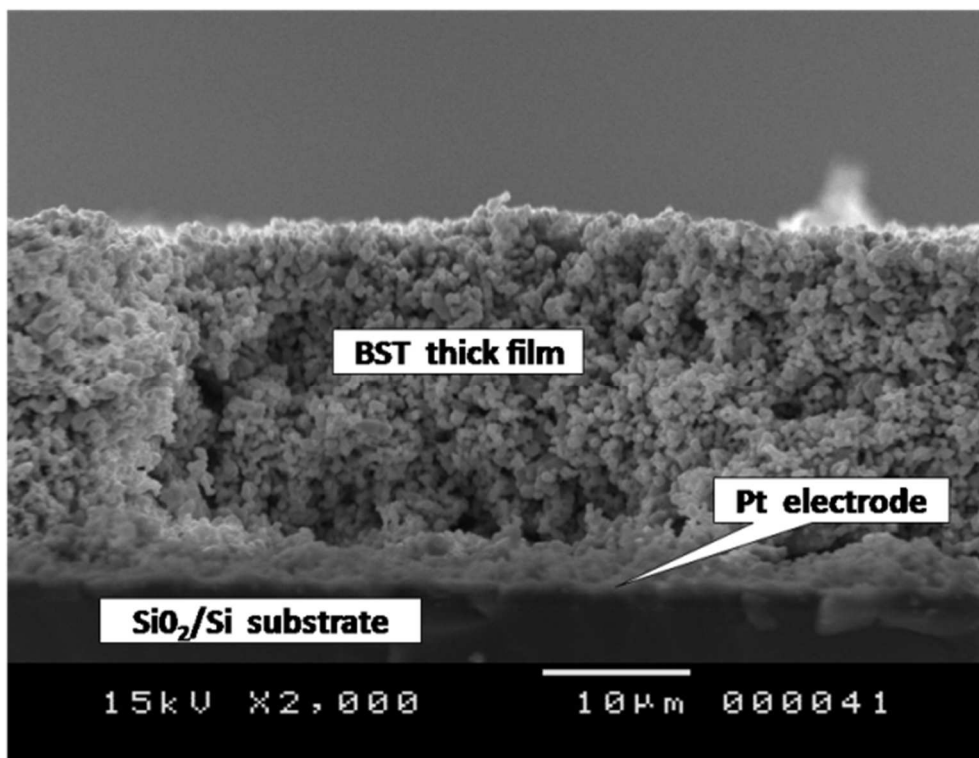
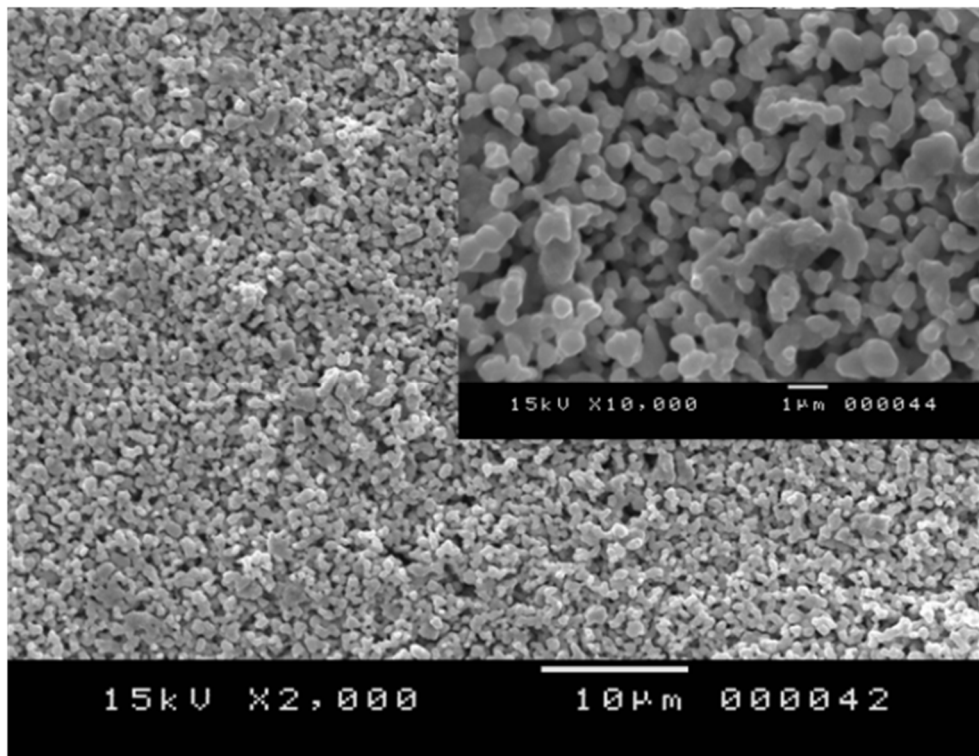


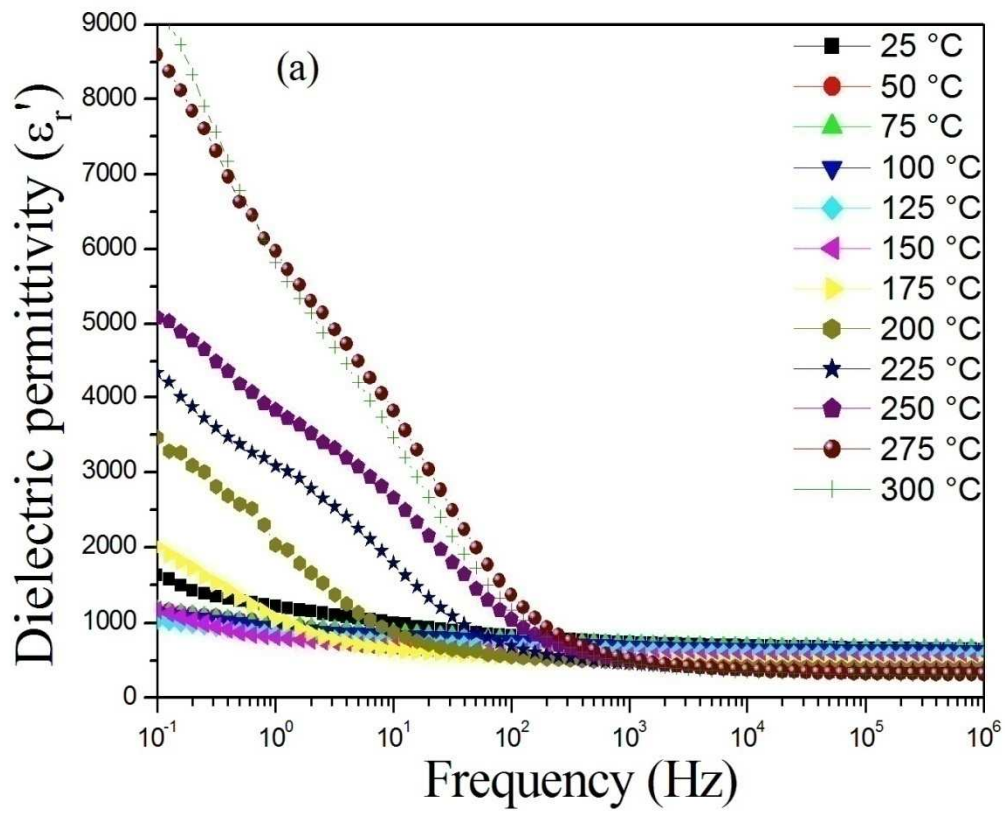
Fig. 3.

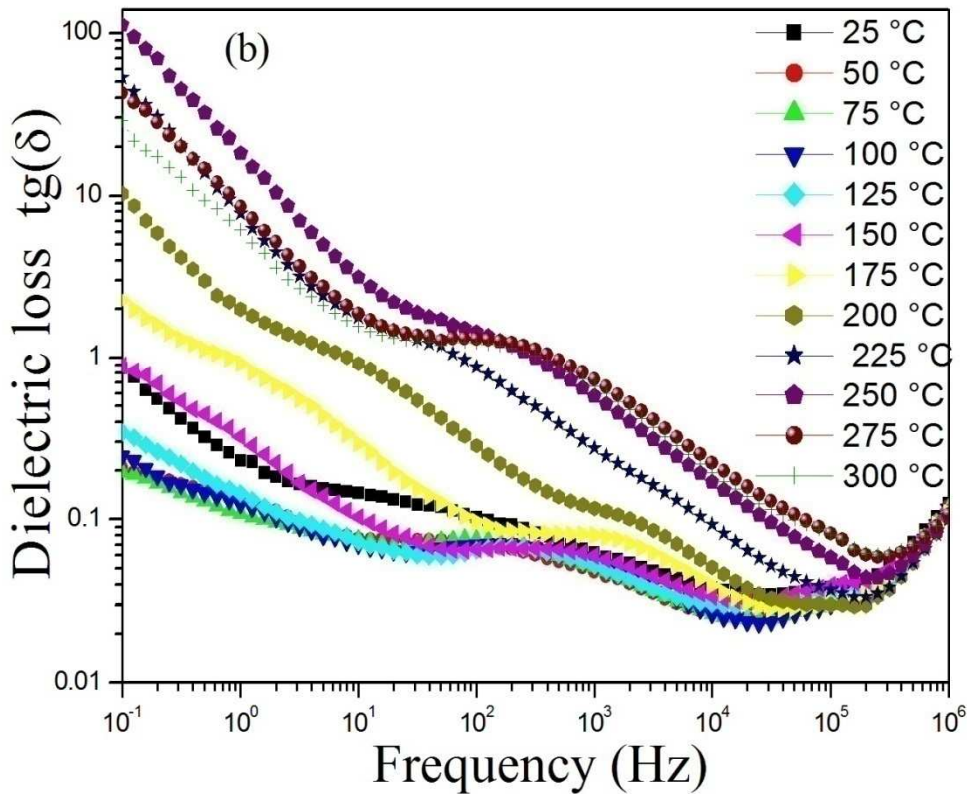


## 3.2 Dielectric Analysis

### 3.2.1. Permittivity and loss studies

Dielectric measurements of BST thick film **are** studied as a function of **frequency** ( $10^{-1}$  Hz to  $10^6$  Hz) and temperature ( $25^{\circ}\text{C}$ - $300^{\circ}\text{C}$ ). The variation of dielectric permittivity ( $\epsilon'_r$ ) is shown in [Fig. 4.a](#). It's clear that for all temperatures, the  $\epsilon'_r$  decrease **greatly** in the low-frequency and then **is** more or less **constant** at high frequency ( $F > 500\text{Hz}$ ) **which** constitute a dielectric dispersion phenomena. This behavior is a common feature for ferroelectric oxides materials [\[17\]](#) and can be due to the Maxwell-Wagner polarization [\[18, 19\]](#). It is interesting to note that the elaborated BST 85/15 thick films show a high dielectric permittivity compared to BST 85/15 thick films deposited on alumina substrates by screen-printing process [\[2\]](#) **and compared to** BST 85/15 thin films deposited by sol-gel on Pt/Ti/SiO<sub>2</sub>/Si substrates published in our earlier work [\[20\]](#). The frequency dependence of **the** dielectric loss factor ( $\text{tg}(\delta) = \epsilon''/\epsilon'$ ) is also measured in the same **ranges of temperature and frequency and presented** in [Figure 4b](#). The dielectric loss decrease with frequency displaying dispersion behavior due to Maxwell–Wagner interfacial polarization. The observed peak in  $\text{tg}(\delta)$  confirms the existence of a dielectric relaxation in **our** BST **films** and also **signifies** that the hopping of charge carriers plays an important role in **the** transport mechanism [\[21\]](#) .

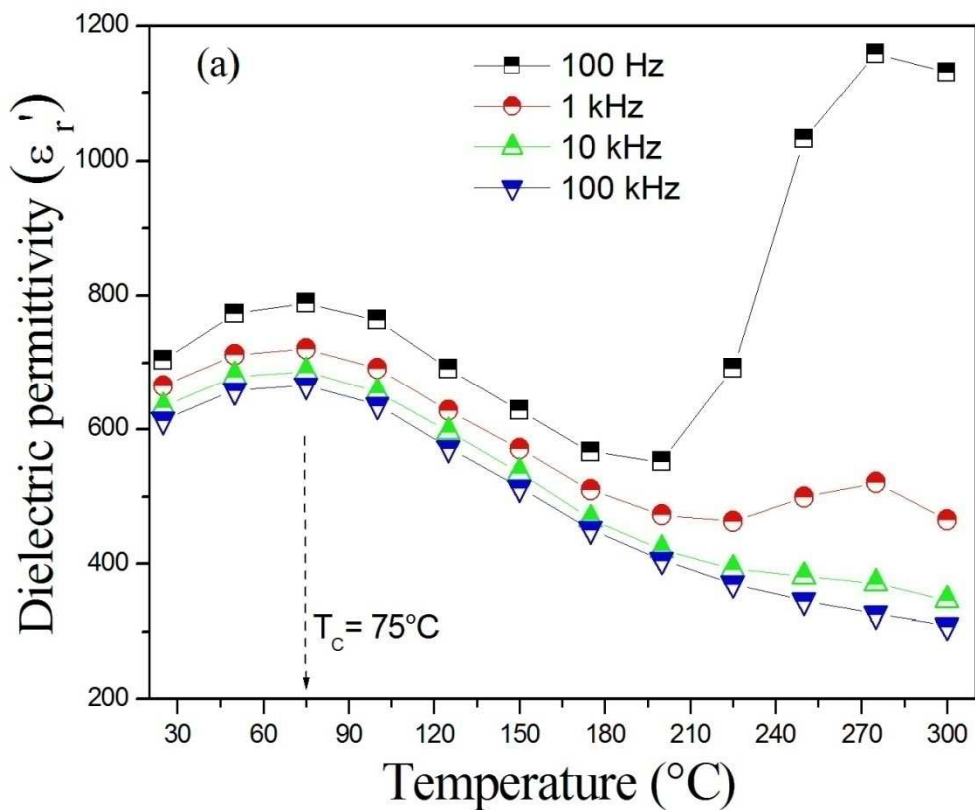




**Fig. 4.**

The variation of  $\epsilon_r'$  of BST thick film with temperature for selected frequencies is presented in the Fig. 5a. The curve shows two maxima in the dielectric permittivity. The first peak is at  $75^\circ\text{C} = T_c$  the Curie temperature which corresponds to a ferroelectric–paraelectric phase transition in the BST system and the second peak is observed clearly at high temperatures around  $270^\circ\text{C}$  for low frequencies under 10 kHz. The appearance of second peak at high temperatures in  $\epsilon_r'$  curves as a function of temperature is similar to that found in our earlier published work on sol-gel BST thin films [20], this behavior can be associated to the existence of nano/micropolar phases in the BST films in the paraelectric state. The obtained  $T_c$  value is different from some of the ones published on BST thick films deposited by screen printing process, for example X. Zhang et al;  $T_c = -2^\circ\text{C}$  for BST 60/40 [14], X. U. Qing et al;  $T_c = 10^\circ\text{C}$  for BST 60/40 [13] and B. Su et al;  $T_c = -30^\circ\text{C}$ ,  $2^\circ\text{C}$  and  $30^\circ\text{C}$  for BST 50/50, BST 60/40 and BST 70/30 respectively [22]. This difference

**is clearly** related to a difference of composition **since the temperature**  $T_C$  is very sensitive to the Sr and Ba ratio and **is** possibly associated to the substrate effect. The value of  $T_C$  found on our BST (85/15) thick films show that the material is in ferroelectric state at **room temperature**. This result is encouraging for several applications **such as** random access memories (RAM), radio-frequency identification (RFID) and ferroelectric capacitors, etc... The variation of **dielectric loss  $\tan(\delta)$**  depending on the temperature for different frequency (Fig. 5b) present low values **inferior to** 0.05 in a large temperature range, ranging from **25 °C** to 170°C. Above 170 °C the dielectric **loss** exhibits a remarkable increase, mainly due to **the** motion of oxygen vacancies. In fact, oxygen vacancies are thermally activated and **become** more mobile which **increases** the polarization [2]. The high values of the dielectric permittivity combined with the low loss values obtained **in** a wide temperature range makes the  $Ba_{0.85}Sr_{0.15}TiO_3$  thick films **a potential** candidates for many modern microelectronic applications.



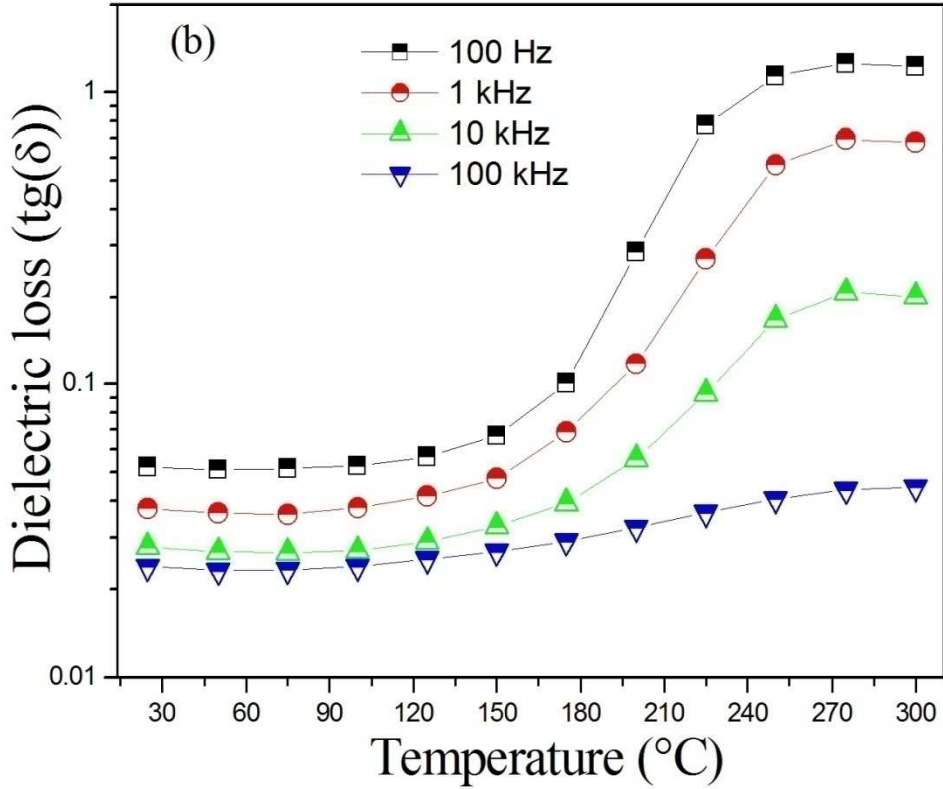


Fig. 5.

### 3.2.2. AC Conductivity Analysis

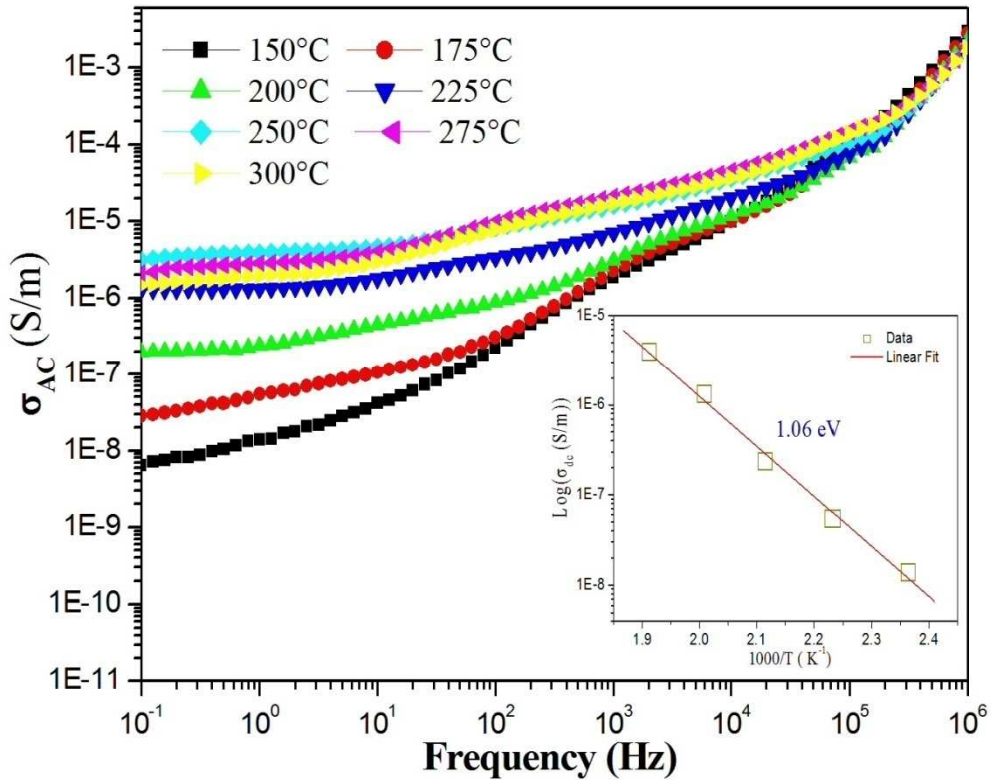
Using the  $\epsilon'_r$  and  $\tan(\delta)$  measurements, the ac electrical conductivity ( $\sigma_{ac}$ ) of our BST thick films can be **determined** from the empirical expression:  $\sigma_{ac} = \omega \epsilon' \epsilon_0 \tan(\delta)$ , Where  $\omega$  is the angular frequency and  $\epsilon_0$  is the vacuum permittivity. Fig. 6 presents the frequency dependence of  $\sigma_{ac}$  at various temperatures (150°C-300°C). **On this graph we** can observe two regions, at low frequencies; the conductivity is practically **constant** with frequency **which** corresponds to the dc bulk conductivity ( $\sigma_{dc}$ ). But **while** in the high frequencies region, the  $\sigma_{ac}$  varies greatly with frequency. Therefore the ac conductivity can be investigated by Jonscher's power law equation:

$$\sigma_{ac} = \sigma_{dc} + A\omega^n \quad (1)$$

Where, A is a pre-exponential constant, and n is the power-law exponent [23]. It is clear from figure that the  $\sigma_{dc}$  conduction process is thermally activated, so  $\sigma_{dc}$  can be obeys the Arrhenius law:

$$\sigma_{dc} = \sigma_0 e^{\left[\frac{-E_\sigma}{k_B T}\right]} \quad (2)$$

Where  $k_B$  is the Boltzmann constant,  $T$  is the temperature in Kelvin,  $\sigma_0$  is the pre-exponential factor, and  $E_\sigma$  is the dc conductivity activation energy. We **have drawn**  $\sigma_{dc}$  versus  $1000/T$  at **0.1 Hz in the inset of Fig.6**. The obtained  $E_\sigma$  value is about  **$1.06 \pm 0.05\text{eV}$**  which can be **attributed to** the migration of oxygen vacancies in BST [21]. **This value is of the same order of magnitude as we published for thin BST85/15 films from 200 to 400 nm thick deposited on Pt/Ti/SiO<sub>2</sub>/Si substrates [20].**



**Fig. 6.**

### 3.3. Tunability and hysteresis behavior

Fig. 7a illustrates the C–V curves obtained at room temperature at 10 kHz from -20 to +20V. The appearance of a butterfly form in the curves and the strongly nonlinear voltage confirm the good ferroelectric characteristics of the BST thick film occurring from domain switching. The dielectric tunability is defined by the following expression [24, 25]:

$$\eta_r = \left(1 - \frac{C_{\min}}{C_{\max}}\right) \times 100 \quad (3)$$

As shown in Fig.7a,  $C_{\max}$  is the maximum value of the capacitance which in our case is about 275pf either at +5V or at -5V and  $C_{\min}$  is the minimum value of the capacitance, which in our case is about 50pf either at +20 V or at -20 V. The tunability at 20 Volts is about 81%, this value is higher than a recent published one on BST thick film of the same composition and same preparation technique deposited on alumina substrate [2]. To the best of our knowledge, the value of 81% is the uppermost reported for a BaSrTiO<sub>3</sub> in different forms of films (thin / thick) and in ceramics. A comparison with published tunability in the literature is given in the following table.

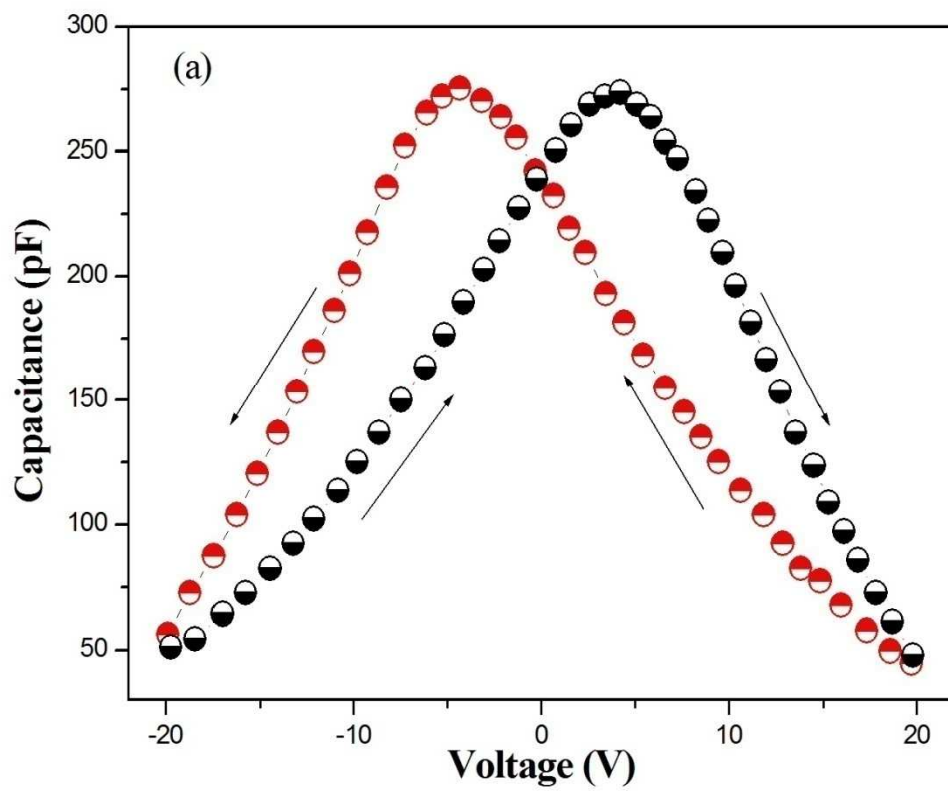
**Table 1.**

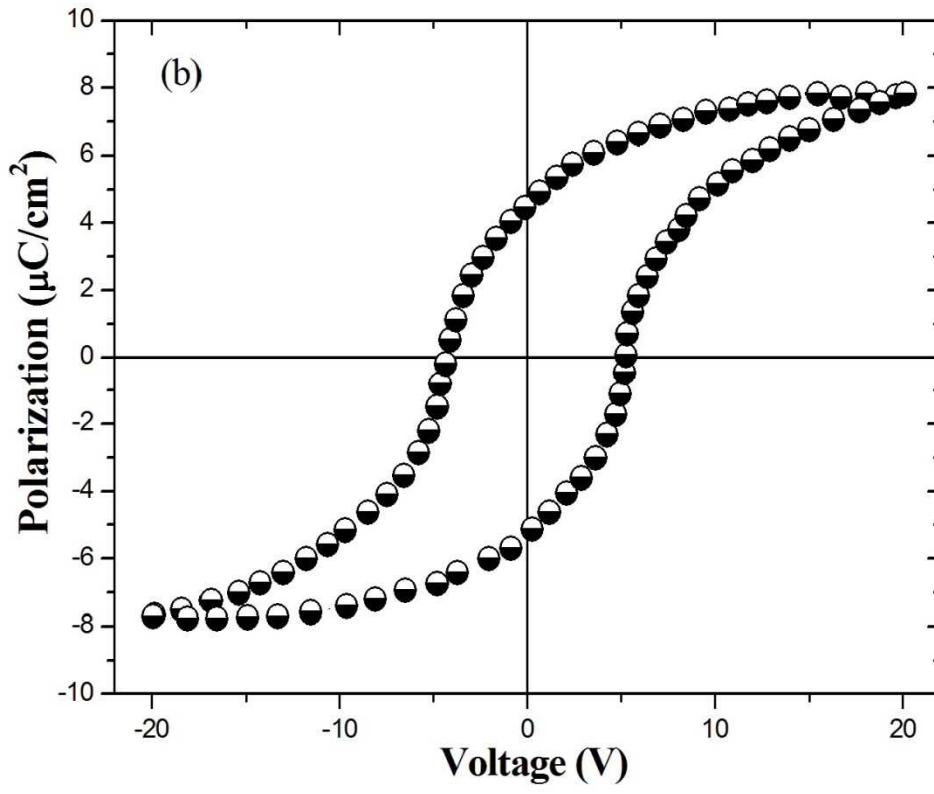
<b>Composition</b>	<b>Form</b>	<b>Tunability @ Electric field</b>	<b>References</b>
BST (85/15)	Thick film	81% @ 10kV/cm	This work
BST (85/15)	Thick film	67% @ 60kV/cm	[2]
BST (70/30)	Ceramic	69% @ 17kV/cm	[8]
BST (70/30)	Thin film	42% @ 600kV/cm	[8]
BST (60/40)	Ceramic	40% @ 66kV/cm	[13]
BST (70/30)	Thick film	43% @ 66kV/cm	[13]
BST (70/30)	Thick film	29% @ 66kV/cm	[13]
BST(60/40)	Thick film	60% @ 57kV/cm	[14]
BST (90/10)	Thin film	30% @ 125kV/cm	[24]

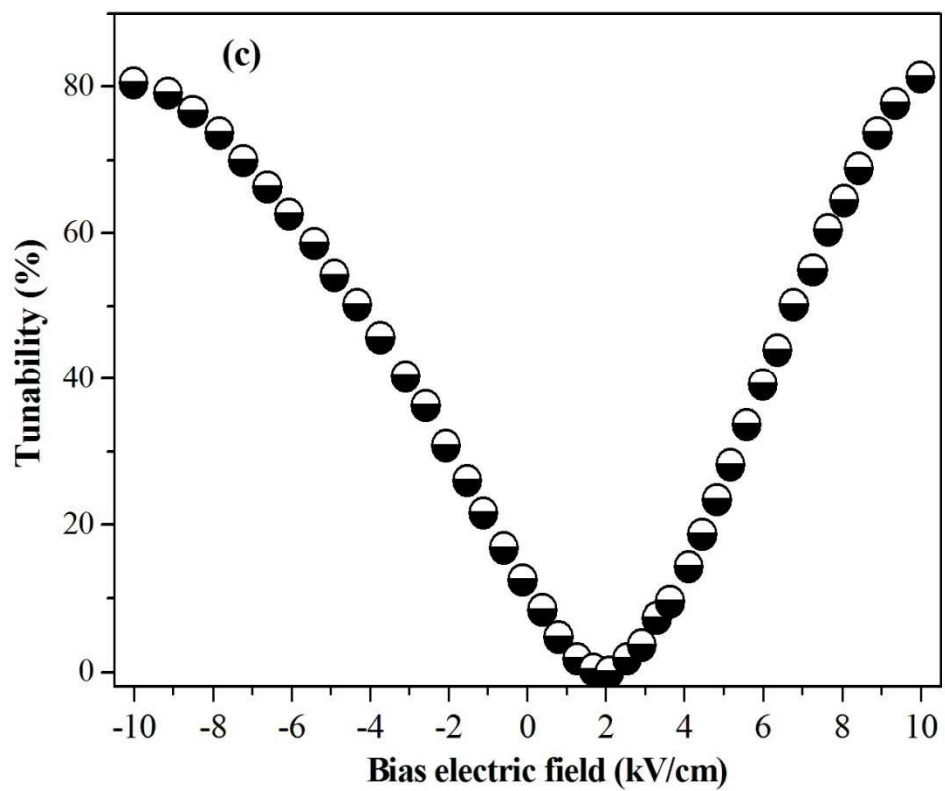
Our obtained tunability of 81% corresponds to an increase by a factor of **5.3** of the capacitance (and therefore of the **dielectric** permittivity). Such a variation can be well compared to the one of a tunable varactor diode and is very attractive for tunable microwaves applications such as filters, antenna beam steering, and tunable capacitor **as well as for tunable optical applications such as refractive index, filters and MEMS.** The evolution of the tenability as a function of the electric field is given in Fig. 7c. The tenability curve is not symmetrical which can be attributed to the different nature of

**the used electrodes (Au and Pt).** The ferroelectric hysteresis loop of the Au/BST (thick film)/Pt **is** measured at room temperature at 10 kHz (Fig. 7b). The existence of a remnant polarization confirms that our film is in a ferroelectric phase. **Moreover, the** remnant polarization ( $P_r$ ) value is estimated to be  $5\mu\text{C}/\text{cm}^2$ . This result is **almost ten times better than** those for screen printed BST thick film deposited on alumina substrate  $P_r = 0.57\mu\text{C}/\text{cm}^2$  reported by X.-F. Zhang et al [14]. **Then,** the remnant polarization of  $5\mu\text{C}/\text{cm}^2$  is encouraging for FeRAM technology [26]. **It can be also observed that the saturation polarization is  $P_{\text{sat}}=8\mu\text{C}/\text{cm}^2$  and the coercive voltage is  $E_c= 5\text{ V}$  which corresponds to an electric field of  $2.5\text{ kV}/\text{cm}$ . This low voltage value allows to switch a ferroelectric domain corresponding to a bit in a FeRAM.**









**Fig. 7.**

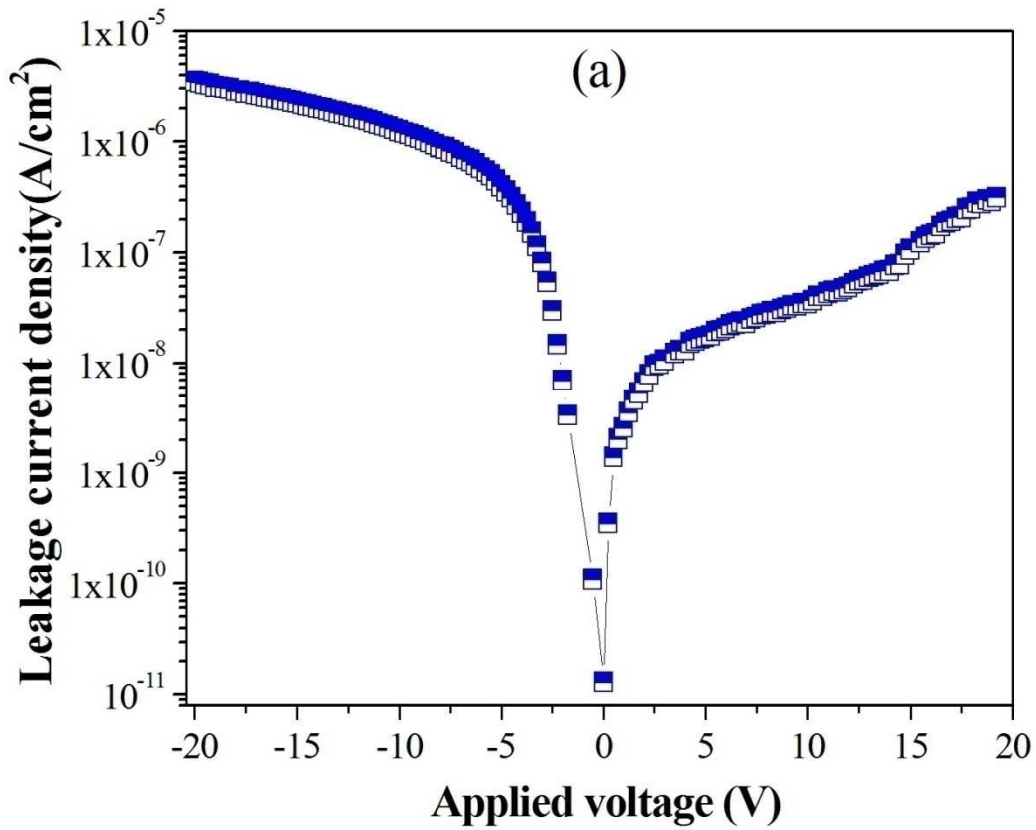
### 3.4. Leakage current measurements

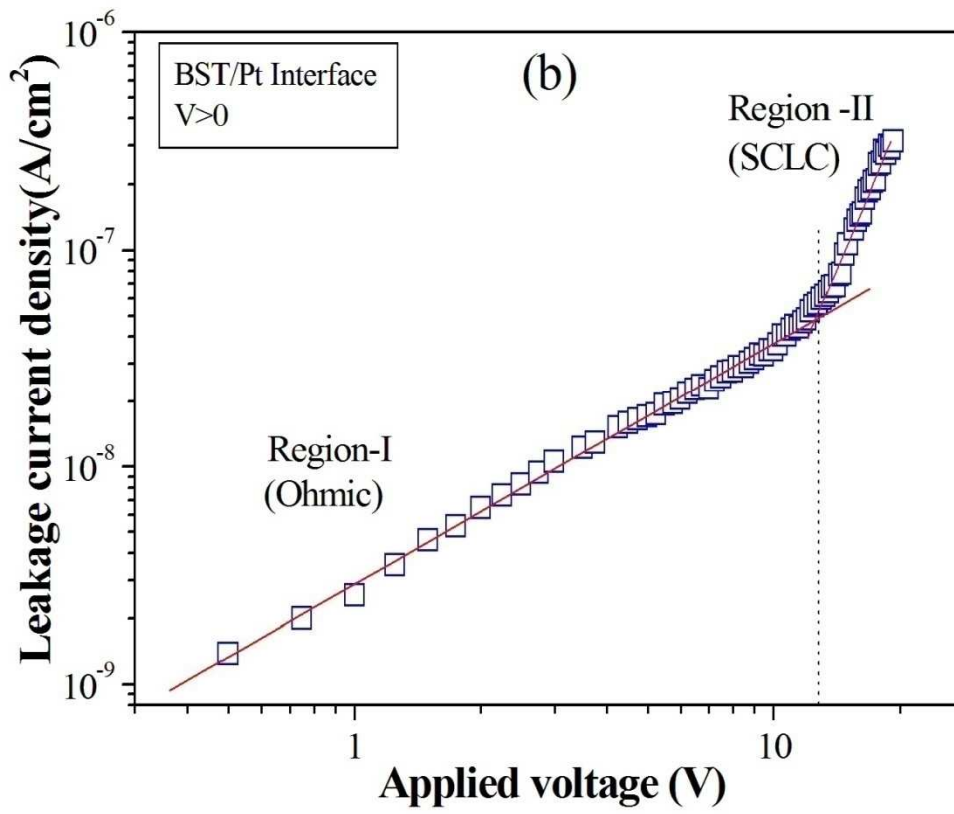
Fig. 8a display the leakage current density of BST capacitor as a function of applied voltage in the (-20V – +20V) range. The leakage current density (J) is smaller than  $10^{-5}$  A/cm<sup>2</sup> in the whole voltage range. The leakage current density curves are not symmetrical which can be attributed to the different nature of the used electrodes (Au and Pt). The J-V behavior reveals that various leakage mechanisms **occurring** in the negative and positive bias region. Generally, the **conduction process can be governed by several mechanisms**, including Poole frenkel effect, Schottky emission, Ohm's law and SCLC emission [9, 21, 27]. These different mechanisms **can either contribute simultaneously or one of them playing the dominant role in the conduction**. To investigate the conduction mechanism in our Ba<sub>0.85</sub>Sr<sub>0.15</sub>TiO<sub>3</sub> **films** in positive applied voltage, the leakage current density (J) is **plotted against applied voltage (V) in a log(J) versus log(V) graph as shown** in Fig. 8b. It can be **clearly observed** that there are two separate regions. For low voltages, the log(J) versus log(V) curve has slope of 1.11, which is value very close to 1, indicating that Ohm's law plays a major role in the conduction mechanism in this voltage region [28]. At high voltage, **the curve** reveals a slope close to 2, which is in **a** good agreement with (SCLC) **model** [29]. For high voltages i.e. high electric field, with a negatively biased Au electrode, the Schottky model can be employed to study the leakage current measurement. The current density J is then given by [30].

$$J = A^{**}T^2 e^{\left(-\frac{q\phi_B}{KT}\right)} \times e^{\left(\frac{q}{KT} \sqrt{\frac{qE}{4\pi\epsilon_0\epsilon}}\right)} \quad (4)$$

Where  $A^{**}$ ,  $E$ ,  $\phi_B$ ,  $q$ ,  $\epsilon_0$ ,  $\epsilon$ ,  $k$ , are **respectively** the effective Richardson constant, **the** applied electric field, **the** Schottky barrier height at the Au electrode, **the** electronic charge, **the dielectric permittivity of the** free space, **the optical** dielectric constant and the Boltzmann constant, respectively. If the conduction mechanism at high electric field is related to Schottky emission, the log J versus  $\sqrt{E}$  plot **should** to be a straight line and the slope **would** provide the optical dielectric constant of BST. **We found in Fig.8c a value of 3.55 which is in accordance** with the square of the refractive index n [28, 31].

In Fig. 8c,  $\log J$  is traced as a function of  $\sqrt{E}$ . It's clear that for applied electric field higher than a threshold value, the Schottky equation provides a good fit with the leakage current curve, as presented by the solid line in Fig. 8c. The value 3.55 is comparable to the known optical dielectric constant value of about 4. So, at a high negative voltage, the leakage current of BST thick films can be well described by the Schottky equation.





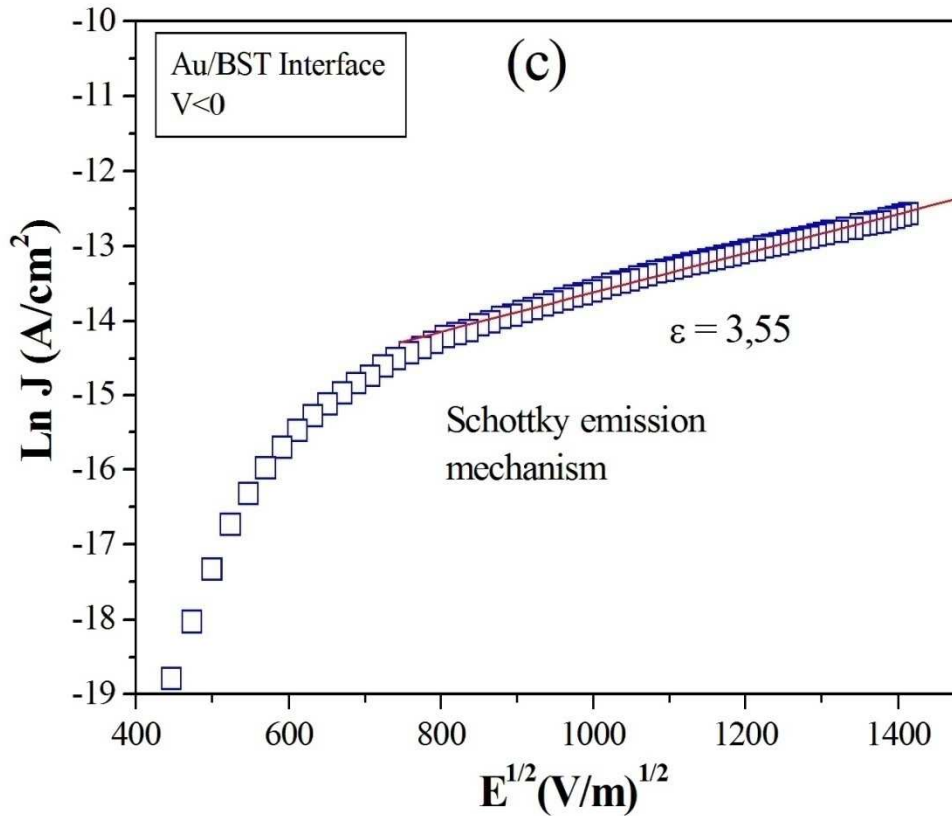


Fig.8

#### 4. Conclusions

In this work, high-quality  $\text{Ba}_{0.85}\text{Sr}_{0.15}\text{TiO}_3$  thick films ( $20\mu\text{m}$ ) are successfully fabricated by screen-printing technique on Pt/Ti/SiO<sub>2</sub>/Si substrates. AC and DC measurements are carried out. The temperature dependence of the dielectric constant shows two peaks: the first one appears at  $75^\circ\text{C}$  and is attributed to the ferroelectric-paraelectric transition (Curie temperature), while the origin of the second peak observed at high temperature ( $270^\circ\text{C}$ ) and low frequency can be related to the existence of nano/micropolar phases in the BST films. The leakage current in Au/BST/Pt capacitor reveals an asymmetric behavior at positive and negative bias. At a high electric field, in the positive bias region the leakage current shows SCLC behavior. In the negative bias region, the leakage current is dominated by Schottky emission. The

high values of the dielectric permittivity combined with low values of the dielectric loss and leakage currents show high performance of our Au/Ba<sub>0.85</sub>Sr<sub>0.15</sub>TiO<sub>3</sub>/Pt capacitors. This structure is very encouraging in the areas of thick film technology and presents a substitute for Pb-based materials. Moreover, the high tunability (81%) makes Ba<sub>0.85</sub>Sr<sub>0.15</sub>TiO<sub>3</sub> thick films a potential candidate for functional applications like tunable varactor diode and tunable capacitor devices.

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