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High tunability inlead-free $Ba_{0.85}Sr_{0.15}TiO_3$thick films for microwave tunable applications

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Abstract

Lead free ferroelectric **$Ba_{0.85}Sr_{0.15}TiO_3$(BST)**thick films were successfully deposited by screen printing process on Ag-Pd/Al_2O_3 substrates. The microstructure of the samples was analyzed by scanning electron microscope (SEM)and X-ray diffraction (XRD). **The thick films** showed a good adhesion to the Ag-Pd/Al_2O_3 substrate, a porous microstructure and uniform **thicknesses**of about10 µm. The dielectric **properties**ofBST thick films were studied over a wide frequency range (10^{-1} Hz to 10^6 Hz) and **in a large temperature range from**25 °C to 350 °C. The AC conductivity in**BST** thick films was systematically investigated.Ferroelectric characterizations and tunability were also studied at room temperature by polarization cycles P(E) and**c**apacitance C(E) measurements. The high values of dielectric permittivity and tunability (67%) combined with low dielectric loss (tgδ) make $Ba_{0.85}Sr_{0.15}TiO_3$ thick films one of the encouraging candidates for microwave tunable applications.

***Keywords*:Barium strontium titanate; Thick films;** Screen printing; Dielectric properties;ac conductivity;Tunability.

1. Introduction

Ferroelectric oxide materials have been studied for a long time, but only recently have they been deposited in the form of thin and thick films with a thickness lower than 1µm and 500µm respectively. The electrical characteristics of ferroelectric materials **arecurrentlyused to** realize electronic components [1-3]. Their principal**propertiesare**large dielectric **permittivity**, high tunability, low microwave losses and non-linear polarization. Lead zirconatetitanate($PbZrTiO_3$or PZT) is the most important **ferroelectric**material used in microelectronic. In fact, it has many applications such as pyroelectric infrared detectors, ferroelectric memories, capacitors, tunable microwave components, filters,etc…However, its **main drawback**is to contain lead which is strictly forbidden since July 2014 by the European directive RoHS.**This directive bans the use of several polluting materials in electronic and microelectronic devices, including lead.Nevertheless,lead oxide based ferroelectric materials are yetused for their unique properties but in thenear future they will be prohibited.** The search to discover an alternative to **PZT**has started **in many**laboratories but **it** mainly concerns the ceramic form. **Consequently, there is an urgent need to find alternative materials to PZT. Our research is focusedon lead-free $Ba_{0.85}Sr_{0.15}TiO_3$(BST), a good candidate to replace PZT in several uses**[4].**This composition (BST85/15) is a good trade-off to have very good ferroelectric and dielectric properties for applications at temperatures close to room temperature. So, $Ba_xSr_{1-x}TiO_3$is the most extensively investigated material to date thanks to the high dielectric permittivity, small dielectric losses,low leakage-current density and other excellent physical properties.For applications** in electronic devices,**$Ba_xSr_{1-x}TiO_3$ h**as been investigated in varied material forms, including thin films (t< 1µm), thick films 1µm<t<500 µm) and ceramics t>500 µm) [5,6]. At present, the majority of**the**works on ferroelectric oxides is oriented towards the form of thin films and**/or**ceramics. So far, BST thick films have been less reported. **Yet** BST thick films present a potential benefit of lower fabrication cost in comparison with that of thin film forms and low bias voltages required **for tunability in relation to the thickness of ceramics.** BST thick films also strongly fill the technological difference between thin films and ceramics.Several techniques have been successfully **used** to elaborate BST thick films including sol-gel, pulsed laser deposition, and screen printing [7–9]. **Among** these different processes, screen printing is a viable, low-cost and practical procedure for wide area elaboration of thick films. This technique is **suitable** for coating surfaces of whatever geometry and morphology and has been applied to produce several ferroelectric thick films such as PZT [10], BZN [11] and BST [9].

In this study, $Ba_{0.85}Sr_{0.15}TiO_3$ thick filmswere successfully deposited by screen printing on alumina substrates covered with a silver-palladium electrode. The dielectric **properties** of $Ba_{0.85}Sr_{0.15}TiO_3$ thick films were studied as a function of temperature and frequency. Various AC electrical **measurements** such as dielectric permittivity,dielectric loss tangent (tanδ) and AC conductivity **were made**in order to better **understand** the correlation between the microstructure of the thick films and the dielectric response. Ferroelectric properties of BST thick film have also been analyzed using Capacitance-Electric field (C-E) and Polarization-Electric field (P-E) measurements. It is shown that **our**BST thick films present hightunability (67%)and good dielectric properties **which** compare **well with BST both** in thin films and **in** ceramics.

2. Experimental

2.1 Thick film preparation

$Ba_{0.85}Sr_{0.15}TiO_3$ thick films were elaborated by using a screen-printingdevice (TIFLEX - HL570S).BST powders were synthesized by the solid-state procedure from strontium oxide (SrO), barium oxide (BaO) and titanium oxide (TiO_2). The ink for screen printing **was prepared by mixing BST** powders with organic vehicles. The organic vehicles were composed of {alpha-terpineol ($C_{10}H_{18}O$), polyvinyl butyral $(C_8H_{14}O_2)_n$,2-(2-butoxyethoxy)-ethyl acetate ($C_{10}H_{20}O_4$) and polyethylene glycol (C_2H_4O)**from ARCOS ORGANICS**. The ink was mixed and milled by a roller to obtain a perfectuniform homogeneity. Initially polyvinyl butyral **was** dissolved in α-terpineol at 60°C with vigorous stirring for 30 minutes. The $Ba_{0.85}Sr_{0.15}TiO_3$ powder (previously dissolved in 2-(2-Butoxyethoxy) ethyl acetate) was added and the polyethylene glycol was added to stabilize the mixture. After 1 hour, the ink was heated at 100 °C for 1 hour to evaporate the solvent. Finally, the ink passes to ultrasound and between the three-cylinder roller in order to break the agglomerates. **High**purity alumina substrates were covered by a silver-palladium**paste**to **realize**the **bottom**electrode for the electrical characterizations. The Ag-Pd paste was deposited via**a**spin-coater on alumina substrates and heated at 1100°C for 1 h. The ink was deposited by screen printingon the Ag-Pd and **heated** at 100 °C for 1 h. This **procedure**was repeated two times. Thereafter, the BST thick films were sintered in air for 2 h at 1100 °C. Finally, gold circular electrodes (**top**electrodes) with different diameters,**from**0.1mm **to**2mm **were**evaporated through a shadow mask on the BST thick films to obtain MIM **(metal-**

insulator-metal)structures for the electrical measurements. The differentmain stepsof preparation are shown in Fig. 1.

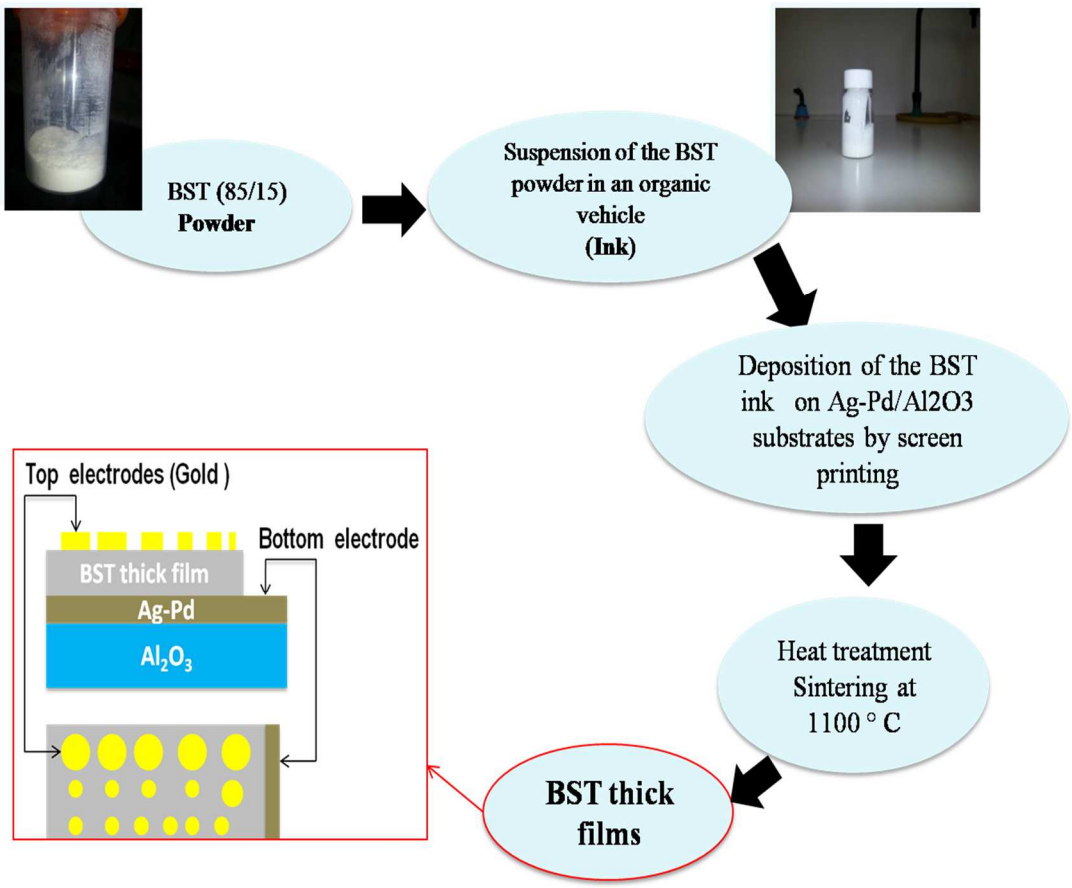


Figure 1.

2.2 Characterizations techniques

The crystallographic microstructure and the morphology of the samples were characterized by X-Ray Diffraction **with a Philips diffractometer (model PW1710)**and Scanning Electron Microscopy**(JEOL JSM-5400)**. The electrical properties ofBST were measured over a large frequency range (10^{-1} to 10^{6} Hz) in the 25°C-350°C **temperature** range using a Solartron 1260A**Impedance Analyzer with a Dielectric Interface 1296**.Capacitance-Voltage (C-E) measurements of the Au/BST/Ag–Pd capacitor**were** performed using an Agilent LCR meter E4980A.**Thehysteresis cycles were recorded with a modified Sawyer Tower circuit at a frequency of 1 kHz.**

3. Results and discussion

3.1Structural and morphological investigation

Fig. 2shows the XRD diffractogram of the BST thick film. It can be observed from the graph the existence of diffraction peaks **indexed to** (100), (110), (111), (200), (210) and (211) **planes. They are characteristicsof BST (JCPDS File no. 39-1395)**in the (10°-60°) 2θ range. In addition, the calculated a-axis and c-axis lattice values are 3.9942 Å and 4.0171 Å respectively. This suggests that the film is crystallized with the tetragonal structural phase. Small alumina and silver phases were also detected, which are associated with the substrateand the bottom electrode respectively.

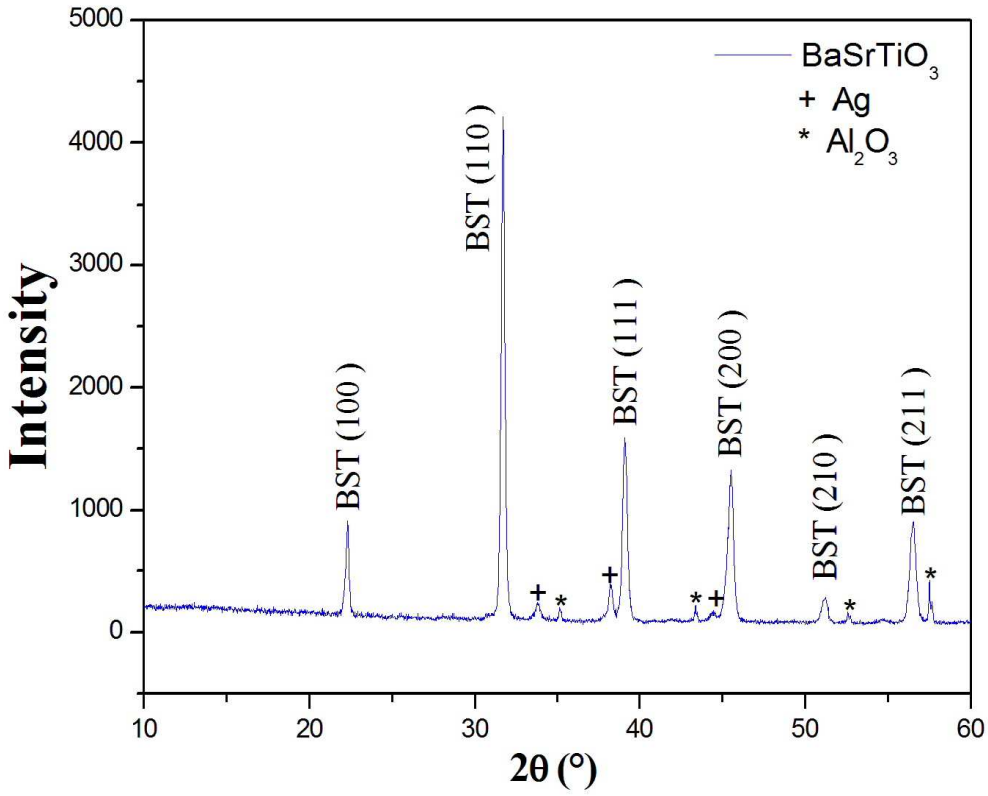


Figure 2.

Fig. 3exhibitsthe SEM images of the BST thick film. Figs. 3a and 3b **show**the cross-section view and a portrait zoom on the BST layer, respectively. It is clear that no cracks on the BST film were observed. The image zoom on BST **presents** a porous microstructure and the

average gain size is0.4 µm.The cross-section view shows a generally uniform thickness of about 9µm and anexcellent adhesion to the Ag-Pd/Al_2O_3 substrate.

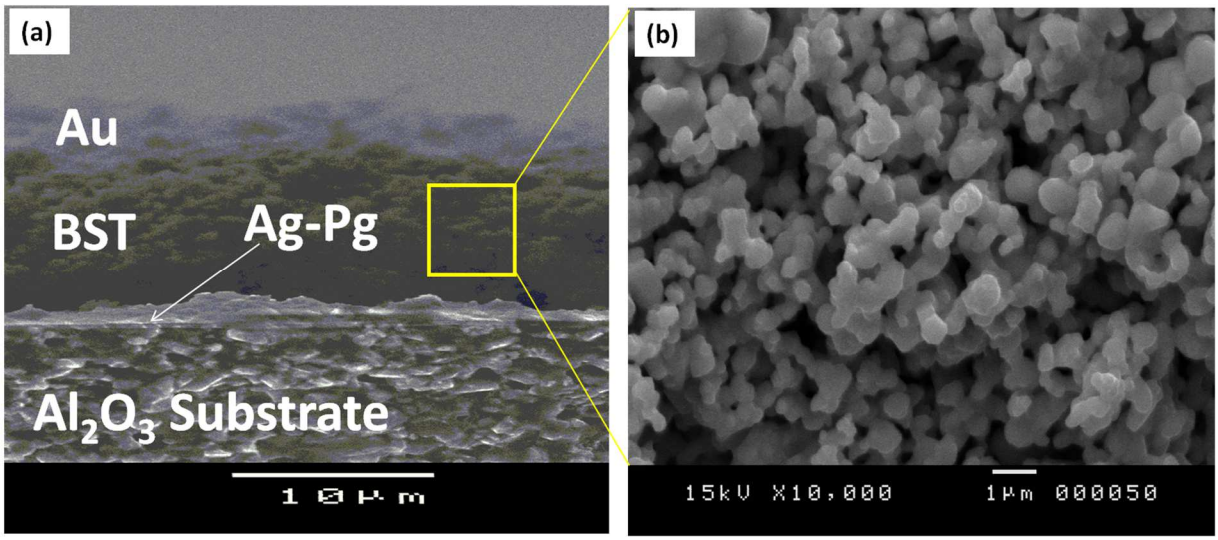


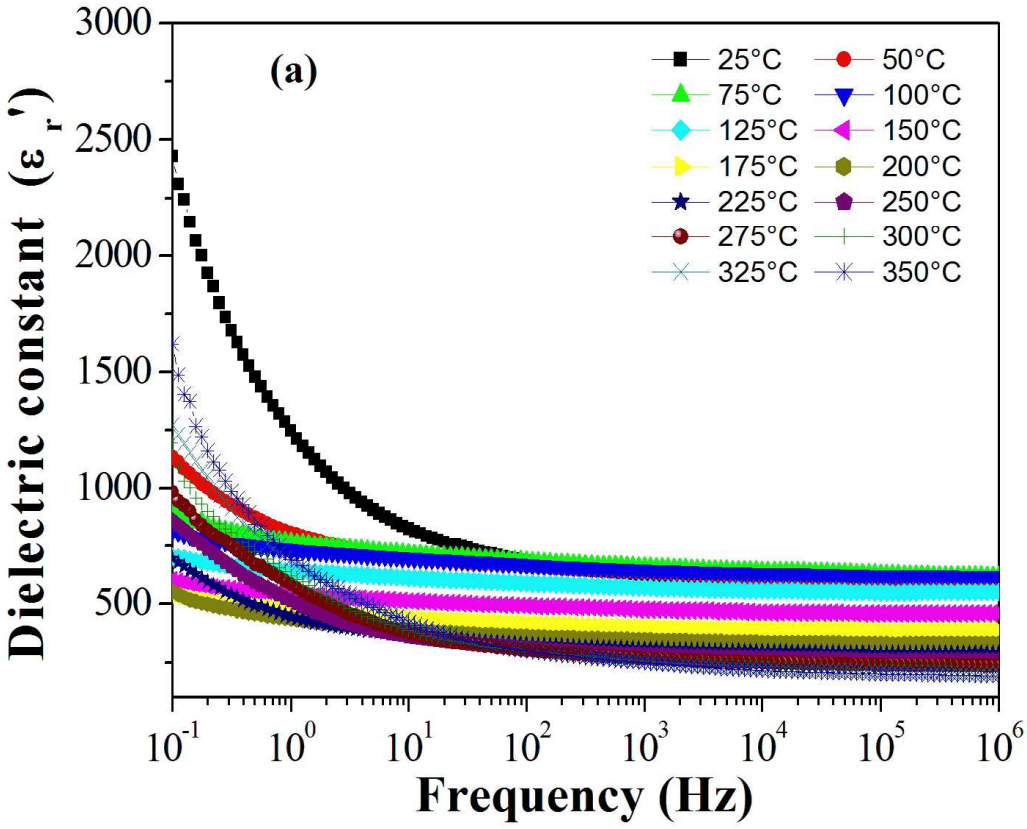
Figure 3.

3.2 Dielectric Analysis

3.2.1. Permittivity and loss studies

Dielectric properties of BST thick film were studied **as a function of** frequencies (0.1 Hz to 1 MHz) and **in the** temperature range 25°C-350°C. The evolution of **the** dielectric **permittivity**ε'_r from the complex permittivity $\varepsilon^* = \varepsilon'_r - i\varepsilon''_r$[12] of **the BST film** is shown in Fig. 4.a. **It is clear that for all temperaturethe dielectric permittivitydecreases rapidly in the low-frequency region and then go more or less stable at higher frequencies.The high value of the dielectric permittivity and the dispersion behavior at lower frequencies can be explained by the dominance of grain boundaries effect. This is an extrinsic contribution associatedto the Maxwell-Wagner interfacial polarization in agreement with Koops phenomenological theory [13]. According to this theory, a dielectric oxide material is made up of conducting grains segregated by highly resistive grain boundaries. The grain boundaries are more effective at low frequencies while the grains are found to be more effective in the high frequencies. Under the effect of the applied electric field, there is an accumulation of electric charge at the interfacesof the grains and the grain boundaries (interfacial polarization). After the initial great value at low frequencies,** ε'_r **becomes nearly constant at frequencies higher than 100Hz.This is due to**

an intrinsic contribution of the ferroelectric domains in the grains of the BST film. It is important to note that the dielectric permittivity showsa comparable value, **above 100 Hz**,to **data** found on BST ceramic **[14]** and **also** presents anexcellent stability in a large frequency range. This result is heartening for microwaves applications.**The dielectric loss tangent (tgδ= ε''$_r$/ε'$_r$) has also been measured atdifferent temperatures.** The curves obtained **for tgδ are presented in**Fig.4b.These **curves** show the same behavior **that**of the dielectric permittivity: for each temperaturethe dielectric losses **decreasewhen the frequency increasesdue** to Maxwell-Wagner interfacial polarization **[7]**.



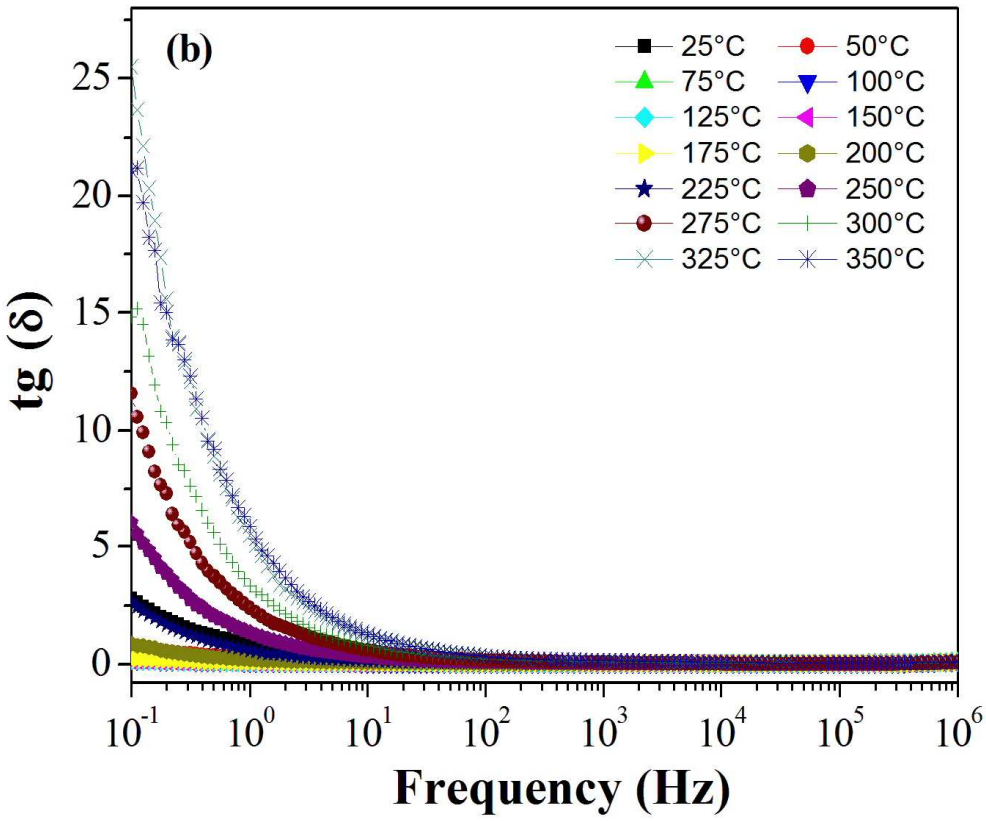
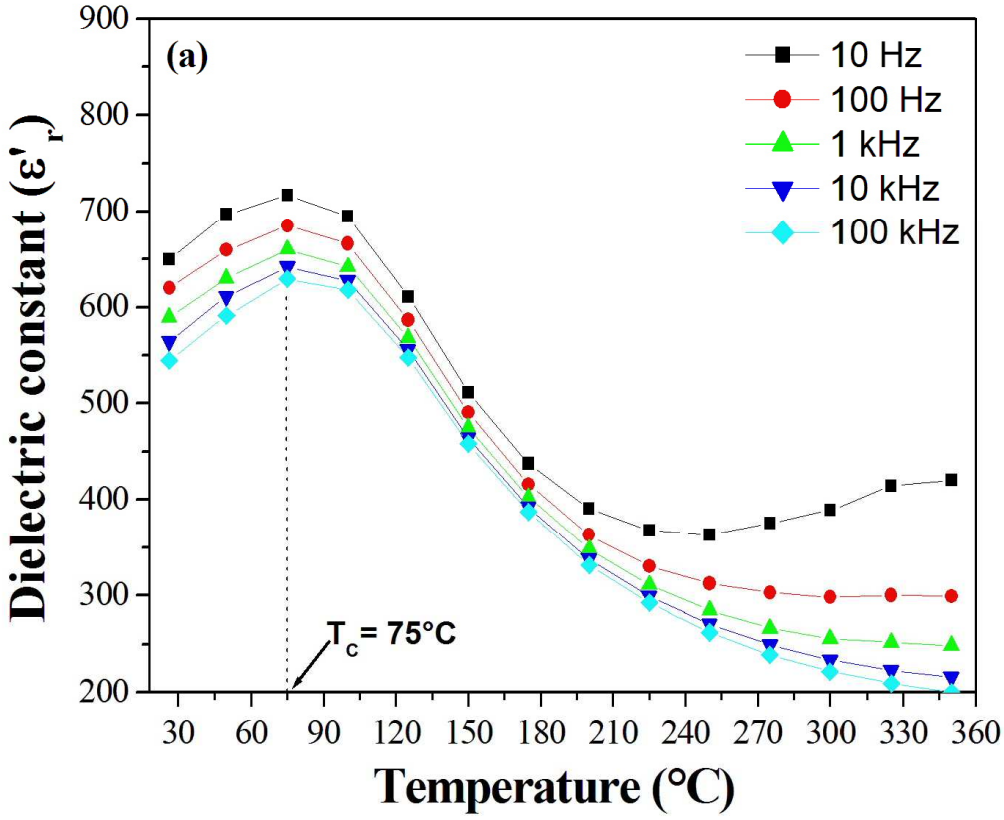


Figure 4.

Thevariation of ε'_r andtanδ of BST thick filmwith temperature for several **frequencies** is represented in Fig. 5.For all frequency values, the dielectric**permittivity**shows,**in Fig. 5a**,a large peak at **a Curie temperature**T_C=+75°C.Thisobserved peakcorresponds to a ferroelectric–paraelectric phase transition in the BST system.**From 25 °C to 75 °C it is admitted that the increase of ε'_r characterizes the ferroelectric state while above 75 °C the monotonous decrease of ε'_r characterizes the paraelectric state where the ferroelectric domains progressively disappeared when the temperature increases.** Moreover, the value of $T_C$is smaller than the one found on a BST in the ceramics form**[14]** where T_C =+101 °C. This variance**is** possibly associated to the small grain size ina thick film compared with a ceramic and may also be related to a little difference of composition as the $T_C$is very sensitive to the Ba/Sr ratio which is **here**90/10 for the ceramic. **For instance,** our value of T_C=+75 °C is for a 85/15 ratio while T_C=-2 °C for a 60/40 ratio on a BST thick film also deposited by screen printing [6]. The value of T_C found on our thick films **confirms**that the BST is in ferroelectric phase at **room temperature**.This result is encouraging for several

applications including high dielectric constant capacitors, super-capacitors and ferroelectric **random-access** memories (FRAM). **The thermal evolution of the dielectric loss tangentfor different frequencies(Fig. 5b) gives low values (tanδ<0.1) in alarge temperature, ranging from 25 °C to 150°C. Above 150 °C the dielectric lossesshow animportant increase which can be attributed tothe motion of oxygen vacancies.In fact, oxygen vacancies are thermally activated and then can make hopping in the grains and grains boundaries which increase the losses [15]. The high values of the dielectric permittivity and the associated low losses values obtained over a wide temperature range (25-150 °C) make the $Ba_{0.85}Sr_{0.15}TiO_3$ thick films one of the possible candidates for the different applications mentioned above.**



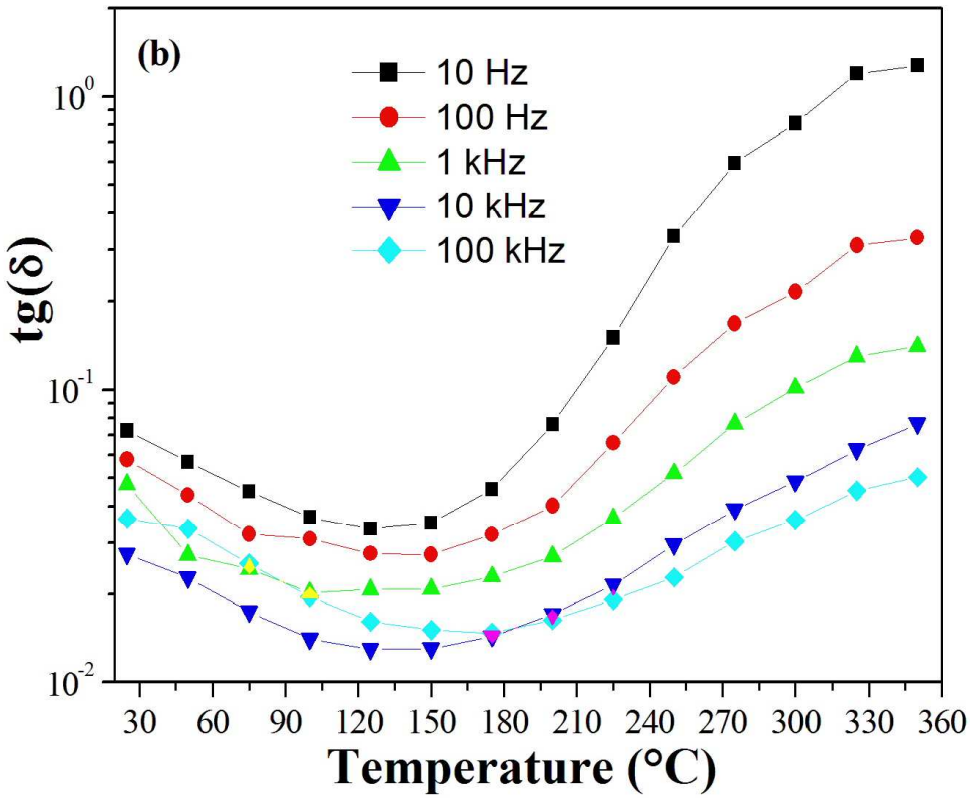


Figure 5.

3.2.2.AC Conductivity Analysis

The AC conductivity (σ_{ac}) of the BST thick film **was** obtained from the **determination of** ε'_r and tanδ and calculated with the **following** formula **[16]**:

$$\sigma_{ac} = \omega \varepsilon'_r \varepsilon_0 \tan(\delta) \qquad (1)$$

Whereε_0 is the vacuum permittivity and $\omega = 2\pi f$is the angular frequency.

Frequency dependent AC conductivity was measured in the **frequency**range 0.1Hz ≤ F ≤ 10^6Hzat varioustemperatures (225°C-350°C) and **is** shown in Fig. 6. In the low frequency region,for all temperatures,the conductivity is practically independent of frequency**[17]**, this **is**associated with the **DC** conductivity **(σ_{dc})**of the BST.**Above about 1 Hz and up to 1 MHz, the AC conductivity increases strongly, for example of 4 decades at 300 °C.** The **AC**conductivity of BSTcan be interpreted by the Jonscher's law of the Universal Dielectric Response (UDR) model **[18]**:

$$\sigma_{ac} = \sigma_{dc} + A\omega^{n} \qquad (2)$$

Fig.6**presents**the theoretical fits **of**the experimental data of σ_{ac}**with** equation (2). From this figure, it **can be**seen that the experimental data **are**well fitted **with**the **UDR** model. In order to specify the principal conduction mechanism of the ACconductivity, the variation of'n' with temperature reveals that theexponent 'n' decreases **when** the temperature increases**(not shown here)**.This trend showsthat the AC conduction in BST thick film follows the Correlated Barrier Hopping (CBH) model**[19]**.Also, from **Fig. 6**and the σ_{dc} values obtained **with** the fit, it's clear that the stationary regime is thermally activated so the conductivity σ_{dc} can be represented by the Arrhenius law **[16]**:

$$\sigma_{dc} = \sigma_0 \, e^{\left[\frac{-E_\sigma}{K_B T}\right]} \qquad (3)$$

where T is the temperature in Kelvin, k_B is the Boltzmann constant,σ_0 is the pre-exponential factor and E_σ is the DC conductivity activation energy.The extracted activation energy value for **DC**conduction **was**found to be around 0.5 eV. According to the literature, this value is comparable to the energy required for a polaron to jump **[14,20,21]**through the grain boundaries.**These polarons can be constituted by electrons associated with singly and doubly positively charged oxygen vacancies O_O° and $O_O^{\circ\circ}$ (with Kroger-Vink notation). As stated in the previous paragraph, these jumps are at the origin of the dielectric losses observed above 150 °C in the measurement frequency range. So, it can be concluded that at high temperature, typically above 150 °C, the DC conductivity and the dielectric losstanδ have the same origin that is to say the hopping of polarons.**

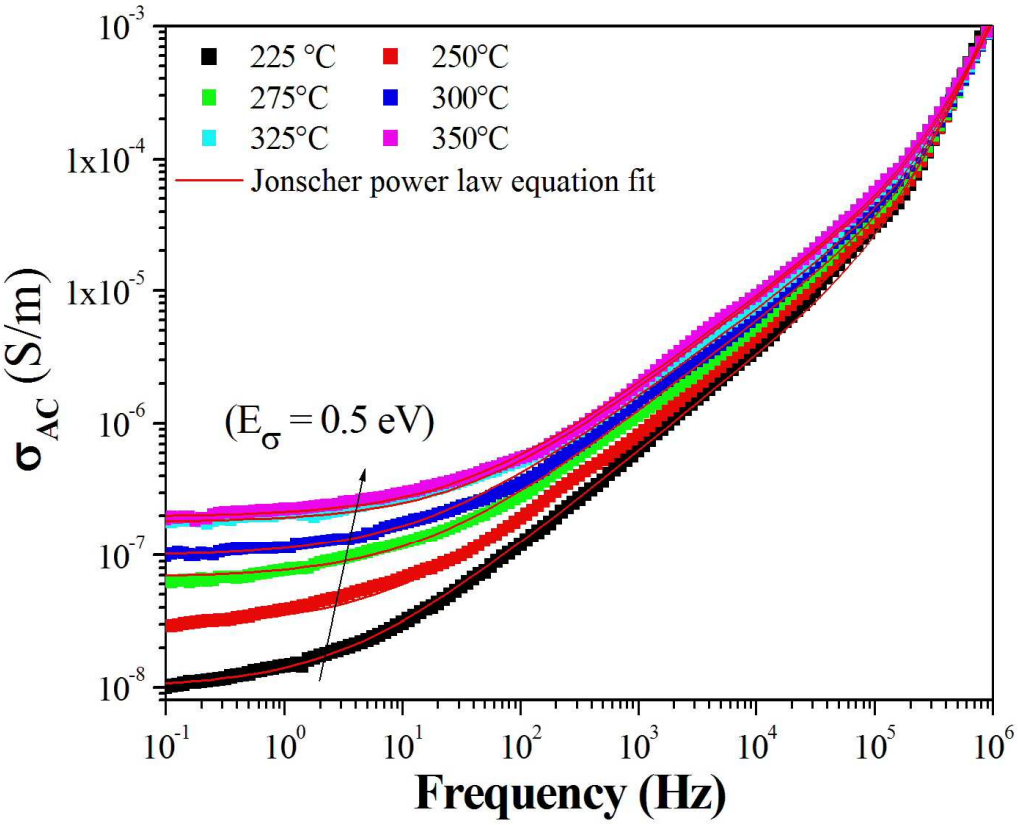


Figure 6.

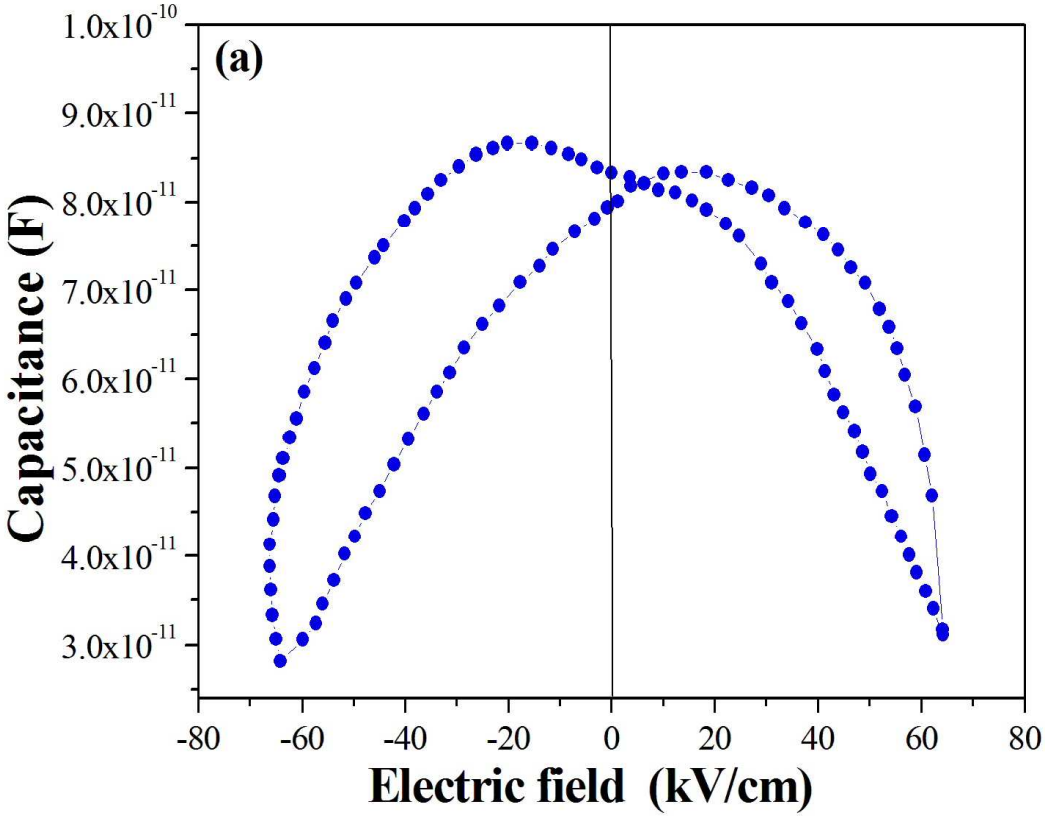
3.3.Capacitance-electric field (C-E) and polarization-electric field (P-E)measurements

Fig. 7ashows the capacitance- electric field response at room temperature **for a** BST thick film. The butterfly form of the curve **confirms** the ferroelectric behavior of the film. The dielectric tunability was obtained using the following **formula [22]**:

$$\frac{C(0) - C(E)}{C(0)} * 100 \quad (4)$$

where C(0) is the capacitance at 0 kV/cm and C(E) is the capacitance for an electric field of value E. The tunability at 12Volts and at 1kHz is about 67%:this value is higher than those recently published **with** BST thin films**[4, 23, 24]** and comparable to that published on BST ceramics**[25]**.**This value**highlight that the high tunability of **our**BST thick films is very encouraging formicroelectronic applications such as tunable capacitors **in radiofrequencies**.Thehysteresis **cycle**of the Au/BST(thick film)/Ag-Pd structure was **recorded**at room temperatureat 1 kHz (Fig. 7b). The existence of a hysteresis loop confirms that the sample is ferroelectric in nature.From **the** hysteresis loop, the remanent polarization

value is estimated to be 0.92μC/cm^2, the maximum polarization is 3.5 μC/cm^2 and the coercive field is about 6 kV/cm at room temperature.



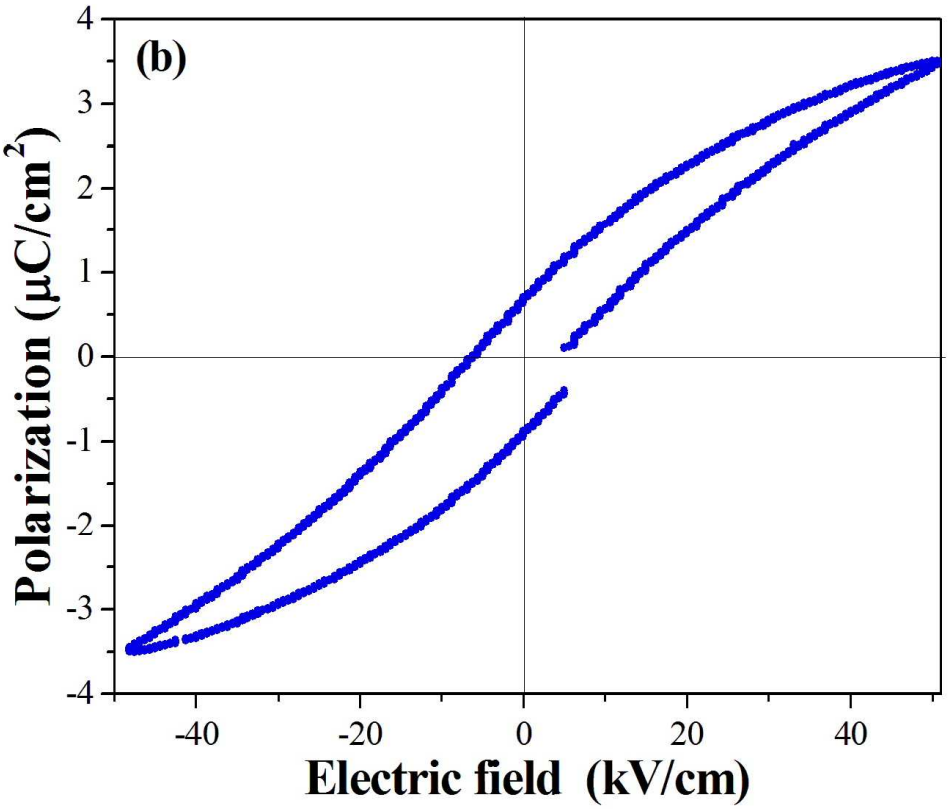


Figure 7.

4. Conclusions

We have shown that it is feasible to elaborate by a low-cost technique,namely a conventional screen-printing method, good quality lead free $Ba_{0.85}Sr_{0.15}TiO_3$ films**of at least 10 µm thick. The electrical characteristics of our BST films** are comparable and sometimes better than the ones of BST thin films and ceramics. This material is very **interesting**in the area of thick film technology and presents an alternative **to**Pb-based **ferroelectric**materials.The high values of dielectric permittivity and tunability (67%) combined with low dielectric loss **tangent**make $Ba_{0.85}Sr_{0.15}TiO_3$ thick films **usable**for functional applications like tunable ferroelectric capacitors**in radiofrequencies and in microwaves**.

Acknowledgments

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