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FREQUENCY AGILE BST MATERIALS FOR MICROWAVE APPLICATIONS

A. Ioachim*, M. I. Toacsan, M. G. Banciu, L. Nedelcu, C. Plapcianu, H. Alexandru[a], C. Berbecaru[a], D. Ghetu[a], G. Stoica[b], R. Ramer[c]

National Institute for Materials Physics, Str. Atomistilor no.105, P.O.Box MG - 7, RO - 76900, Bucharest-Magurele, Romania
[a]University of Bucharest, Faculty of Physics, Bucharest – Magurele, Romania
[b]I.P.E.E.- S.A. Curtea de Arges, Romania
[c]University of New South Wales, Faculty of Electrical Engineering and Telecommunications, Sydney, NSW 2052, Australia

$(Ba_{1-x}Sr_x)TiO_3$ solid solution (BST), with different molar compositions (*x = 0.25, 0.5, 0.75, 0.9)* were prepared by conventional solid-state reaction from raw materials. Structural parameters, crystallite sizes and bulk densities were determined. Perovskite type polycrystalline structure of the BST ceramics was revealed by X-ray diffraction (XRD) data. The dependence of permittivity and losses at low frequency (1 kHz) was analyzed. The microwave investigations at room temperature revealed dielectric constant around 1000 and loss smaller than 1% at 1.1 GHz. The results indicate that some BST dielectric ceramics are suitable in paraelectric phase for microwave devices. The dielectric constant can be adjusted by using a DC-bias field and such materials are appropriate for manufacturing electric field controlled components such as resonators, phase shifters etc.

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1. Introduction

Ferroelectric materials are frequently used for several applications in electronics as dynamic random access memories because their hysteresis, or as capacitors due to their high dielectric constant. Moreover, the ferroelectric materials present a great potential for electrically tuneable microwave devices such as phase shifters, mixers and parametric amplifiers [1]. These materials are mostly used in paraelectric phase when there is no thermal hysteresis. The ferroelectrics in paraelectric phase may exhibit high dielectric constant, low losses for high frequencies, and a temperature coefficient of resonant frequency, which can be adapted to the specific application.

The dielectric constant of the ternary compounds $BaO-SrO-TiO_2$, also known as BST, can be modified by applying a DC bias electric field [2-5]. In the present paper, BST compositions with the chemical formula $Ba_{1-x}\ Sr_x\ TiO_3$ were prepared by standard solid-state reaction. Structural parameters, bulk densities and morphology were investigated. Moreover, Curie temperature, low-frequency and microwave dielectric parameters were determined in order to select the optimum BST material for development of electric field controlled components such as resonators, tuneable filters, phase shifters, varactors, etc.

* Corresponding author: ioachim@alpha1.infim.ro

2. Experimental

The $Ba_{1-x}Sr_xTiO_3$ samples were prepared by standard ceramic technology. Four BST compositions were investigated: BST25 ($x = 0.25$), BST50 ($x = 0.5$), BST75 ($x = 0.75$) and BST90 ($x = 0.9$). The starting materials were $BaCO_3$, $SrCO_3$ and TiO_2, powders of purity higher than 99.9%. The raw material was weighted out in stoichiometric proportions, ball-milled in water for 2 hours, dried and then calcined at T=1150^{0}C for 2 hours. The powder was crushed and then milled again for 2 hours. Mixing was carried out for 2 hours in an agate mortar containing agate balls. The mixture was pressed into pellets of 11 mm diameter and 12 mm thickness, before the sintering for 2 hours at temperatures 1230 ^{o}C and 1260 ^{o}C. The relative densities of the sintered samples were measured using a water immersion technique.

The $Ba_{1-x}Sr_xTiO_3$ polycrystalline samples were characterized by X-ray diffraction (XRD), on a Seifert Debye Flex 2002 Diffractometer in the 2θ-θ mode using Cu K_α radiation and Ni-filter. The sample morphology was investigated by using Scanning Electronic Microscopy (SEM).

Low frequency dielectric measurements were performed on the samples of 1 mm thick by using a self-acting RLC bridge at 1 kHz. The obtained samples were cut, cleaned and then treated at 150 ^{o}C for 15 hours in order to eliminate residual water from the porous structure. Then, silver paste was painted on the polished samples. A special treatment at 120 ^{o}C was applied for 3 hours before every set of measurements. Some samples were measured in the range –200 ^{o}C to + 100 ^{o}C.

Microwave characteristics were obtained by using the Hakki–Coleman method. Samples of 9 mm diameter and 7.5 mm height were positioned in a Courtney holder. The microwave measurements were performed using a computer aided measurement system in the 1÷12 GHz frequency bandwidth. The system includes a HP 8757C network analyzer and a HP 8350B sweep generator.

3. Results and discussions

Following the sintering process at two different sintering temperatures T_s = 1230 ^{o}C and T_s = 1260 ^{o}C, samples with a good compactness were obtained for x = 0.25, 0.5, 0.75 as shown in Table 1. Exceptions are the samples with a very high strontium content (x = 0.9), which would have required a sintering temperature higher than 1260 ^{o}C.

Table 1. BST samples.

Sample	Sr content (x)	Sintering Temperature T_S (^{o}C)	Density (g/cm^3)
BST 25	0.25	1260	5.17
BST 50	0.50	1260	5.15
BST 75	0.75	1260	4.67
BST 90	0.90	1260	2.95
BST 25	0.25	1230	5.13
BST 50	0.50	1230	4.86
BST 75	0.75	1230	4.85
BST 90	0.90	1230	2.85

The X-ray diffraction (XRD) patterns confirmed the perovskite structure of the BST ceramics. Morphological investigation by scanning electron microscopy (SEM) revealed a ceramic material with glassy aspect in fracture, with grains incorporated in vitreous mass as shown in Fig. 1. The grains with irregular, mostly spherical shapes have a granular distribution with sizes in the range 3÷5 μm.

They are arranged in conglomerates with dimensions between 10 and 50 μm. Two kinds of pores are present: spherical pores with diameter of 3÷5 μm and long pores with length of 10÷20 μm and width of 2÷4 μm.

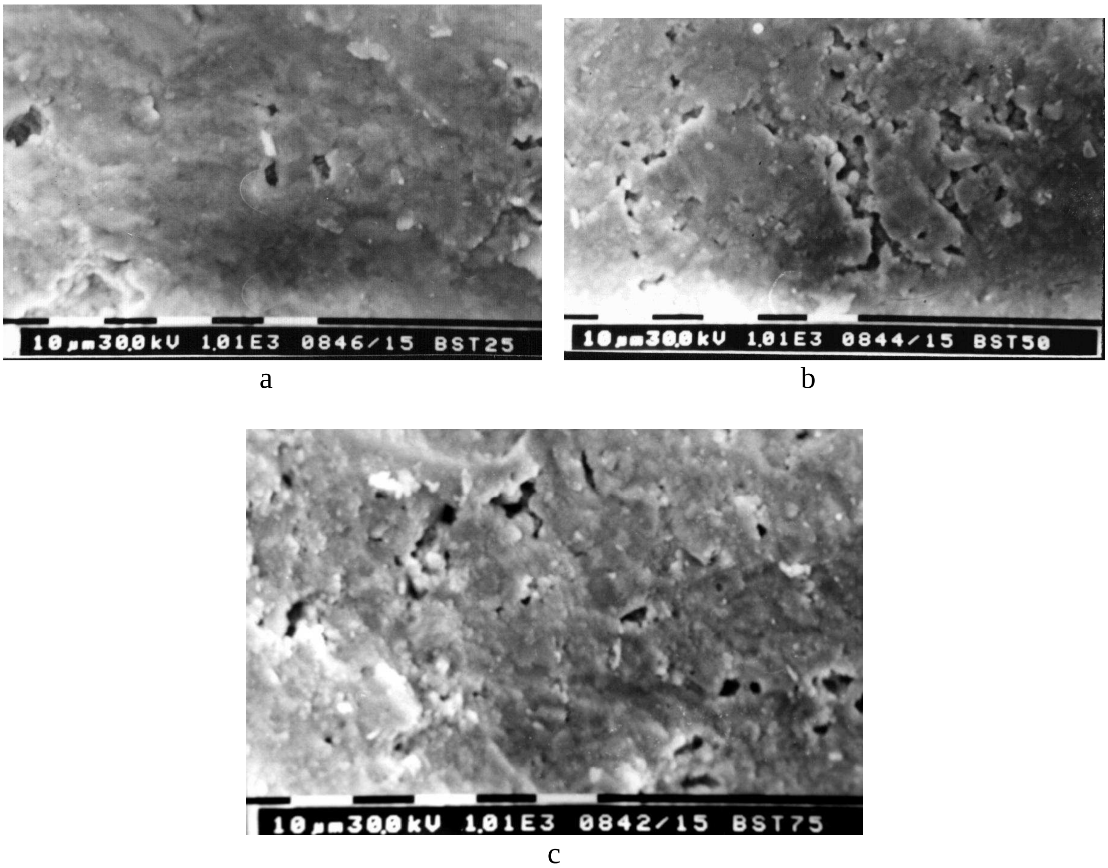

Fig. 1. SEM images for BST samples sintered at TS=1260 °C / 2h; (a) BST25; (b) BST50; c) BST75. Markers are of 10 μm.

Dielectric measurements show that Curie temperature slightly increases with the sintering temperature at the same Sr concentration. Two distinct peak values of the permittivity appear for BST 25 samples sintered at 1260 °C as shown in Fig. 2. The effect seems to be related to the distribution of crystallite size [7], centered on two main dimensions 3÷5 μm and 10÷50 μm. This effect decreases with the Sr concentration increase as shown in Fig. 2.

The dependence of the dielectric constant on temperature shows in Fig. 2 that the peak dielectric constant at 1 kHz increases with the Sr concentration decrease. The peak dielectric constant measured on samples sintered at T_s=1260 °C decreases from $\varepsilon_{r\ peak}$ = 2769 for BST 25 (x = 0.25) to $\varepsilon_{r\ peak}$ = 2337 for BST 75 (x= 0.75) as shown in the plots on the right side of Fig. 2. Also, the peak dielectric constant increases with the sintering temperature increase. For BST50 sample, $\varepsilon_{r\ peak}$ increases from 1737 to 2450, when the sintering temperature increases from T_S=1230 °C to T_S = 1260 °C.

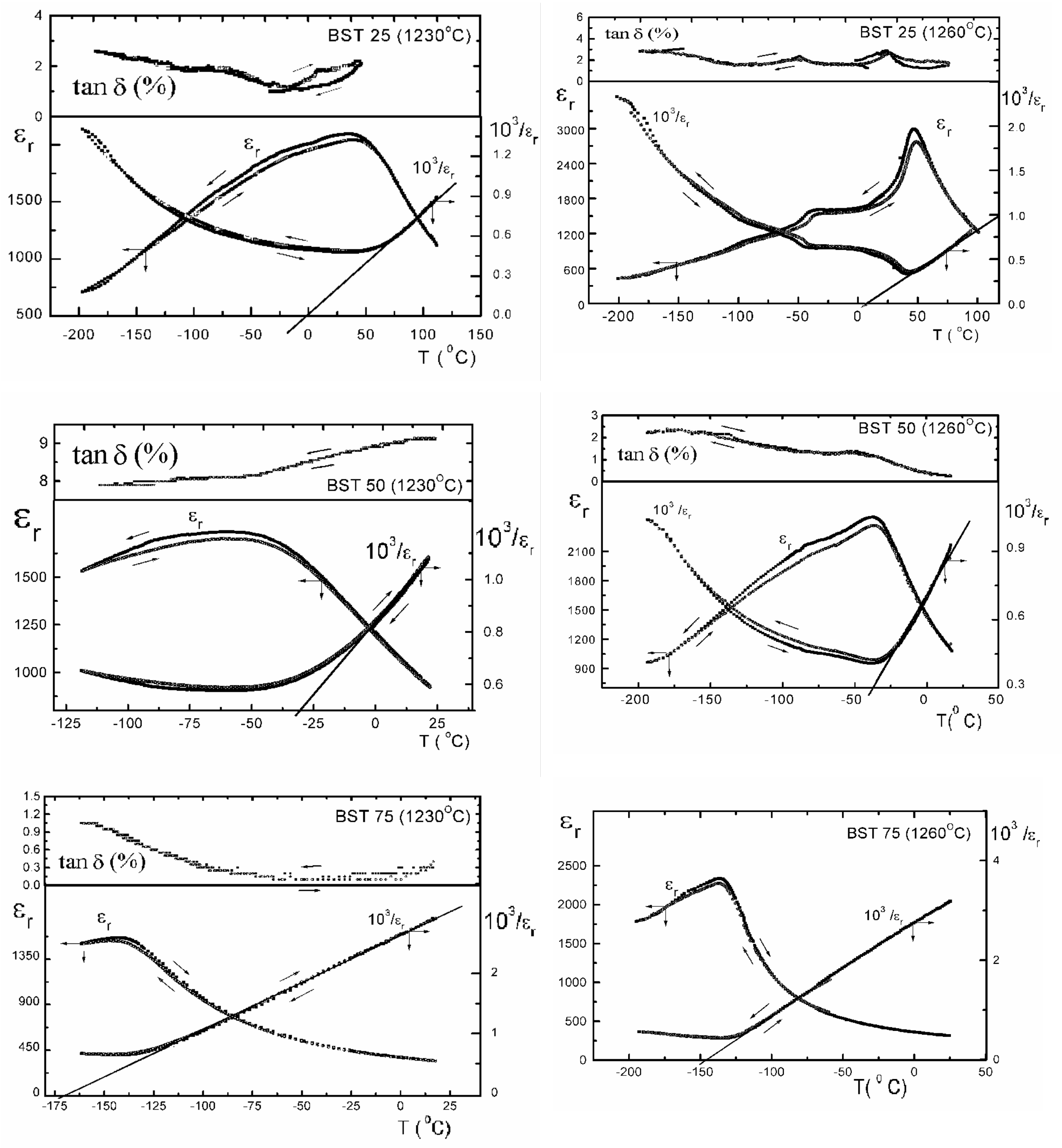

Fig. 2. Dielectric measurements at 1 kHz for BST samples sintered at Ts=1230 °C (left) and for samples sintered at Ts=1260 °C (right).

The microwave investigations on BST samples revealed high dielectric constant for samples with low strontium content up to ε_r = 1746 for BST25 and up to ε_r = 1074 for BST50 as shown in Table 2. However, the BST25 samples exhibit high losses 0.12÷0.14. Therefore, the samples BST50 and BST75 seem to offer a good compromise between dielectric constant as high as 320÷1074 and dielectric loss as low as $(1.8÷4.6)\times10^{-3}$ measured at 1.1 GHz.

Table 2. Dielectric parameters at room temperature.

Sample	ε_r (1 kHz)	tg δ, (%) (1 kHz)	T_c (^{o}C)	ε_r (1GHz)	tg δ, (%) (1 GHz)
BST 25 (T_s=1230 ^{o}C)	2080	0.12	37	1746	12
BST 50 (T_s=1230 ^{o}C)	928	0.91	-59	910	0.82
BST 75 (T_s=1230 ^{o}C)	343	0.08	-143	320	0.18
BST 25 (T_s=1260 ^{o}C)	1873	0.125	49	1530	14
BST 50 (T_s=1260 ^{o}C)	1077	0.25	- 37	1074	0.46
BST 75 (T_s=1260 ^{o}C)	314	0.20	- 136.5	208	0.20

4. Conclusions

Ternary system BaO-SrO-TiO_2, with the molar formula $(Ba_{1-x}Sr_x)TiO_3$ were prepared by solid-state reaction from raw materials, with strontium content x = 0.25, 0.50, 0.75, 0.9.

BST samples sintered at 1230^{o}C and 1260^{o}C exhibit a good compactness with the exception of BST90 samples, which do not show resonance mode in microwave and would require a sintering temperature higher than 1260 ^{o}C.

The low frequency (1 kHz) dielectric measurements revealed the decrease of the Curie temperature and of the peak permittivity with the increase of strontium concentration. Also, higher Curie temperatures were measured for BST samples sintered at 1260^{o}C compared to BST probes sintered at 1230 ^{o}C.

Microwave measurements at 1.1 GHz on dielectric resonators made of BST materials revealed high dielectric constants for samples with low strontium content (x = 0.25, 0.50). However, samples with x = 0.25 exhibit high dielectric loss 0.12-0.14 at 1.1 GHz. Materials with high Q-factors, such as BST50 with tan $\delta = 4.6\times10^{-3}$ or BST75 tan $\delta = 1.8\times10^{-3}$, offer an attractive solution for applications, such as tuneable microwave devices.

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