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$BaTiO_3$ (BTO)–$CaCu_3Ti_4O_{12}$ (CCTO) substrates for microwave devices and antennas

A. F. L. Almeida · P. B. A. Fechine · L. C. Kretly · A. S. B. Sombra

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Abstract The solid state procedure was used to produce bulk ceramics of BTO ($BaTiO_3$), CCTO ($CaCu_3Ti_4O_{12}$) and $BTO_{0.5}$–$CCTO_{0.5}$ that were studied in the medium-frequency (MF) range (100 Hz–1 MHz) and in the microwave range of frequencies. The presence of BTO is decreasing the dielectric constant (ε_r) of the BTO–CCTO composite. The CCTO and BTO samples present a strong tendency to the increase of the loss with frequency. The BTO substrates are presenting higher values of the ε_r in the range of 1–4 GHz (around 140). For pure CCTO the dielectric constant is around 37.6. Similar behaviour observed at the MF range, that the higher dielectric constant is also associated to the higher loss is also present in the microwave region. The study of a planar microstrip antenna, that uses the BTO_X–$CCTO_{(1-X)}$ ceramic as a high ε_r substrate was done. Therefore, these measurements confirm the potential use of such materials for small high dielectric planar antennas (HDA). These materials are also very promising for capacitor applications and certainly for microelectronics, microwave devices (cell mobile phones for example), where the miniaturization of the devices is crucial.

A. F. L. Almeida · P. B. A. Fechine
Departamento de Química Orgânica e Inorgânica, Centro de Ciências, UFC, Fortaleza, Ceará, Brazil

L. C. Kretly
Departamento de Microondas e Óptica (DMO), Faculdade de Engenharia Elétrica e Computação (FEEC), UNICAMP, SaoPaulo, Brazil

A. S. B. Sombra (✉) · A. F. L. Almeida · P. B. A. Fechine
Laboratório de Telecomunicações e Ciência e Engenharia dos Materiais (LOCEM), Departamento de Física, UFC Campus do Pici, Caixa Postal 6030, CEP 60 455-760 Fortaleza, CE, Brazil
e-mail: sombra@fisica.ufc.br

Introduction

High dielectric constants have been found in oxides of the type $CaCu_3Ti_4O_{12}$ [1–3] (CCTO) which shows a dielectric constant (ε_r) at 1 kHz of about 10,000 that is nearly constant from room temperature to 300 °C. Oxides with the perovskite structure are well stabilised by their high dielectric constants (ε_r) which lead these class of materials to a big number of technological applications [4]. However, this behaviour is generally associated to ferroelectric or relaxor properties. In these cases the highest value of ε_r is obtained during a phase transition (as a function of temperature) presented by the material.

The existence of the transition temperature event is generally a problem when one is talking about applications of these materials. The reported results for CCTO shows that the ε_r is high but with small dependence on the temperature [3].

Such material is very promising for capacitor applications and certainly for microelectronics, microwave devices (cell mobile phones for example), where the miniaturization of the devices are crucial.

High ε_r ceramics make it possible to noticeably miniaturize passive microwave devices. Their size can typically be reduced in comparing with classical resonators and filters by a factor of $\sqrt{\varepsilon_r}$ (relative dielectric constant).

The structure of CCTO was previously determined from neutron powder diffraction data [5]. It belongs to space group Im3 (No. 204) [5]. Recently we propose the use of mechanical alloying to produce CCTO [6]. The mechanical alloying is proving to be a powerful technique to obtain any quantity of powder with controlled microstructure [7]. Recently a polymeric citrate precursor route was used to produce CCTO [8]. It was observed dielectric constant of 3000 and loss around 0.3–0.35 at 1 kHz.

Barium titanate ($BaTiO_3$-BTO), a well known ferroelectric material, has a high dielectric constant (ε_r) around 6000 at a fine grain size of ~1 μm, and of 1500–2000 at a coarse grain size [9, 10]. The ε_r value of BTO at the Curie temperature of 120 °C can reach a peak value as high as 10,000, but decrease as the temperature decreases. Chemical additives or so called shifters have been applied to BTO to move the Curie peak value towards room temperature to improve the ε_r value, and to smooth the Curie peak to obtain a lower temperature coefficient of the ε_r.

Dielectric properties of BTO ceramics are highly dependent upon the grain size, phase content of the ceramic body and also the type of dopants used. For BTO powder, its behavior is also related to the particle size, phase content, and the dopants added. Cubic BTO (c-BTO) powder transforms to the tetragonal phase (t-BTO) if its size is larger than 30 nm [11]. Powder with a size of 40–80 nm contains a single domain. Crystallites with a size larger than 80 nm will be miltidomain t-BTO [12]. The increase of the dielectric constant is possibly caused by a summation of the domain size and the stress effect [13]. It is also mentioned that the width of ferroelectric 90° domains decrease proportional to the square root of the grain diameter for grains with size <10 μm [13]. The removal of grain boundaries, i.e. elimination of constrained forces from neighboring grains and a drop in domain density as the particle size decreases, may reduce the ε_r value of the BTO powders. With increasing particle size, unsintered powder eventually becomes a ceramic-like body and possesses similar properties as the ceramics [14]. Therefore, ceramics and powders of BTO can show a different dielectric behavior, depending on the particle size.

In this paper we report the preparation of the composite ceramic of BTO_X–$CCTO_{1-X}$ (with X = 0, 0.5 and 1 where it represents the presence of each phase (100 × %)) through the solid state route and use the bulk ceramic as a substrate for a planar microwave antenna application. The produced samples were studied using X-ray diffraction and the dielectric constant and loss were also studied in the MF range (100 Hz to 1 MHz) and in the microwave range.

The production, and the study of the properties of the BTO–CCTO ceramics is important in view of possible applications as bulk devices like microwave resonators and oscillators, thick and thin high ε_r films.

Experimental procedure

Sample preparation

Commercial oxides $Ca(OH)_2$ (Vetec, 97% with 3% of $CaCO_3$), titanium oxide (TiO_2) (Aldrich, 99%), CuO (Aldrich, 99%) were used in the CCTO preparation. The material was ground on a Fritsch Pulverisette 6 planetary mill with the proportionality of $Ca(OH)_2$–3CuO–$4TiO_2$. Milling was performed in sealed stainless steel vials and balls under air. Mechanical alloying was performed for 1 h of milling. In this case the milling was used only to give a good homogeneity of the powder. However, we already showed in the literature that for 100 h of milling the complete production of CCTO is possible [6]. The reaction occurring during milling can be summarised as:

$$Ca(OH)_2 + 3CuO + 4TiO_2 \xrightarrow{\text{Impacts}} CaCu_3Ti_4O_{12} + H_2O$$

The ceramic was submitted to calcination and sintering in air in the range of 900–1100 °C for 12 h. This ceramic is called CCTOCS (calcination + sintering). The pellets were then sintered at 1050 °C for 24 h. The denomination for these samples will be CCTO (see Table 1).

BTO powder preparation

The used BTO powder was a commercial product ($BaTiO_3$-Aldrich). This ceramic is called BTO.

In this paper one has three types of bulk ceramics: BTO (100% BTO), CCTO (100% CCTO) and $BTO_{0.5}$–$CCTO_{0.5}$ (In this ceramic one has 50% of each component) where the percentage is given in weight %. The BTO bulk ceramic was prepared from the commercial powder and the ceramic was sintered at 980 °C for 24 h. For the composite ceramic ($BTO_{0.5}$–$CCTO_{0.5}$) the samples were also sintered at 980 °C for 24 h.

XRD

The X-ray diffraction (XRD) patterns were done using a Siemens D5000 equipment with CuK_α radiation in a Bragg–Brentano geometry at room temperature (300 K) by step scanning using powdered samples. We used five seconds for each step of counting time, with a CuK_α tube at 40 kV and 25 mA.

Table 1 Medium frequency(MF) measurements of the dielectric constant and dielectric loss of the samples(f =1 kHz)

Sample	Thickness e (mm)	Electrode diammeter ϕ (cm)	Dielectric loss (D) (10^{-2}) $D = \varepsilon''/\varepsilon'$ 1 kHz	Dielectric constant $\varepsilon_r = \varepsilon'/\varepsilon_0$ 1 kHz
CCTO-a	2.12	3.6	20	7370
CCTO-b	2.12	4.2	22	7073
$BTO_{0.5}$–$CCTO_{0.5}$-a	0.91	1.09	19	668
$BTO_{0.5}$–$CCTO_{0.5}$-b	1.05	1.07	45	596
BTO	1.04	1.15	1.5	552

Electrical measurements

The dielectric and loss measurements were obtained from a HP 4291A Impedance Analyzer, which cover the region of 100 Hz–1 MHz. The dielectric permittivity (ε_r) and dielectric loss (D) measurements, were performed using a parallel plate capacitor arrangement. The samples were formed into thin circular disks with diameter around ϕ = 1–5 cm and thickness around e = 0.9–2.2 mm (see Table 1). The Ag (circular electrodes) were screen printed at each surface and fired at 400 °C for 1 h.

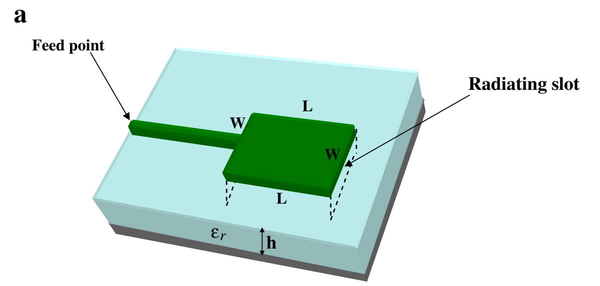
The resonance measurements in the range of 9 kHz–4 GHz range was done in a conventional set up, measuring the S_{11} parameter using a HP 8714 ET or ZVRE (Rohde & Schwarz) Network Analyzers.

The used samples for the low frequency region (100 Hz–1 MHz) and for the microwave region are described in Tables 1 and 2. For the three different samples: BTO, CCTO and $BTO_{0.5}$–$CCTO_{0.5}$, one has different geometries for the measurements. For the samples in the same phase, the preparation procedure was the same. The only difference is associated to the sample geometry (like CCTO-a and CCTO-1).

In the low frequency region we have samples CCTO-a, and CCTO-b, $BTO_{0.5}$–$CCTO_{0.5}$-a, $BTO_{0.5}$–$CCTO_{0.5}$-b and BTO (see Table 1). For the microwave region we have samples CCTO-1, 2 and 3 and $BTO_{0.5}$–$CCTO_{0.5}$-1, 2, 3 and BTO-1, 2, 3 (see Table 2).

Model for microstrip radiator

The test structure used for dielectric constant measurements is a resonant-style radiator shown in Fig. 1a, b. The patch antenna is fed by a microstrip line in direct contact to the patch conductor of length L and width W. There are several models related to the planar patch antenna analysis. Among them are: transmission line model, cavity model and full-wave analysis [12]. Microstrip antennas resemble dielectric loaded cavities. A well accepted model detailed in [12, 13]. Due to the field configuration at the edges, the antenna has two radiation edges and two resonant edges. The radiation of the antenna takes place from the two slots comprising between the W edges and ground plane. The L edges are the resonant dimensions (see Fig. 1a). The transmission line model approaches the microstrip antenna as two slots separated by a low impedance transmission line (the patch) of length L. For the dominant mode TM_{010} which is the dominant mode with the lowest frequency, the resonant frequency is given by [13]



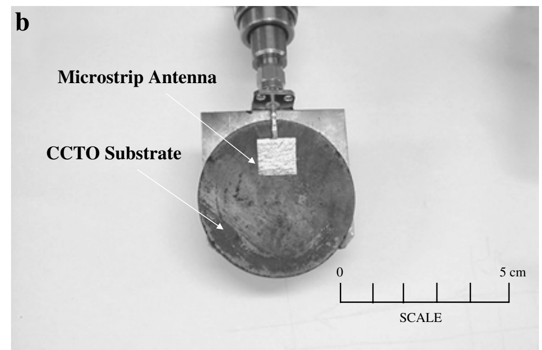


Fig. 1 (**a**) The microstrip antenna configuration for modeling and analysis. (**b**) Planar microstrip antenna on CCTO substrate for 3 GHz operation

$$f_r = \frac{c}{2(L + 2\Delta L)\sqrt{\varepsilon_{\mathrm{reff}}}} \tag{1}$$

where ΔL is the additional line length to account the fringing fields which has a practical approximation given by [13]:

Table 2 Microwave measurements of the samples

Planar antenna measurements Samples	Thickness e (mm)	Sample diameter ϕ (cm)	Resonant frequency—f_R (GHz)	$\varepsilon_{\mathrm{eff}}$	ε_r	Q^{-1} (10^{-2})
CCTO–1	2.61	4.7	2.95	30.8	41.6	10
CCTO-2	2.29	4.7	2.9	25	33.7	3
$BTO_{0.5}$–$CCTO_{0.5}$-1	2.47	4.63	1.93	60.4	80	13.9
$BTO_{0.5}$–$CCTO_{0.5}$-2	2.46	4.66	2.02	55.1	73	20.7
$BTO_{0.5}$–$CCTO_{0.5}$-3	2.53	4.68	1.87	64.3	85.5	13
BTO-1	2.44	4.98	1.29	135.2	179.3	113
BTO-2	2.3	5	1.55	93.6	123.2	172
BTO-3	2.46	4.98	1.59	88.9	118	167

$$\frac{\Delta L}{h} = 0.412\frac{(\varepsilon_{\mathrm{reff}}+0.3)\left(\frac{W}{h}+0.264\right)}{(\varepsilon_{\mathrm{reff}}-0.258)\left(\frac{W}{h}+0.8\right)} \tag{2}$$

and the $\varepsilon_{\mathrm{reff}}$ is the effective dielectric constant (where effective means: a composite value of the overall dielectrics in the system which involves the dielectric constant of material itself and the air). In general $1 < \varepsilon_{\mathrm{reff}} < \varepsilon_{\mathrm{r}}$, the fringing effects on the microstrip gives:

$$\varepsilon_{\mathrm{reff}}(W) = \frac{\varepsilon_{\mathrm{r}}+1}{2} + \frac{\varepsilon_{\mathrm{r}}-1}{2}F(W/h) \tag{3}$$

and

$$F(W/h) = \begin{cases} (1+12h/W)^{-1/2} + 0.04(1-W/h)^2 & W/h \le 1 \\ (1+12h/W)^{-1/2} & W/h \ge 1 \end{cases} \tag{4}$$

For the measurements in this work we have used a square microstrip antenna with $W = L = 1$ cm.

The resonant frequency is precisely measured using a Network Analyser and measuring the return loss (in dB) through the S_{11} scattering parameter of the antenna under test. The very precise resonant frequency identification is done trough the lowermost point in the S_{11} curve (see Fig. 2). The S_{11} test is the main test for antennas. This parameter shows the reflections of the signal at the feeding point, the entrance of the antenna. High negative values (typically between −12 and −40 dB) in the dip (see

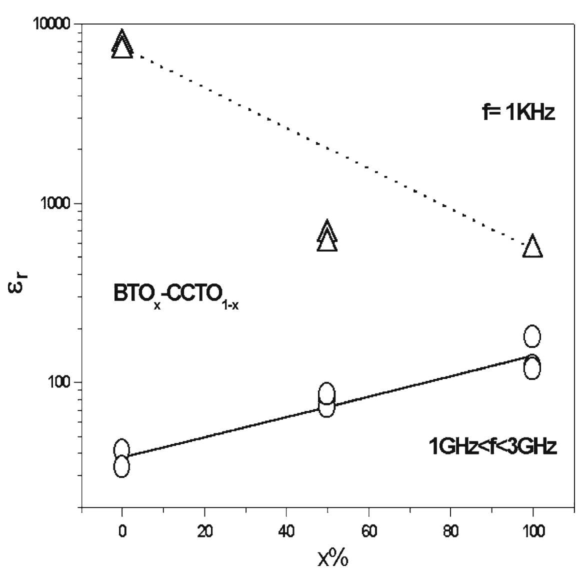


Fig. 2 Dielectric permittivity for 1 kHz and in the microwave range. Dotted line (f = 1 kHz) and continuous line (1 GHz < f < 3 GHz) were obtained from equation. Experimental points are (Δ) for 1 kHz, and in the microwave range (O), 1 GHz < f < 3 GHz

Fig. 3a–c) indicate that the electromagnetic waves were not reflected. It means that the antenna is radiating. In the −10 dB line, there is an antenna bandwidth, BW, (almost equal to VSWR : 2:1). With this data the antenna personnel might be able to estimate the radiation performance to a certain degree.

The ε_{r} value is extracted from the expressions above with the correction related to the fringing fields. There are other methods for ε_{r} measurements but this one is straightforward because the test structure is close to the final application structure.

Essentially ε_{r} is determined from the resonant frequency at f_{R} and Q^{-1} (the loss factor) is determined from $\Delta f/f_{\mathrm{R}}$, where Δf (BW) is the 10 dB bandwidth of the resonance (see Table 1).

Results and discussion

Figure 4 shows the X-ray diffraction (XRD) patterns of the substrates BTO, CCTO and $BTO_{0.5}$–$CCTO_{0.5}$ together with the XRD of the references (JCPDS), obtained from the literature. In the same figure one starts with the XRD of the BTO substrate. For sample $BTO_{0.5}$–$CCTO_{0.5}$ the presence of BTO and CCTO is easily identified and for the ceramic CCTO, the presence of CCTO is quite clear

In Figs. 5 and 6 one has the dielectric permittivity (ε_{r}) of all the samples (CCTO-1,CCTO-2, $BTO_{0.5}$–$CCTO_{0.5}$-a, $BTO_{0.5}$–$CCTO_{0.5}$-b, and BTO) in the range of 100 Hz–1 MHz. For all the samples there is a decrease of the ε_{r} value for this range of frequency. For the CCTO-1 and CCTO-2 samples, the dielectric constant is high. For samples CCTO-1 and CCTO-2 the value at 100 Hz, ε_{r} ~10,000 decreases to 4000 at 1 MHz. In Fig. 7 one has the dielectric loss for these two samples. At low frequencies (1 kHz) the samples CCTO-1 and CCTO-2 present the loss tangent (D) around 0.2 (see also Table 1). The loss is increasing with the frequency and is around 0.35 and 0.6 for the same samples around 1 MHz. The increase of the loss for the CCTO samples with frequency was reported in the literature by others authors [8]. In Table 1 one has the dielectric constant and loss for all the samples at 1 kHz.

In Fig. 6 one has the dielectric permittivity (ε_{r}) of the samples ($BTO_{0.5}$–$CCTO_{0.5}$-a, $BTO_{0.5}$–$CCTO_{0.5}$-b, and BTO) in the range of 100 Hz to 1 MHz. One can observe that for the BTO sample the dielectric constant (ε_{r}) is quite stable with frequency. It is around 552 at 1 kHz. However, for the samples where the BTO and CCTO form a composite ceramic, the dielectric constant is also decreasing with frequency. One can conclude that the presence of the BTO is decreasing the dielectric constant (see Table 1) for f = 1 kHz. However, the presence of BTO is giving to the

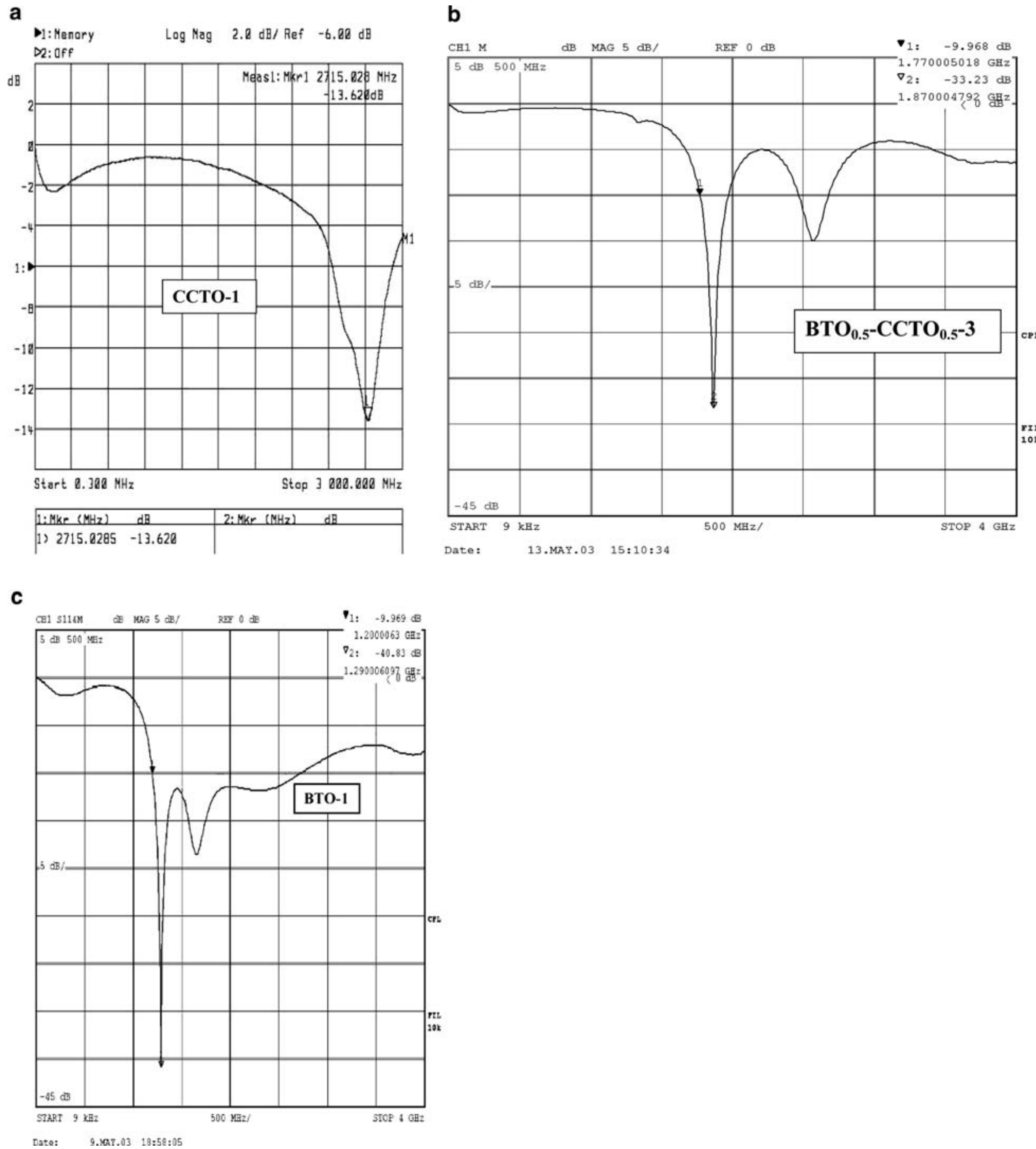


Fig. 3 (**a**) The resonant frequency of the microstrip antenna of sample CCTO-1, measured trough the S_{11} parameter (return loss). The bandwidth is obtained at −10 dB which correspond to VSWR < 2:1. (**b**) The resonant frequency of the microstrip antenna of sample $BTO_{0.5}$ $CCTO_{0.5}$-3, measured trough the S_{11} parameter (return loss). The bandwidth is obtained at −10 dB which correspond to VSWR < 2:1. (**c**) The resonant frequency of the microstrip antenna of sample BTO-1, measured trough the S_{11} parameter (return loss). The bandwidth is obtained at −10 dB which correspond to VSWR < 2:1

CCTO a low decrease rate for the dielectric constant with frequency. Around 10 kHz there is an inversion of values for ε_r. The BTO values are higher compared to the BTO–CCTO samples for higher frequencies. One can expect that at higher frequencies the BTO values should be bigger. This kind of behavior will be very critical at higher frequencies (microwave range).

In Fig. 8 one has the dielectric loss for these three samples. At low frequencies the samples $BTO_{0.5}$–$CCTO_{0.5}$-a and $BTO_{0.5}$–$CCTO_{0.5}$-b present higher loss compared to

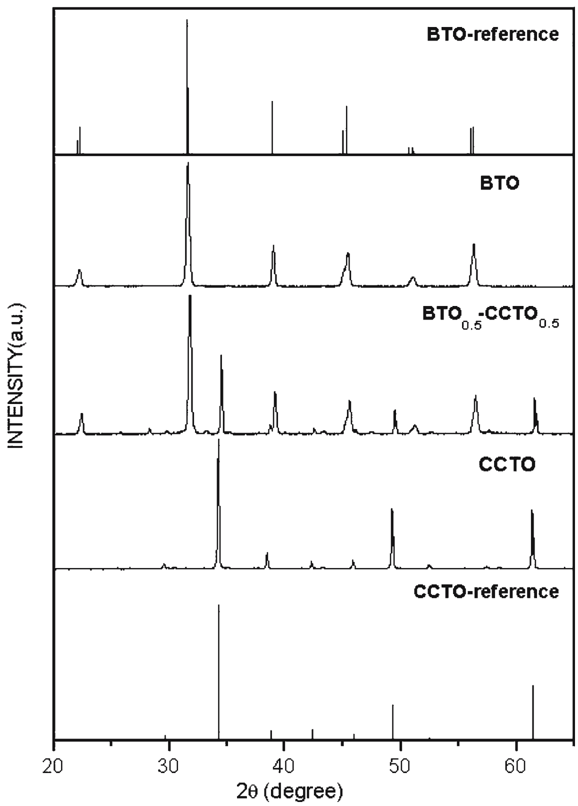


Fig. 4 Comparison of the XRD of the samples BTO, CCTO and $BTO_{0.5}$–$CCTO_{0.5}$ with the BTO and CCTO references

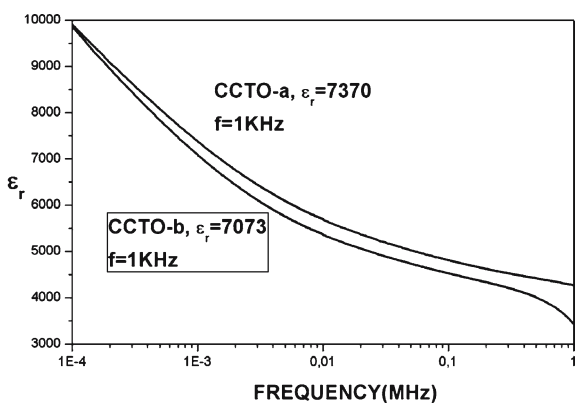


Fig. 5 Dielectric constant (ε_r) of samples CCTO-a and CCTO-b in the frequency range of 100 Hz–1 MHz

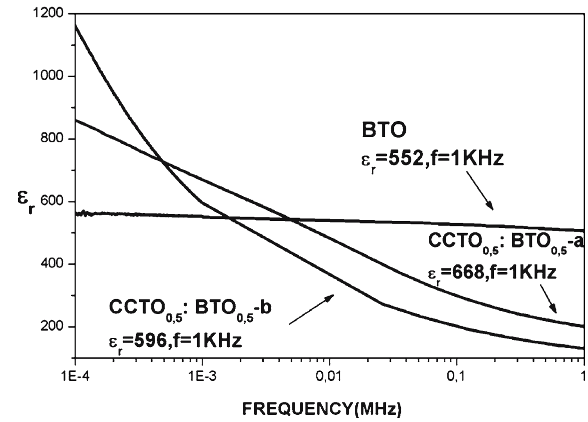


Fig. 6 Dielectric constant (ε_r) of samples $BTO_{0.5}$ $CCTO_{0.5}$-a, $BTO_{0.5}$ $CCTO_{0.5}$-b and BTO in the frequency range of 100 Hz–1 MHz

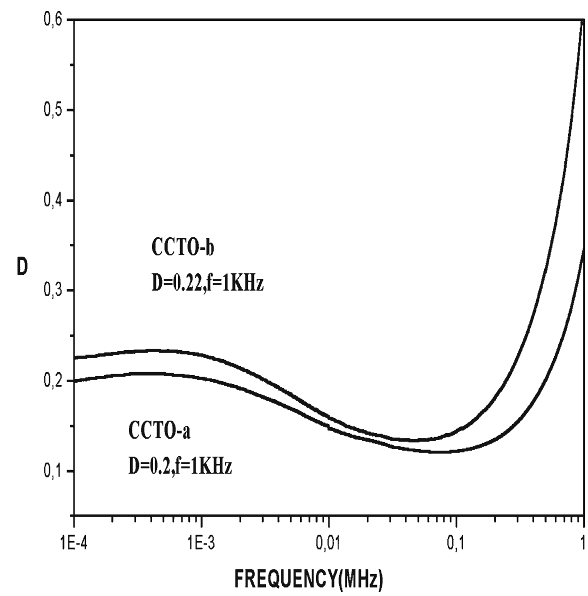


Fig. 7 Dielectric loss ($D = tg\alpha$) of samples CCTO-a and CCTO-b in the frequency range of 100 Hz–1 MHz

the BTO sample. However, the previous behavior obtained for the CCTO samples where the loss was increasing with frequency is no longer observed.

The loss tangent (D) is decreasing with the frequency for samples $BTO_{0.5}$–$CCTO_{0.5}$-a and $BTO_{0.5}$–$CCTO_{0.5}$-b. The BTO sample present the lowest loss obtained for all the samples, it is around 0.015 at 1 kHz. However, there is a little tendency of increasing with frequency. Comparing the BTO sample and the BTO–CCTO samples, one can expect that at higher frequencies the BTO substrate could have higher loss compared to the composite ceramic.

In Table 1 one has the dielectric constant and loss for all the samples at 1 kHz. In summary one can say that the presence of BTO is decreasing the dielectric constant (ε_r) from an average value of 7222 (CCTO) to an intermediate value of 632($BTO_{0.5}$–$CCTO_{0.5}$), and finally to 552(BTO).

The CCTO samples present a strong tendency to the increase of the loss with frequency, which is also happen to the BTO sample however with much lower velocity and from very low values of this loss. The composite ceramic ($BTO_{0.5}$–$CCTO_{0.5}$) is presenting a tendency to decrease the loss with frequency, which seems to be a mixing effect of the two phases.

To investigate the potential application of the BTO_X–$CCTO_{1-X}$ materials for microwave planar devices, a series

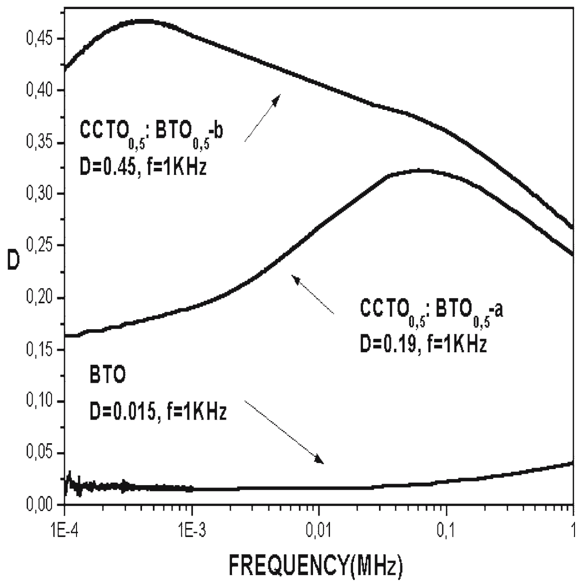


Fig. 8 Dielectric loss ($D = tg\alpha$) of samples $BTO_{0.5}$ $CCTO_{0.5}$-a, $BTO_{0.5}$ $CCTO_{0.5}$ -b and BTO in the frequency range of 100 Hz–1 MHz

of planar microstrip antennas were done. The demand for new mobile communication systems will push hard to miniaturization and low volume devices and equipment. Due to these requirements in portable, or repeater stations in a mobile communication for cellular systems, for example, high dielectric constant materials can be used to effectively reduce the size of planar microstrip antennas [11].

The performance of a planar antenna is related to the L and W dimensions of the patch and the dielectric constant ε of the substrate. For the best compromise between antenna gain, efficiency, bandwidth and volume, an adequate material with a high ε must be found for low volume. Of course, there is a trade off: high dielectric constant materials give a low volume antenna but it imposes low bandwidth and gain. Moreover, there is a demand for monolithic integration of antennas and associated circuitry and these requirements claim compatible high dielectric constant materials.

The simple rectangular antenna prototypes were designed on substrate samples with nominally 4–5 cm in diameter and 2–3 mm thick (see Table 2 and Fig. 1a, b). The resonant frequency (f_R) was obtained in the range of 1–4 GHz, based on the dielectric constant available data [1–3, 8]. One of the experimental antennas is shown in Fig. 1b (sample CCTO).

All the samples show the electromagnetic radiator potential properties, that is, antenna. This is proved when the dips in the S_{11} measurements reach lower values than −10 dB. The −10 dB point in the S_{11} parameter corresponds to a VSWR of 2:1. The S_{11} parameters for the antennas (return loss) were measured, and the results are shown in Fig. 3a–c for the CCTO-1, $BTO_{0.5}$–$CCTO_{0.5}$ -3 and BTO-1 antennas (as examples of all the eight studied antennas). The VSWR < 2:1 (−10 dB line) criterion was used to identify the antenna bandwidth and the dip in the S_{11} measurement below −10 dB is a preliminary indicator of the electromagnetic radiation properties of the device.

In Table 2 one has the general characteristics of all the studied antennas. In this table one can find the geometry of the substrate (eight samples). In Table 2 one has also the values of the ε_r (in the microwave region), ε_{eff} (effective dielectric constant), and the loss factor Q^{-1} (obtained from the S_{11} and is given by $Q = f_R/BW$).

From Table 2 one can conclude that the higher values of the ε_r in the range of 1–4 GHz antennas is presented by the BTO substrates. For the three BTO samples the average value for ε_r is around 140. With the presence of CCTO in the samples ($BTO_{0.5}$–$CCTO_{0.5}$) this average value of the ε_r decrease to 79.5. For pure CCTO the dielectric constant is around 37.6. The highest value for the dielectric constant was obtained for the BTO-1 sample, which was around 179. The lowest one was obtained for the CCTO-2 sample (33.7), see Table 2. This sample also presents the lowest value of the loss factor (Q^{-1}) which is around $3 \cdot 10^{-2}$.

It was observed that the highest value for the dielectric constant is also associated with high loss factor (Q^{-1}). The highest loss factor (Q^{-1}) was obtained for the BTO-2 sample, which is around $172 \cdot 10^{-2}$ (see Table 2).

Similar behaviour observed at low frequencies, that the higher dielectric constant is also associated to the higher loss is also present in the microwave region.

The higher value for ε_r obtained for the BTO substrate compared to the BTO–CCTO substrate is in agreement with the low frequency measurements (see Fig. 6) where the BTO value for ε_r was bigger compared to the BTO–CCTO substrate for frequencies over ~10 kHz. The higher value obtained for the loss factor (Q^{-1}) obtained for BTO compared to the BTO–CCTO substrates is also in agreement to the low frequency measurements that shows a tendency of increasing and decreasing loss (D) with frequency, for BTO and BTO–CCTO, respectively.

One can conclude that the presence of CCTO in the BTO ceramic is presenting quite distinct behaviour when one compares the MF range and microwave regions of the spectra. In the low frequency region the increase of the CCTO presence is increasing the dielectric constant (up to 1 kHz frequency). However, in the microwave region one has the opposite behaviour. The presence of the CCTO phase is decreasing the ε_r value (see Tables 1 and 2) and the loss factor.

We have to emphasize that the loss factor obtained in the present device is associated to an open cavity. In this case the substrate loss and the radiating loss are operating together.

The classic treatment applied for two or more phases present in a dielectric are associated to the dielectric mixing rules that fix a limit to the dielectric constant that can be achieved with the mixed phase. The empirical logarithmic rule for the dielectric constant (ε_r) and the dielectric constants (ε_{rI}) of the individual phases is given by [14]

$$\log \varepsilon_r = \sum_I V_I \log \varepsilon_{rI} \qquad (5)$$

In Fig. 2 one has the plot of the dielectric constant as a function of the substrate composition for the microwave region and for $f = 1$ kHz, based in Eq. 5.

The dotted line and continuous line are associated to the value of ε_r obtained from Eq. 5, at 1 kHz and in the microwave region, respectively.

The experimental points of the samples are also indicated in the figure. One can easily conclude that the addition of CCTO is increasing the ε_r of the substrate at 1 of frequency. However, for $x = 0.5$ the experimental value of the dielectric constant is much lower than (average value of $\varepsilon_r \sim 632$) compared to the expected value suggested in Eq. 5 ($\varepsilon_r \sim 2021$). This is an unexpected behaviour. It means that the composite is not following the linear regime of the model.

However, in the microwave range the addition of CCTO is decreasing the ε_r value and all the samples are in good agreement with the proposed model in the composite ceramic substrate (see Eq. 5 and Fig. 2).

In summary, three different substrates of BTO, CCTO and $BTO_{0.5}$–$CCTO_{0.5}$ were studied in the medium frequency (MF) and microwave range of frequencies.

The dielectric permittivity (ε_r) of all the samples in the range of 100 Hz–1 MHz were studied (MF). For all the samples there is a decrease of the ε_r value with frequency for this range of studied frequencies. For samples CCTO-1 and CCTO-2 the value at 100 Hz, $\varepsilon_r \sim 10{,}000$ decreases to 4000 at 1 MHz. At low frequencies these samples present the loss tangent (D) around 0.2. The loss is increasing with the frequency and is around 0.35 and 0.6 for the same samples around 1 MHz.

The dielectric permittivity (ε_r) of the samples ($BTO_{0.5}$–$CCTO_{0.5}$-a, $BTO_{0.5}$–$CCTO_{0.5}$-b, and BTO) were studied in the range of 100 Hz to 1 MHz. One can observe that for the BTO sample the dielectric constant (ε_r) is quite stable with frequency. It is around 552 at 1 kHz. However, for the samples where the BTO and CCTO form a composite ceramic, the dielectric constant is also decreasing with frequency. One can conclude that the presence of the BTO is decreasing the dielectric constant for $f = 1$ kHz. However, the presence of BTO is giving to the CCTO a lower decrease rate for the dielectric constant with frequency. Around 10 kHz there is an inversion of values for ε_r. The BTO values are higher compared to the BTO–CCTO samples.

The dielectric loss for the samples $BTO_{0.5}$–$CCTO_{0.5}$-a and $BTO_{0.5}$–$CCTO_{0.5}$-b are higher compared to the BTO sample.

Comparing the loss behavior with frequency for the BTO and the BTO–CCTO samples, one can expect that at higher frequencies the BTO substrate could have higher loss compared to the composite ceramic.

From the analysis of the antenna operation of the samples, one can conclude that the higher values of the ε_r in the range of 1–4 GHz antennas is presented by the BTO substrates. For the three BTO samples the average value for ε_r is around 140. With the presence of CCTO in the samples ($BTO_{0.5}$–$CCTO_{0.5}$) this average value of the ε_r decrease to 79.5. For pure CCTO the dielectric constant is around 37.6. The highest value for the dielectric constant was obtained for the BTO-1 sample, which was around 179. The lowest one was obtained for the CCTO-2 sample (33.7). This sample also presents the lowest value of the loss factor (Q^{-1}) which is around $3 \cdot 10^{-2}$.

The amazing result of this series is that the highest value for the dielectric constant is also associated with high loss factor (Q^{-1}). Similar behaviour observed at low frequencies, that the higher dielectric constant is also associated to the higher loss is also present in the microwave region.

Considering the classic treatment applied for two or more phases present in a dielectric and the empirical logarithmic rule for the dielectric constant (ε_r) and the dielectric constants (ε_{rI}) of the individual phases one conclude that in the microwave region of the spectra the value of the dielectric constant is in good agreement with the rule. However, in the low frequency region of the spectra the deviation is quite strong.

Conclusions

In conclusion, the traditional ceramic procedure was used to produce bulk ceramics of BTO ($BaTiO_3$), CCTO ($CaCu_3Ti_4O_{12}$) and $BTO_{0.5}$–$CCTO_{0.5}$ and were studied in the MF and microwave range of frequencies.

For all the samples there is a decrease of the ε_r value with frequency for the MF range of the of studied frequencies. For samples CCTO-1 and CCTO-2 the value at 100 Hz, $\varepsilon_r \sim 10{,}000$ decreases to 4000 at 1 MHz. At low frequencies these samples present the loss tangent (D) around 0.2. The loss is increasing with the frequency and is around 0.35 and 0.6 for the same samples around 1 MHz.

One can observe that for the BTO sample the dielectric constant (ε_r) is quite stable with frequency. It is around 552 at 1 kHz. However, for the samples where the BTO and CCTO form a composite ceramic, the dielectric constant is also decreasing with frequency. One can conclude that the presence of the BTO is decreasing the dielectric constant for $f = 1$ kHz. However, the presence of BTO is giving to the CCTO a lower decrease rate for the dielectric constant with frequency. Around 10 kHz there is an inversion of values for ε_r. The BTO values are higher compared to the BTO–CCTO samples. The dielectric loss for the samples $BTO_{0.5}$–$CCTO_{0.5}$-a and $BTO_{0.5}$–$CCTO_{0.5}$-b are higher compared to the BTO sample.

From the microstrip antenna operation of the samples, the highest value for the dielectric constant was obtained for the BTO-1 sample, which was around 179. The lowest one was obtained for the CCTO-2 sample (33.7). This sample also presents the lowest value of the loss factor (Q^{-1}) which is around $3 \cdot 10^{-2}$.

One can say that the presence of CCTO in the BTO ceramic is presenting quite distinct behaviour when one compares the MF range and microwave regions of the spectra. In the low frequency region the increase of the CCTO presence is increasing the dielectric constant (up to 1 kHz frequency). However, in the microwave region one has the opposite behaviour. The presence of the CCTO phase is decreasing the ε_r value and the loss factor.

In summary the performance of a planar microstrip antenna, that uses the BTO_X–$CCTO_{(1-X)}$ ceramic as a high ε_r substrate was examined. Therefore, these measurements confirm the potential use of such materials for small high dielectric planar antennas (HDA). These materials are also very promising for capacitor applications and certainly for microelectronics, microwave devices (cell mobile phones for example), where the miniaturization of the devices is crucial.

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