In this work the magnetic and dielectric properties of ceramic-ceramic and ceramic-polymer composites with BiNbO_4, SrBi_2Nb_2O_9 (SBN), BaBiTi_4O_15 (BBT), Na_2Nb_2O_6 (SCN) and ferrites BaFe_{12}O_{19} and Y_3Fe_5O_{12} (YIG) was studied for RF and microwave applications. New configurations of magneto-dielectric composites and blends structures for high frequency applications were done. A new method for the measurement of the temperature coefficient of resonant frequency ($\tau_f$) is presented. The traditional method (based on the Courteney method) present some limitations of measuring the values of $\tau_f$, for samples with high dielectric loss.
Final accomplishments:

In this work the magnetic and dielectric properties of ceramic-ceramic and ceramic-polymer composites with BiNbO$_4$, SrBi$_2$Nb$_2$O$_9$ (SBN), BaBi$_4$Ti$_4$O$_{15}$ (BBT), Na$_2$Nb$_4$O$_{11}$ (NNO), Sr$_2$CoNbO$_6$ (SCN) and ferrites BaFe$_{12}$O$_{19}$ and Y$_3$Fe$_5$O$_{12}$ (YIG) was studied for RF and microwave applications. New configurations of magneto-dielectric composites and blends structures for high frequency applications was done. The 0-3 type dielectric and magnetic composites with homogenously distributed ceramic inclusions was fabricated in a polymer matrix. Magnetic Yttrium Iron Garnet (YIG) and (SBN) powders were used to enhance the permittivity and permeability of the composites. This group of dielectric and magnetic phases was studied in the RF and microwave region. The microstructure, high frequency dielectric and magnetic properties of individual layers and 2-2 composites was investigated and measured.

A new method for the measurement of the temperature coefficient of resonant frequency ($\tau_f$), is presented. The traditional method (based on the Courtney method) present some limitations of measuring the values of $\tau_f$, for samples with high dielectric loss due to their inability to observe clearly the TE$_{011}$ mode. The new experimental setup, to measure the $\tau_f$ value, is based on the variation of the temperature of the dominant mode of a dielectric resonator antenna (DRA).

The study of the thermal stability of magneto-dielectric composites is important for applications at the microwave band and in the millimeter and near millimeter region (100-300GHz) where the thermal stability of the resonators is fundamental. In this project we are investigating experimentally and numerically this new method to measure the thermal stability of layered dielectric and magnetic composite structures for RF and Microwave Applications.

In the area of communication is important that the devices, responsible for transmitting/receiving data have its characteristics preserved in whatever temperature environment they are submitted. This new method for the measurement of the temperature coefficient of resonant frequency ($\tau_f$), is presented. The traditional Courtney method, present some limitations of measuring the values of $\tau_f$ for samples with high dielectric loss due to their inability to observe clearly the TE011 mode. The new experimental setup (figure below), to measure the $\tau_f$ value, is based on the variation of the temperature of the dominant mode of a dielectric resonator antenna.
Modified setup, for the measurement of $\tau_f$

A new method to measure the microwave thermal stability coefficient $\tau_f$

$$\tau_f = \frac{1}{f_i} \times \frac{\Delta f}{\Delta T} \times 10^6,$$

To use this new method a group of traditional materials were used to compare the traditional and new method

| TABLE I. TE$_{01\delta}$, HE$_{11\delta}$ e TM$_{01\delta}$ modes and dielectric parameters of CTO, Al$_2$O$_3$, and BTNO dielectrics. |
|-----------------|-----------------|-----------------|
|                  | CaTiO$_3$       | Al$_2$O$_3$     | BTNO            |
| a (mm)           | 7.48            | 12.70           | 7.31            |
| h (mm)           | 8.04            | 12.70           | 7.38            |
| a(mm)/h(mm)      | 0.93            | 1.00            | 0.99            |
| $\varepsilon_R$  | 92.25           | 9.80            | 63.68           |
| tan $\delta$     | $5.81 \times 10^{-4}$ | $1.11 \times 10^{-4}$ | $5.61 \times 10^{-2}$ |
| $f_{\text{monopole}}$ (GHz) measured | 1.888 | 3.089 | 2.439 |
| $f_{\text{HE11}}$ (GHz) calculated | 1.837 | 3.147 | 2.328 |
| $f_{\text{TE01}}$ (GHz) calculated | 1.830 | 3.201 | 2.288 |
| $f_{\text{TM01}}$ (GHz) calculated | 2.695 | 4.527 | 3.357 |

Used samples in the measurements

The comparative between the two systems for measurement of $\tau_f$ values, show excellent agreement, as observed in Figure 4. In the Courtney procedure the obtained value is 621.10 ppm/$^\circ$C and compares to 624.32 ppm/$^\circ$C obtained in the DRA procedure. Both measurements exhibit the same linearity and angular coefficient (see TableII and Figure4).
The frequency evolution of the HE$_{11d}$ mode with increasing temperature for DRA procedure is showed in Figure 5, where the HE$_{11d}$ mode is isolated and well defined. The decrease in the return loss (in modulus) is associated to impedance matching variation due to volumetric expansion and the change in value of dielectric permittivity the DRA with temperature. The measurement of $\tau_f$ for the BTNO phase was not reported in the literature. We believe that the reason is the high dielectric loss, which almost do not allows to use the Courtney method. In this case, the resonances are too broad. Considering the Courtney geometry, the quality factor for TE$_{011}$ mode is low, leading to a broad band. The monitoring of the resonant frequency shift with temperature is quite difficult with the enlargement of this band and a very poor mode visualization, see Figure 7.

In the present proposed new method, the measurement of the $\tau_f$ for BTNO is quite satisfactory. The HE$_{11d}$ mode is quite strong and well defined. The value of $\tau_f = -104.19$ ppm/$^\circ$C (Table II) was obtained for the first time. The linearity for frequency shift with temperature increase is showed in Figure 9, where a good linear agreement of the frequency with temperature was obtained.
FIG. 5. Frequency variation of the HE_{11a} mode for DRA based on CTO with increasing temperature.

FIG. 9. Measurement of $\tau_f$ for a DRA based on BTNO by the alternative method (HE_{11a}).
In conclusion a new experimental configuration to measure the temperature coefficient of resonant frequency ($\tau_f$) in dielectric resonators was presented. The new experimental setup, to measure the $\tau_f$ value, is based on the frequency variation with the temperature of the HE$_{11d}$ mode of a DRA. The method is quite compatible with the measurement of $\tau_f$ of the Courtney method. The obtained results by measuring the $\tau_f$ value of CTO and Al$_2$O$_3$, in this proposed method, is presenting excellent agreement when compared to the traditional Courtney method. The dielectric loss is less affected in this method and this is the most important advantage that was obtained. In the tests, the $\tau_f$ of the sample with higher loss ($>10^{-2}$) was obtained. In this case, the $\tau_f$ value for the BTNO resonator was $-104.19$ ppm$^\circ$C$^{-1}$. The analysis of the temperature coefficient of resonant frequency ($\tau_f$) in dielectric
resonators is an important property for the development of high frequency electronic devices, considering that this is a fundamental parameter, for the production of new components like filters, oscillators and antennas, with high thermal stability.

M.A.S. Silva, T.S. M. Fernandes and A.S.B. Sombra
doi:10.1063/1.4755799

Archival publications (published) during reporting period:

Supervision of PhD Thesis


2- Study of thermal and structural stability of Ca (Nb12Y12) xTi1-xO3 Y Bi, Fe and Ca (Nb23Li13) xTi1-xO3-δ and its use in dielectric resonator antennas (DRAs) Antonia Daniele Souza Bruno Costa, Programa de Pós Graduação em Engenharia de Teleinformática (2012)

3- STUDY of THERMAL STABILITY of microwave Resonant frequency of (τf) of DIELETRICS and SYNTHESIS of CERAMIC MATERIALS with NEAR-ZERO τf Marcelo Antonio Santos da Silva, Programa de Pós Graduação em Química (2012)

4- STUDY OF DIELECTRIC PROPERTIES OF CERAMIC MATRIX SrBi2Nb2O9 (SBN) FOR USE IN RF AND MICROWAVE DEVICES, EMMANUELLE DE OLIVEIRA SANCHO PROGRAMA DE PÓS-GRADUAÇÃO EM ENGENHARIA E CIÊNCIA DE MATERIAIS- Universidade Federal do Ceará-UFC (2013)


6- STUDY OF THE EFFECTS OF THE ADDITION OF TiO2 AND V2O5 IN DIELECTRIC PROPERTIES OF CERAMIC MATRIX Na2Nb4O11 (NN00) AND ITS APPLICATIONS IN DIELECTRIC RESONATORS. Mairton Cavalcante Romeu,
Supervision of MSc Thesis

1- Study of dielectric and magnetic properties of the composite matrix: SrBi$_2$Nb$_2$O$_9$(SBN)$_x$-BaFe$_{12}$O$_{19}$(BFO)$_{1-x}$ Klara Rhaissa Burlamaqui Theophilo
Departamento de Física/UFC Programa de Pós Graduação em Física, (2011)

2- EFFECTS OF ADDITIVES FOR COPPER (Cu) AND ZINC (Zn), ON THE DIELECTRIC PROPERTIES OF BiNbO$_4$ ARRAY (BNO), FOR APPLICATIONS IN RADIO-FREQUENCY (RF) AND MICROWAVE (MW) Antonio Jefferson Mangueira Sales Departamento de Teleinformática/Universidade Federal do Ceará Programa de Pós Graduação em Teleinformática, (2011)

3- Study of thermal stability of Ca (Nb12Bi12) xTi1-xO3: B2O3) y for use in Dielectric Resonator Antenna (DRA), Tatiana Sainara Maia Fernandes
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7-ANTENNAS FOR RADIO FREQUENCY IDENTIFICATION SYSTEMS MADE BY REUSING AND CONSTRUCTION WASTE RECYCLING Edmilson Carneiro Moreira Universidade Federal do Ceará Programa de Pós-Graduação em Engenharia de Teleinformática (2012)

8- EXPERIMENTAL AND NUMERICAL STUDY OF DIELECTRIC RESONATOR ANTENNA (DRA) BASED ON Sr$_2$CoNbO$_6$
José Eduardo Vasconcelos de Morais-- Universidade Federal do Ceará, Programa de Pós-Graduação em Engenharia de Teleinformática (2014)
9- Study of dielectric properties of Ceramic Matrix FeNbTiO6: (ZnO) x for applications in Radio-frequency (RF) and microwave range. Armando José Neves de Castro--- Universidade Federal do Ceará, Programa de Pós-Graduação em Engenharia de Teleinformática (2014)

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7- HIGH THERMAL STABILITY OF MICROWAVE DIELECTRIC PROPERTIES OF CaTi₁₋ₓ(Nb₁/₂FeLi₁/₂)ₓO₃ CERAMICS A. D. S. BRUNO COSTA, M. C. ROMEU,R. C. S. COSTA, T. S. M. FERNANDES,F. W. DE O. AMARANTE, M. A. S. DA SILVA,
8- Morphological, structural, optical and dielectric properties of 91SiO2:4Li2O:4Nb2O5:1Dy2O3 (% mole) glass prepared by sol–gel

9- Study of the structural and dielectric properties of Bi2O3 and PbO addition on NbO4 ceramic matrix for RF applications

Materials Sciences and Applications, 3(1) (2012) 6–17 (Scientific Research Publishing )
doi: 10.4236/msa.2012.31002

Microwave and Optical Technology Letters 54(1)(2012), 18–23(Wiley) doi: 10.1002/mop.26486

Physica Scripta 86 (2012) 025701 -025710 (IOP)
doi: 10.1088/0031-8949/86/02/025701


DOI: 10.1007/s11664-012-2428-4

16- Yttrium Iron Garnet: Properties and Applications Review
17-High dielectric permittivity of SrBi$_2$Nb$_2$O$_9$ (SBN) added Bi$_2$O$_3$ and La$_2$O$_3$
Journal of Electroceramics Vol 30(3) (2013) 119-128
Springer DOI 10.1007/s10832-012-9772-x

18-Experimental and numerical investigation of dielectric resonator antenna based on the BiFeO$_3$ ceramic matrix added with Bi$_2$O$_3$ or PbO
Journal of Alloys and Compounds 576 (2013) 324-331 ELSEVIER
DOI http://dx.doi.org/10.1016/j.jallcom.2013.06.009

19-A Review on Ba$_x$Sr$_{1-x}$Fe$_{12}$O$_{19}$ Hexagonal Ferrites for use in Electronic Devices
doi:10.4028/www.scientific.net/SSP.202.1

20-High dielectric permittivity and low loss of SrBi$_4$Ti$_4$O$_{15}$ with PbO and V$_2$O$_5$ additions for RF and microwave applications
C. A. Rodrigues Jr., J. M. S. Filho, P. M. O. Silva, M. A. S. Silva, C. C. M. Junqueira, A. S. B. Sombra
DOI 10.1007/s10854-013-1271-6

21-Preparation of Bi$_4$Ti$_3$O$_{12}$(BIT) Ceramics via a High-Energy Ball Milling Process Doped with Multi-Walled Carbon Nanotubes (MWNTs)
A. G. Pinheiro, G. D. Saraiva, J. M. Filho, A. S. B. Sombra
Materials Sciences and Applications, 4(9) 2013 pp 549-555 (Scirp)
http://dx.doi.org/10.4236/msa.2013.49067

22- Impedance spectroscopy study of TiO$_2$ addition on the ceramic matrix Na$_2$Nb$_3$O$_{11}$
M. C. Romeu, R. G. M. Oliveira, A. J. M. Sales, P. M. O. Silva, J. M. S. Filho, M. M. Costa, A. S. B. Sombra
DOI 10.1007/s10854-013-1514-6

23- Impedance spectroscopy study of Na$_2$Nb$_4$O$_{11}$ ceramic matrix by the addition of Bi$_2$O$_3$
R. G. M. Oliveira, M. C. Romeu, M. M. Costa, P. M. O Silva, J. M. S. Filho, C. C. M. Junqueira, A. S. B. Sombra
http://dx.doi.org/10.1016/j.jallcom.2013.08.208

24-Radiofrequency and microwave properties study of the electroceramic BaBi$_4$Ti$_4$O$_{15}$
P. M. O. Silva, T. S. M. Fernandes, R. M. G. Oliveira, M. A. S. Silva, A. S. B. Sombra
Materials Science and Engineering B 182 (2014) 37–44 ELSEVIER
http://dx.doi.org/10.1016/j.mseb.2013.11.017
Changes in research objectives, if any: xxxxx
Change in AFOSR program manager, if any: xxxxx
Extensions granted or milestones slipped, if any: xxxxx
The study of layered magneto-dielectric composites structures is important for applications at higher frequencies where the use of metals is leading to higher loss. This kind of component based in a new configuration and using a new group of magneto-dielectric composites and blends is expected to present better bandwidth, low loss, high impedance matching that will open the possibility to be used in radars, communication devices, navigation equipments, and so on.

The use of special structures based in composites and blends is important for components operating at high frequencies.

In this work the magnetic and dielectric properties of ceramic-ceramic and ceramic-polymer composites with BiNbO$_4$, SrBi$_2$Nb$_2$O$_9$ (SBN), BaBi$_4$Ti$_4$O$_{15}$ (BBT), Na$_2$Nb$_4$O$_{11}$ (NNO), Sr$_2$CoNbO$_6$ (SCN), FeNbTiO$_6$, BiFeO$_3$, CaTi$_{1-x}$(Nb$_{1/2}$Fe$_{1/2}$)$_x$O$_3$ and ferrites BaFe$_{12}$O$_{19}$ Ba$_2$Co$_2$Fe$_{12}$O$_{22}$ (Co$_2$Y) and Y$_3$Fe$_5$O$_{12}$ (YIG) was studied for RF and microwave applications.
The study of the thermal stability of magneto-dielectric composites is important for applications at the microwave band and in the millimeter and near millimeter region (100-300GHz) where the thermal stability of the resonators is fundamental.

In this presentation we will discuss

--- A study in the structural and microwave properties of the alloy matrix of CaTi$_{1-x}$(Nb$_{1/2}$Fe$_{1/2}$)$_x$O$_3$

--- Ferrimagnetism and Ferroelectricity of the Composite Matrix: SrBi$_2$Nb$_2$O$_9$(SBN)$_x$-BaFe$_{12}$O$_{19}$(BFO)$_{100-x}$

--- A new method to measure the microwave thermal stability coefficient $\tau_f$ of materials
In this work, we studied and discussed the structural and microwave dielectric properties of the B-site modified calcium titanate ceramics. The compounds were prepared by a new procedure in the conventional solid-state method. They were properly studied, using X-ray diffraction (XRD), Raman Scattering spectroscopy, and microwave dielectric properties. Therefore, the refinement analysis of the XRD was presented and discussed.

![XRD patterns of CTO and CNFTOX series.](image1)

![Graph showing unit-cell parameters (10^-1 nm) for CNFTOX series.](image2)

![Table 5. Microwave Dielectric Properties for samples ball-milled with ratio of 1 ball/g, calcinated at 900°C for 3 and 5 h, and sintered at 1100°C, for 3 h.](image3)
Figure 2.4  Schematic sketch of Courtney setup for measuring the dielectric constant under end shorted condition (after Ref. [12]).

Table 5. Microwave Dielectric Properties for samples ball-milled with ratio of 1 ball/g, calcinated at 900°C for 3 and 5 h, and sintered at 1100°C, for 3 h.

<table>
<thead>
<tr>
<th>Sample</th>
<th>Calcination condition</th>
<th>$f_r$ (GHz)</th>
<th>$\varepsilon_r$</th>
<th>$\tan \delta$</th>
<th>$Q \times f$ (GHz)</th>
</tr>
</thead>
<tbody>
<tr>
<td>CNFT01</td>
<td>900°C/3h</td>
<td>4.451</td>
<td>30.42</td>
<td>$6.4 \times 10^{-3}$</td>
<td>681.14</td>
</tr>
<tr>
<td>CNFT01</td>
<td>900°C/5h</td>
<td>3.619</td>
<td>58.00</td>
<td>$3 \times 10^{-3}$</td>
<td>1067.86</td>
</tr>
<tr>
<td>CNFT02</td>
<td>900°C/3h</td>
<td>4.804</td>
<td>25.72</td>
<td>$3 \times 10^{-3}$</td>
<td>1535.70</td>
</tr>
<tr>
<td>CNFT02</td>
<td>900°C/5h</td>
<td>4.365</td>
<td>38.83</td>
<td>$4 \times 10^{-3}$</td>
<td>979.81</td>
</tr>
</tbody>
</table>

Table 6. Microwave Dielectric Properties for samples calcinated at 900°C (for 5 h), and sintered at 1100°C (for 3 h).

<table>
<thead>
<tr>
<th>Sample</th>
<th>Balls/Mass ratio</th>
<th>$f_r$ (GHz)</th>
<th>$\varepsilon_r$</th>
<th>$\tan \delta$</th>
<th>$Q \times f$ (GHz)</th>
</tr>
</thead>
<tbody>
<tr>
<td>CNFT01</td>
<td>1 ball/g</td>
<td>3.619</td>
<td>58.00</td>
<td>$3.0 \times 10^{-3}$</td>
<td>1067.86</td>
</tr>
<tr>
<td>CNFT01</td>
<td>2.4 balls/g</td>
<td>2.937</td>
<td>78.11</td>
<td>$1.5 \times 10^{-3}$</td>
<td>1794.89</td>
</tr>
<tr>
<td>CNFT02</td>
<td>1 ball/g</td>
<td>4.365</td>
<td>38.83</td>
<td>$4.0 \times 10^{-3}$</td>
<td>979.81</td>
</tr>
<tr>
<td>CNFT02</td>
<td>2.4 balls/g</td>
<td>3.435</td>
<td>56.05</td>
<td>$9.4 \times 10^{-3}$</td>
<td>358.61</td>
</tr>
</tbody>
</table>
The temperature coefficient of resonant frequency ($\tau_f$) measures, the variation of the resonance frequency of the dielectric resonator with temperature variation, as seen in below equation $^{1,4}$

$$\tau_f = \frac{1}{f_i} \frac{\Delta f}{\Delta T} \times 10^6,$$

(1)

Table 7. Microwave Dielectric Properties for samples ball-milled with ratio of 2.4 balls/g, calcinated at 900°C (for 5 h) and sintered 1100°C, for 3 h.

<table>
<thead>
<tr>
<th>Sample</th>
<th>$f_r$ (GHz)</th>
<th>$\varepsilon_r$</th>
<th>$T_g \delta$</th>
<th>$\tau_f$ (ppm/°C)</th>
<th>$Q \times f$ (GHz)</th>
</tr>
</thead>
<tbody>
<tr>
<td>CTO</td>
<td>2.659</td>
<td>101.33</td>
<td>$2.1 \times 10^{-3}$</td>
<td>1022.909</td>
<td>1266.19</td>
</tr>
<tr>
<td>CNFTO1</td>
<td>2.937</td>
<td>78.11</td>
<td>$2.3 \times 10^{-3}$</td>
<td>518.676</td>
<td>1275.65</td>
</tr>
<tr>
<td>CNFTO2</td>
<td>3.435</td>
<td>56.05</td>
<td>$9.4 \times 10^{-3}$</td>
<td>422.987</td>
<td>365.43</td>
</tr>
<tr>
<td>CNFTO3</td>
<td>3.964</td>
<td>40.66</td>
<td>$5.5 \times 10^{-3}$</td>
<td>264.635</td>
<td>720.73</td>
</tr>
<tr>
<td>CNFTO4</td>
<td>4.335</td>
<td>34.16</td>
<td>$5.1 \times 10^{-3}$</td>
<td>412.154</td>
<td>850.00</td>
</tr>
<tr>
<td>CNFTO5</td>
<td>4.889</td>
<td>26.52</td>
<td>$4.8 \times 10^{-3}$</td>
<td>58.478</td>
<td>1017.92</td>
</tr>
<tr>
<td>CNFTO6</td>
<td>4.831</td>
<td>28.23</td>
<td>$4.2 \times 10^{-3}$</td>
<td>2.866</td>
<td>1150.24</td>
</tr>
<tr>
<td>CNFTO7</td>
<td>5.280</td>
<td>24.60</td>
<td>$3.6 \times 10^{-3}$</td>
<td>-32.574</td>
<td>1466.94</td>
</tr>
<tr>
<td>CNFTO8</td>
<td>5.381</td>
<td>22.62</td>
<td>$4.6 \times 10^{-3}$</td>
<td>-44.744</td>
<td>1169.78</td>
</tr>
<tr>
<td>CNFTO9</td>
<td>5.771</td>
<td>22.18</td>
<td>$1.0 \times 10^{-3}$</td>
<td>-71.318</td>
<td>577.10</td>
</tr>
<tr>
<td>CNFO</td>
<td>5.723</td>
<td>21.28</td>
<td>$4.8 \times 10^{-3}$</td>
<td>-88.231</td>
<td>1192.29</td>
</tr>
</tbody>
</table>

Fig. 4. Dielectric permittivity of CNFTO (0 ≤ $x$ ≤ 1) for ball milled samples with ball/mass ratio of 2.4, calcinated at 900°C (for 5 h), and sintered at 1100°C (for 3 h).

Fig. 6. $\tau_f$ of CNFTO (0 ≤ $x$ ≤ 1) for ball milled sample with ball/mass ratio of 2.4, calcinated at 900°C (for 5 h), and sintered at 1100°C (for 3 h).
Results showed that the samples belong to the Pbnm spatial group. The microwave dielectric properties of the Ca\([(Fe_{1/2}Nb_{1/2})_xTi_{1-x}]O_3\) for ball-milled samples (with ratios of 1 and 2.4 balls/g), calcinated at 900°C (with different time of exposure – 3 and 5 h), and sintered at 1100°C (for 3h) were investigated. Dielectric permittivity values in the range of 20 to 80 were obtained.

Regarding the studied samples, the quality factor values increased with the decrease of the titanium substitution in the region from \(x = 0.2\) to 0.7. Considering the increase of the \(x\) value (titanium substitution), we observe the decrease of the temperature coefficient of resonant frequency (\(\tau_f\)). The CNFTO has excellent microwave properties at \(x = 0.6\), with a temperature coefficient of resonant frequency (\(\tau_f\)) almost zero (\(\tau_f = 2.8 \text{ ppm/}^\circ\text{C}\)). At \(x = 0.7\), the \(\tau_f\) values became negative and Q.f decreases.
In this work, our main goal is to develop a dielectric material that is able to respond to both electric and magnetic stimulus, i.e. that is ferroelectric and ferromagnetic.

To do so, we use the Aurivillius ceramic SrBi$_2$Nb$_2$O$_9$ and the Hexaferrite BaFe$_{12}$O$_{19}$. Such a material could be applied in the same way that common dielectrics (as dielectric resonator antennas, for example) but opening a wide range of possibilities to make the application of ceramics to electronic devices, memories and telecommunications more useful and powerful.
Figure 1. X-Ray diffractograms for SBN100, BFO50, BFO100 samples.

Table 1. Rietveld refinement parameters.

<table>
<thead>
<tr>
<th>Sample</th>
<th>SBN 100</th>
<th>BFO100</th>
</tr>
</thead>
<tbody>
<tr>
<td>a (nm)</td>
<td>0.5515</td>
<td>0.5868</td>
</tr>
<tr>
<td>b (nm)</td>
<td>0.5513</td>
<td>0.5868</td>
</tr>
<tr>
<td>c (nm)</td>
<td>2.5024</td>
<td>2.3106</td>
</tr>
<tr>
<td>Density (g/cm³)</td>
<td>7.293</td>
<td>5.358</td>
</tr>
<tr>
<td>Volume (nm³)</td>
<td>0.761062</td>
<td>0.689074</td>
</tr>
<tr>
<td>Rp</td>
<td>10.74%</td>
<td>27.43%</td>
</tr>
<tr>
<td>Rap</td>
<td>14.7%</td>
<td>34.96%</td>
</tr>
<tr>
<td>Rep</td>
<td>11.6%</td>
<td>22.73%</td>
</tr>
<tr>
<td>S</td>
<td>1.27</td>
<td>1.54</td>
</tr>
</tbody>
</table>

Table 2. Relative density of the samples obtained from the Archimedes method.

<table>
<thead>
<tr>
<th>Binder &amp; Sample</th>
<th>Relative Density</th>
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<tr>
<td>BFO100 T</td>
<td>93.83%</td>
</tr>
<tr>
<td>BFO75 T</td>
<td>83.09%</td>
</tr>
<tr>
<td>BFO50 T</td>
<td>91.88%</td>
</tr>
<tr>
<td>BFO25 T</td>
<td>87.19%</td>
</tr>
<tr>
<td>SBN100 T</td>
<td>82.70%</td>
</tr>
<tr>
<td>BFO100 P</td>
<td>83.09%</td>
</tr>
<tr>
<td>BFO75 P</td>
<td>80.77%</td>
</tr>
<tr>
<td>BFO50 P</td>
<td>85.60%</td>
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<tr>
<td>BFO25 P</td>
<td>78.11%</td>
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<tr>
<td>SBN100 P</td>
<td>67.92%</td>
</tr>
<tr>
<td>BFO100 G</td>
<td>66.04%</td>
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<td>BFO75 G</td>
<td>81.02%</td>
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<td>Glycerin</td>
<td></td>
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<tr>
<td>BFO50 G</td>
<td>83.61%</td>
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<tr>
<td>BFO25 G</td>
<td>83.32%</td>
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<tr>
<td>SBN100 G</td>
<td>80.30%</td>
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Figure 4. Mössbauer Spectrum for the BFO100 sample.

Table 3. Hyperfine parameters of the Mössbauer measurements.

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<tr>
<th>Sample</th>
<th>Sites</th>
<th>Coordination</th>
<th>IS (mm/s)</th>
<th>QS (mm/s)</th>
<th>$H_M$ (T)</th>
<th>$R_\chi$ (%)</th>
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<tr>
<td>12k</td>
<td>octahedral</td>
<td>0.351</td>
<td>0.401</td>
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<td>41.09</td>
<td>36%</td>
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<tr>
<td>4fl</td>
<td>tetrahedral</td>
<td>0.334</td>
<td>0.097</td>
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<td>49.00</td>
<td>18%</td>
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<td>BFO100</td>
<td>4f2</td>
<td>octahedral</td>
<td>0.372</td>
<td>-0.099</td>
<td>52.16</td>
<td>15%</td>
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<tr>
<td></td>
<td>2a</td>
<td>octahedral</td>
<td>0.371</td>
<td>-0.098</td>
<td>50.79</td>
<td>24%</td>
</tr>
<tr>
<td></td>
<td>2b</td>
<td>trigonal bipyramidal</td>
<td>0.326</td>
<td>2.310</td>
<td>40.65</td>
<td>6%</td>
</tr>
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</table>
Figure 6. RF measurements of (a) Permittivity and (b) Loss tangent for TEOS samples.

<table>
<thead>
<tr>
<th>Samples</th>
<th>100 MHz</th>
<th>100 MHz</th>
<th>500 MHz</th>
<th>1 GHz</th>
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<tr>
<td></td>
<td>ε</td>
<td>tanδ</td>
<td>ε</td>
<td>tanδ</td>
</tr>
<tr>
<td>BFO 100 G</td>
<td>12.38</td>
<td>0.0072</td>
<td>12.58</td>
<td>0.02</td>
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<tr>
<td>BFO 75 G</td>
<td>23.17</td>
<td>0.0374</td>
<td>22.80</td>
<td>0.023</td>
</tr>
<tr>
<td>BFO 50 G</td>
<td>69.75</td>
<td>0.167</td>
<td>48.71</td>
<td>0.212</td>
</tr>
<tr>
<td>BFO 25 G</td>
<td>36.12</td>
<td>0.0043</td>
<td>36.84</td>
<td>0.117</td>
</tr>
<tr>
<td>SBN 100 G</td>
<td>35.46</td>
<td>0.048</td>
<td>37.51</td>
<td>0.246</td>
</tr>
<tr>
<td>BFO 100 P</td>
<td>13.19</td>
<td>0.0061</td>
<td>13.43</td>
<td>0.036</td>
</tr>
<tr>
<td>BFO 75 P</td>
<td>77.04</td>
<td>0.0484</td>
<td>78.03</td>
<td>0.068</td>
</tr>
<tr>
<td>BFO 50 P</td>
<td>33.10</td>
<td>0.0854</td>
<td>31.07</td>
<td>0.0217</td>
</tr>
<tr>
<td>BFO 25 P</td>
<td>42.42</td>
<td>0.0115</td>
<td>44.02</td>
<td>0.174</td>
</tr>
<tr>
<td>SBN 100 P</td>
<td>27.89</td>
<td>0.0037</td>
<td>28.03</td>
<td>0.0027</td>
</tr>
<tr>
<td>BFO 100 T</td>
<td>8.04</td>
<td>0.0055</td>
<td>8.05</td>
<td>0.002</td>
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<tr>
<td>BFO 75 T</td>
<td>96.68</td>
<td>0.0046</td>
<td>104.8</td>
<td>0.238</td>
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<tr>
<td>BFO 50 T</td>
<td>108.94</td>
<td>0.128</td>
<td>91.44</td>
<td>0.231</td>
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<tr>
<td>BFO 25 T</td>
<td>35.35</td>
<td>0.0055</td>
<td>36.47</td>
<td>0.146</td>
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<tr>
<td>SBN 100 T</td>
<td>39.05</td>
<td>0.0086</td>
<td>39.19</td>
<td>0.008</td>
</tr>
</tbody>
</table>
Figure 7. Magnetic hysteresis loops for (a) TEOS and (b) Glycerin samples.

Figure 8. Electric hysteresis loops recorded at 1 Hz frequency for (a) SBN100T sample and (b) BFO25P, BFO50P and BFO75P composites.
Table 5. Magnetic Hysteresis parameters of the samples.

<table>
<thead>
<tr>
<th>Samples</th>
<th>Remanent Magnetization (emu/g)</th>
<th>Coercive Field (Oe)</th>
<th>Saturation Magnetization (emu/g)</th>
<th>Maximum Field (Oe)</th>
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</thead>
<tbody>
<tr>
<td>BFO 100 T</td>
<td>23.98</td>
<td>3744.4</td>
<td>34.97</td>
<td>7680</td>
</tr>
<tr>
<td>BFO 75 T</td>
<td>10.07</td>
<td>932.8</td>
<td>20.39</td>
<td>7676.4</td>
</tr>
<tr>
<td>BFO 50 T</td>
<td>7.60</td>
<td>813.6</td>
<td>17.39</td>
<td>7539</td>
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<tr>
<td>BFO 25 T</td>
<td>2.34</td>
<td>725</td>
<td>5.99</td>
<td>7388</td>
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<tr>
<td>BFO 100 P</td>
<td>21.13</td>
<td>3699.4</td>
<td>31.33</td>
<td>7578</td>
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<tr>
<td>BFO 75 P</td>
<td>15.84</td>
<td>1110</td>
<td>27.34</td>
<td>7418.4</td>
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<td>BFO 50 P</td>
<td>8.73</td>
<td>633.8</td>
<td>18.13</td>
<td>7629</td>
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<tr>
<td>BFO50 P</td>
<td>3.76</td>
<td>923</td>
<td>6.46</td>
<td>7648.6</td>
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<tr>
<td>BFO 100 G</td>
<td>19.99</td>
<td>3460.4</td>
<td>28.90</td>
<td>7692.8</td>
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<tr>
<td>BFO 75 G</td>
<td>13.86</td>
<td>833</td>
<td>26.24</td>
<td>7731.6</td>
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<td>14.10</td>
<td>868</td>
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<td>BFO 25 G</td>
<td>4.01</td>
<td>747.4</td>
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Conclusions

A complex behavior was observed for the loss tangent over the radio frequency range, which means that less lossy samples could not help keeping this characteristic over entire frequency range. The magnetic hysteresis loops showed that composite samples preserve the ferrimagnetism for hexaferrite when SBN is added to the composite, although they become less coercive. For electric hysteresis the density of the samples are not high enough to define the true behavior of ferroelectricity in composite samples. For further works, the properties over microwave frequency range, thermal influences on the dielectric properties will be investigated for possible applications of the composite.
Objectives and Approach

A new method to measure the microwave thermal stability coefficient $\tau_f$

\[
\tau_f = \frac{1}{f_i} \times \frac{\Delta f}{\Delta T} \times 10^6,
\]

Modified setup, for the measurement of $\tau_f$

| Table I. TE$_{01\delta}$, HE$_{11\delta}$ e TM$_{01\delta}$ modes and dielectric parameters of CTO, Al$_2$O$_3$, and BTNO dielectrics. |
|---------------------|---------------------|---------------------|
|                     | CaTiO$_3$ | Al$_2$O$_3$ | BTNO     |
| a (mm)              | 7.48      | 12.70      | 7.31     |
| h (mm)              | 8.04      | 12.70      | 7.38     |
| a(mm)/h(mm)         | 0.93      | 1          | 0.99     |
| $\varepsilon_R$      | 92.25     | 9.80       | 63.68    |
| tan $\delta$        | $5.81 \times 10^{-4}$ | $1.11 \times 10^{-4}$ | $5.61 \times 10^{-2}$ |
| $f_{\text{monopole}}$ (GHz) measured | 1.888     | 3.089      | 2.439    |
| $f_{\text{HE11\delta}}$ (GHz) calculated | 1.837     | 3.147      | 2.328    |
| $f_{\text{TE01\delta}}$ (GHz) calculated | 1.830     | 3.201      | 2.288    |
| $f_{\text{TM01\delta}}$ (GHz) calculated | 2.695     | 4.527      | 3.357    |

Used samples in the measurements
FIG. 4. Measurement of $\tau_f$ for a DRA based on CTO: □ alternative method (HE$_{11\delta}$) and ○ Courtney method (TE$_{011}$).

FIG. 5. Frequency variation of the HE$_{11\delta}$ mode for DRA based on CTO with increasing temperature.

FIG. 7. Measurement of transmission by the Courtney method for the BTNO resonator.

FIG. 9. Measurement of $\tau_f$ for a DRA based on BTNO by the alternative method (HE$_{11\delta}$).
In this work a new experimental configuration to measure the temperature coefficient of resonant frequency ($\tau_f$) in dielectric resonators was presented. The new experimental setup, to measure the $\tau_f$ value, is based on the frequency variation with the temperature of the HE11dmode of a DRA. The method is quite compatible with the measurement of $\tau_f$ of the Courtney method. The obtained results by measuring the $\tau_f$ value of CTO and Al2O3, in this proposed method, is presenting excellent agreement when compared to the traditional Courtney method. The dielectric loss is less affected in this method and this is the most important advantage that was obtained. In the tests, the $\tau_f$ of the sample with higher loss ($>10^{-2}$) was obtained. In this case, the $\tau_f$ value for the BTNO resonator was $-104.19$ ppm. C$^{-1}$. The analysis of the temperature coefficient of resonant frequency ($\tau_f$) in dielectric resonators is an important property for the development of high frequency electronic devices, considering that this is a fundamental parameter, for the production of new components like filters, oscillators and antennas, with high thermal stability.

M.A.S. Silva, T.S. M. Fernandes and A.S.B. Sombra
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   Final Report

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   asbsombra@gmail.com

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   Contact phone number if there is a problem with the report
   55-85-99690713

Organization / Institution name
   Universidade Federal do Ceara

Grant/Contract Title
   The full title of the funded effort.
   ELECTRICAL AND STRUCTURAL PROPERTIES STUDY OF LAYERED DIELECTRIC AND MAGNETIC COMPOSITES AND BLENDS STRUCTURES FOR RF AND MICROWAVE APPLICATIONS

Grant/Contract Number
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   FA9550-11-1-0095

Principal Investigator Name
   The full name of the principal investigator on the grant or contract.
   Antonio sergio Bezerra Sombra

Program Manager
   The AFOSR Program Manager currently assigned to the award
   James Fillerup

Reporting Period Start Date
   05/15/2011

Reporting Period End Date
   06/05/2014

Abstract
   In this work the magnetic and dielectric properties of ceramic-ceramic and ceramic-polymer composites with BiNbO4, SrBi2Nb2O9 (SBN), BaBi4Ti4O15 (BBT), Na2Nb4O11(NNO), Sr2CoNbO6 (SCN) and ferrites BaFe12O19 and Y3Fe5O12 (YIG) was studied for RF and microwave applications. New configurations of magneto-dielectric composites and blends structures for high frequency applications was done. The 0-3 type dielectric and magnetic composites with homogenously distributed ceramic inclusions was fabricated in a polymer matrix. Magnetic Yttrium Iron Garnet (YIG) and (SBN) powders were used to enhance the permittivity and permeability of the composites. This group of dielectric and magnetic phases was studied in the RF and microwave region. The microstructure, high frequency dielectric and magnetic properties of individual layers and 2-2 composites was investigated and measured.
A new method for the measurement of the temperature coefficient of resonant frequency ($\tau_f$), is presented. The traditional method (based on the Courtney method) present some limitations of measuring the values of $\tau_f$, for samples with high dielectric loss due to their inability to observe clearly the TE011 mode. The new experimental setup, to measure the $\tau_f$ value, is based on the variation of the temperature of the dominant mode of a dielectric resonator antenna (DRA).


Changes in research objectives (if any):

Change in AFOSR Program Manager, if any:

Extensions granted or milestones slipped, if any:

AFOSR LRIR Number

LRIR Title

Reporting Period

Laboratory Task Manager

Program Officer

Research Objectives

Technical Summary

Funding Summary by Cost Category (by FY, $K)

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Report Document

Appendix Documents
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