Characterization of dielectric properties of screen-printed MgTiO$_3$–CaTiO$_3$ composite thick films in the microwave frequency range

J. I. Marulanda, R. A. A. Lima, M. C. R. Carvalho
CETUC / PUC-Rio
Centro de Estudos em Telecomunicações
Pontifícia Universidade Católica do Rio de Janeiro
Rio de Janeiro, Brazil
jmarulan@ctec.puc-puc-rio.br

A. F. L. Almeida, A. B. S. Sombra
Departamento de Física
Universidade Federal do Ceará
Fortaleza, Brazil
sergio@fisica.ufc.br

L. S. Demenicis
Seção de Engenharia Elétrica
Instituto Militar de Engenharia
Rio de Janeiro, Brazil
luciene@ime.eb.br

Abstract—Dielectric characterization of MgTiO$_3$, CaTiO$_3$ and MgTiO$_3$(x)–CaTiO$_3$(1-x) composite thick films with different concentrations (x = 0.95, 0.50, and 0.20) in the microwave frequency range at room temperature is presented. The films were fabricated by screen-printed method with thickness between 105 and 165 µm. Dielectric constant values between 4.2 and 17.5 and loss tangents between 0.0064 and 0.0098 were measured for frequencies in the range from 3.22 to 3.89 GHz using the coplanar waveguide (CPW) resonators technique. A relationship between the concentration ratio of MTO-CTO in the films and the dielectric constant is also presented.

Keywords-microwave ceramic films; thick films; high dielectric constant; MTO; CTO

I. INTRODUCTION

There is a growing demand for high dielectric constant ceramics due to successful applications in microelectronics and microwave industry. The use of high dielectric constant materials allows the miniaturization of microwave structures [1]. In this context, a ceramic must exhibit three basic properties: high dielectric constant, low losses and stable temperature coefficient. Both MgTiO$_3$ (MTO) and CaTiO$_3$ (CTO) are well known as low loss dielectric ceramics and have wide applications in communications systems, especially as base materials to elaborate resonators, filters, antennas, radar and global positioning systems operating at microwave frequencies [2, 3].

High dielectric constant has been reported for MTO and CTO bulk ceramics [4] and for sol-gel prepared thin films at microwave frequencies [5]. However, those values depend on many fabrication variables, such as the film deposition method, film thickness, thermal treatments after film deposition etc. For annealing temperatures of 600, 700, and 800 °C, CTO films of 0.4-µm thickness have exhibited dielectric constant values of 83, 125, and 160 respectively at 6 GHz, showing the strong dependence on the conditions of the post-deposition thermal treatment [5]. Multilayered complex structures of MTO-CTO, consisting in depositing intercalated layers of pure MTO and CTO with different layer thickness, using a solid state reaction process, have been designed to obtain low loss dielectric ceramics with dielectric constant between 19 and 179 at 1 MHz [6, 7], but there are no reported values of these properties in the microwave range for those structures.

Some MTO(x)–CTO(1-x) composite dielectric properties have been studied previously using different characterization methods [8-10]. A comparison of techniques for microwave characterization, including coplanar waveguides, coplanar resonators and interdigital capacitors is presented in [11], founding a good agreement between results for barium strontium titanate (BST) thin films. In this work, the characterization of screen-printed MTO-CTO composite thick films, regarding the concentration ratio of the materials, is assessed for the first time with measurements in the microwave frequency range.

The dielectric properties of MTO, CTO and MTO(x)–CTO(1-x) composite screen-printed thick films at room temperature were measured in the frequency range from 50 MHz to 20 GHz using the coplanar waveguide (CPW) resonator method described in [12]. In this characterization technique, a CPW resonator coated with a deposited high dielectric constant film presents a displacement of the resonance frequency peaks towards lower frequencies when compared to an identical resonator without film (a reference resonator). Once the thickness of the film and the fundamental peak frequency of the film-coated resonator (measured with a network analyzer) are known, the film dielectric constant can be obtained through interpolation among a set of curves simulated for CPW resonators with the same dimensions and different values for film dielectric constant and thickness. The
losses at a given resonance frequency are calculated from the
insertion loss and loaded quality factor $Q_L$ of the resonance
peak [13].

II. EXPERIMENTAL RESULTS

A. Circuit fabrication and film deposition

The resonators were fabricated on Piconics® thin-film
polished alumina substrate ($\varepsilon_r = 9.8$, $\tan\delta = 0.0001$), using a
conventional photolithography process. Schematics and
physical parameters of the considered resonators are shown in
Fig. 1. The cross-section of the resonators can be observed in
Fig. 1 (a). The height and width of the alumina substrate are
respectively $H = 635 \, \mu m$ and $A = 5.0 \, mm$. The thickness $d$ of
the film is measured above the circuit gold metallization.
According to the top view of the resonator, depicted in Fig. 1
(b), the total length of the circuit is $L = 25.4 \, mm$, while the
length of the resonator middle-section is $L_1 = 15.3 \, mm$ and the
coupling gap is $D = 100 \, \mu m$. Further, the width of the CPW
central line and lateral gap are respectively $W = 500 \, \mu m$ and
$G = 210 \, \mu m$.

![Schematics and parameters of the resonators. (a) cross-section, and
(b) top view.](image)

All films used in this work were prepared by the screen
printed method with double deposition layer over the prepared
CPW resonators. After the deposition of the first layer, the
films were heated at a rate of $1^\circ C/min$ from environment
temperature to $400^\circ C$. Afterwards, the temperature was raised
from $400^\circ C$ to $900^\circ C$ at a rate of $5^\circ C/min$. Once the film had
cooled down to room temperature, a second layer was screen-
printed over the first one. The same thermal treatment was
performed for the second deposited layer. The film thickness
($d$) was measured with a precision micrometer. The obtained
films and their measured thickness are presented in Table I.

![Measured Thickness](image)

B. Microwave measurements

The fundamental resonance peak of each film-coated
resonator was measured using a vector network analyzer (HP
8720C; 50 MHz ~ 20 GHz). Fig. 2 shows the experimental
insertion loss peaks obtained for the film-coated resonators ($\delta_1$
parameter) in comparison with a reference CPW resonator
(with the same physical dimensions but without film coating).

![Measured fundamental resonance peaks for the analyzed
resonators.](image)

As can be seen from this figure, starting with pure MTO
and increasing the CTO concentrations, a downshift of
resonance peaks is observed.

As mentioned previously, having measured the
fundamental resonance frequency peak and since the thickness
of the film is known, the relative dielectric constant of the
ceramic material constituent of the film can be obtained from
interpolation in the set of theoretical (simulated) curves, shown
in Fig. 3. These curves of fundamental resonance peak as a
function of the relative dielectric constant were obtained from
the results of multiple simulations of CPW resonators for films
with different dielectric constant and thickness using a
commercially available microwave-frequency structure
simulator – CST Microwave Studio®.

To refine the value of the relative dielectric constant
obtained from interpolation, an optimization of the structure
simulation was carried out. By making fine adjustments in the
relative dielectric constant, it was possible to find the value for the best match between theoretical and experimental results. This optimization procedure was carried out for each fabricated film. As an example, Fig. 4 shows the measured and simulated insertion loss for the resonator with MTO(0.2)–CTO(0.8). It can be observed that both results are in very good agreement.

The highest dielectric constant value measured was 17.5 for the CTO film and the lowest one was 4.2 for the MTO film. A strong relationship between the film relative dielectric constant and the concentration of CTO (or alternatively MTO) in the composite was observed. Fig. 5 displays the measured values for the dielectric constant according to the relative concentrations of CTO and MTO. An exponential fitting was applied to these data and the resulting curve is depicted also in Fig. 5, laying emphasis on this relationship. This characteristic could be used as a guide to formulate a particular material with a specific dielectric constant at some given frequency.

The film loss at a given resonance frequency can be determined from the insertion loss and loaded quality factor $Q_L$ of the measured resonance peak. The value of $Q_L$ is extracted from the $S_{21}$ parameter measurement by means of the expression:

$$Q_L = \frac{f_R}{\Delta f_{3DB}}$$

(1)

where $f_R$ is the peak frequency and $\Delta f_{3DB}$ is the 3 dB bandwidth of the resonance peak. The unloaded quality factor $Q_0$ can be calculated from [13]:

$$Q_0(f_R) = \frac{Q_L(f_R)}{\frac{\epsilon_r}{\epsilon_r + 1}}$$

(2)

Finally, the dielectric loss can be calculated with the following approximate expression:

$$\tan \delta \approx \frac{1}{Q_0}$$

(3)

Expression (3) is used assuming that dielectric losses are considerably higher than conductor and radiation losses [14]. The results found for dielectric loss tangent of the films remained between 0.0064 and 0.0098, for frequencies in the range from 3.22 to 3.89 GHz. The dielectric properties of the films analyzed in this work are summarized in Table II. These values are proximate to the measuring sensibility of the method, considering the dimensions of the resonator used, indicating that the actual values of film loss are probably lower than the ones observed.

The quality factor of ceramic films can be improved by means of a diversity of methods. Doping MTO with niobium increases the dielectric constant and helps eliminate defects [15]. Annealing process and the use of complex layered structures of MTO-CTO are other techniques to improve the dielectric properties of ceramic films [16, 17]. The list of variations that can be tentatively applied in the fabrication processes in order to obtain materials with higher dielectric constant and lower losses is extensive.

### Table II. Experimental results for the deposited films.

<table>
<thead>
<tr>
<th>Film Composition</th>
<th>Fundamental peak frequency (GHz)</th>
<th>$\epsilon_r$</th>
<th>$\tan \delta$</th>
</tr>
</thead>
<tbody>
<tr>
<td>MTO</td>
<td>3.89</td>
<td>4.2</td>
<td>0.0064</td>
</tr>
<tr>
<td>MTO(0.95)–CTO(0.05)</td>
<td>3.88</td>
<td>5.1</td>
<td>0.0082</td>
</tr>
<tr>
<td>MTO(0.50)–CTO(0.50)</td>
<td>3.66</td>
<td>8.3</td>
<td>0.0066</td>
</tr>
<tr>
<td>MTO(0.20)–CTO(0.80)</td>
<td>3.38</td>
<td>12.4</td>
<td>0.0090</td>
</tr>
<tr>
<td>CTO</td>
<td>3.22</td>
<td>17.5</td>
<td>0.0090</td>
</tr>
</tbody>
</table>

![Figure 3. Theoretical prediction for the relationship between the fundamental resonance frequency and the film relative dielectric constant for different thickness ($d$ in µm) for the dimensions of the elaborated CPW resonators.](image)

![Figure 4. Measured and simulated insertion loss for the resonator with MTO(0.2)–CTO(0.8).](image)
Dielectric constant and losses of MTO, CTO and MTO(x)–CTO(1–x) composite screen-printed thick films at room temperature were measured for the first time in the microwave frequency range using the CPW resonator method. Experimental results are in a very good agreement with theoretical analysis. The highest dielectric constant value measured was 17.5 for the CTO film; the dielectric loss tangent of the films observed was between 0.0064 and 0.0098, for frequencies in the range from 3.22 to 3.89 GHz. A strong dependence of the dielectric constant value at a specific frequency.

III. CONCLUSIONS

Dielectric constant and losses of MTO, CTO and MTO(x)–CTO(1–x) composite screen-printed thick films at room temperature were measured for the first time in the microwave frequency range using the CPW resonator method. Experimental results are in a very good agreement with theoretical analysis. The highest dielectric constant value measured was 17.5 for the CTO film; the dielectric loss tangent of the films observed was between 0.0064 and 0.0098, for frequencies in the range from 3.22 to 3.89 GHz. A strong dependence of the dielectric constant with the MTO-CTO composite concentrations in the films was observed and can be eventually used as a method to elaborate some material with a determinate dielectric constant value at a specific frequency.

ACKNOWLEDGMENT

This work was supported by Conselho Nacional de Desenvolvimento Científico e Tecnológico (CNPq), the Fundação Carlos Chagas Filho de Amparo à Pesquisa do Estado do Rio de Janeiro (FAPERJ) and the Fundação Cearense de Apoio ao Desenvolvimento Científico e Tecnológico (FUNCAP), Brazil.

The ceramic films were fabricated by the group of Dr. A. S. B. Sombra, Department of Physics, UFC, Brazil. The resonators were printed by photolithography at LabSem (Semiconductor Laboratory), PUC-Rio, Brazil. All microwave measurements were carried out at CETUC/PUC-Rio, Brazil.

The authors acknowledge the use of FDTD software CST Microwave Studio® under the CST Cooperation License Agreement with CETUC/PUC-Rio. All simulations of the microwave structures (resonators) presented in this work were carried out in this platform.

REFERENCES