Characterization of PMMA/BaTiO₃ Composite Layers Through Printed Capacitor Structures for Microwave Frequency Applications

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Abstract—This paper presents the extraction of microwave properties of low-temperature cured inorganic composite materials based on barium titanate (BaTiO₃). These composite materials exhibit attractive features such that when the volume fraction of the filler contents varied, its electrical properties of high permittivity and moderately low loss tangent can be manipulated to suit different areas of applications. For the extraction of the permittivity and the loss tangent, three different ink particles were developed and printed on the top of interdigital-shaped microwave capacitor. The properties of the inks were extracted from measured results through computer simulations. The obtained results were verified with several types of interdigital capacitor structures of different fingers and linewidths. The effect of the thickness of the ink layer materials on the top of the capacitor structures was likewise investigated. The results show relative permittivity (ϵ_r) values of 30, 25, and 27 for composite layers printed using inks with Pr. A shape at 67.4 wt% (percentage by weight), Pr. B shape at 66.3 wt%, and Pr. C shape at 67.1 wt% of BaTiO₃, respectively, at 2 GHz. Corresponding loss tangents ($\tan \delta$) were 0.065, 0.040, and 0.025. The dielectric properties of the composite materials are influenced by the thickness variation of the ink layers on the capacitor structures. This novel capacitor composite materials would be a promising candidate for printed application in mobile telecommunication operations, especially in the frequency range of 0.5–3 GHz.

Index Terms—Barium titanate (BaTiO₃), microwave, printed interdigital capacitor, RF passives, screen printing.

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I. INTRODUCTION

THE use of printing as a technique for the realization of large-area and low-cost electronic systems has recently gained a remarkable level of interest in the microelectronics industry.

Printed electronics offers several benefits over traditional electronics such as transparency, flexibility, and less costly fabrication, and it is growing more rapidly. Printing is expected to further support the realization of electronics on flexible and considerably cheap substrates such as metal foils and plastic [1]. Essentially, the increased need for energy-efficient products with tremendous performance and cost-effective manufacturing is stimulating the advancement of overall printed electronics market [2]. The technology is deployed and under investigations in several applications such as sensors [3]–[5], radio frequency circuits [6]–[9], solar cells [10], antennas [11], [12], and other electronic components like displays [13]. To realize these systems, it is necessary to develop printable materials with low loss and sufficient permittivity values, especially for high-frequency applications [14]. Functional materials with suitable low loss and high relative permittivity do already exist, but they are high-temperature sintered [15], [16]. The high-temperature sintering or sputtering methods limit their practical usage on the flexible organic substrate. Thus, low curing temperature materials with considerable electrical properties are needed.

Lately, several studies have been carried out in the field of dielectric properties of polymer–ferroelectric ceramic composites [17]–[23]. Some of the possible commercially available of polymer–ceramic composites are polystyrene, polyethylene, peek, PTFE as matrix materials with fillers such as alumina, glasses, titania, and silica [15]. Wang *et al.* [24] prepared polyimide/barium titanate (BaTiO₃) composite with different BaTiO₃ contents from 0 to 90 wt% (0–67.5 vol.%), which yielded relative permittivity ranging from 3.53 to 46.50 and loss tangent from 0.005 to 0.015 at the frequency of 10 kHz. Balaraman *et al.* [25] utilized hydrothermal process (<100 °C) for the production of BaTiO₃ on titanium foils and measured to have a capacitance per area varying from 20 to 600 nF/cm² at 100 kHz with a loss tangent ranging from 0.03 to 0.8. Chameswary *et al.* [15] reported the dielectric properties of BaTiO₃ butyl rubber

filled composite processed at temperature 200 °C. The composite exhibited relative permittivity of 3.5, loss tangent of 0.005 with 10 vol.%, permittivity of 8, and loss tangent of 0.02 with 30 vol.% at 5 GHz [15]. Composite ink properties of BaTiO₃—poly (vinylidene fluoride—trifluoroethylene) were reported in [26]. The realized permittivity and loss tangent of the composite material measured at 1 MHz were 46 and 0.15, respectively. Balaraman *et al.* [25] reported S_{Rx}La_(1-x)Ti_xAl_(1-x)O₃ ceramic with CaO–B₂O₃ glass additive shown lower sintering temperature from 1550 °C to 1100 °C and had a dielectric permittivity, quality factor, and temperature dependence of permittivity ($\epsilon_r \sim 43.3$, $Q \times f \sim 10780$ GHz, and $\tau f \sim 20.8$ ppm/°C). Popielarz *et al.* [28] investigated also the dielectric properties of BaTiO₃–polymer composites over a wide frequency range from 100 Hz to 10 GHz at temperature range from –140 to +150 °C.

The motivation behind the study presented in this paper is to develop composite material with low loss and low curing temperature for radio and microwave field. In addition, composite material could also be manipulated into preferred properties and structures depending on the type of the component under design. This kind of material would be desirable for electronics miniaturization purposes and for improving the electrical properties of printed components and devices, such as antenna, matching circuits, filters, transmission lines, and resonators.

In this paper, BaTiO₃ was chosen as an integrating material based on its high-performance properties of low loss tangent, and high relative permittivity at room temperature, which makes it most widely used material especially in electronics device like ceramic capacitors [21]. Three different shaped and processed BaTiO₃ particles (denoted by Pr. A, Pr. B, and Pr. C) in poly (methyl methacrylate) (PMMA) matrix were used for the performance improvement of capacitive properties of low-loss microwave capacitors. Special attention in this paper was in operation frequencies of mobile telecommunication devices, i.e., from 0.5 to 3 GHz, due to the amount of potential applications. The performance of capacitor structures was investigated with these new materials. This paper has been organized and presented from the computer modeling point of view by considering capacitor structures and emphasizing the modeling of RF characteristics of new materials. Section II presents the development of dielectric inks utilizing different shape particles as fillers. Section III describes a method to accurately characterize the dielectric composite materials based on interdigital-shaped capacitors. The results and discussion are in Section IV. Last, Section V provides the conclusion.

II. DEVELOPMENT OF DIELECTRIC INKS

Solid barium hydroxide octahydrate, Ba(OH)₂ · 8H₂O, was reacted with sodium titanate slurry at 90 °C for 3 and 30 h to form sample Pr. A and Pr. C, respectively. With ion exchange reaction, the sodium ions in the titanate were replaced by barium ions forming small crystal size BaTiO₃ [1]. The BaTiO₃ slurry was filtered and washed (sodium OFF) with water, and the filter cake was dried at 110 °C. The dry sample was calcined in a rotary kiln at 800 °C for 1.5 h. Then added

Fig. 1. FESEM picture of Pr. A. sample slurry at 90 °C for 3 h and calcinated at 800 °C (average size 90 nm).

Fig. 2. Pr. C sample slurry at 90 °C for 30 h and calcinated at 800 °C (average size 150 nm, dendrite structures).

Fig. 3. Pr. B. sample slurry at 90 °C for 3 h and calcinated at 300 °C (average size 60 nm).

into water, filtrated, washed with water, dried, and dry milled. Morphology of the particles, which were characterized by field emission scanning electron microscopy (FESEM), is shown in Fig. 1 (Pr. A) and Fig. 2 (Pr. C).

In the case of Pr. B sample with FESEM picture in Fig. 3, solid barium hydroxide monohydrate, Ba(OH)₂ · H₂O, and strontium hydroxide octahydrate, Sr(OH)₂ · 8H₂O, were reacted with sodium titanate slurry at 90 °C for 3 h. With ion exchange reaction, the sodium ions in the titanate were replaced by barium and strontium ions forming small crystal size barium strontium titanate. Similarly, the slurry was filtered and washed (sodium OFF) with water, and the filter cake was dried at 110 °C. In this case, the dry sample was calcined in a rotary kiln at 300 °C for 1.5 h. Afterward, it was added into water,

TABLE I
COMPOSITIONS WITH BaTiO₃ INKS

Filler	BaTiO ₃					
	mass [g]	wt. %	mass [g]	wt. %	mass [g]	wt. %
Pr. A	5	67.4	-	-	-	-
Pr. B	-	-	5	66.3	-	-
Pr. C	-	-	-	-	5	67.1
PMMA	1.8	24.2	1.8	23.9	1.8	24.2
OKB	0.6	8.3	0.7	9.8	0.65	8.7
Final layer contains 3.5 wt. % of polymer						

filtrated, washed with water, dried, and dry milled. Molar ratio of the Sr/Ba in the material was 2:5, and the amount of elements was measured by utilizing X-ray fluorescent.

Compositions of screen-printed inks with BaTiO₃ are reported in Table I, where the vehicles composition is 10 wt% of PMMA ($M_w \sim 350\,000$) and diethylene glycol n-butyl ether acetate solvent denoted as OKB.

It is noteworthy to mention that the silver conductor layer with thickness of 15 μm was printed on the substrate adopting screen printing techniques. Similarly, the same screen printing technique was utilized to create patterns on the top of the conductive pattern forming interdigital-shaped capacitors, which were used for the characterization of the BaTiO₃ composite ink. The BaTiO₃ composite inks were printed on the capacitor structures with 200 mesh stainless steel screens and cured in 120 °C for 30 min. Alumina (96% Al₂O₃) was used for stable substrate with high accuracy and repeatability.

III. MEASUREMENT AND CHARACTERIZATION OF PRINTED DIELECTRIC POLYMER LAYERS

In this section, a dielectric material characterization method based on interdigital-shaped capacitor is presented. The aim of the characterization was to obtain the frequency dependent permittivity ϵ_r and dielectric loss tangent of the printed composite layers by simulating the whole component structures including the composite materials. Thus, the characterization takes into account the final form of the material behavior (film thickness, density, surface roughness) including fabrication tolerance and processing parameters that can be different when analyzed using TE_{01 δ} dielectric resonator method in [29]. To accurately define the characteristics of the new materials, three sets of interdigital-shaped capacitor structures with different number of linewidths and fingers were fabricated and measured. Three different shapes were used for statistical reasons, and the material properties can be extracted from the capacitor component when components have been fabricated with and without the BaTiO₃ ink filling. Thus, the order of research involved the design, fabrication, and measurement of capacitor circuits. It followed by the components simulation and finally simulated the materials on the components to extract the electrical properties of the materials.

The detailed capacitor component structures, with and without the composite layers, are given in Fig. 4. The components were designed in a 4 × 4 mm² area. Linewidths and number of fingers for the interdigital capacitors were 180 μm and five

Fig. 4. Different screen printed interdigital capacitor component types in a 4 × 4 mm² area with linewidth of (a) 180 μm and five fingers, (b) 200 μm and six fingers, and (c) 240 μm and seven fingers. The structures of the printed dielectric polymer layers on the capacitor components indicated as (d) Pr. A filler, (e) Pr. B filler, and (f) Pr. C filler.

fingers, 200 μm and six fingers, and 240 μm and seven fingers for component type a, b, and c [see Fig. 4(a)–(c)], respectively. In Fig. 4(d)–(f), the thickness of the composite layers printed on the capacitor varies from 33 to 39 μm , 20 to 31 μm , and 23 to 28 μm for Pr. A, Pr. B, and Pr. C shape filler layers, respectively.

Furthermore, effects of varied thickness of the dielectric materials pasted on the top of the fabricated capacitor structures were considered for the permittivity and loss tangent calculations during the simulation of all the component types. The values of the permittivity and dielectric loss tangent were calculated by simulating the interdigital-shaped capacitor structures with and without the composite materials with an electromagnetic solver. In this paper, CST Microwave Studio (MWS) was utilized. An initial guess value was set for both the dielectric constant and the loss tangent. Both the measured and simulated magnitudes of S-parameters at resonant frequency was compared until the simulated parameters were properly correlated with that of measured parameters. Thus, an optimized value of dielectric and loss tangent was obtained. As a comparison, Han *et al.* [30] had reported a meander-shaped inductor component for nanocomposite material characterization. The material properties extraction method and fabrication technique, described in [30], which have the benefits of material usage minimization and uncomplicated fabrication, are similar in this paper. Since the electrical properties of capacitor component depend on the physical dimension of the component and its material properties, essentially, the capacitance and Q -factor of the passive component with and without the ink were also measured.

The properties of the capacitors were measured with 8530A Agilent Technologies vector network analyzer, and the calibration of the measurement system was performed using the line-reflect-reflect-match method. The measurement results were obtained in the frequency range from 0.5 to 3 GHz as the component self-resonance was limiting the highest applicable frequencies. Both finite-element method (FEM) design and circuit model simulations of the interdigital-shaped capacitors (Fig. 5) were performed using CST MWS. The first-order circuit model presented in Fig. 5(b) describes the characteristics of the interdigital capacitor type b with six fingers given in Fig 5(b). The parallel elements of the circuit model consist of Csub1 and 2 (0.001 pF), which represent the capacitive coupling between the conductor and the substrate. In addition, the series branch of the circuit consists of parasitic series inductance L_s (3.8 nH), series resistance R_s (5 Ω), and the desired set capacitance C , which value varies with different ink layer materials. Finally, the results of both the FEM and the circuit model were compared to that

TABLE II
PROPERTIES OF DIFFERENT COMPONENT TYPES WITH AND WITHOUT DIELECTRIC INK LAYER

Component types	Measured capacitance, C(pF)								Measured Q-Factor							
	Reference		Pr. A		Pr. B		Pr. C		Reference		Pr. A		Pr. B		Pr. C	
	1 G	2 G	1 G	2 G	1 G	2 G	1 G	2 G	1 G	2 G	1 G	2 G	1 G	2 G	2 G	2 G
a	0.4	0.7	0.8	1.8	0.8	-	0.6	1.2	45.0	19.2	35.5	10.2	23.2	-	42.1	14.5
b	0.6	1.0	1.0	2.6	0.8	1.9	0.8	1.7	43.4	17.4	34.0	8.5	37.1	11.0	40.0	12.5
c	0.8	2.0	1.4	18.1	1.2	7.0	1.2	5.1	41.0	12.0	40.0	1.7	33.5	4.0	36.5	5.7
1 G = @1 GHz, 2 G = @2 GHz

Fig. 5. (a) Model of the type b capacitor for the FEM analysis. (b) First-order circuit model simulations of capacitor type b (see Fig. 4) using CST MWS.

of the measurement results, and the dielectric polymer layer properties were obtained from the results.

IV. RESULTS AND DISCUSSION

Two-port scattering parameters (S-parameters) graph results of measurement, FEM design, and circuit model are presented in Fig. 6 by means of capacitor component type b [see Fig. 4(b)]. Fig. 6(a) shows reference without filling layer, Fig. 6(b) shows filling layer Pr. A, Fig. 6(c) shows filling layer Pr. B, and Fig. 6(d) shows filling layer Pr. C, taken at frequency from 0.5 to 3 GHz. It can be noticed that the frequency responses are relatively close to each other in Fig. 6(a)–(d) behaving such as capacitor's typical characteristic before its self-resonance frequency. As shown in the graphs, the measurement response characteristics are in agreement (S_{12} inaccuracy below 5%) with both the CST FEM design and the circuit model. From S-parameters of the measurement results, electrical parameters of capacitor, such as capacitance C and Q -factor, were extracted by admittance (Y) matrix parameter calculations. The Q -factor values for capacitor were calculated from matrix parameters as

$$Q = \frac{|\text{Im}(Y_{21})|}{\text{Re}(Y_{21})}. \quad (1)$$

Likewise, the capacitance values (pF) of the capacitor were realized by using (2), where f is the frequency

$$C = \frac{10^{12}}{2\pi f \times \text{Im}(1/Y_{21})} \text{ pF}. \quad (2)$$

Plots of the measurement results of capacitance and Q -factor for component type b are reported as a function of frequency

in Fig. 7(a) and (b). The capacitance values started from 0.5 pF at 0.5 GHz increasing up to 0.8 pF with printed filling. The self-resonance of the component b was around 2.9 GHz the reference component, and it decreased to 2.6, 2.55, and 2.4 GHz with filling Pr. C, Pr. B, and Pr. A, respectively. The Q -factors started from 85 at 0.5 GHz decreasing down to 65 with printed layer. Q -factor decreased to zero at the self-resonance according to (1). The component types a and c (see Fig. 4) also followed similar patterns.

In Table II, all the three component types (a, b, and c) are presented on the first column followed by eight columns presenting reference and three printed filling materials measured capacitances. Next eight columns are presenting measured Q -factors of corresponding cases. The properties are presented at 1 and 2 GHz, which are widely used in telecommunications frequencies. The capacitance values measured at 1 GHz were from 0.4 to 0.8 pF with reference components. However, the capacitance values were found to increase from 0.6 to 1.4 pF with printed layers (Pr. A–C). As observed from Table II, the printed layer, acting as filler materials, improved the capacitance values where Pr. A ink layer had the highest effect. In addition, the capacitance values measured at 2 GHz were from 0.7 to 2 pF in reference components. With printed layer materials, the capacitance values increased significantly from 1.2 to 18.1 pF. Again, at 2 GHz, Pr. A material had the highest effect. Materials (Pr. A–C) improved the capacitance at 2 GHz more than at 1 GHz. The reason can be attributed to the self-resonance, which increased the capacitive values at 2 GHz, especially with component type c that had the highest capacitance. The Q -factor values (1 GHz) were from 41 to 45 with no materials. With materials (Pr. A–C), the values were from 23 to 42. All materials decreased the Q -factor. Among the materials, Pr. C has the highest Q -factor. Measured at 2 GHz, the Q -factor values were from 12 to 19 with no materials. With materials, the values were from 1.7 to 14.5. Similarly, Pr. C material has the highest Q -factor. Notably, the self-resonance decreased the Q -factor at 2 GHz, especially with component type c.

In Fig. 8, the simulated relative permittivity values for the different ink layers across frequency band 0.5 to 3 GHz are presented. The permittivity results extracted for each of the component types were based on the method explained in Section III. It is noticed in Fig. 8 that the permittivity plots for each of the ink materials decreased at a constant rate as the frequency increases. The reason is that constant-fit-tan-delta model was utilized for the simulation. In the future

Fig. 6. Linear S-parameters of component type b, given in Fig. 4. (a) Without filling layer (reference). (b) With filling layer Pr. A shape. (c) With Pr. B shape. (d) With Pr. C BaTiO₃ filled layers.

Fig. 7. Frequency dependence of measured (a) capacitance and (b) Q -factor results for component type b.

project, it will be on plan that other nonlinear models will be considered, and in that case, the material would be simulated together with the component structures for higher frequencies, up to 6 GHz, a useful frequency band for 5G telecommunication applications. It is found during the simulation that the variation of ink layer thickness pasted on the top of the capacitor affected the performance of the permittivity properties. The lower the thickness of the material layer, the higher the values of permittivity. By considering the minimum values of the measured ink layer thicknesses of 33, 31, and 23 μm for Pr. A, Pr. B, and Pr. C, respectively, the extracted relative

permittivity values of 30, 25, and 27 were obtained for all the component types, taken at 2 GHz. The variation of permittivity values as the thickness increases is about $\pm 8\%$ at the predefined values of the measured material thickness that is in the range of similar studies presented in the field. It is noticed from Fig. 8 that Pr. A has the highest relative permittivity while Pr. B has the lowest value. This may be due to the presence of higher ceramic filler contents of 67.4 wt% (see Table I) in Pr. A material than that of 66.3 and 67.1 wt% in Pr. B and Pr. C, respectively. Similar trend can be observed in [15] and [24] as relative permittivity and loss tangent increased with increasing

Fig. 8. Characterized relative permittivity and loss tangent as a function of frequency.

BaTiO₃ contents in butyl rubber-BaTiO₃ composites. It has been reported likewise in [31] that the dielectric constant of BaTiO₃-polyvinylidene fluoride composite film increases with an increase in the particle volume fraction from 20 to 30 vol.%. A similar BaTiO₃ particle content effect resulted to an increase in the dielectric constant of the materials.

The loss tangents for the ink layer materials printed on component type b [see Fig 4(e)] were likewise investigated. The same method in Section III was considered for loss tangent properties extraction. As shown in Fig. 8, the loss tangents of all the three ink materials were below 0.08. Typically, in manufacturing of electronic components like a capacitor, the magnitude of loss tangent should be as low as possible. The simulated loss tangent values of the three inks taken at 2 GHz were 0.065, 0.040, and 0.025 for Pr. A, Pr. B, and Pr. C, respectively. The results revealed appreciable low loss performance for all the ink materials. Among the different inks, Pr. C shape particle had the lowest loss tangent value. This characteristic behavior exhibited in the Pr. C ink material might be attributed to its filler size and content. When compared with the BaTiO₃/Polymer composites in [28] taken at 2 GHz, the loss tangent of the composite materials obtained in this paper, especially with Pr. C, is lower than the loss tangent of 0.04 for BaTiO₃/TDDMA, 0.05 for BaTiO₃/TMPTA, and 0.2 for BiTiO₃/PEGDA composites in [28] at constant 30% of BaTiO₃.

The above results suggested that as the volume fraction of the filler contents in the composite material (PMMA/BaTiO₃) varied, the dielectric properties and loss tangent of the material can be manipulated based on the area of applications. Thus, for high-frequency application where loss tangent should be as low as possible, Pr. C shape particle with the minimum loss tangent has higher preference over Pr. A and Pr. B. On the other hand, Pr. A would be highly considered among the ink materials for microwave applications of electronic components that require moderate loss tangent but high properties of capacitance and permittivity.

V. CONCLUSION

In this paper, characterization of a novel composite material using printed interdigital-shaped capacitor structures

was presented. Inks consisting of Pr. A-, Pr. B-, and Pr. C-shaped BaTiO₃ particles in PMMA matrix were developed and printed on the top of the capacitors for the extraction of the material properties. The dielectric properties of the composite materials were influenced by the thickness variation of the ink layers on the capacitor structures. The results show moderate values of relative permittivity 30 for inks with Pr. A shape at 67.4 wt%, 25 for Pr. B shape at 66.3 wt%, and 27 for Pr. C shape at 67.1 wt% of BaTiO₃ particles at 2 GHz. In addition, low dielectric losses of 0.065, 0.040, and 0.025 were obtained, respectively. These new capacitor composite materials would be suitable for printed application in mobile telecommunication systems, especially in the frequency range of 0.5 to 3 GHz. The results further suggested that as the volume fraction of the filler contents in the composite materials varied, the dielectric properties and loss tangent of the materials can be manipulated in accordance to the area of applications.

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