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Innovative and Cost-Effective Fabrication Technique for Dielectric Composite Material

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ABSTRACT

In this paper, the fabrication process of epoxy resin and nanocomposite barium titanate using a cost-effective apparatus is presented along with measurements of complex permittivity. The material is prepared by mixing epoxy resin and barium titanate powder which has high permittivity. Measurement of complex permittivity of materials is performed using a waveguide technique in the G-band with a frequency between 4 and 6 GHz. The results are then compared with the previous works that use advanced and precision instrumentation. The results indicate that the material's permittivity, determined using our proposed technique, performs admirably. Specifically, at a frequency of 5 GHz, the permittivity is 7.32 with a loss tangent of 0.025 for a filler concentration of 20%. These values not only meet but also closely match those obtained with high-end equipment. Therefore, the proposed technique could be a good potential as an alternative technique for dielectric material preparation which is suitable to be used as a substrate antenna.

1. Introduction

The microwave field has a wide range of applications in biomedical, such as thermotherapy by Mustafa *et al.,* [1], laser ablation by Schena *et al.,* [2], microwave imaging by Alsawaftah *et al.,* [3], and photo-acoustic imaging by Fadhel *et al.,* [4]. To create small, lightweight, and strong designs for microwave applications, materials with high permittivity are required [5]. New material could be beneficial to the evolvement of microwave applications, where the electrical properties could be easily tailored based on the applications. Composite material, which is created by mixing or combining two or more different materials with various electrical and physical properties, is a promising solution [6]. One of these materials is known as the matrix, and the other main material is reinforcement in the form of fibers or particles to improve the matrix characteristics [7,8]. Examples of composite matrix can be polymer, metal, or ceramic. Epoxy resin is used as a polymer matrix in many high-technology applications due to its cost-effective material and high chemical resistance [9].

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Furthermore, ceramic nanoparticles such as barium titanate are considered very promising fillers in improving the dielectric and functional properties of nanocomposites [10]. In the medical sector, low dielectric permittivity values tend to produce higher characteristic impedance, increasing the reflection coefficient value [11]. To have a good value of reflection coefficient, high permittivity material is needed to enhance the properties of dielectric material for existing composite materials [12]. Ferroelectric ceramics are frequently employed to enhance the permittivity of composite materials due to their high permittivity coupled with minimal dielectric loss [13,14].

Composite material fabrication involves three main steps, which are mixing, degassing, and curing. In the mixing phase, filler and matrix are combined using machines like overhead stirrers [5-10] or mechanical stirrers [15]. Dissolved gases are eliminated through degassing, achieved using vacuum chambers or vacuum desiccators [16]. Curing can be achieved through methods such as a vacuum oven or rotary heater treatment followed by room temperature curing. Additionally, curing agents like diamino diphenyl methane (DDM) and 3-(Dodecylamino) propane nitrile can be employed [15-17]. Previous studies by Hasan *et al.,* [5] used similar epoxy-barium titanate composition but using high-end equipment. They employed an overhead stirrer and vacuum chamber for stirring and degassing, respectively, followed by vacuum oven curing and room temperature for final curing. Another study by Lévêque *et al.,* [18] emphasized multiple steps, including high-speed mixing, evaporation, vacuum desiccator degassing, and two-stage high-temperature curing for enhanced composite properties.

This paper focuses on innovative ways of making new composite materials that have higher permittivity value, cost-effective, and high functionality. Section 2 provides a detailed demonstration of the fabrication and characterization of the dielectric composite material while Section 3 presents the findings related to the properties of the dielectric material. Finally, this work is concluded in Section 4.

2. Methodology

The methodology employed in this study involves fabrication techniques for composite material and characterization techniques for high frequency covering the range from 4 GHz to 6 GHz. The composite material is formulated by enforcing nano-sized barium titanate powder into the epoxy system which is made of epoxy resin with a medium hardener. These materials were selected due to their compatibility, ease of processing, and potential for achieving the desired electrical properties. For the characterization procedure, a vector network analyzer (VNA) is employed to collect the scattering parameter data. After that, the technique proposed by Karim *et al.,* [19,20] is used to extract important dielectric properties, which are permittivity and loss tangent.

2.1 Preparation of Composite Material

Figure 1 shows the process of making composite material. The procedure begins by adding 60 g of barium titanate into 39 g of epoxy solution. The 60 g of barium titanate corresponds to a 20% filler concentration, determined by the following formulation [21]:

$$V_f = \frac{\frac{m_f}{\rho_f}}{\left(\frac{m_m}{\rho_m} + \frac{m_f}{\rho_f}\right)} \tag{1}$$

where V_f, m_f, m_m, ρ_f, and ρ_m stand for percent of filler, volume filler, volume matrix, density filler, and density of matrix, respectively. The epoxy resin used is EpoxAmite™ 120 medium hardener, with a matrix density of 1.25 g/cm^3 and a 50 g volume. Barium titanate has a density of 6 g/cm^3. The solution is then stirred using a USB portable electric foam maker. After mixing barium titanate and epoxy resin for three minutes, 11 g of epoxy hardener is added to the mixture, and the mixture is further mixed for another two minutes.

The mixing process can cause an air bubble in the mixture solution. Trapped air bubbles influence the electrical properties of the composite material. The air bubbles in the composite material can lessen the value of dielectric permittivity. In this study, the heating process is utilized to eliminate the air bubble in the mixture of epoxy resin and barium titanate ($BaTiO_3$). All the mixing procedures to prepare the composite material are carried out on a heated surface. In this study, the temperature of a hotbed is set at 60°C. Finding the right balance is crucial when setting the hotbed temperature. If it is too high, it introduces more bubbles during mixing, while too low might not effectively eliminate bubbles.

For the curing technique, the composite material is filled into the mold and allowed to rest overnight at an ambient temperature of 25°C for a 24-hour duration. After 24 hours, the composite material is removed from the mold and prepared for measurement of complex permittivity.

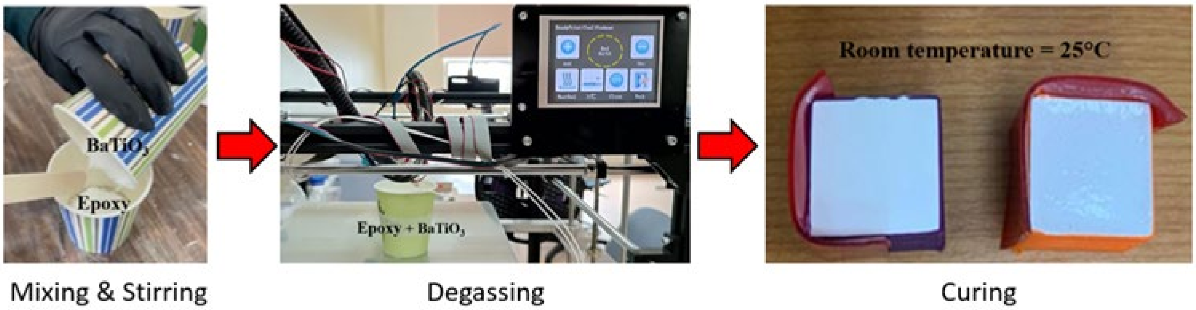

Fig. 1. Preparation of composite material

2.2 Characterization of Composite Material

In this study, the complex permittivity of composite material is measured based on the transmission-line method which uses a rectangular G-band waveguide rather than mono-frequency resonant methods [22]. This waveguide covers frequencies from 3.95 GHz to 5.85 GHz [23]. The waveguides are linked to a VNA via a coaxial cable and coaxial waveguide adapter as illustrated in Figure 2. The accuracy of the VNA is ensured through a comprehensive Short-Open-Load-Thru (SOLT) calibration of the two process ports. Then, the specimen is positioned in the middle of the rectangular waveguide [24,25], and the magnitude and phase of scattering parameters, S_{21}, are evaluated. The reason why S_{21} is used in this technique is that the reflection coefficient, S_{11}, is too small and can easily be affected by noise.

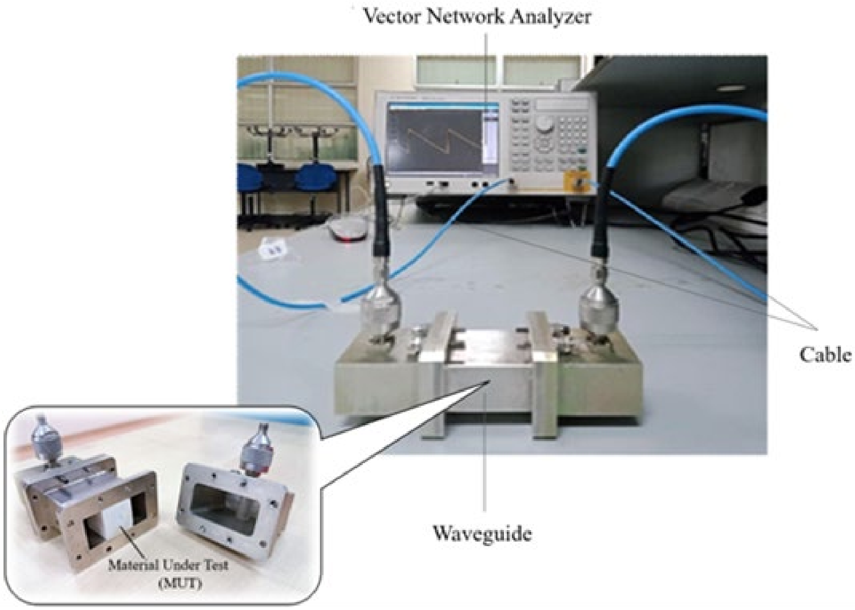

Fig. 2. Measurement setup

The electrical properties in terms of complex permittivity are obtained by using the adopted technique in [24] where the difference between the measured and calculated S_{21} is minimized by changing the complex permittivity. When the difference falls beneath the tolerance value, the final change value of the complex permittivity is endorsed as the electrical property of the specimen. Full illustrations of composite material characterization techniques are presented in Figure 3.

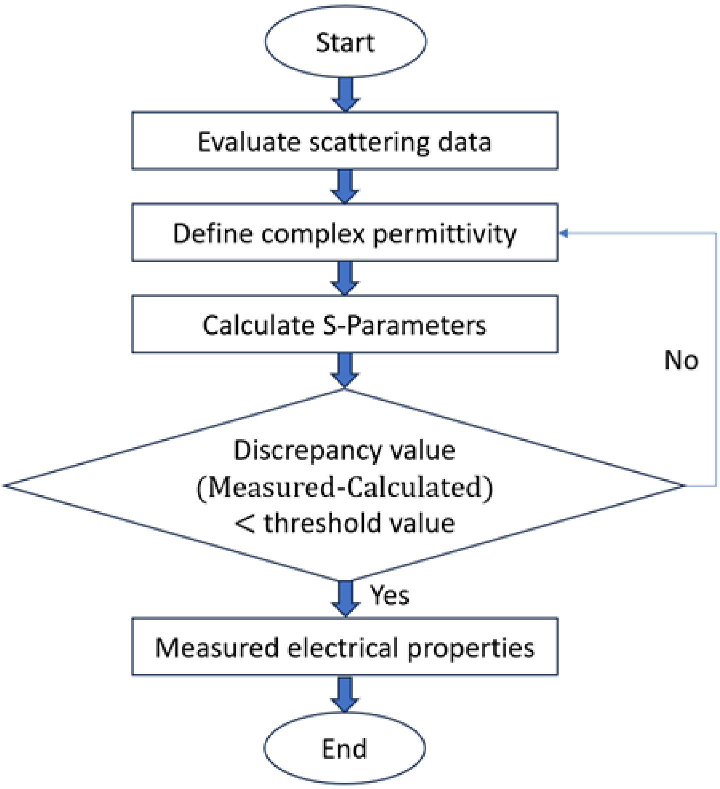

Fig. 3. Material characterization technique

3. Results

The complex permittivity of composite materials is measured between 4 and 6 GHz using the waveguide technique, and the value is compared with the previous study on epoxy-barium titanate composite. During the measurement, the composite material is measured several times to get a precise reading. In this study, the composite material is specifically evaluated at the frequency of 5 GHz. Most of the previous studies in microwave technology have shown that 5 GHz is the critical frequency range for certain applications, especially in communication systems and biomedical. In addition, 5 GHz is within a frequency range that is well-suited for testing and experimentation in laboratory settings.

Figure 4 shows the measurement result of the complex transmission coefficient, S_{21}, for the composite material of epoxy-barium titanate. The magnitude of S_{21} represents signal attenuation, where smaller values indicate higher signal loss and reduced transmission efficiency. Conversely, as the magnitude increases, it diminishes signal attenuation and enhances transmission efficiency. The phase reflects the degree of phase shift or time delay experienced by the signal, with larger values indicating more significant shifts. These parameters offer insights into how the composite material impacts signal strength and timing. The magnitude and phase of composite material for 201 points are recorded between frequencies 4 and 6 GHz. These values are then used to find the complex permittivity by using a technique by [24]. Figure 5 illustrates the permittivity and loss tangent for composite material between 4 and 6 GHz. The permittivity and loss tangent obtained at 5 GHz is 7.32 and 0.025, respectively.

The value is compared with the previous study and prediction model as stated in Table 1. Hasan *et al.,* [5] and Lévêque *et al.,* [18] utilizing the same epoxy-barium titanate composition focuses on a 20% filler concentration in composite fabrication. The resulting material displays permittivity and loss tangent values of 7.03 and 0.044 for Hasan *et al.,* [5], and 12.50 and 0.013 for Lévêque *et al.,* [18]. On the other hand, the composite material in this study exhibits higher permittivity (7.32) and lower loss tangent (0.025) than Hasan *et al.,* [5] at 20% concentration making it ideal for compact antennas, waveguides, and microwave applications due to improved signal propagation and energy efficiency. A low-loss tangent contributes to minimal signal distortion, reduced energy loss, and enhanced performance in applications like communication systems, antennas, and wireless power transfer, ultimately bolstering the reliability and effectiveness of electromagnetic applications.

In addition, this study also includes the data from the prediction model. The Jayasundere-Smith model [5] and the symmetric Bruggeman model [26] are used to calculate the permittivity value and the values obtained are 5.96 and 6.71, respectively. Eq. (2) and Eq. (3) show the theoretical model of Jayasundere-Smith and Bruggeman model, respectively.

$$\varepsilon_c = \frac{\varepsilon_m\left(1-V_f\right)+\varepsilon_f V_f \times \dfrac{3\varepsilon_m}{\varepsilon_f+2\varepsilon_m} \times \left[1+\dfrac{3V_f\left(\varepsilon_f-\varepsilon_m\right)}{\varepsilon_f+2\varepsilon_m}\right]}{1-V_f+\dfrac{3V_f\varepsilon_m}{\varepsilon_f+2\varepsilon_m} \times \left[1+\dfrac{3V_f\left(\varepsilon_f-\varepsilon_m\right)}{\varepsilon_f+2\varepsilon_m}\right]} \tag{2}$$

$$\varepsilon_c = \frac{1}{4}\left[3V_f\left(\varepsilon_f-\varepsilon_m\right)+2\varepsilon_m-\varepsilon_f+\sqrt{\left(1-3V_f\right)^2\varepsilon_f^2+2\left(2+9V_f-9V_f^2\right)\varepsilon_f\varepsilon_m+\left(3V_f-2\right)^2\varepsilon_m^2}\right] \tag{3}$$

where ε_c is permittivity of composite material, ε_m is the permittivity of matrix, ε_f and V_f are the permittivity and volume fraction of filler, respectively. The permittivity produced in this study, which is 7.32, is within an acceptable range when compared to these theoretical values. In fact, this study is not only within this acceptable range but also exceeded the predicted value, indicating a higher permittivity value than others.

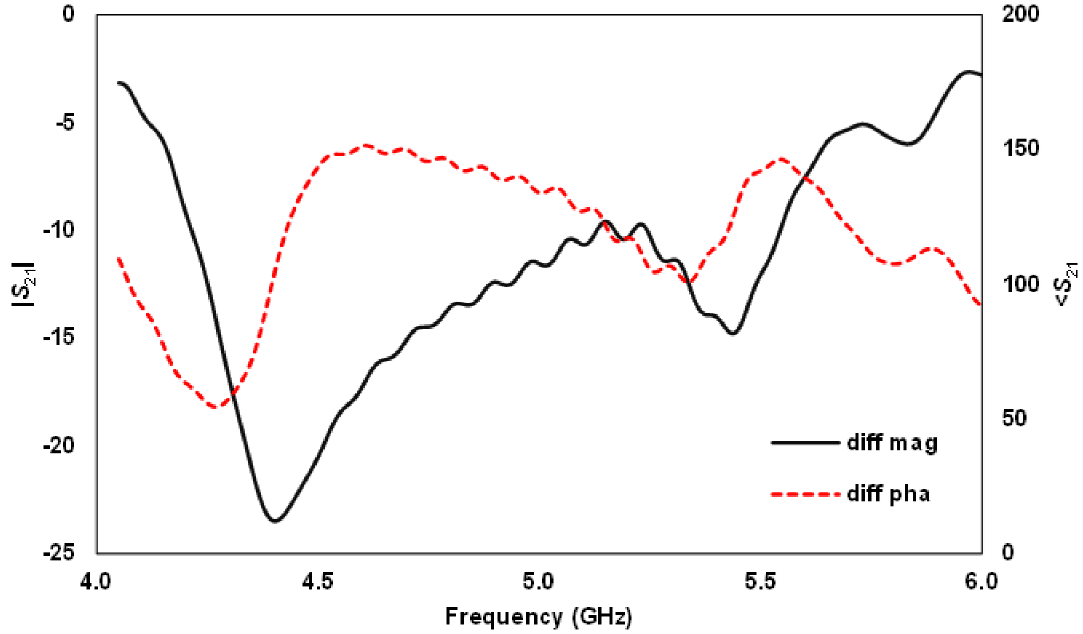

Fig. 4. Magnitude and phase of scattering parameter

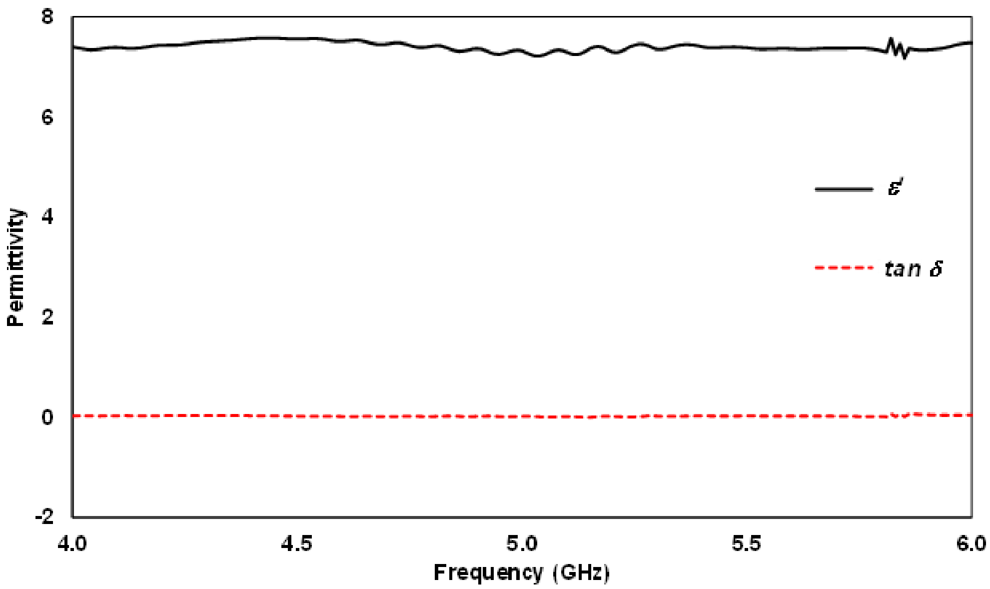

Fig. 5. The permittivity of composite material

Table 1
Comparison of dielectric properties with a different technique

Technique	Filler concentration	Frequency	Permittivity	Loss Tangent
This study	20%	5 GHz	7.32	0.025
Hasan *et al.,* [5]	20%	5 GHz	7.03	0.044
Lévêque *et al.,* [18]	20%	1 kHz	12.50	0.013
Jayasundere-Smith model [5]	20%	-	5.96	-
Symmetric Bruggeman model [26]	20%	-	6.71	-

4. Conclusions

Nanocomposite materials from epoxy and barium titanate nanopowder were fabricated for antenna application. The composite material was measured using VNA by using the waveguide technique at G-band frequency (4 to 6 GHz). The results show the permittivity of composite material in this study compared to Hasan *et al.,* [5] increased by 4.13% from 7.03 to 7.32. Meanwhile, loss tangent can be suppressed to be lower by 43.18% from 0.044 to 0.025. This occurs due to the rise in permittivity of epoxy when a higher concentration of barium titanate nanopowder filler is added to it. Thus, the result of permittivity is better and acceptable while the value of tangent loss is smallest and almost zero indicating that the material possesses low loss characteristics and is well-suited for use as an antenna substrate. Even though this method uses a cost-effective apparatus, it still can achieve higher permittivity and lower loss tangent.

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