



# Dielectric properties of epoxy–barium titanate composite for 5 GHz microstrip antenna design

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## Abstract

To fulfill the demands for robust, compact microstrip antenna for wireless communication, miniaturization is necessary. To achieve this, conventional dielectric microstrip antenna substrate can be replaced with high-permittivity composite dielectric material. Epoxy–barium titanate composite has potential to be used as antenna substrate. This paper focuses on fabrication of epoxy–barium titanate composite at different filler loadings. Then, the permittivity of the composite at G-band frequencies (4–6 GHz) is measured using waveguide technique. The effect of filler concentration to permittivity is observed at 5 GHz, the intended resonant frequency of the antenna. Waveguide technique determines the complex permittivity by analyzing only the measured transmission coefficient of the material, and easily noise affected reflection coefficient is not used. The experimental results show that the permittivity of epoxy–barium titanate increases steadily as the filler volume increases. At the highest filler volume (20%), the permittivity of the composite at 5 GHz is at 6.67. The results obtained are in good agreement with theoretically predicted values.

**Keywords** Permittivity · Polymer–ceramic composite · Microstrip antenna · G-band frequencies · Waveguide

## 1 Introduction

Wireless communication systems today are mostly compact, portable and multi-functional. These communication devices require small, lightweight and robust antennas to support fast and secure data transmission between devices. Microstrip antennas meet these requirements as they are robust, lightweight and have low profile. Since its discoveries in the late 1970s, microstrip antenna technology has been critically studied to improve its performance to suit the evolution of wireless communication technology, including studies on miniaturize microstrip antenna. There are several ways to meet the current demands for miniaturized microstrip antenna, and one effective way is by using high-permittivity dielectric material as antenna

substrate [1–3]. The substrates' permittivity controls the physical size, the radiation, the efficiency and the bandwidth of the antenna [4]. Commonly used dielectric substrates are FR-4 and RT-Duroid, which have permittivity of 4.4 and 2.2, respectively [4]. Thus, by substituting common antenna substrate with a higher-permittivity dielectric, the overall size of the antenna can be reduced. This can be achieved by using composite material as dielectric substrate.

Composite materials are developed by adding fillers into base materials such as polymer and epoxy resin [5]. Common fillers include ceramic powders, carbon fibers, glass fibers, hollow microspheres, graphene and carbon nanotubes. The permittivity  $\epsilon_r$  of composite material depends on the volume of filler material added to it. By

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varying the ratio of filler substance volume, permittivity  $\epsilon_r$  can be tailored to the desired value. High-permittivity materials have long been used in electronic devices. But some of these materials are unsuitable to be used as antenna substrate. Ceramics, for example, has high permittivity and widely used in microwave. However, they are fragile and need high sintering temperature, making the processing difficult and costly [6]. One solution to this problem is by mixing the high-permittivity material with low-permittivity base material and developing a balanced new composite material. This method allows the adjustment of the permittivity value of the new material, resulting to the antenna miniaturization with optimum potential.

In this paper, composite substrates consisting of epoxy resin–barium titanate is formulated at different filler concentrations. Epoxy resin is commonly used as based material for polymer–ceramic composite and has low-permittivity value of from 2 to 5 [7]. The permittivity of cured epoxy resin in G-Band has been measured in the previous work [8]. The value is around 2.7–3. Barium titanate on the other hand is a ferroelectric oxide material and is widely used in many electronic components [9, 10]. It could be in different shapes of crystals, cubic, tetragonal, orthorhombic and rhombohedral. The permittivity of barium titanate is usually very high depending on shape and size of the crystals, impurities and its processing techniques [9]. At room temperature, the permittivity for nanosized barium titanate particles is between 3500 and 6000 [7]. According to Ertuğ [10], the permittivity for nanosized barium titanate is 3279. The main factor that affects the permittivity of epoxy–barium titanate composite is the concentration of barium titanate filler [11–13]. However, the permittivity value can also be affected by other factors such as filler grain size, impurities of the filler material and the composite fabrication technique used [14–16]. This paper focuses on the effect of filler concentration to the permittivity of epoxy resin–barium titanate composite at 5 GHz.

Permittivity of epoxy resin–barium titanate composite varies at different frequencies. Singha and Thomas [17] describe that permittivity and loss tangent of a polymer system depend on the number of orientable dipoles in the molecular chains and their ability to orient under an applied electric field of certain frequency. In general, like other different types of polymer-based composite, the permittivity of epoxy resin–barium titanate composite decreases with frequency. The dipoles' ability to orient according to applied electric field is better at low frequencies compared to high frequencies [18]. This applies for both composites fabricated with micro-sized and

nanosized barium titanate filler [11, 15, 18, 19]. Hyun et al. [19] discovered that the permittivity of epoxy resin–barium titanate composite embedded capacitor films (ECFs) decreases when measured across frequency span between 1 MHz and 0.5 GHz. Similar findings are presented by Patsidis and Psarras [20] which concluded that the permittivity of epoxy resin–barium titanate composite decreases when measured across frequencies between  $10^{-1}$  and  $10^7$  Hz. Most of the open literatures are focused on measuring epoxy resin–BaTiO<sub>3</sub> permittivity at up to 1 GHz. Not many researchers are investigating the permittivity at microwave band. This paper investigates the permittivity of the epoxy resin–barium titanate composite at G-band frequencies (4–6 GHz). For this study, measurement was kept at room temperature, the size of filler particle was kept constant, and the influence of particles distribution and interfacial interactions between matrix and filler are not considered.

There are several ways to measure permittivity and loss tangent of materials, such as open-ended coaxial probe method, resonance method and waveguide method among others. Each technique has their own advantages and disadvantages such as limited frequency span for measurements and the type of material that can be measured [21]. The selection of which method to use depends on the types of material to be measured and the intended operating frequency. Permittivity of a dielectric material is expressed using complex number:

$$\epsilon = \epsilon'_r + \epsilon''_r$$

The real part represents the quantity of stored energy in the dielectric material from the external electrical field, while the imaginary part represents the amount of energy loss from the material due to an external electric field [21, 22]. Energy loss in a dielectric material is more commonly represented using loss tangent (also known as dissipation factor or loss factor). Mathematically, loss tangent ( $\tan \delta$ ) is the ratio of the imaginary part to the real part of the complex relative permittivity. In this paper, the complex permittivity of epoxy resin–BaTiO<sub>3</sub> composite is measured using rectangular waveguide technique. This technique was previously used to measure the complex permittivity of pure cured epoxy resin [8]. The advantages of using this technique are that the material under test (MUT) is less complex, where it does not need to perfectly fit the cross section of the waveguide. Moreover, this technique only utilizes the transmission coefficient,  $S_{21}$ , thus eliminating the use of reflection coefficient,  $S_{11}$ . Using  $S_{11}$  to obtain complex permittivity is prone to error, since  $S_{11}$  is too small and easily affected by measurement noise. Therefore, using  $S_{21}$  is the better alternative.

## 2 Methodology

### 2.1 Material

Commercially available epoxy resin and nanosized barium titanate powder were used in this study. The epoxy resin used is EpoxAmite™ epoxy system, with medium hardener. The weight ratio of 100:29 (100 g of epoxy resin and 29 g of hardening agent) is applied for the epoxy resin system, as suggested by the manufacturer. The filler used is nanosized barium titanate, with particle size of  $< 3 \mu\text{m}$  and 98.0% purity. The density of epoxy resin and nanosized barium titanate powder is  $1.25$  and  $6 \text{ g/cm}^3$ , respectively.

### 2.2 Sample preparation

Four different epoxy resin–barium titanate composite samples were fabricated, at the following volume ratio: 5 vol%, 10 vol%, 15 vol% and 20 vol%. The weight ratio (wt%) for base matrix and filler is then calculated based on the filler volume ratio. Fabrication process starts with mixing the appropriate amount of both epoxy resin and barium titanate nanosized powder in a container. Next, the mixture was stirred thoroughly using an overhead stirrer (WiseStir HS-30D) for 2 min. The stirrer speed was kept constant at 500 rpm throughout the process. Stirring was carried out at high speed, to allow the materials to mix well and minimize the agglomeration of the filler particles. Agglomeration would form large filler particles in the composite. This would eventually reduce the filler surface area to volume ratio, which would compromise the unique nanoparticle properties in the polymer [17]. The stirring process was kept at a short time, to reduce entrapment of air bubbles in the mixture [23]. Trapped air bubbles would affect the

mechanical properties of the composite [24]. To remove trapped air bubbles in the mixture, vacuum degassing technique is used. In this process, the mixture is placed inside a vacuum chamber (30 in.-Hg) for 2 min. This allows the air that is trapped within the mixture escape. The mixture is then carefully poured into a mold which has dimension of  $47.55 \times 22.15 \text{ mm}$  and thickness of  $22.10 \text{ mm}$ . This size is chosen as it is the same size of the waveguide that is used to measure the permittivity. The composite mixture is then let to cure inside vacuum oven at  $60^\circ\text{C}$  for 30 min. The epoxy resin system manufacturer suggests that curing time can be reduced by applying mild heat. High curing temperature also increases the heat deflection temperature of the composite. Curing at higher temperature also improves the mechanical properties of composite [25]. The composite mixture is further cured at room temperature for 24 h. The fabrication process was repeated for each different filler volume. Composite preparation process is depicted in Fig. 1.

### 2.3 Permittivity measurement

The waveguide used in this study is a G-band waveguide, which supports frequencies between 4 and 6 GHz. The measurement of  $S_{21}$  is taken using Agilent Vector Network Analyzer (VNA), which is connected to the waveguide by using coaxial cables and coax-waveguide adapters. The temperature during measurement was kept at room temperature. Before measurement can take place, the VNA is first calibrated using a full two-port Short-Open-Load-Thru (SOLT) calibration. This step is important to ensure accurate measurements. Then, the MUT with dimension of approximately  $22.10 \times 21.44 \text{ mm}$  is placed in the cavity of a waveguide as shown in Fig. 2.

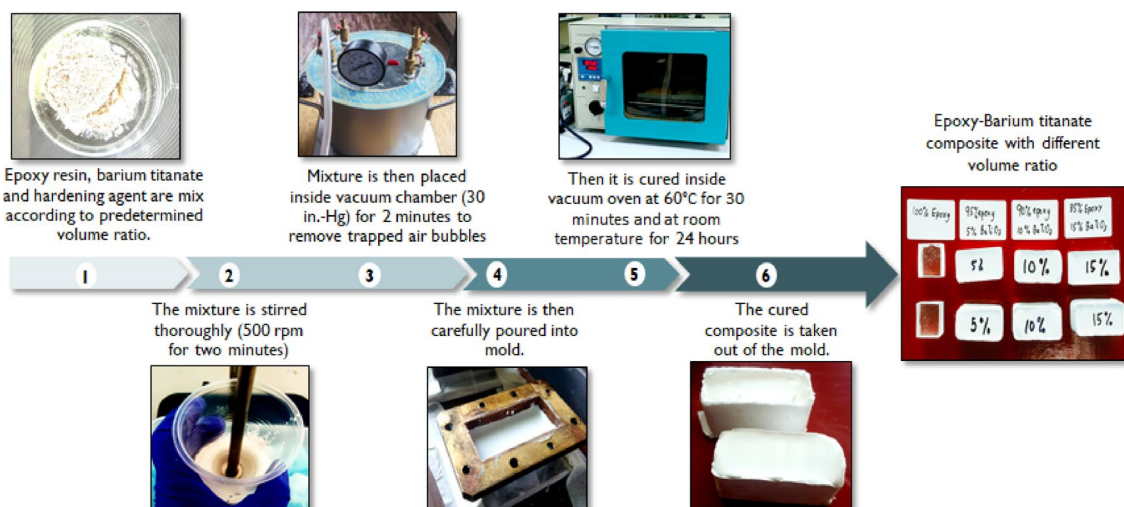


Fig. 1 Material preparation process

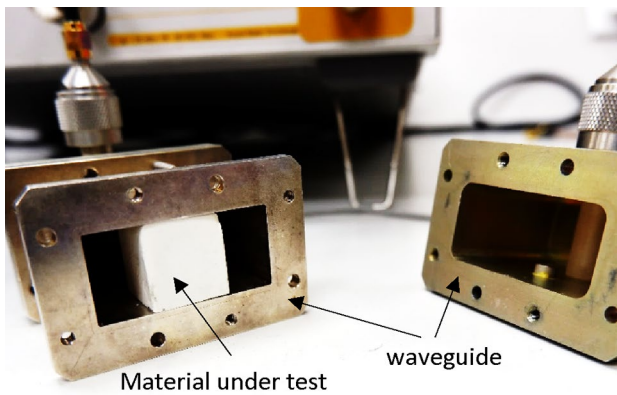


Fig. 2 MUT inside the cavity of G-band waveguide



Fig. 3 Complete measurement setup

Figure 3 shows the complete measurement setup. Then, the transmission coefficient,  $S_{21}$ , is measured across the waveguide. Both the magnitude and phase are measured in order to determine the real and imaginary part of complex permittivity. The complex permittivity is calculated by applying an EM field analytical method proposed by Miyagawa et al. [26, 27] and further improved by [28].

### 2.4 Mixing model/prediction

In order to estimate the permittivity of a composite mixture, theoretical models can be used. Some models are simple and straight forward, such as volume fraction model, in which the permittivity of the new composite material can be predicted using:

$$\epsilon_c = v_m \epsilon_m + v_f \epsilon_f \tag{1}$$

where  $\epsilon_m$  and  $v_m$  are permittivity and volume fraction of polymer matrix, respectively, while  $\epsilon_f$  and  $v_f$  are permittivity and volume fraction of ceramic filler, respectively.

This model, however, is inaccurate [7, 29] compared to more complex models listed as follows [11, 13, 19, 30–33]:

- i. Maxwell–Garnett model:

$$\frac{\epsilon_c - \epsilon_m}{\epsilon_c + \epsilon_m} = V_m \frac{\epsilon_f - \epsilon_m}{\epsilon_f + 2\epsilon_m} \tag{2}$$

- ii. Lichtenecker model:

$$\log \epsilon_c = V_f \log \epsilon_f + (1 - V_f) \log \epsilon_m \tag{3}$$

- iii. Jayasundere–Smith model

$$\epsilon_c = \frac{\epsilon_m(1 - V_f) + \epsilon_f V_f \times \frac{3\epsilon_m}{\epsilon_f + 2\epsilon_m} \times \left[ 1 + \frac{3V_f(\epsilon_f - \epsilon_m)}{\epsilon_f + 2\epsilon_m} \right]}{1 - V_f + \frac{3V_f \epsilon_m}{\epsilon_f + 2\epsilon_m} \times \left[ 1 + \frac{3V_f(\epsilon_f - \epsilon_m)}{\epsilon_f + 2\epsilon_m} \right]} \tag{4}$$

- iv. Wiener upper limit:

$$\epsilon_c = V_f \epsilon_f + (1 - V_f) \epsilon_m \tag{5}$$

- v. Wiener lower limit:

$$\epsilon_c = \frac{\epsilon_f \epsilon_m}{V_f \epsilon_f + (1 - V_f) \epsilon_m} \tag{6}$$

- vi. Symmetric Bruggeman model

$$\epsilon_c = \frac{1}{4} \left[ 3V_f(\epsilon_f - \epsilon_m) + 2\epsilon_m - \epsilon_f + \sqrt{(1 - 3V_f)^2 \epsilon_f^2 + 2(2 + 9V_f - 9V_f^2) \epsilon_f \epsilon_m + (3V_f - 2)^2 \epsilon_m^2} \right] \tag{7}$$

Permittivity obtained from the measurements is then compared to mixing model value, to observe the accuracy.

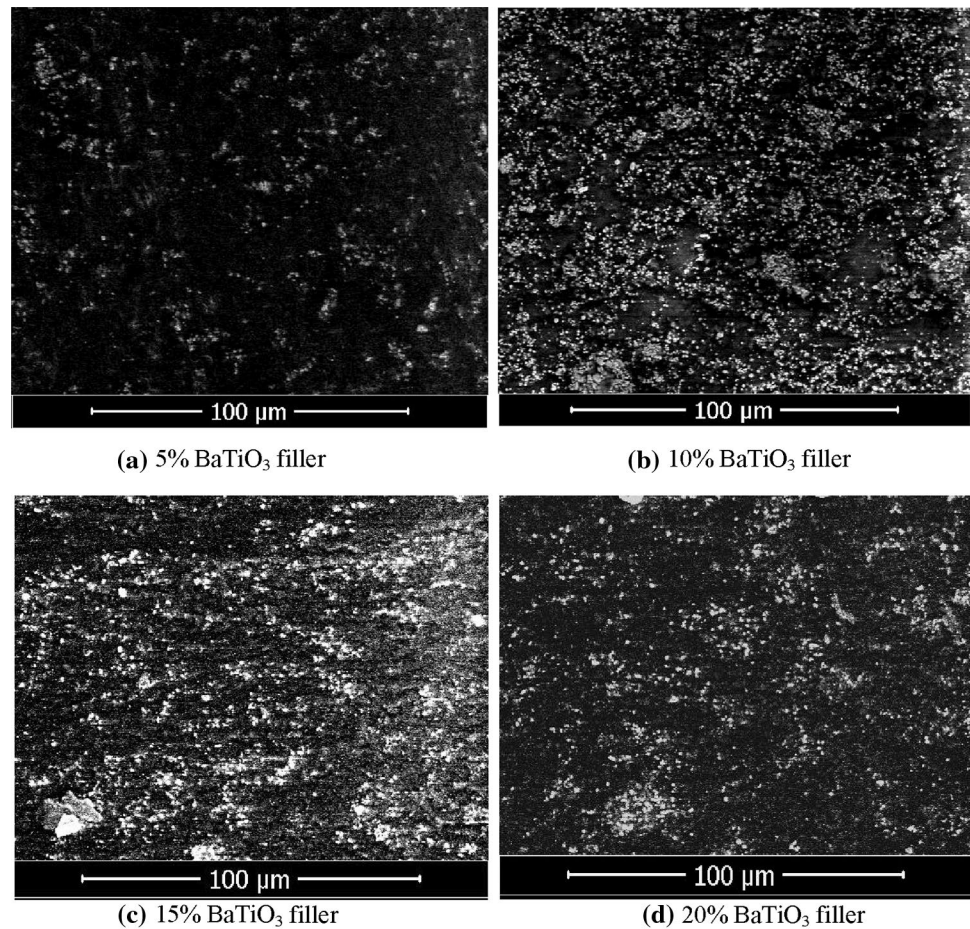
## 3 Results

### 3.1 SEM micrographs

Figure 4a–d shows the cross-sectional SEM of the epoxy resin–BaTiO<sub>3</sub> composites with varying filler content. Dispersions of barium titanate nanopowder can be clearly seen in the SEM micrographs, where light-colored dots represent barium titanate nanopowders and the dark background is epoxy resin matrix. In general, it can be seen that for all filler concentrations, filler particles were dispersed well within epoxy resin matrix, with only small trails of clusters and pores found. However, more clusters of filler particles can be found in composite with 15 vol% and 20 vol% filler volume ratios, compared to 5 vol% and 10 vol%. This is expected because of the higher concentration of filler particles in the composite. As the concentration of



**Fig. 4** SEM micrographs of epoxy resin–barium titanate composite at various filler contents

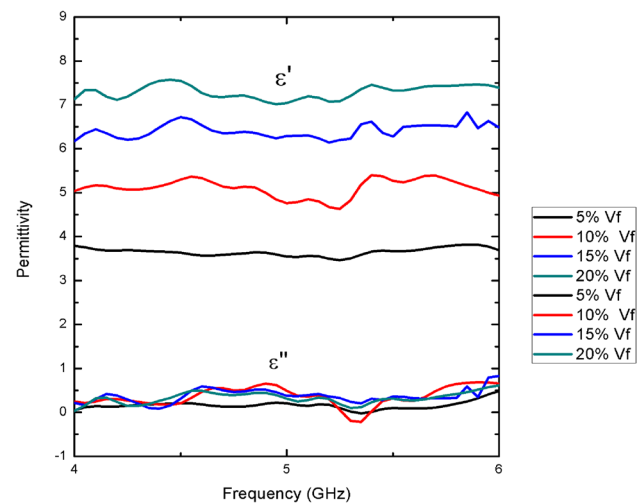


nanofiller increases, the concentration of epoxy resin decreases, thus causing the nanosized filler particles to cluster together.

The images of composites with higher filler loading show more bright dots. The particle sizes of the filler are among a few micrometers. With the increase of ceramic filler, slight aggregation appears in certain areas, which illustrates that the volume fraction of filler could not be larger, because that might lead to poor mechanical properties as a result of the agglomeration of the fillers (Homogenous).

### 3.2 Electrical properties: permittivity, loss tangent and permeability

Figure 5 shows the real and imaginary values of permittivity for epoxy resin–barium titanate composite, measured between 4 and 6 GHz. At 5 vol% filler, the permittivity is around 3.5–4. As the filler volume ratio is increased, the permittivity of the composite material increases steadily. At the highest filler concentration (20 vol%), the permittivity is around 7–7.5. For all filler volume ratio, the permittivity remains stable throughout the G-Band frequencies.



**Fig. 5** Permittivity versus frequency

The imaginary values for all composite samples exhibit the same pattern except for 10 vol%. The imaginary permittivity value for 10 vol% shows significant drop at frequencies around 5.4 GHz. Therefore, the loss tangent also follows

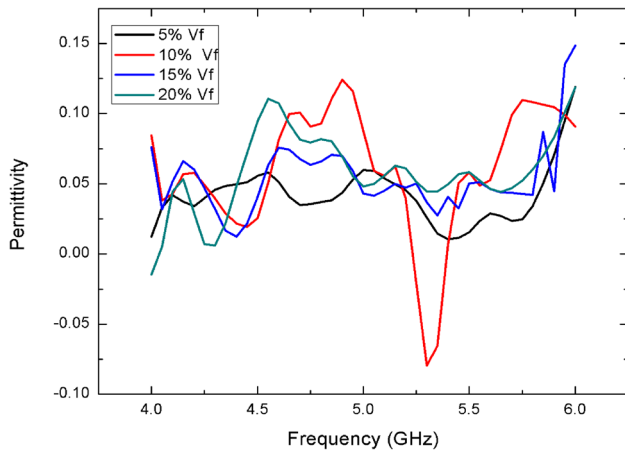


Fig. 6 Loss tangent versus frequency

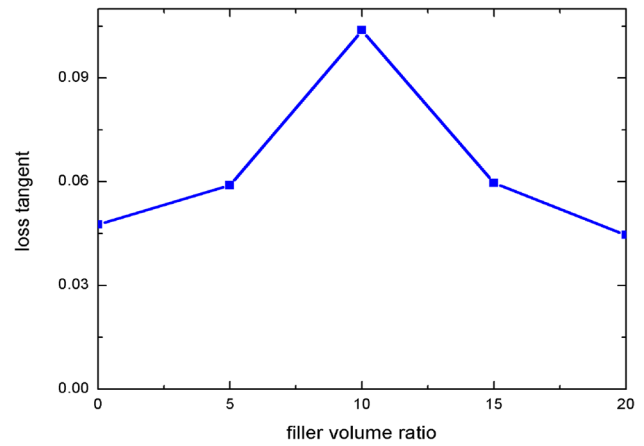


Fig. 8 Loss tangent versus filler volume (vol%)

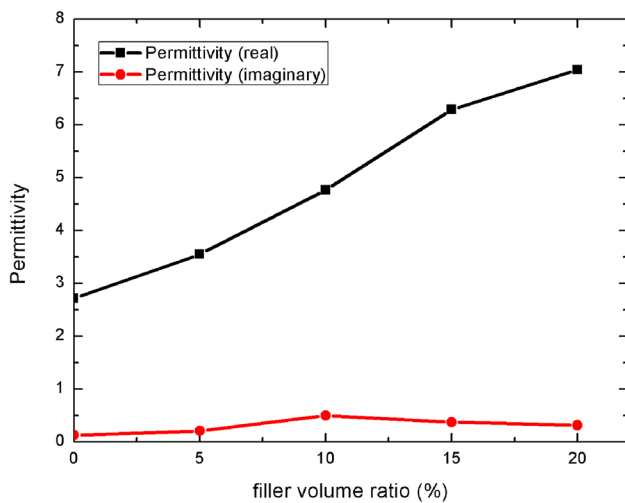


Fig. 7 Permittivity versus filler volume (vol%)

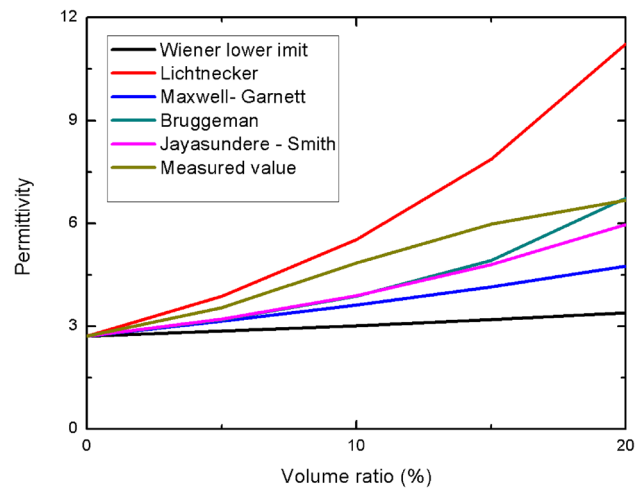


Fig. 9 Permittivity prediction and experimental value comparison

this pattern as shown in Fig. 6. This may be caused by error in measurement or with sample preparation. However, the loss tangent values for all the composites are still considered low and acceptable.

Figure 7 shows the permittivity of epoxy resin–barium titanate composite at 5 GHz. The real value of permittivity increases almost linearly with the filler volume, while the imaginary value only increases slightly. The resultant loss tangent of epoxy resin–barium titanate composite at 5 GHz is shown in Fig. 8. The loss tangent shows increasing trend at the beginning, with the highest at 10% filler volume. Then, the value decreases, with the lowest value of 0.043 at 20% filler volume.

### 3.3 Comparison with models for effective permittivity

Figure 9 shows the comparison of experimental measured permittivity at 5 GHz with theoretical value. It is observed that the experimentally measured permittivity shows the same trends with all the predictions except for Wiener lower limit and Lichtenecker. Based on the results, the measured permittivity can be considered as the valid results. Therefore, the composite process is considered successful. Of all the theoretical predictions, Jayasundere–Smith and Maxwell Garnet models show minimal discrepancies to the measured value.

### 3.4 Comparison with other materials

Table 1 shows comparisons of proposed epoxy resin–barium titanate composite’s permittivity and loss tangent

**Table 1** Comparison of permittivity with other materials

Material	Frequency	Permittivity	Loss tangent	References
Proposed material: (20% filler vol. ratio) Filler particle size 300 nm	5 GHz	7.039186	0.044609	This work
FR-406	5 GHz	3.92	0.0172	Datasheet [34]
RT/Duroid 5870	1 MHz–10 GHz	2.33 ± 0.02	0.0012	Datasheet [35]
RT/Duroid 5880	1 MHz–10 GHz	2.20 ± 0.02	0.0009	Datasheet
PDMS–BaSrTiO <sub>3</sub> (26% filler vol.ratio)	4 GHz	11.69	0.0380	Castro et al. [36]
Epoxy–BaTiO <sub>3</sub> (40% filler vol. ratio) Filler particle size 100 nm	5 GHz	~ 11	~ 0.04	Yang et al. [13]
Epoxy–BaTiO <sub>3</sub> (40% filler vol.ratio) Filler particle size 200 nm	5 GHz	~ 15	~ 0.06	Yang et al. [13]
Polyethylene (HDPE)-polystyrene-coated BaO– Nd <sub>2</sub> O <sub>3</sub> –TiO <sub>2</sub> (BNT) (20% filler vol. ratio)	1 GHz	~ 4	Not mentioned	Zhang et al. [6]
PLA	1–10 GHz	3.549	– 0.001	Huber et al. [37]
Epoxy resin	5 GHz	2.71652	0.047607	Karim et al. [8]

with other materials. The composite materials in this work have higher dielectric permittivity compared to PLA, FR-406 and RT/Duroid substrates. The loss tangent of the proposed composite, however, is higher. The table also compared the permittivity and loss tangent of the composite material with cured epoxy resin. In this case, the permittivity is higher compared to cured epoxy resin, and the loss tangent has the same value. Finally, the proposed composite properties are on par with the other types of composites listed in Table 1.

## 4 Conclusion

High-permittivity substrate is desired to achieve antenna miniaturization. This can be achieved with epoxy resin–barium titanate composite. The results show the permittivity of epoxy resin is increased when barium titanate nanopowder is added to it. Higher concentration of filler leads to higher value of permittivity. However, composite with high content filler volume is more prone to agglomeration and degrade the performance of the material. Accurate and reliable permittivity measurement is a crucial step that needs to be done before a dielectric material can be used as antenna substrate; thus, waveguide technique is suitable since it is accurate and reliable.

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## Compliance with ethical standards

**Conflict of interest** On behalf of all authors, the corresponding author states that there is no conflict of interest.

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