IOP Conference Series: Materials Science and Engineering

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To cite this article: Nurulfadzilah Hasan *et al* 2021 *IOP Conf. Ser.: Mater. Sci. Eng.* **1045** 012012

Fabrication and characterization of epoxy resin–barium titanate at G-band using waveguide technique

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Abstract. In this paper, fabrication process of epoxy resin-barium titanate nanocomposite and measurement of its complex permittivity are presented. The material is prepared by mechanical mixing of epoxy resin and barium titanate nanopowder. The nanocomposite is intended to be used as high permittivity microstrip antenna substrate, which requires accurate measurement of its electrical characteristics. Thus, characterization of materials is done using waveguide technique, which does not require a precise machining of sample's width and thickness, and does not utilize small reflection coefficient, which can cause error in measurement. The complex permittivity of the nanocomposite is measured in G-band (4 to 6 GHz). Then, the measured values are compared with prediction method, Lichtnecker and Maxwell-Garnet method. The results show that the measured permittivity of composite materials are in good range with prediction method, while the measurements of loss tangent show that the developed materials are low-loss and suitable to be used as substrate of antenna.

Keywords: antenna substrate, barium titanate, epoxy resin, material characterization, material fabrication

1. Introduction

In our modern society, microwave materials have been used widely by researcher in a lot of advanced communication devices. For this reason, a wide variety of new materials have been developed especially for communication system [1], radar system [2], energy storage device [3], and industrial waste [4]. These devices require high permittivity of microwave material to develop a small, lightweight and robust design [5]. The dielectric permittivity of the composite can be enhanced by adding ferroelectric ceramics, such as barium titanate ($BaTiO_3$) and lead magnesium niobate ($MgNb_2O_9Pb_3$)[6]. They are used as fillers in dielectric composites.

In this study, a technique to prepare and characterize composite substrate of antenna, which is made of epoxy resin and barium titanate is presented. Epoxy resin has been widely used due to their low cost, high moisture, and high chemical resistance [7], while barium titanate is well known as high permittivity material [8]. Thus, the mix of these two materials can afford high value of permittivity in order to be used in compact and small antenna design.

Prediction methods such as Lichtnecker [9]-[11], Maxwell-Garnett [9]–[12], Jayasundere-Smith [11], effective medium theory [11], and Yamada model [10] have been used to guess the value of permittivity for composite material involving volume fraction of base matrix and ceramic material. In microwave region, the permittivity is decreasing as the frequency increases. Hence, these methods are not applicable to estimate permittivity for antenna applications, as the methods not consider frequency changes but yet important to get initial idea of permittivity during fabrication of material. In the past, many techniques have been carried out to determine complex permittivity (permittivity and loss tangent) of material in microwave measurement such as free-space method [13], resonant method [14]-[15], and transmission-line method [16]–[18]. Measurement for free-space method is among the easiest setup, which is typically, consists of two horn antennas as transmitter and receiver while the sample is placed between the antennas. However, the measurement using this method is easily affected by surrounding and cause inaccuracy determination of complex permittivity. Alternatively, resonant method able to provide high accuracy of measurement, but the measurement can be done only at one point of frequency. In this paper, the fabricated substrate is measured using transmission-line method in order to provide wide-frequency measurement while the accuracy is comparable with resonant method. G-band waveguide, frequency range from 4 to 6 GHz, is used in this method to measure scattering parameters. Then the complex permittivity will be determined from scattering parameters using an accurate and effective hybrid electromagnetic method as proposed by [19].

The rest of this paper is organized as follows. Section 2 explains the prediction of permittivity and sample fabrication of nanocomposite materials. Then, the measurement and complex permittivity estimation of fabricated materials is described in Section 3. Then, the cumulative results are presented in Section 4. Finally, Section 5 concludes our works.

2. Nanocomposite Sample Preparation

2.1. Permittivity Prediction Method

In general, adding high permittivity materials such as barium titanate to a low permittivity material, such as epoxy resin will increase the permittivity of the composite. Furthermore, the permittivity increases as the concentration of filler is increase. Thus, the permittivity of a composite material can be predicted by using theoretical models such as Lichtnecker, Maxwell-Garnett, Jayasundere-Smith, effective medium theory, and Yamada model. The theoretical models relate the permittivity of the new composite material, with the permittivity and the volume fractions of base matrix and filler. In this work, commonly used Lichtnecker and Maxwell-Garnett model are adopted to verify our permittivity's measurement. The relationship between volume fraction of base matrix and ceramic material, and permittivity in Lichtnecker model [9] is given as following;

$$\ln \varepsilon_{reff} = v_1 \ln \varepsilon_{r1} + v_2 \ln \varepsilon_{r2} \tag{1}$$

where, ε_{reff}, ε_{r1} and ε_{r2} are the permittivity of the composite, base, and ceramic material, respectively. v_1 and v_2 are the volume fraction of the base and ceramic material, respectively.

Meanwhile, according to Maxwell-Garnett model [9], the relationship can be represented as;

$$\frac{\varepsilon_{reff} - \varepsilon_{r2}}{\varepsilon_{reff} + 2\varepsilon_{r2}} = v_1 \frac{\varepsilon_{r1} - \varepsilon_{r2}}{\varepsilon_{r1} + 2\varepsilon_{r2}} \tag{2}$$

2.2. Sample Fabrication Technique

Base matrix of the composite is EpoxAmite™, epoxy resin system with medium hardener. The epoxy resin system has density of 1.25 g/cm³ and permittivity between 2.7 and 3 [16]. Filler of the composite is barium titanate nanopowder, with particle size of less than 3 μm and 98.0% purity. The density is 6 g/cm³, while the permittivity is 3279 [20].

To prepare composite material with 5 vol.% filler, first the weight ratio (wt.%) for base matrix and filler is calculated based on the filler volume ratio and density of the materials. Fabrication process starts with adding 12.63 g barium titanate nanopowder to 50 g epoxy resin system. The mixture is then stirred thoroughly using an overhead stirrer (WiseStir HS-30D) at 500 rpm for two minutes. Stirring at high speed is necessary to ensure the filler particles are well dispersed within the base matrix. However, it must be done within a short period to minimize trapped air bubbles in the mixture. Next, the mixture is placed inside a vacuum chamber (30 in.-Hg) for 2 min to remove trapped air bubbles. Then mixture is carefully poured into mold with dimension of 47.55 mm × 22.15 mm and thickness of 22.10 mm. Curing of the composite is done in two stages. First, the mixture is let to cure inside vacuum oven at 60°C for 30 minutes. This is done to remove trapped air in the mixture and improves the mechanical properties of composite [21]. The composite mixture is then further cured at room temperature for 24 hours. Finally, the cured composite is taken out of the mold and prepared for permittivity measurement. Composite preparation process is depicted in Figure 1.

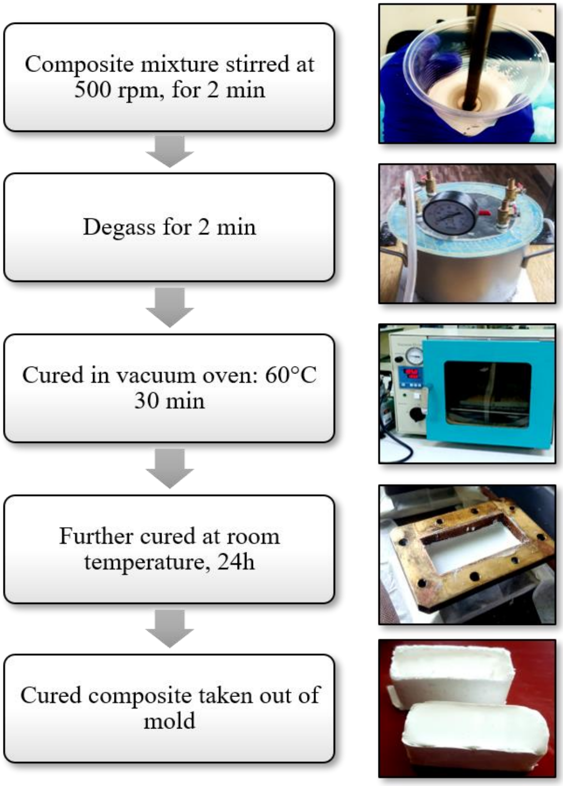


Figure 1 Material preparation process.

3. Material Characterization Based on Waveguide Technique

3.1. Sample Preparation and Measurement

Three fabricated nanocomposite samples are machined to fit the height of G-band waveguide, i.e., 22.15 mm, while the width and length can be varied in order to ease the processes of sample preparation and measurement. Figure 2 shows the sample material under test (MUT). The dimensions for MUT 1, MUT 2 and MUT 3 are 20.94 mm × 21.96 mm, 21.12 mm × 21.77 mm, and 21.15 mm × 16.57 mm, respectively.

Figure 3 shows the experimental setup to measure the scattering parameters. First, MUT is partially placed inside the waveguide. Then, the measurement is done by connecting the waveguide to the network analyzer using low-noise RF cable and waveguide-to-coax adapters. Partially loading MUT into the waveguide is beneficial, since TE_{10} mode characteristic; where the electric field is stronger at the center of the waveguide can be fully utilized. For instance, if the sample absorb high level of energy, the transmission parameter will be very low and lead to inaccurate measurement. Thus, to overcome this problem, the sample might be placed at the sidewall of the waveguide where the electric field is weak so that less energy will be absorbed by the sample.

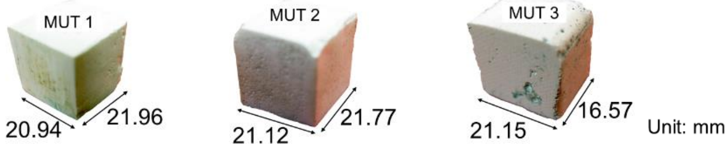


Figure 2 Material under test

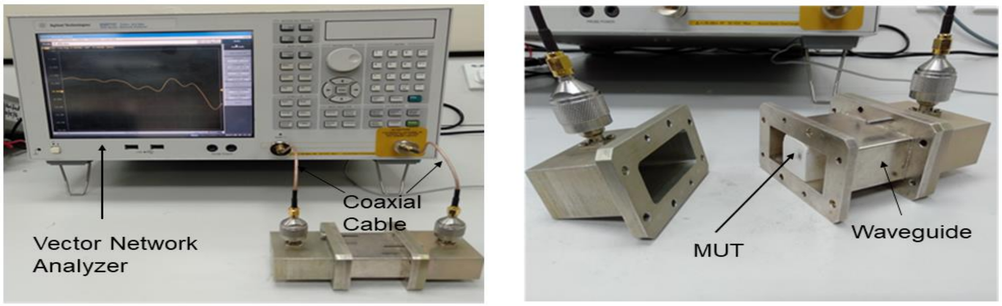


Figure 3 Measurement setup

3.2. *Material Characterization Technique*

To enhance the accuracy of material characterization, systematic errors is suppressed by utilizing calibration-free technique of waveguide measurement [22]. It is done by taking difference of complex transmission parameters (magnitude and phase) of waveguide measurement with and without MUT. Hence, this technique is able to eliminate electrical disturbances from measurement setup such as noise of cable and vector network analyzer (VNA).

Electrical properties in terms of complex permittivity are derived by resorting inverse technique [14] where the difference of measured and calculated complex transmission parameters are minimized by changing the complex permittivity. Once the difference is below than tolerance value, the last changed value of complex permittivity is adopted as the electrical properties of MUT. The full illustration for the material characterization technique is shown in Figure 4.

4. Results and Discussions

Permittivity of composite materials are measured using waveguide technique and the compared with prediction methods as mentioned above. Meanwhile, the measurement of loss (tangent delta) will be presented even the developed composite material is low loss, yet important for an accurate antenna design. However, there is no comparison between measurement and prediction data for tangent delta since the prediction value will lead to almost zero value due to low loss properties.

Figure 5 shows the measured complex transmission parameters (magnitude and phase), S_{21}, for all three MUTs within frequency range of 4 to 6 GHz. There is no significant resonance of magnitude of S_{21}, so it is very promising to obtain an accurate measurement of complex permittivity. Complex permittivity is determined from complex transmission parameter by resorting the inverse technique explained above (section 3).

4.1. *Permittivity/Dielectric Constant*

In this section, the prediction value of permittivity of epoxy resin with 5% filler of $BaTiO_3$ is compared with the measurement value done using waveguide technique. Figure 6 depicts the measured permittivity of all nanocomposite materials. The prediction values using Lichtnecker and Maxwell-Garnett model are included for comparison. All measured values are in good range with the predicted value. However, the measured value for each MUT is slightly different due to the difficulty to keep the same conditions during fabricating and percentage of trapped air bubbles might different. Hence, each sample has its own characteristics and measurement for each sample is necessary before using it for design purposes. It should be noted that the predicted values are not frequency-dependent. Therefore, it can be a reference for developing material but cannot be used for design purposes.

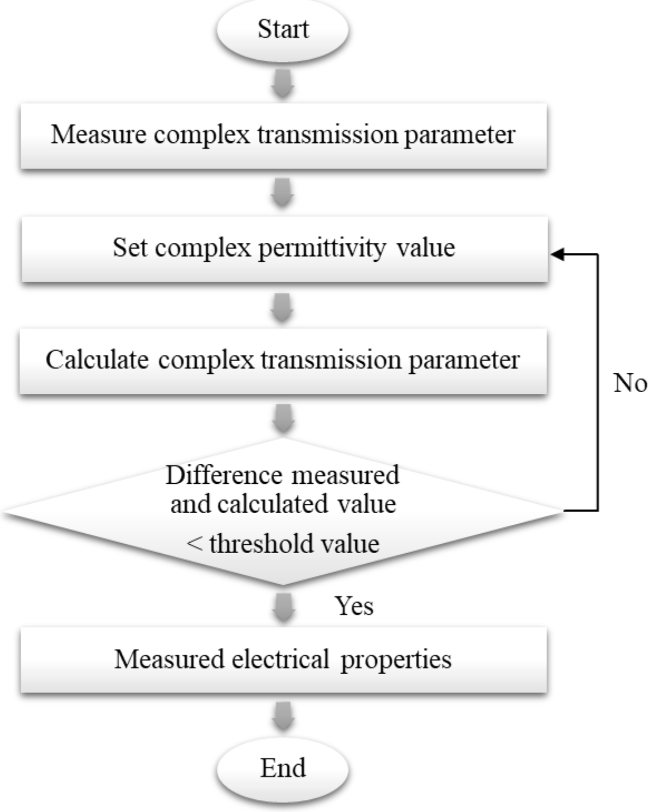


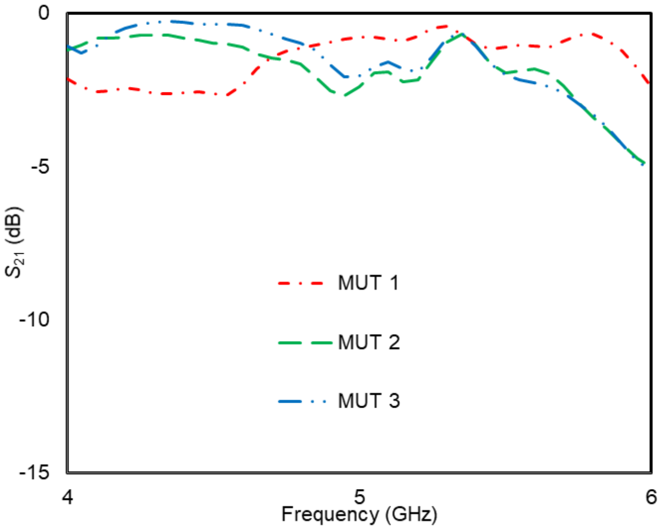
Figure 4 Material characterization flow.

4.2. Tangent Delta

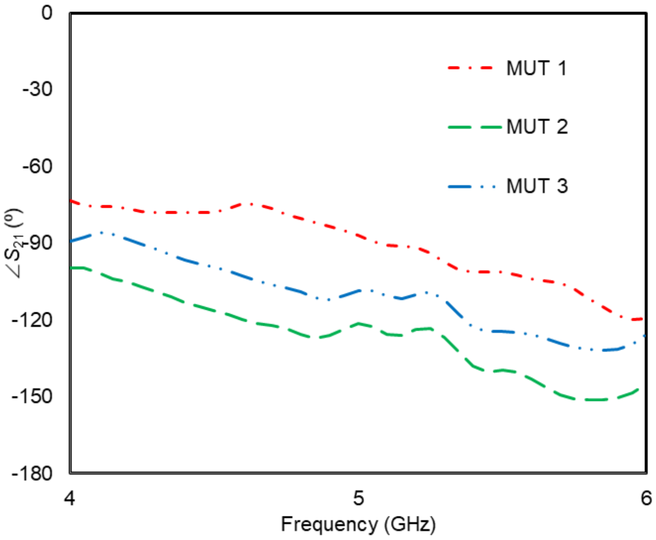
Value of tangent delta for these nanocomposites are small, yet important for design purposes. Figure 7 shows the measured value of tangent delta. All three samples have tangent delta values that are consistently close to zero. Small value of tangent delta is desirable for an antenna substrate, because the loss is small.

5. Conclusions

Nanocomposite materials made of epoxy resin system as base matrix and barium titanate nanopowder as filler were fabricated for antenna applications. The necessary electrical properties for antenna designing were measured using waveguide technique at 4 to 6 GHz. This measurement method was promising since there is no resonant for measurement of scattering parameters. The measured values of permittivity were compared with two prediction methods, Lichtnecker, and Maxwell-Garnett model. They are in good range with the predicted value. Meanwhile, the measured values of loss tangent were almost zero, which is indicated that the fabricated nanocomposite material is low loss and suitable to be used as substrate of antenna.



(a) Magnitude of transmission parameter, S_{21}



(b) Phase of transmission parameter, S_{21}

Figure 5 Measured complex transmission parameter.

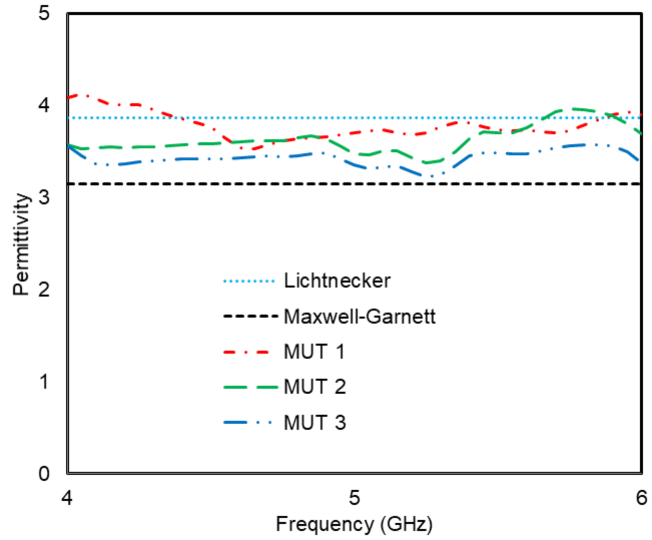


Figure 6 Comparison of measured and predicted permittivity.

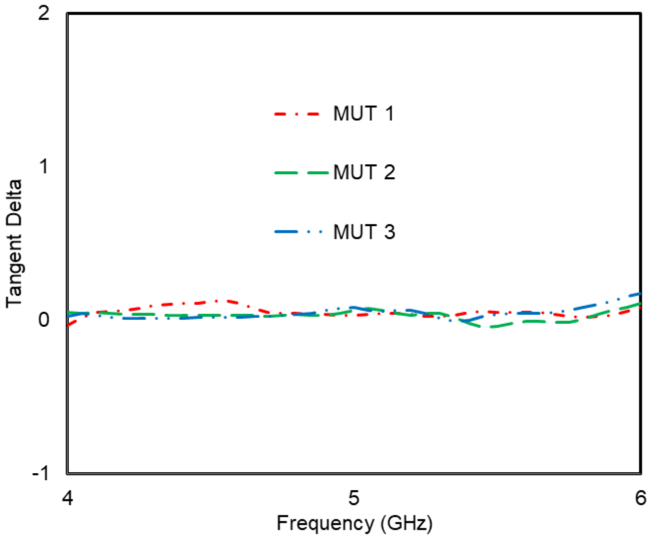


Figure 7 Measured values of tangent delta (loss tangent).

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Acknowledgments

This work was supported in part by the Universiti Malaysia Pahang and Ministry of Education (MOE) Malaysia under Fundamental Research Grant Scheme (FRGS) with reference no. FRGS/1/2018/STG02/UMP/02/1 or RDU190140. This work also was partly supported by the Research and Innovation Department, Universiti Malaysia Pahang under Postgraduate Research Grants Scheme (PGRS) with reference no. PGRS200315.