Design and characterization of a magneto-dielectric composite in high frequency with aligned magnetite powders

L A Lara¹, D L Mancipe¹, Y Pineda¹, J J Moreno² and G Peña-Rodríguez³
¹ Universidad Pedagógica y Tecnológica de Colombia, Sogamoso, Colombia
² Centre of High Frequency Engineering Cardiff University, Cardiff, U.K.
³ Universidad Francisco de Paula Santander, San José de Cúcuta, Colombia

E-mail: luisangel.lara@uptc.edu.co

Abstract. A magneto-dielectric material composed of a polyester resin-based microstrip (P115A), copper sheets and magnetite powders was designed in concentrations of 10, 20 and 30% Wt and with filters of 200, 325 and 500. The particles were aligned vertically and horizontally during the curing process using 300 mT magnetic fields. From a complete factorial design of 3³, 27 microstrip-type circuits of 4 mm width, 70 mm length and 0.93 mm thickness were manufactured, characterized by scanning electron microscopy and vector network analysis. The cross-matrix analysis determined an optimal circuit response from the magneto-dielectric material with a concentration of 20% magnetite and an average particle size of 21.48 µm in horizontal alignment to the applied magnetic field and to the transmission line, obtaining a relative dielectric constant of Er 3.88 with a low dielectric loss of 0.054, within an operating range of 150 KHz to 4 GHz.

1. Introduction

The objective of this research work was to verify the effect of the concentration, size and distribution of magnetite particles on the properties of a magneto-dielectric composite, characterized by vector network analysis (VNA). This type of research has gained relevance because advances in information and communication technologies have required the development of new electronic devices and components in order to increase the speed of the transmission of large volumes of information [1]. That, in turn, has multifunctional benefits, such as high permeability and low losses [2], in addition to inductive and capacitive properties [3], which enable the manufacture of next generation microwave communication devices [4]. This type of material is known as a magneto-dielectric composite [5], the functionality of which is closely linked to the relationship between the matrix and the particles [6], the size and distribution of which should be studied in order to improve the performance of this type of material [7,8].

Regarding the above, the manufacturing method of a magneto-dielectric material made of P115A polyester resin and micrometric particles of magnetite is here presented, which were elaborated in three configurations according to the disposition of the particles in the polymeric matrix [9], arranged randomly, aligned in vertical and horizontal positions with respect to a constant magnetic field of 300 mT during the curing process [10]. Microstrips were made by adding two layers of conductive copper while curing in the presence of the magnetic field.

The characterization of the material was carried out by means of a Rohde & Schwarz ZVB8 Networks vector network analyzer, through measurements on a transmission line of 4 mm width, 70 mm length
and 0.3 mm thickness on the substrate. Twenty-seven samples were characterized under a complete factorial design of $3^3$ variable particle sizes, concentrations and magnetite dispersions in the polymer matrix, reporting the dielectric properties obtained from the analysis of the parameters S of the 27 transmission lines designed, obtaining an optimized model for the configuration with particles aligned horizontally with respect to the magnetic field, in a concentration of 20% Wt by weight of magnetite, with an average particle size of 21.48 microns.

2. Material and Methods

2.1. Materials

As raw materials for the preparation of the composites, magnetite powders supplied by Green Magnetite S.A. of reference G-Mag A® were used, with a density of 5100 Kg / m$^3$ and a particle size of less than 75 μm, filter 200. The polymer matrix was elaborated from orthophthalic polyester resin (P115-A) with a density of 1148 Kg/m$^3$ supplied by Novasuin S.A.S. As a conductor, 0.92 mm copper sheets were used. A SuperCat S-960® catalyst supplied by NovaSuin was used as a catalyst, in concentrations of 1% by weight.

2.2. Preparation of composites

2.2.1. Preparation of the microstrip. The manufacturing process started by weighing 50 grams of each resin at room temperature (17 °C). The concentration by weight of magnetite in the polymer matrix was defined in weight percentages of 10%, 20% and 30%, as reported by Ngo [11] and Husain [12]. Taking these parameters into consideration, 5, 10 and 20 grams of magnetite were weighed for each particle size. Subsequently, the filling material was added to the resin by stirring the mixture for 5 minutes at a frequency of 100 rpm by using a DLAB OS20-S stirrer.

Acrylic molds 7 cm wide, 7 cm long, with a thickness of 1.72 mm were used, which were covered with a release agent. A 0.94 mm thick copper sheet was placed on their base.

Once the mixture was homogenized, a 1% catalyst (SuperCat S-960®) was added with respect to the volume of resin, stirring for approximately 1 minute, after which the mixture was poured into the acrylic molds, filling them up to the top. At the end of this process, a second copper sheet was placed on the surface with the same dimensions as the base in a sandwich-type configuration. In order to guarantee the adhesion of the sheets with the polyester resin loaded with magnetite powders and the homogeneity in the dimensions of the composites, an acrylic press was used.

2.2.2. Filled Aligned. The magnetite particles in the filling were aligned by the use of ferrite magnets with a constant magnetic field of 300mT. The samples were prepared in three configurations, scattered random particles, without field effect, aligned horizontally (Figure 1), and aligned vertically to the magnetic field lines (Figure 2) [13]. All samples of composite material were cured at room temperature for 24 hours.

![Figure 1. Horizontal alignment.](image1)

![Figure 2. Vertical Alignment.](image2)
2.2.3. Preparation of transmission line. Once the material was cured and the demolding process of the microstrips was carried out, the transmission lines were prepared, for which a 4 mm thick portion of copper was isolated through the samples. A total of 27 samples were immersed in ferric chloride at a concentration of 40 vol%. Subsequent to the chemical reaction, microstrip type circuits were obtained on which SMA connectors were soldered on each of the terminals of the circuit in order to configure each of the transmission lines as shown in Figure 3.

![Figure 3. Configuration of the microstrip transmission line.](image)

2.3. Experimental design
A complete factorial design of $3^3$ was constructed with three factors and three levels per factor [14], matrix alignment (Mi), size of TTI filter with particle sizes Tp and percentage by weight of Wti filling. The levels for each factor are shown in Table 1. The complete factorial design involved 27 samples. The behaviour in high frequency and the response of the dielectric magneto compound was carried out by means of a vector network analyzer of Rohde & Schwarz ZVB8 Networks.

| Levels            | Low         | Medium     | High        |
|-------------------|-------------|------------|-------------|
| Alignment (Mi)    | Random (M1) | Vertical (M2) | Horizontal (M3) |
| Size (TTi)        | 200 (Tp <75 µm) | 325 (Tp <45 µm) | 500 (Tp <25 µm) |
| % filling (Wti)   | 10          | 20         | 30          |

3. Results and discussion

3.1. Microstructural analysis of magnetite
Figure 4 shows the morphology of the magneto-dielectric composites, analysed by scanning electron microscopy (SEM), in which the effect of the application of the magnetic field was observed, which aligned the magnetite particles according to the direction of the field lines.

The above was consistent with that reported by Z. Varga [13], who argued that if the polymer with ferromagnetic particles is cured in the presence of a magnetic field, the magnetizable particles form columnar structures along the field direction producing a polymer under the magnetorheological effect [15].
Figure 4. SEM images for the samples of magneto-dielectric composites in a concentration of 10% Fe$_3$O$_4$, (a) particles randomly dispersed, (b) vertically aligned with respect to the magnetic field and (c) horizontally aligned with respect to the magnetic field.

From the micrographic analysis, using Image J® software, the information regarding the size of each of the particles was extracted. This information was analysed statistically with Minitab 18®. The results reveal a mean particle size of 21.48 μm. for sieve 200 with a standard deviation of 13.43 μm. and a maximum value of 58.99 μm. The average values for sieve # 325 are 16.74 μm. with a standard deviation of 8.43 μm and a maximum size of 44 μm, and for sieve # 500 an average value of 8.36 μm was found, with a standard deviation of 3.68 μm and a maximum value of 18.62 μm.

3.2. VNA characterization
The VNA was calibrated within a frequency range of 150 KHz to 4 GHz [16] in order to measure each of the samples, thus, obtaining the dispersion parameters.

Figure 5 shows a simulation model of the transmission line, each of the 27 responses was compared with the reflection coefficient S (1.1), so as to determine if the manufactured composites are applicable in the design of high frequency circuits.

Figure 5. Simulation model of the microstrip line.

Figure 6 shows the results obtained in the pilot experiment (M8020TT325H). The magneto-dielectric material with a concentration of 20% magnetite, a particle size of less than 45 microns, aligned horizontally with respect to the magnetic field and the transmission line, which was selected at random, where the adjustment between the simulated model (black line) and the experimental response (magenta line) was verified.
Figure 6. Comparison of simulated S parameters versus experimental data. Pilot sample M8020TT325H.

Once the pilot test was carried out and it was verified that the designed magneto-dielectric material behaved well at high frequencies, we proceeded to compare the repetitiveness of the frequency, the period of the repetition and that the resulting coupling was less than -10 dB, which placed the samples within the range of desirable acceptability, through the analysis of the Smith chart.

Taking into account that the experiment requires a high volume of data, it was necessary to consider all the interactions present among the factors of the model. For this reason, the data was organized in a matrix in order to facilitate the comparison of the experimental responses, selecting those samples with a characteristic impedance close to 50 Ω, in the established frequency range of 150 KHz to 4 GHz.

3.3. Cross-matrix analysis
Figure 7 presents the results obtained from the cross-matrix analysis performed, which was based on a defined criteria, setting the particle size factor for each row and the concentration factor in the columns. As a result, the best 9 responses per row and per column were obtained: a total of 18 responses. The alignment of the particles was determined as a free variable.

The 18 outstanding responses were organized in row and column vectors in order to carry out a second analysis of the reduced matrixes, obtaining six objective answers. The final iteration of the model showed that the best response in high frequency was presented by the manufactured magneto - dielectric composite material with 20% magnetite, an average particle size of 21.48 μm, aligned horizontally to the magnetic field and to the transmission line. The result of this mixture was evaluated through a
simulation process. Figure 8 shows a good correlation between the simulated model and the experimental data.

![Simulation vs Experimental Data](image)

**Figure 7.** Smith chart for the results obtained from cross-matrix analysis, based on the defined decision criteria: characteristic impedance close to 50 Ω, in the established frequency range of 150 KHz to 4 GHz.

![Comparison of S Parameters](image)

**Figure 8.** Comparison of simulated S parameters versus experimental data for the best sample obtained from the cross-matrix analysis of the magneto-dielectric composite material with 20% magnetite, average particle size of 21.48 μm, aligned horizontally to the magnetic field and to the transmission line.

Table 2 presents the results of the application of a cross-matrix analysis for the magneto-dielectric compound, which optimized the high-frequency response for the following variables: relative dielectric constant $E_r$, effective dielectric constant $E_{eff}$, characteristic impedance $Z_0$ and dielectric loss.

| Material     | $E_r$ | $E_{eff}$ | $Z_0$    | Dielectric loss |
|--------------|-------|-----------|----------|-----------------|
| M8020TT200H  | 3.88  | 2.48      | 43.73    | 0.05            |
4. Conclusions
The magneto-dielectric composite material was composed of unsaturated P155A polyester resin and magnetite powders, varying the concentration, size and alignment in the filling.

The cross-matrix analysis obtained, from a complete factorial design of $3^3$, the combination of values that optimized the dielectric properties of a composite material with 20% magnetite, average particle size of 21.48 μm, aligned horizontally to the magnetic field and to the transmission line.

The horizontal alignment of the particles in the functional filling of magnetite Fe₃O₄ improved the response of the dielectric properties of the composite material in high frequency within a range of 150 KHz to 4 GHz.

The size, concentration and alignment of the magnetite particles in the filling of a magneto-dielectric composite material allowed for obtaining different responses in the dielectric constant, opening an important path in the investigation of this type of material and its applicability in the design of high frequency circuits.

Acknowledgments
This work was supported by the European Union’s Horizon 2020 Research and Innovation Programme through the Marie Curie Skłodowska-Curie agreement under grant 793529.

References
[1] Yang H, Bai L, Lin L, Wang F and Wang T 2017 Ceramics International. 43 2903–2909
[2] Yang T I, Brown R N C, Kempel L C and Kofinas P 2008 J. Magn. Magn. Mater. 320 2714–2720
[3] Lin Y, Liu X, Yang H, Wang F and Liu C 2017 Mater. Res. Bull. 86 101–6
[4] Lin Y, Liu X, Yang H, Wang F, Liu C, and Wang X 2016 J. Alloys Compd. 688 571–76
[5] Teo M L S, Kong L B, Li Z W, Lin G Q and Gan Y B 2008 J. Alloys Compd. 459 557–66
[6] Li Q, Chen Y and Harris V G 2018 J. Magn. Magn. Mater. 453 44–7
[7] Park T J, Papaefthymiou G C, Viescas A J, Moodenbaugh A R and Wong S S, Nano Lett. 7 766–72
[8] Mattei J L, Guen E Le, Chevalier A and Tarot A C 2015 J. Magn. Magn. Mater. 374, 762–68
[9] Varga Z, Filipcei G and Zrínyi M 2005 Polymer 46 7779–7787
[10] Philippova O, Barabanova A, Molchanov V and Khokhlov A 2011 Eur. Polym. J. 47 542–59
[11] Ngo I L, Jeon S, and Byon C 2016 Int. J. Heat Mass Transf. 98, pp. 219–26
[12] Hussain A R J, Alahyari A A, Eastman S A, Thibaud-Erkey C, Johnston S, and Sobkowicz M J 2017 Appl. Therm. Eng. 113 1118–27
[13] González L A L, Peña-Rodríguez G and Triana Y P 2019 AIMS Materials Science, 6 549–558
[14] Boon M S and Mariatti M 2014 J. Magn. Magn. Mater. 355 319–324
[15] Schubert G and Harrison P 2015 Polym. Test. 42 122–34
[16] Heuermann H 2008 IEEE Trans. Microw. Theory Tech.56 2505–2510