Chapter 2

Material selection, preparation and characterization

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References
2.1 Introduction

Particulate composites with nano magnetic inclusions are developed as magnetodielectric material for microstrip antenna substrate applications, in X-band. The chapter initially discusses the selection of nano magnetic material and polymer for operations in microwave frequency followed by synthesizing process for the inclusions and composite preparation. Confirmation of size of synthesized inclusions is done using X-ray diffraction (XRD) and transmission electron microscopy (TEM). The inclusions are reinforced into the polymer matrix and homogeneity of distribution is observed using scanning electron microscopy (SEM). The material is tested for its thermal, physical and water absorbance property. Hysteresis measurements are conducted on the composite systems to determine its saturation magnetization and area of the loop. The results are reported in the subsequent sections.

2.2 Material Selection and Synthesis

The materials to be used as substrate for microstrip antennas should have desired functional requirements, hence, choice of host materials are very crucial. Processing technique employed should be able to control the particle size, reduce agglomeration and assure homogeneity for propagation of microwave through the synthesized materials. In addition, the fabrication process should be easy and cost effective.

2.2.1 Selection of inclusions and host matrix

Properties of magnetodielectric materials can be varied with compositional variation of simple ferrites like \( \text{MnFe}_2\text{O}_4 \), \( \text{CoFe}_2\text{O}_4 \), \( \text{NiFe}_2\text{O}_4 \), \( \text{MgFe}_2\text{O}_4 \) etc. Synthesis of nano ferrites in the present work, are carried out using two basic ferrite series: nickel ferrites and cobalt ferrites. Nickel ferrites have high Curie

\[ \text{temperature } \sim 525^0\text{C} \]

which makes other magnetic properties independent of temperature variation. Additionally, nickel ferrites are soft ferrite [1-3] and can be easily tuned with application of magnetic field. \( \text{Co}^{2+} \) ions have degeneracy in
\(d_{xy}\) and \(d_{yz}\) orbital that makes cobalt ferrites a fast relaxers and useful at high frequencies [4].

The polymer base of the magnetodielectric composite systems give shape flexibility, reduces weight and are cost effective. Low density polyethylene (LDPE) is a solid polymer having melting point 383 K (110 °C) [5]. LDPE contains the chemical elements carbon and hydrogen (figure 2.1). Branching of LDPE makes the intermolecular forces (instantaneous-dipole induced-dipole attraction) weaker, lowers the tensile strength and increases its elasticity. This helps in homogeneous distribution of reinforcing metal oxide filler particles with. Also, since its molecules are less tightly packed and less crystalline because of the side branches, its density is lower [6]. It is not reactive at room temperatures, except by strong oxidizing agents, and some solvents cause swelling. Polymer gets dissolved in solvents like carbon tetrachloride, toluene, at 80°C, and subsequently precipitates out as the solution cools down to room temperature, which is advantageous for composite synthesis. The mechanical properties of LDPE are between those of rigid materials like polyester and limped polymers like vinyls. In addition, it has low dielectric loss at microwave frequencies and thus is appropriate host matrix for microwave substrate application. The details of relevant properties are given in table 2.1

<table>
<thead>
<tr>
<th>Materials</th>
<th>Microwave property</th>
<th>Physical property</th>
<th>Thermal property</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Dielectric constant 10GHz</td>
<td>Dielectric loss</td>
<td>Density (gm/cc)</td>
</tr>
<tr>
<td>*LDPE</td>
<td>3.1</td>
<td>0.0002</td>
<td>0.91 - 0.94</td>
</tr>
</tbody>
</table>

*Microwave Material, "V R K Murthy, S Sundaram, B Viswanathan", Narosa Publishing House
Figure 2.1  Flowchart for ferrite and magnetodielectric composite synthesis procedure
2.2.2 Synthesis of nickel and cobalt ferrite nanoparticles

Cobalt (II) nitrate hexahydrate (98% pure, Co\(\text{(NO}_3\text{)}_{2.6}\text{H}_2\text{O}\)) for cobalt ferrite and for nickel ferrites, nickel (II) nitrate hexahydrate (98% pure, Ni\(\text{(NO}_3\text{)}_{2.6}\text{H}_2\text{O}\)), with iron (III) nitrate nonahydrate (≥98% pure, Fe\(\text{(NO}_3\text{)}_{2.9}\text{H}_2\text{O}\)) are used as precursors. Desired compositions is obtained by using stoichiometric amounts of Ni\(\text{(NO}_3\text{)}_{2.6}\text{H}_2\text{O}\)/Co\(\text{(NO}_3\text{)}_{2.6}\text{H}_2\text{O}\) and Fe\(\text{(NO}_3\text{)}_{2.9}\text{H}_2\text{O}\). The salts are dissolved in ultra-pure, deionized water. Keeping the temperature fixed at 80°C the solution is stirred for 2.5 hours. 3M sodium hydroxide (NaOH) is added to the solution till precipitation. The pH of the reaction is kept at 12. Few drops of oleic acid (C\(_{17}\)-H\(_{33}\)COOH) are added as surfactant. The precipitate is washed with double distilled water to remove traces of sodium and nitrate ions and dried for about 12 hrs at a temperature of 100°C. The dried polycrystalline precipitate is crushed and sintered at three temperatures 600°C, 700°C and 800°C for 2 hours and cooled at room temperature. The sintered samples are microstructurally tested for ferrite formation, shape and size.

2.2.3 Fabrication of magnetodielectric material

In particulate composite the electrical and magnetic properties of the materials can be controlled by a small variation of the inclusion content [7-8] in the polymer matrix. LDPE is dissolved in cyclohexane (99%) and stirred continuously at 70°C to make a homogeneous mixture. Cobalt ferrite and nickel ferrite nano particles are added in different percentage volume fraction in the polymer matrix. The volume fraction (VF) is defined as the ratio between the volumes of the filler to the total volume of the (polymer + filler) mixture. Mathematically,

\[
x\% = \frac{V_f}{V_f + V_p} \times 100\%
\]

where, \(V_f\) = volume of the fillers, \(V_p\) = volume of the polymer

The composite is stirred for 2-3 hours to get uniform blending. The homogeneous liquid is then poured in desired shaped mold and allowed to cool gradually at
room temperature. During preparation the environment; conditions are kept same for each batch of samples. The processing chart is given in figure 2.1.

Beyond 5% VF the composite becomes brittle and fragile and shrinkage is observed as the inclusions reach the percolation limit. For further investigations magneto-dielectric composite with upto 5% VF is considered.

2.3 Microstructural Studies

2.3.1 X-ray diffraction

XRD of developed ferrite are recorded at 2θ values from 10° to 80° using Rigaku X-Ray Diffractometer, model: Miniflex using Cu Kα line of wavelength \( \lambda = 1.541841 \) \( \text{Å} \). The patterns of cobalt ferrite and nickel ferrite particles, sintered at temperatures 600°C, 700°C and 800°C, are shown in figure 2.3(a-f). The reflection plane (311) in the XRD pattern is indications of the formation of ferrite. The
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(c) 
- CoFe$_2$O$_4$
- Fe$_2$O$_3$

(d) 
NiFe$_2$O$_4$

(e) 
- NiFe$_2$O$_4$
- Hematite
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Figure 2.2 (a), (b) and (c) XRD patterns for cobalt ferrite nanoparticles sintered at 600°C, 700°C and 800°C and (d), (e) and (f) XRD patterns for nickel ferrite nanoparticles sintered at 600°C, 700°C and 800°C respectively.

The value of the lattice parameter, \( a \), for NiFe\(_2\)O\(_4\) is 8.339 Å and for CoFe\(_2\)O\(_4\), is 8.381 Å, which agrees with the reported values in JCPDS card numbers 10-325 and 22-1086, respectively, thus confirming the formation of cubic spinel structure.

The diffraction peaks 220, 311, 422, can be perfectly indexed to the cubic spinel structure. Some peaks of impurities are detected in the XRD pattern possibly due to presence of traces of precursors used in synthesizing the ferrites.

The average crystallite size using first two strong peaks, is determined by the Debye-Scherrer’s formula [9]

\[
D = \frac{0.9 \lambda}{\beta \cos \theta}
\]

where, \( D \) is the average crystallite size, \( \lambda \) is the wavelength of Cu \( K_\alpha \), \( \beta \) is the full width at half maximum (FWHM) of the diffraction peaks, and \( \theta \) is the Bragg’s angle. The average crystallite size is estimated to be about 24.2 nm in diameter.

2.3.2 Transmission electron microscopy

High resolution transmission electron micrographs (HRTEM) of the nano ferrite particles are taken using JEOL-TEM-100 CXII (Collidion Coated Copper Grids) in
Figure 2.3  (a),(b) and (c) TEM images of cobalt ferrite nanoparticles sintered at 600°C, 700°C and 800°C, respectively and (d),(e) and (f) TEM images of nickel ferrite nanoparticles sintered at 600°C, 700°C and 800°C, respectively RSIC, NEHU, Shillong, India. Figures 2.3 (a-c) and 2.3 (d-f) show HRTEM images. CoFe₂O₄ and NiFe₂O₄ nanoparticles sintered at 700°C and 800°C show an average particle size of around 40 nm and 30 nm, respectively. The particle size for sintering temperature 600°C of CoFe₂O₄ and NiFe₂O₄ is found to be 20 nm
and 30 nm, respectively and are used for making composite in further investigations. Most of the particles appear spherical in shape.

2.3.3 Scanning electron microscopy

SEM of particulate magnetodielectric composite is taken by JEOL-JSM-6390. The surface of each samples are platinum coated before taking the micrographs. The micrographs are taken at $10^{-11}$ Å probe current and 20 KV accelerating voltage at different resolutions. Figure 2.3 (a-c) and 2.3 (d-f) show the SEM micrographs of 1%, 3% and 5% VF cobalt ferrite/LDPE composite and 1%, 3% and 5% VF nickel ferrite/LDPE composite, respectively.

Most of the particle appear spherical in shape and almost homogeneously distributed over the matrix as seen from the SEM images.
2.4 Density and Water Absorbance

The density of a material is equal to its weight per unit volume.

\[ d_s = \frac{W}{V} \]  \hspace{1cm} (2.3)

where \( d_s \) = Density in gm/cc
\( W \) = Weight in gms
\( V \) = Volume in cm\(^3\)

The densities of the samples are measured with Archimedes’s principle. The samples are cut into rectangular shape of 1 cm\(^2\) area and tied with a thin inelastic thread and its weight in air is measured. The specimen is then completely dipped in water and weighed. Since the buoyancy force is equal to the difference between these two weights, the volume of the sample is equal to the weight difference divided by the density of water. The weight of the thread is also taken and duly subtracted to get the correct weight of the sample in air. If,

\[ W_{air} = \text{Weight of the sample in air + weight of thread (gm)} \]
\[ W_{water} = \text{Weight of the sample in water + weight of thread (gm)} \]
\[ W_{thread} = \text{Weight of thread (gm)} \]
\[ D_{water} = \text{Density of water (=1 gm/cc)} \]
Then

\[ d_s = \frac{W_{\text{air}} - W_{\text{thread}}}{W_{\text{air}} - W_{\text{water}}} \times D_{\text{water}} \]  

(2.4)

Percentage of water absorbance is studied by immersing the composite for 48 hours at room temperatures and measuring its weight (upto three decimal places). The density and percentage of water absorbance determined are tabulated in table 2.2.

**Table 2.2** Density and percentage of water absorbance of 1%-5% VF of CoFe₂O₄/LDPE and NiFe₂O₄/LDPE composite

<table>
<thead>
<tr>
<th>Sample composition (VF)</th>
<th>CoFe₂O₄-LDPE</th>
<th>NiFe₂O₄-LDPE</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Density (gm/cc)</td>
<td>% Water absorbance</td>
</tr>
<tr>
<td>1%</td>
<td>1.60</td>
<td>0</td>
</tr>
<tr>
<td>2%</td>
<td>1.70</td>
<td>0</td>
</tr>
<tr>
<td>3%</td>
<td>1.75</td>
<td>0</td>
</tr>
<tr>
<td>4%</td>
<td>1.80</td>
<td>0</td>
</tr>
<tr>
<td>5%</td>
<td>1.90</td>
<td>0</td>
</tr>
</tbody>
</table>

The results show that both 1%-5% VF of CoFe₂O₄/LDPE and NiFe₂O₄/LDPE composites do not absorb any water at room temperatures and can be used in high humidity environment.

A marginal increase in density of the composite is observed as volume fraction increases from 1% to 5% showing its applicability as alternate light-weight material.

### 2.5 Thermal conductivity

Effective thermal properties of composite materials depend on the intrinsic microscopic properties of the filler, homogeneity of distribution and compactness of filler in the matrix [10-11]. There are different techniques for thermal conductivity measurements in insulators. Lee’s method is the most popular one, but it suffers from the need of bulky steam system for heating and high possibility of radial heat flow. The set up is indigenously designed and fabricated in the laboratory by using more robust components like a guarded hot plate, solid state transducer and amplifier for acquiring data. The theory of the
instrument is based on Lee's method [12]. The instrument consists of two brass slabs where one acts as heat source and other as sink. The sample to be tested is placed between the source and the sink. The exposed portion of the source and sink is covered with heat insulating-seal to ensure no radial heat flow. A thermostatic electric heater is used to heat the source and the sink is kept at room temperature. A spring system is attached to the other end of sink to ensure good contact between the source, the sample and the sink and easy placement of the sample. A non-conducting casing is used for mounting the system. The schematic diagram of the instrument is shown in figure 2.7.

Temperature of the slabs is measured using a transistor as a transducer element. It works based on Peltier effect. The system can measure both absolute and relative temperatures of the slabs using a switch.

The area of sample is kept same as the area of the slab. The source is heated and heat conducts through the sample to the sink. The rate at which heat, $Q$, conducted through the specimen is equal to the rate at which the sink losses heat and is

$$Q = \frac{K A (\theta_1 - \theta_2)}{d}$$

(2.5)

Where,

- $K$ is the coefficient of thermal conductivity of the specimen
- $A$ is the area of sample
- $d$ is thickness of sample
- $\theta_1$ and $\theta_2$ are temperatures of the source and sink at steady state, respectively

If $M$ is the mass of the slab, $S$, the specific heat of the slab material, the rate of cooling of the sink at $\theta_2$ is

$$Q = MS \left( \frac{d\theta}{dt} \right)_{\theta = \theta_2}$$

(2.6)

where, $d\theta/dt$ is the rate of fall of temperature at $\theta_2$
Figure 2.5  Schematic diagram of thermal conductivity measurement set-up

From equations 2.5 and 2.6

\[
\frac{KA(\theta_1 - \theta_2)}{d} = MS \left( \frac{d\theta}{dt} \right)_{\theta = \theta_2}
\] (2.7)

\[
K = \frac{dMS \left( \frac{d\theta}{dt} \right)_{\theta = \theta_2}}{A(\theta_1 - \theta_2)}
\] (2.8)

The rate of cooling is found by heating the sink to a temperature above \( \theta_2 \). The sample is then placed between the source and sink and source is heated. It is allowed to reach an equilibrium temperature with the sink. The time of fall of temperature, \( t \), of the sink is noted at regular intervals. The slope of the tangent drawn to this plot gives the value of rate of fall of temperature.

The thermal conductivity of the prepared samples measured with the above technique is tabulated in table 2.3.

The thermal conductivity increases with increasing filler content in the composite. The 5% VF of both the composites shows an enhanced thermal conductivity. [13-14]. The effective thermal properties of composite materials depend on the intrinsic microscopic properties of the filler dielectrics, homogeneity of distribution and compactness of the composite.
Table 2.3 Thermal conductivity of CoFe$_2$O$_4$/LDPE and NiFe$_2$O$_4$/LDPE composite for different volume fraction

<table>
<thead>
<tr>
<th>Sample composition</th>
<th>1% volume fraction (W/cm°K)</th>
<th>2% volume fraction (W/cm°K)</th>
<th>3% volume fraction (W/cm°K)</th>
<th>4% volume fraction (W/cm°K)</th>
<th>5% volume fraction (W/cm°K)</th>
</tr>
</thead>
<tbody>
<tr>
<td>CoFe$_2$O$_4$-LDPE</td>
<td>0.0033</td>
<td>0.0133</td>
<td>0.0243</td>
<td>0.0373</td>
<td>0.3933</td>
</tr>
<tr>
<td>NiFe$_2$O$_4$-LDPE</td>
<td>0.0108</td>
<td>0.0156</td>
<td>0.0198</td>
<td>0.0233</td>
<td>0.0376</td>
</tr>
</tbody>
</table>

Particles reinforced in polymer matrix forms an interacting interface [15]. Increase in number of inclusions with increasing %VF increases the effective area of interaction between the filler-phase and polymer-phase. Heat transfer in such composites probably takes place via the polymer-filler interface [16]. The filler particles may begin to touch each other, or form agglomerates and conductive chains in the direction of heat flow, resulting in increase in thermal conductivity of the composites with increasing number of reinforcing particles.

2.6 Vibrating sample magnetometry

Magnetization properties of the magnetodielectric composites have been studied using Vibrating Sample Magnetometer (VSM), Make: Lake Shore Cryotronics Inc., USA, Model: 7410.

The basic principle of operation for a VSM is that, a change in magnetic flux induces a voltage in a pickup coil. The time-dependent induced voltage is given by the following equation:

$$V_{coil} = \frac{d\Phi}{dt} = \left(\frac{d\Phi}{dz}\right)\left(\frac{dz}{dt}\right)$$ (2.9)

$\Phi$ is the magnetic flux enclosed by the pickup coil, $z$ is the vertical position of the sample with respect to the coil and $t$ is time of oscillation. For a sinusoidally oscillating sample position, the voltage is found from the following equation:

$$V_{coil} = 2\pi f C m A \sin(2\pi ft)$$ (2.10)

where, $C$ is a coupling constant, $m$ is the dc magnetic moment of the sample, $A$ is the amplitude of oscillation, and $f$ is the frequency of oscillation.
The density of the samples is taken from table 2.2, for emu/gm to Gauss conversion. The Saturation magnetization ($4\pi M_s$) measurements are carried out for 1%, 3%, 5% VF of cobalt ferrite/LDPE and nickel ferrite/LDPE composites at room temperature with a maximum applied field up to 20 kGauss. The M-H plot for both the composite systems is plotted in figure 2.6 (a) and (b) respectively. The saturation magnetization results for cobalt ferrite/LDPE and nickel ferrite/LDPE composites are given in table 2.4

Table 2.4 Tabulation of saturation magnetization values for different VF of the magneto-dielectric composite

<table>
<thead>
<tr>
<th>Volume fraction of composite</th>
<th>$4\pi M_s$ (G)</th>
</tr>
</thead>
<tbody>
<tr>
<td>1% CoFe$_2$O$_4$/LDPE</td>
<td>8.489</td>
</tr>
<tr>
<td>3% CoFe$_2$O$_4$/LDPE</td>
<td>8.956</td>
</tr>
<tr>
<td>5% CoFe$_2$O$_4$/LDPE</td>
<td>8.356</td>
</tr>
<tr>
<td>1% NiFe$_2$O$_4$/LDPE</td>
<td>18.835</td>
</tr>
<tr>
<td>3% NiFe$_2$O$_4$/LDPE</td>
<td>18.231</td>
</tr>
<tr>
<td>5% NiFe$_2$O$_4$/LDPE</td>
<td>17.565</td>
</tr>
</tbody>
</table>

![Figure 2.6](image)

Figure 2.6 VSM curve for 1%, 3% and 5% VF (a) cobalt ferrite/LDPE composite (b) nickel ferrite/LDPE composite

For cobalt ferrite/LDPE composite the $4\pi M_s$ is found to be 0.7365 emu/gm for 5% VF composite. Small coercive field, $H_C = 143.88$ Gauss shows that synthesized cobalt ferrite/LDPE composite is not hard magnetic material, unlike bulk cobalt ferrite. The remnant magnetization, $M_r$, is 0.0815 emu/gm is quite low in
comparison to that of 31.7 emu/gm of cobalt ferrite nanoparticles [17]. This shows that the composite relaxes faster than the cobalt ferrite nanoparticles.

The low value of saturation magnetization for the cobalt ferrite/LDPE composite as compared to the bulk cobalt ferrite composites (580 emu/cm$^3$ as reported in [18]), can be due to surface spin canting [19]. The schematic to explain the phenomenon is shown in figure 2.7. The pronounced growth of magnetic anisotropy inhibits the alignment of the moment in an applied field affecting the relatively inert surface layer that has low magnetization [20]. Usually, the saturation magnetization increases consistently with particle size. The decrease in $4\pi M_s$ value can be explained on the basis of this surface effect, which becomes less significant with increase in size of the particles [21]. Canting angle ($\sin \gamma = M_y/M_x$) of 12.2543 suggests that the surface layer contains spins that are not collinear with the magnetic field [22].

The lowest $4\pi M_s$ is found to be 17.565 G for 5% VF nickel ferrite/LDPE composite. It is observed that this composite has a coercive field of $H_c = 273.344$ Gauss. The $M_r$, of nickel ferrite/LDPE is 0.337 emu/g, which is also quite low in comparison to that of nickel ferrite nanoparticles (10 emu/gm is reported in [22]).

![Spin orientation](image)

**Figure 2.7** Schematic showing spin canting mechanism

The narrow M-H loop suggests very low magnetic loss. The low values of $4\pi M_s$ can be due to exchange interaction and spin canting. The average distance between two neighboring inclusions dispersed in the polymer matrix as observed from the SEM is $\sim 3\mu$m. This sufficiently isolates the particles from each other, and thus the exchange interaction energy is low in this case, leading to lower $H_c$. 
values. Usually, for these type of isolated particles, spin canting should be less and the canting angle \( \sin \gamma = M_x / M_s \) should be of the order of \( \sim 30 \). The canting angle found in the present study is \( \sim 10.3820 \). If no agglomeration of inclusions in matrix takes place, the transition region is large and the Neel wall thickness, \( T \), is large, hence the critical value of surface anisotropy field, \( H_{Anis}^C \), \[ \text{[23]}, \] given below will be large.

\[
H_{Anis}^C = 4\pi M_s (T + 1)
\]

The SEM picture of the composite shows some agglomeration of inclusions in the matrix, which may increase surface anisotropy field, \( H_s \) and lowers \( T \). Thus canting increases and thereof spin canting angle decreases, which accounts for low saturation magnetization \[ \text{[24]}, \] as observed.

2.7 Conclusions

Synthesized CoFe\(_2\)O\(_4\) and NiFe\(_2\)O\(_4\) nanoparticles show a cubic spinel structure. The ferrites annealed at 600\( ^\circ \)C show minimum average crystallite diameter, ranging from 7.36 - 10.76 nm. TEM pictures show the shape to be generally spherical and average particle size is \( \sim 20 \) nm for CoFe\(_2\)O\(_4\) and \( \sim 30 \) nm for NiFe\(_2\)O\(_4\). Magnetodielectric composite shows homogeneous distribution of inclusions, as seen from SEM. The sizes of the reinforced particles are smaller than the probing wavelength (\( \sim 3 \) cm) hence, the scattering losses can be assumed to be negligible. However, some agglomeration of inclusions is seen for composites VF beyond 3\%, where the particles form a semi contiguous geometry.

Both the composite systems are light weighted and compact as seen from density measurements. The thermal conductivity of the magneto-dielectric composite is higher than that of pure LDPE matrix and 5\% VF shows maximum value of 0.3933 W/cmK. Higher thermal conductivity implies better heat dissipation in the circuit, which is an essential substrate property for microwave transreceiving devices like microstrip patch antenna. The hydrophobic nature of the magnetodielectric composites, show possibility of its use in high humidity environment.
The magnetic measurements confirm the magnetic nature of the composites at room temperature. The saturation magnetization of the developed magnetodielectric composite is very low. The narrow loop suggests that magnetic losses are very low.

The synthesized nano magnetodielectric material shows all the desired properties which are essential for its use as substrate material. Microwave characterizations of the synthesized material are carried out in the following chapter.
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References


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