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STRONTIUM AND TITANIA INFUSED LLDPE NANOCOMPOSITES FOR X-BAND ELECTROMAGNETIC ABSORPTION

S. Ozah*
Department of Physics, DKD College,
Dergaon, Assam, India

Abstract: An absorption study is carried out on a blended composite of strontium ferrite and titania particles in linear low density polyethylene (LLDPE) matrix with 60 wt.% in the frequency range 8.2 to 12.4 GHz. X-ray diffraction pattern studies confirms the formation of hexagonal structure of the strontium ferrite particles. The crystallite size of the strontium and titania particles are in nanorange with 2.2 nm and 104 nm, respectively. The complex permittivity and complex permeability of the ferrite nanocomposite is measured at X-band and found to increase on mixing with titania. Based on transmission line theory, an absorber of the strontium/titania/LLDPE nanoccomposite is fabricated on a metal backing. A comparative absorption study is conducted by varying the texture of the fabricated absorber. The maximum insertion loss is observed for the rough surface of the strontium/EG/LLDPE nanoccomposite with -21.67 dB at 12.4 GHz with > -18 dB absorption bandwidth over the X-band.

Keywords: Microwave absorber, barium ferrite, reflection loss, complex permittivity, complex permeability

1. INTRODUCTION

Electromagnetic interference (EMI) is a detrimental and uncontrolled consequence of explosive growth of electronics and widespread use of transient power sources. To overcome this, shielding materials are employed to minimize the influence of external electromagnetic waves on device functioning, which also operate as camouflage materials in defense applications. Over the past few decades, extensive research has been carried out for the development of efficient EMI shielding materials, and several studies have been reported in this regard [1–5]. For maximum absorption of electromagnetic (EM) waves, both the electric and magnetic components must be sufficiently attenuated within the absorber thickness. However, in dielectric absorbers, the magnetic loss component is absent, which necessitates a greater thickness of the absorber to achieve effective absorption [5]. On the other hand magnetodielectric absorbers, with both dielectric and magnetic losses, provide enhanced attenuation of EM waves within a relatively smaller thickness, making them more efficient for customised EMI shielding applications [6–8].

The extent of absorption depends on electromagnetic wave interactions with the material properties viz. complex permittivity and complex permeability, size and shape of the filler particles and thickness of the absorber [9]. The huge interaction zone created by the high interfacial area of nanoparticles boosts interfacial polarizations and the magnetic interactive loss mechanism, which improves the dielectric and magnetic characteristics and, ultimately, increases microwave absorption [9-12].

In the present work, the microwave absorption is studied with nanosized hexagonal ferrite infused with titania in Linear Low Density Polyethylene (LLDPE) matrix. Nano sized strontium ferrite particles are synthesized and used as the magnetic inclusions, while titania nanoparticles are used as the dielectric inclusion in the LLDPE matrix. Strontium hexagonal ferrite ($SrFe_{12}O_{19}$) is used for development of X-band absorbers as it has high saturation magnetization, high crystalline anisotropy and is chemically stable [13-16]. Titania particles increase the complex permittivity of the composites which tends to concentrate the electric vector of

microwave and also reduce the absorber thickness [17, 18]. LLDPE is defined by a density range of 0.915–0.925 g/cm^3. LLDPE is a substantially linear polymer (polyethylene), with significant numbers of short branches, commonly made by copolymerization of ethylene with longer-chain olefins. The copolymerization process produces an LLDPE polymer that has a narrower molecular weight distribution than conventional LDPE. It has a property of flexibility with tensile strength 16.5 MPa and elongation at break 705% [19, 20]. For which this polymer is used for making the flexible composite sample.

Microstructural properties of the synthesized M-type strontium ferrite and titania particles and density and water absorption studies of the $SrFe_{12}O_{19}/TiO_2$/LLDPE nanocomposites are studied. Microwave characterisations are conducted on the developed nanocomposite to investigate its performance as microwave absorber. The texture of the fabricated absorber is varied and reflection loss is measured using Transmission/reflection method [21].

2. SYNTHESIS OF THE MAGNETODIELECTRIC MATERIAL

2.1 Preparation of M-type barium ferrite ($BaFe_{12}O_{19}$) and TiO_2 particle:

M-type strontium ferrite ($SrFe_{12}O_{19}$) particles are prepared from nitrate precursors using co-precipitation technique. Strontium nitrate (ε98%) and iron (III) nitrate nonahydrate (ε98%) precursors are used as the base materials to which sodium hydroxide is added dropwise to control the size of the particles. Oleic acid is used as surfactants in order to reduce interparticle interaction and distribution. Aqueous solutions of strontium and iron salts are prepared separately by dissolving the salts in reverse osmosis (RO) deionized water maintaining the molar ratio of barium to ferric nitrate as 1:12 with constant magnetic stirring condition. Then the iron and strontium salt solutions are mixed together and heated at 70° C with continuous magnetic stirring for one hour. 4M (25 ml) solution of sodium hydroxide is prepared separately and slowly added to the salt solution drop wise. The pH of the solution is maintained to a level of 11-12. A few drops (~ 0.1 ml) of oleic acid ($C_{17}H_{33}COOH$) is added to the solution as a surfactant and coating material [22]. The system is cooled to room temperature. Subsequently, the precipitate is washed with distilled water and ethanol to get the precipitate free from sodium and nitrate compounds. Finally, the precipitate is dried at 100°C. The dried powder is crumbled and annealed at 800°C for two hours to get strontium ferrite particles.

TiO_2 particles (supplied by Merck Specialities Private Limited) are mixed with doubly distilled water and stirred at 70^o C for 12 hrs, then autoclaved for 1 hr at 15lb pressure and finally the product is annealed at 600ºC for 3 hrs and then crushed well to get fine titania powder.

2.2 Fabrication of $SrFe_{12}O_{19}/TiO_2$/LLDPE composite

The synthesized ferrite, titania powder and LLDPE powder are mechanically blended at ~ 15000 rpm for 60 weight percentage of filler in the base matrix. Mathematically, if total weight of the composite is Z grams, X_1, X_2 and Y are the weights of strontium ferrite, titania and polymer matrix, respectively, i.e. Z = X + Y, where, $X=X_1+X_2$, then, for *N* wt.% of filler-LLDPE composite, amount of filler present in the composite is given by

$$\text{Filler} = X = (N/100) \cdot Z \text{ grams} \quad (1)$$

and amount of LLDPE present in the composite is given by

$$\text{Polymer matrix} = Y = (Z-X) \text{ grams} \quad (2)$$

Using the relation (1) and (2), a uniform mixture of ferrite, titania and LLDPE powder is obtained with 60 wt. % of filler particles. The mixture is placed in a specially designed three-piece die-mould consisting of a cavity, upper and lower plunger with spacer and initially heated up to 95–100° C. A pressure up to 1.5-2 tons is slowly applied and the fixture with the sample is isothermally heated at 150 °C for 2 hours and then allowed to cool at room temperature.

2.3 Analysis and characterization techniques

Propagation of electromagnetic wave, passing through the composite material depends not only on the intrinsic properties of the constituents, but also on the size of the inclusions and the change in structural configuration of the inclusion particles in the composite matrix. Microstructural studies give the complete picture of the composite deciding its utility as substrate in the microwave technology.

Microstructural characterization: X-ray diffraction patterns (Rigaku, Miniflex 200 X-ray diffractometer) of the prepared materials are obtained using Cu Kα radiation (wavelength, λ= 1.541841 Å) at room temperature.

Microwave characterization: Complex permittivity and complex permeability over the X-band is computed from S_{11} and S_{21} values using Nicolson–Ross method [23]. Reflection loss parameter is measured using

transmission/reflection method, using Agilent WR-90 X11644A rectangular waveguide line compatible with Agilent 8722ES vector network analyzer [21].

3. RESULTS AND DISCUSSION

3.1 Microstructural and water absorbance studies:

3.1.1 X-ray diffraction of M- type strontium hexa ferrite and titania particles

X-ray diffraction (XRD) patterns of the strontium ferrite particles are recorded at 2θ values from 20° to 80°. Diffraction pattern of $SrFe_{12}O_{19}$ particles annealed at T=800^{0}C is shown in fig.1a. The reflection planes: (1 0 2), (1 1 0), (1 0 7), (1 1 4), (2 0 0), (2 0 3), (0 0 10), (2 0 5), (10 12), (3 0 0), (2 1 7), (2 0 11), (2 2 0), (2 0 14), (3 1 6) and (4 0 4) indicate presence of a hexagonal structure. Crystallinity and size of the particles are calculated from the XRD patterns using Debye–Scherrer formula [24].

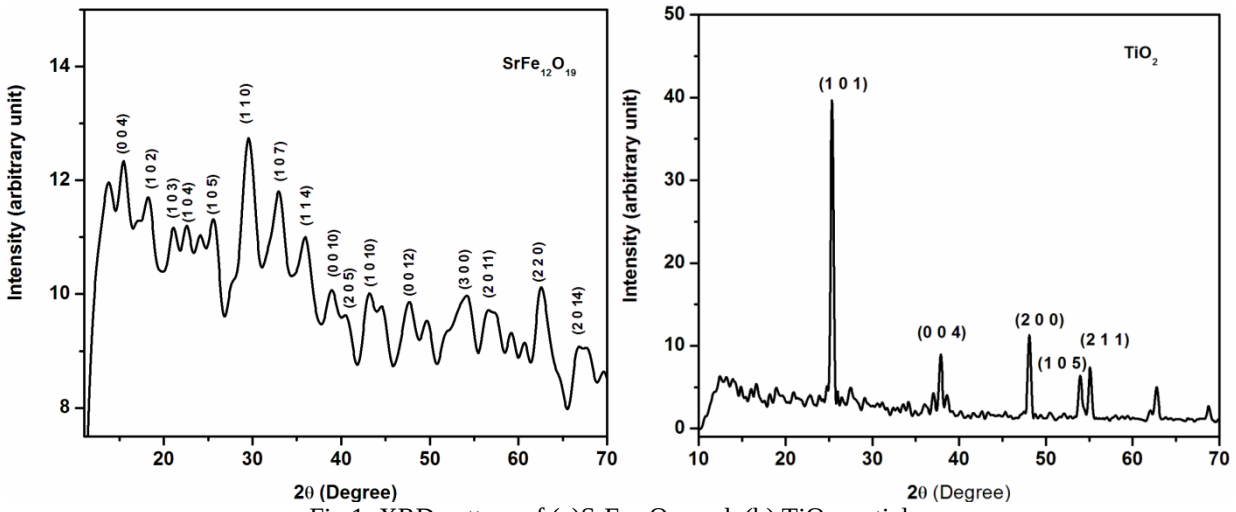


Fig 1: XRD pattern of (a)$SrFe_{12}O_{19}$ and (b) TiO_2 particles

The lattice parameters for the hexagonal magnetoplumbite phases are computed (table 1) using the d-spacings value and the respective (*h k l*) parameters. Interplanar distance, d is given by

$$\frac{1}{d^2} = \frac{4}{3}\left(\frac{h^2 + k^2 + l^2}{a^2}\right) + \frac{l^2}{c^2} \qquad (3)$$

The lattice constants for $SrFe_{12}O_{19}$ particles are, a=5.87 Å and c=23.17 Å agree with those reported in JCPDS card number 43-0002. TiO_2 shows the reflection planes (1 0 1), (0 0 4), (2 0 0), (1 0 5), and (2 1 1) confirming its crystallinity as reported in JCPDS card number 21-1272. The crystallite size of titania particles is 104 nm. The lattice constants, a=3.75 Å and c=9.50 Å.

Table 1: Microstructural parametrs calculated from XRD pattern

Particles	**Crystalline sizes (nm)**	**Lattice parameter '*a*' (angstrom)**	**Lattice parameter 'c' (angstrom)**
$SrFe_{12}O_{19}$	2.2	5.87	23.17
TiO_2	104	3.75	9.5

3.1.2 Density And Water Absorbance studies of $SrFe_{12}O_{19}/TiO_2$/LLDPE nanocomposite

Light weight microwave absorbers are easy to install and this in turn makes them suitable for free space applications. Density of a composite gives an idea of the compactness of the material and its weight. Measurement of density of the ferrite-titania-LLDPE nanocomposites is carried out by using Archimedes's principle [25]. The samples of dimension 10.16 mm · 22.86 mm · 3.77 mm are prepared and weight is measured in air, W_{air}. The composite is suspended in water and apparent immersed weight of the sample is measured, referred as W_{app}. Then the experimental bulk density (d_s) of the composite is measured by the Archimede's principle and is given by

$$d_s = \frac{W_{air}}{W_{air}-W_{app}} \times D_{water} \quad (4)$$

$$\text{or } d_s = \frac{W_{air}}{W} \times D_{water} \quad (5)$$

where

W_{air} = Weight of the sample in air (gm)

W_{app}= Weight of the sample in air - weight of the displaced water (gm)

D_{water} = Density of water at room temperature (=0.997 g/cc at 25 °C)

Water absorbance studies of the material help in determining the porosity of the material in humid and wet environmental conditions in which the system can work without affecting its microwave performance. The sample is inserted in water for 72 hours at room temperature. Thereafter pat dried at room temperature. The percentages of water absorption of the composites are determined according to the expression

$$Waterabsorbance(\%) = \frac{W_t - W_0}{W_0} \times 100 \quad (6)$$

where, W_t and W_0 are the weights of the wet and dry composites respectively.

Table 3.4 Density and percentage of water absorbance of $BaFe_{12}O_{19}$-NPR nanocomposites

Sample	Density (g/cc)	(%)Water absorbance
$SrFe_{12}O_{19}$-TiO_2-LLDPE	1.07	0.01

The bulk density of the magnetodielectric composite is 1.069 g/cc and the percentage of water absorbance is very less. The weight gain is only ~0.01%, confirming its use in high humid environments. The density measurement confirms the compactness of composite system. There is a slight weight gain of maximum ~0.01 % is observed when the sample is placed for 72 hours in water and indicates its use in high humidity environment.

3.2 Microwave characterization of the $SrFe_{12}O_{19}/TiO_2$/LLDPE composite

Complex permittivity and permeability of $SrFe_{12}O_{19}/TiO_2$/LLDPE composite is studied in the X-band. The complex permittivity, ($\varepsilon_r = \varepsilon_r' - j\varepsilon_r''$) spectra of the $SrFe_{12}O_{19}/TiO_2$/LLDPE composite carried out in the frequency range 8.2 GHz to 12.4 GHz is plotted in 2 (a & b).

The plots show that the composite reinforced with strontium ferrite and titania has real (ε_r') and imaginary (ε_r'') part of complex permittivity ranging in 3.0-3.5 and 0.25-3.0. The complex permittivity of the composite is almost constant over the X-band. The enhanced dielectric properties are observed due to the interfacial polarization and intrinsic electric dipole polarization of the ferrite particles. The polarization in ferrites is mainly due to the presence of Fe^{2+} ions. Since, Fe^{2+} ions are easily polarizable, with increase in the number of Fe^{2+} ions, the dielectric constant increases with increase in ferrite inclusions [26].

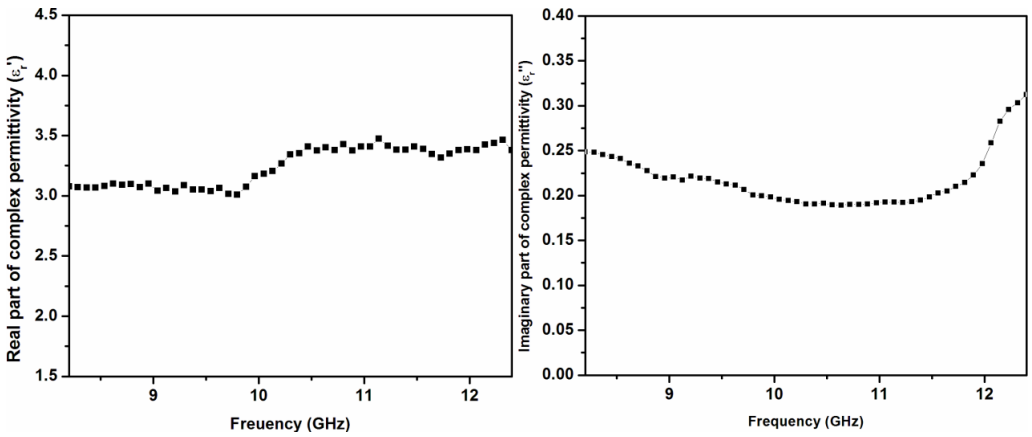


Figure 2: Complex permittivity of $SrFe_{12}O_{19}/TiO_2$/LLDPE nanocomposite, (a) real part and (b) imaginary part

The complex permeability spectra ($\mu_r = \mu_r' - j\mu_r''$), as depicted in figures 3 (a & b), shows resonant behavior for both μ' and magnetic loss, μ''. The dispersion of complex permeability in the magnetic polymer composite is primarily due to the resonance of oscillating domain walls and the resonance of precessing magnetic moments in the domains, which is known as natural ferromagnetic resonance [27]. The peak in μ_r' and μ_r'' corresponds to natural resonance frequency (f_r). The maximum complex permeability of, μ_r' ~1.11 and μ_r''~0.03 is observed for the $SrFe_{12}O_{19}/TiO_2$/LLDPE nanocomposite.

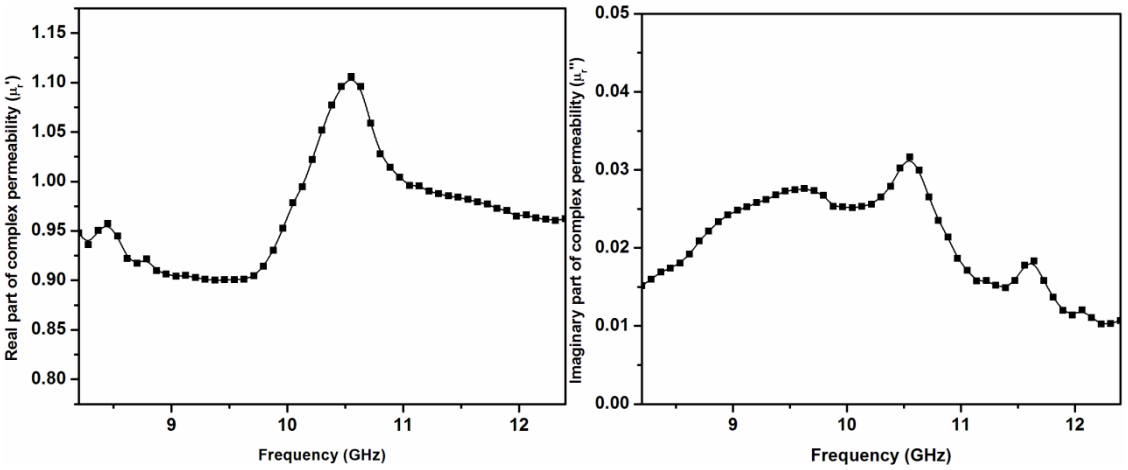


Figure 3: Complex permeability of $SrFe_{12}O_{19}/TiO_2$/LLDPE nanocomposite, (a) real part and (b) imaginary part

3.3 ABSORPTION STUDIES

Transmission/reflection method, using Agilent WR-90 X11644A rectangular waveguide line compatible with Agilent 8722ES vector network analyzer, is used to measure scattering parameter of the $SrFe_{12}O_{19}/TiO_2$/LLDPE nanocomposites [28]. The texture of the developed absorber samples are modified mechanically. Figure 4 shows the reflection loss (dB) versus frequency curve for the composites with rough and smooth surfaces. The $SrFe_{12}O_{19}/TiO_2$/LLDPE nanocomposites with smooth surface show absorption of -12.4 dB at 10.6 GHz, while the rough surface shows -17.03 dB at 11.80 GHz. The rough sample is showing -11 dB (>90% absorption) bandwidth throughout the X band (4.2 GHz) with -15 dB bandwidth of 0.6 GHz and i.e. for the smooth sample is 3.5 GHz. The -11 dB bandwidth for the smooth sample is ~4.0 GHz. Both the samples are showing a very broadband absorption in the X-band. The rough surface of the absorber sample is found to have better absorption capacity than the smooth one.

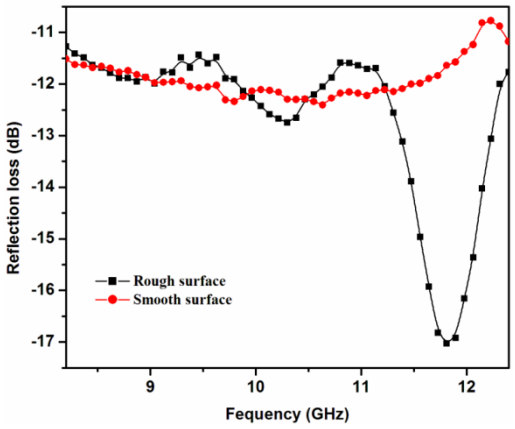


Figure 4. Reflection loss parameter of $SrFe_{12}O_{19}/TiO_2$/LLDPE nanocomposite

4. CONCLUSIONS

Microwave characterization of ferrite-titania-LLDPE nanocomposites are performed over the X-band .The composite reinforced with strontium ferrite and titania shows almost constant value of permittivity in the X-band and a high dielectric loss of ~3.0. The $SrFe_{12}O_{19}/TiO_2$/LLDPE nanocomposite shows resonant behavior in the complex permeability results. Maximum absorption of -17.03 dB at 11.80 GHz is obtained for the developed absorber with a rough surface with >-15 dB absorption of 0.6 GHz. The developed nanocomposite shows very less water absorption and can be used as a flexible absorber in humid environments.

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