

A Performance Study of Doped Strontium Ferrite/LLDPE Nano Composite as X-band Microwave Absorber

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Abstract—This paper presents the design, development and characterization of the M- type nano-sized hexagonal ferrite powder $[SrAl_xFe_{12-x}O_{19}, x=3]$ as magnetic filler, where aluminium is the dopant. The developed ferrite powder is mixed with Linear Low Density Polyethylene (LLDPE) matrix to form a composite. The complex permittivity and permeability of the composites for different weight % are measured in the X-band. The experimental results for a single layer microwave absorber have been reported. It has been found that it shows the broadband characteristics with minimum reflection loss of -26.7 dB and -10 dB bandwidth (i.e. 90% absorption) of ~3.02 GHz for a minimum thickness of 3 mm in over the X-band.

1. INTRODUCTION

Microwave absorbing materials are of two types: dielectric composites and magnetic composites based on the filler material. In order to achieve broadband absorption, the electrical/ magnetic properties of the absorbing materials as well as the structure are modified. In dielectric composites, there is no magnetic loss, so the thickness of the absorber is increased in order to get good absorption. Since in magnetic composites, both magnetic and dielectric losses exist, it helps to attenuate the wave within a smaller thickness. The design of these structures depends on material properties such as complex permittivity, complex permeability and conductivity. Nano- particles have high surface to volume ratio, which creates a large interaction zone and hence increases interfacial polarizations and magnetic interactive loss mechanism, which leads to enhanced dielectric and magnetic properties [1, 2].

Magnetic materials, spinel as well as hexagonal ferrites have been studied for absorption purpose [3, 4]. Spinel ferrites having isotropic properties show absorption up to 3 GHz while anisotropic properties of hexagonal ferrites with increased ferromagnetic resonance enable its use for applications above 3 GHz. M- type hexagonal ferrites have great importance due to their high dielectric and magnetic losses in the microwave frequency range. In order to change the magnetic properties, the hexaferrites are doped with special cations [5].

The intrinsic magnetic properties of hexaferrite can be significantly improved by substituting Fe^{3+} in different sites with other suitable ions, such as Cu^{2+} [5], Cr^{3+} [6], Ti^{4+} [7-9], Al^{3+} [10-14] for Fe^{3+} ions of hexaferrite. The microwave studies on Al^{3+} substituted $SrFe_{12-x}Al_xO_{19}$ are not reported yet. In general, the nonmagnetic Al^{3+} ions substitute the octahedral sites at low Al^{3+} doping level.

In this paper M- type nano- sized aluminium doped strontium hexagonal ferrite powder $[SrAl_xFe_{12-x}O_{19}, x=3]$ is studied as magnetic filler. The developed ferrite powder is mixed with a thermosetting polymer, LLDPE matrix to form composites to be used as X-band microwave absorbers. Subsequently, the complex permittivity, permeability and microwave absorbing performance are investigated in detail.

2. MATERIAL SYNTHESIS AND TESTING METHODS

Strontium nitrate ($\geq 98\%$), aluminium nitrate nonahydrate ($\geq 98\%$) and iron (III) nitrate nonahydrate ($\geq 98\%$) precursors are used as the base materials. NaOH is added dropwise to control the size of the particles. Oleic acid is used as surfactant in order to reduce inter-particle interaction and prevent agglomeration. Aqueous solutions of strontium, aluminium and iron salts are prepared separately by dissolving the salts in stoichiometric proportion in minimum amount of deionized water. The molar ratio of strontium to aluminium to ferric nitrate is set up as 1:3:9. Then the iron, aluminium and strontium salt solutions are mixed together by continuous magnetic stirring on a hot plate for 1 h. The solution is allowed several minutes to cool down to room temperature. Subsequently, the brownish precipitate is washed twice with distilled water and ethanol to get the precipitate free from sodium and nitrate compounds. Finally, the precipitate is dried in the hot-air oven. The dried powder is crushed and annealed at $1000^\circ C$ at $10^\circ C/min$ for 2 hours to get the powder. XRD studies of the synthesized powder is carried out.

Phase and morphology of the doped ferrites were respectively studied by X-ray diffraction analysis. LLDPE is used as the polymer matrix. The composite is prepared in situ by mixing mechanically desired weight % of the filler material into LLDPE powder. Then, the evaluation of the reflection loss of the samples in X- band (8.2-12.4 GHz) was determined by a vector network analyzer.

3. CHARACTERIZATIONS AND DISCUSSIONS

3.1 Microstructural Characterizations

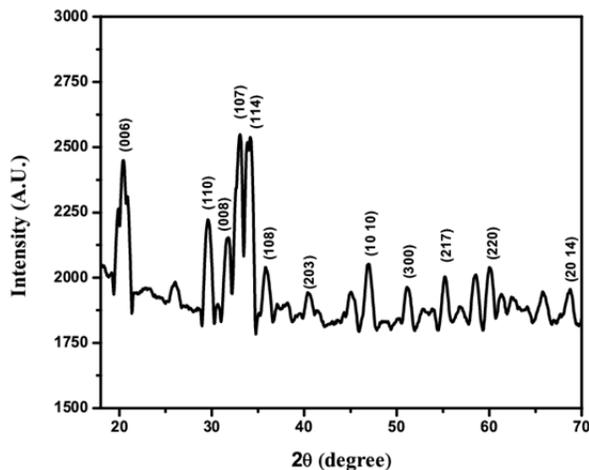


Fig. 1. XRD pattern of $\text{Sr}_3\text{Fe}_9\text{O}_{19}$ annealed at 1000°C .

Fig. 1 shows XRD pattern of $\text{SrFe}_{12}\text{O}_{19}$ annealed at 1000°C . Reflection planes, (1 0 1), (1 0 2), (0 0 6), (1 1 0), (1 0 7), (1 1 4), (2 0 3), (2 0 5), (1 0 10), (2 0 9), (2 0 11), (2 2 0) and (2 0 14) indicates M-type hexagonal structure of the aluminium substituted strontium ferrites. The planes are determined from JCPDS card number 33-1340. No characteristic plane of Al^{3+} ions is observed confirming that the Al^{3+} ions enter the lattice of strontium ferrite as the ionic radius of Al^{3+} ion (0.0535 nm) is less than that of Fe^{3+} ions (0.065 nm) [14]. Crystallinity and size of the particles are calculated using Debye-Scherrer formula [15] and are found to be 28.8 nm.

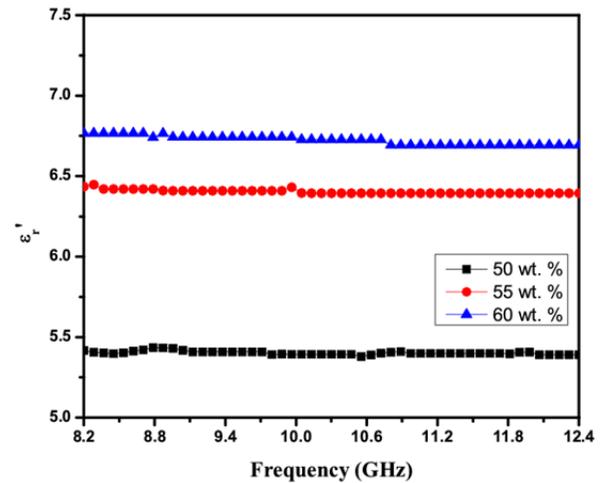
3.2 Microwave Characterizations

Permittivity and permeability of composites are measured using Thru-Reflect-Line (TRL) method [16]. Pellets of dimensions 10.16 mm x 22.86 mm x 3mm, of different weight % of composite is placed in Agilent WR-90 X11644A. The measurements are carried out using Agilent E8362C Vector Network Analyzer, in the frequency range 8-12 GHz. The permittivity is determined using Nicolson-Ross technique.

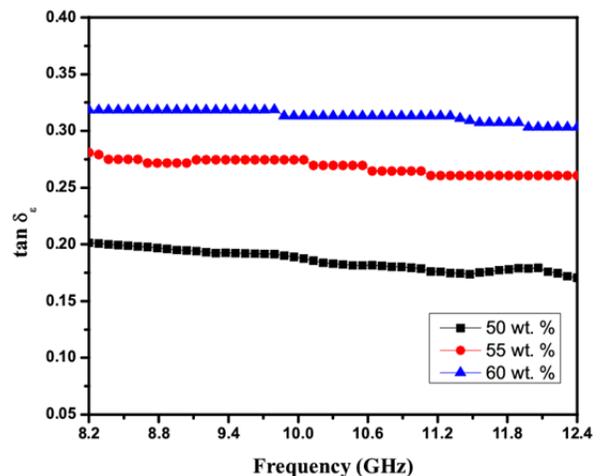
Complex permittivity and complex permeability

Evaluation of complex permittivity ($\epsilon_r = \epsilon_r' - j\epsilon_r''$) and complex permeability ($\mu_r = \mu_r' - j\mu_r''$) of the samples are done in X- band. Fig. 2 (a-d) shows the real part of

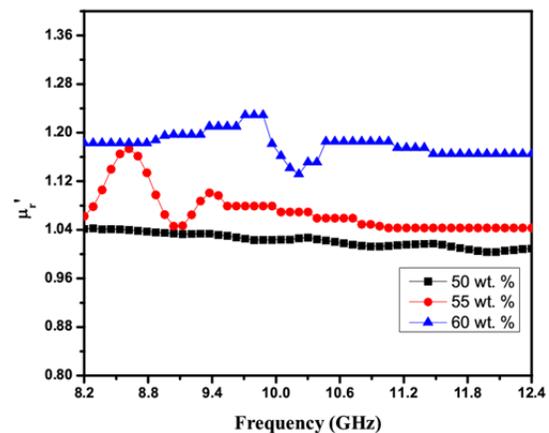
permittivity, dielectric loss tangent, real part of permeability and magnetic loss tangent respectively.



(a)



(b)



(c)

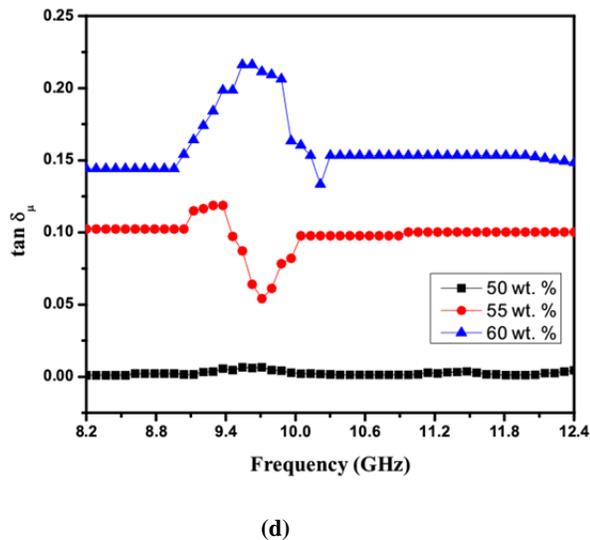


Fig. 2: (a) Real part of complex permittivity (ϵ'_r), (b) Dielectric loss tangent ($\tan \delta_\epsilon$), (c) Real part of complex permeability (μ'_r), (d) Magnetic loss tangent ($\tan \delta_\mu$) of $\text{SrAl}_3\text{Fe}_9\text{O}_{19}$ -LLDPE composite.

Sample with 60wt. % $\text{SrAl}_3\text{Fe}_9\text{O}_{19}$ -LLDPE composite has higher real part of permittivity, permeability and higher loss tangent. The composites with three different weight percentages are fabricated with a thickness of 3 mm and used to measure the reflection loss (RL) experimentally and hence the absorption.

Absorption studies

To determine RL, single layer conductor backed configuration is considered. Using (TRL) method the RL is determined experimentally for all the three composites. When an electromagnetic wave strikes the surface of a metal- backed microwave absorber at normal incidence, the reflection loss is a function of frequency, thickness, permittivity and permeability of the absorbing material. The electromagnetic attenuation offered by absorbing materials may depend on the three mechanisms which are reflection of the incoming wave, absorption of the wave as it passes through the material's thickness, and multiple reflections of the waves at various interfaces.

The reflection loss of the metal-backed single layer absorber is given by,

$$RL = 20 \log |(Z_{in} - Z_0) / (Z_{in} + Z_0)| \quad (1)$$

where $Z_0 = (\mu_0 / \epsilon_0)^{1/2}$ is the free- space impedance, and the input impedance at the air- absorber interface is given as

$$Z_{in} = Z_0 (\mu_r / \epsilon_r)^{1/2} \tanh [j2\pi f d (\mu_r / \epsilon_r)^{1/2} / c] \quad (2)$$

where f is the frequency, c is the velocity of light and d is the thickness of the absorbing material. From the equations it can be seen that how the complex permeability and complex permittivity affect the absorption along with the thickness of the absorbing layer. Fig.3 shows the measured reflection loss

of the fabricated composites. It can be seen that among the three weight fractions, 60 wt. % shows the maximum RL value of -26.7 dB with a -10 dB absorption bandwidth of ~3.02 GHz.

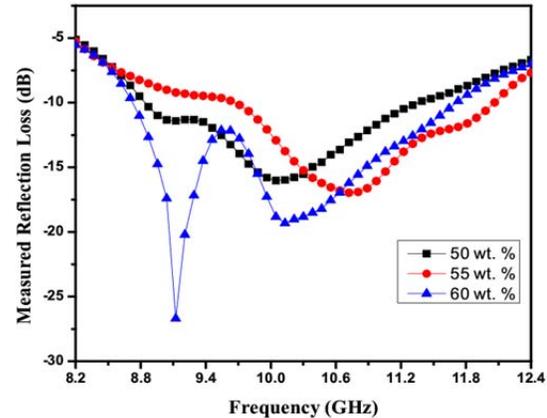


Fig. 3: Measured reflection loss of Reflection Loss (RL) of the conductor backed single layer $\text{SrAl}_3\text{Fe}_9\text{O}_{19}$ -LLDPE absorber for different weight percentages.

4. CONCLUSION

Diffraction pattern shows the formation of cobalt doped strontium ferrite and the size of the particle is found to be in nanometer range. A maximum RL of -26.7 dB and -10 dB absorption bandwidth (i.e. 90% absorption) of 3.02 GHz over the X- band is found for 60 wt. % for $\text{SrAl}_x\text{Fe}_{12-x}\text{O}_{19}$ /LLDPE composite ($x=3$). Substitution of Fe^{3+} with Al^{3+} is found to increase the absorption properties of strontium ferrite. The results of this investigation exhibit the possibilities of using $\text{SrAl}_3\text{Fe}_9\text{O}_{19}$ -LLDPE composites as thin, light weighed and broadband single layer microwave absorbers.

REFERENCES

- [1] Jalalia, M., Dauterstedt, S., Michaud, A. and Wuthrich, R., "Electromagnetic shielding of polymer-matrix composites with metallic nanoparticles", *Composites Part B Engineering*, 42, 6, Sept. 2011, pp. 1420-1426.
- [2] Chen, N., Mu, G., Pan, X., Gan, K. and Gu, M., "Microwave absorption properties of $\text{SrFe}_{12}\text{O}_{19}/\text{ZnFe}_2\text{O}_4$ composite powders", *Material Science Engineering B*, 139, 2-3, May 2007, pp. 256-60.
- [3] Kim, S.-S., Kim, S.-T., Yoon, Y. and Lee, K., "Magnetic, dielectric, and microwave absorbing properties of iron particles dispersed in rubber matrix in gigahertz frequencies," *Applied Physics Letters*, 97, 10, May 2005, pp. 10F905.
- [4] Ghasemi, A., Hossienpour, A., Morisako, A., Saatchi, A. and Salehi, M., "Electromagnetic properties and microwave absorbing characteristics of doped barium hexaferrite", *Journal of Magnetism and Magnetic Materials*, 302, 2, July 2006, pp. 429-435.

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- [5] You, L. Q. and Zheng, J., "The magnetic properties of strontium hexaferrites with La-Cu substitution prepared by SHS method", *Journal of Magnetism and Magnetic Materials*, 318, 1-2, Nov. 2007, pp. 74-78.
- [6] Ounnunkada, S. and Winotai, P., "Properties of Cr-substituted M-type barium ferrites prepared by nitrate-citrate gel-autocombustion process", *Journal of Magnetism and Magnetic Materials*, 301, 2, June 2006, pp. 292-300.
- [7] Clark, T. M., Evans, B. J. and Thomson, G. K., "⁵⁷Fe Mössbauer spectroscopic investigation of complex magnetic structures in Ga, Sc, and In substituted M-type hexagonal ferrites", *Journal of Applied Physics*, 85, 8, Apr. 1999, pp. 5229-5230.
- [8] Castellanos, P. A. M., Rivera, J. A., Fuentes, A. C. and Serrano, R. L., "Magnetic and microstructural properties of the Ti⁴⁺-doped Barium hexaferrite", *Journal of Magnetism and Magnetic Materials*, 280, 2, Sept. 2004, pp. 214-220.
- [9] Albanese, G., Carbucicchio, M., Pareti, L., Rinaldi, S., Lucchini, E. and Slokar, G., "Low temperature specific heat of dilute iron-carbon martensite", *Journal of Magnetism and Magnetic Materials*, 15-18, 3, Jan. 1980, pp. 1453-1454.
- [10] Dhage, V. N., Mane, M. L., Keche, A. P., Birajdar, C. T. and Jadhav, K. M., "Structural and magnetic behaviour of aluminium doped barium hexaferrite nanoparticles synthesized by solution combustion technique", *Physica B*, 406, 4, Feb. 2011, pp. 789-793.
- [11] Eraky, M. R., Beslepkin, A. A. and Kuntsevich, S. P., "Magnetic properties and NMR studies of the SrAlM hexagonal ferrite system", *Materials Letters*, 57, 22-23, July 2003, pp. 3427-3430.
- [12] Bregar, V. B., "Advantages of ferromagnetic nanoparticle composites in microwave absorbers", *IEEE Trans. on Magnetics*, 40, 3, May 2004, pp. 1679-1684.
- [13] Motojima, S., Noda, Y., Hoshiya, S. and Hishikawa, Y., "Electromagnetic wave absorption property of carbon microcoils in 12-110 GHz region", *Journal of Applied Physics*, 94, 4, May 2003, pp. 2325-2330.
- [14] Qiu, J., Gu, M. and Shen, H., "Microwave absorption properties of Al- and Cr substituted M-type barium hexaferrite", *Journal of Magnetism and Magnetic Materials*, 295, 3, Sept. 2005, pp. 263-268.
- [15] Klug, H. P. and Alexander, L.E., *X-ray Diffraction Procedures for Poly Crystalline and Amorphous Materials*, Wiley Interscience, New York, 1974.
- [16] Engen G. F. and Hoer, C. A., "Thru-reflect-line: An improved technique for calibrating the dual six- port automatic network analyzer", *IEEE Trans on Microwave Theory and Techniques*, 27, 12, Dec. 1979, pp. 987-993.