DEVELOPMENT OF AL- DOPED STRONTIUM FERRITE/ LLDPE NANO COMPOSITE AND ITS MICROWAVE ABSORPTION PERFORMANCE IN X-BAND

Soma Chakraborty[1], Nidhi S. Bhattacharyya[2], Satyajib Bhattacharyya[3]

[1]*Dept. of ECE, Tezpur University, Napaam, Assam-784028, India*
[2]*Microwave Engineering Laboratory, Dept. of Physics, Tezpur University, Napaam, Assam-784028, India*
[3]*Dept. of ECE, Tezpur University, Napaam, Assam-784028, India*

Abstract

In the present work, aluminium doped strontium ferrite nanoparticles ($SrAl_xFe_{12-x}O_{19}$, x=1, 2, 3, 4) are used as inclusions in Linear Low Density Polyethylene (LLDPE) matrix to develop a composite. $SrAl_xFe_{12-x}O_{19}$ is prepared for three different annealing temperatures, i.e., 800ºC, 900ºC and 1000ºC. The X-ray diffraction study is conducted on $SrAl_xFe_{12-x}O_{19}$.The complex permittivity and complex permeability is measured at X-band. Reflection loss of $SrAl_xFe_{12-x}O_{19}$/LLDPE composite is determined using transmission line modelling (TLM) from the measured permittivity values. Thickness optimization carried out for the same sample from 1 mm to 4 mm, showed maximum reflection loss for 3 mm thickness and -10 dB bandwidth (i.e. 90% absorption) of ~3.02 GHz (8.70 to 11.72 GHz) for $SrAl_xFe_{12-x}O_{19}$/LLDPE composite (x=3).

---***---

1. INTRODUCTION

Increasing use of wireless communications, radar systems and military applications in the gigahertz frequency range has increased the demand of electromagnetic shields and microwave absorbing materials. To minimize em wave reflections from large structures such as aircraft, ships and tanks and to cover the walls of anechoic chambers microwave absorbing materials (MAMs) have been widely used. In general, RAMs are fabricated in the form of sheets that consist of polymer, like natural rubber, epoxy resin, novalac phenolic resin and magnetic or dielectric loss materials such as ferrite, permalloy, carbon black, and short carbon fiber. Microwave absorption characteristic of material depends on its dielectric properties (complex permittivity, $\varepsilon_r = \varepsilon_r^{'} - j\varepsilon_r^{''}$), magnetic properties (complex permeability, $\mu_r = \mu_r^{'} - j\mu_r^{''}$), thickness and frequency range. Magnetic materials, such as, M-type hexagonal ferrites are known to have high saturation magnetization, large dielectric and magnetic losses, high curie temperature, stability over the time and low synthesis cost.In order to change the magnetic properties, the hexaferrites are doped with special cations [1].

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+}, Cr^{3+}, Ti^{4+}, Al^{3+}[1-9] 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 the present work, synthesis of aluminium doped strontium ferrite nanoparticles ($SrAl_xFe_{12-x}O_{19}$, x=1, 2, 3, 4) is carried out which is used as inclusions in Linear Low Density Polyethylene (LLDPE) matrix to develop a composite, is studied. The developed composite is characterized forcomplex permittivity andcomplex permeability in X-band. Using transmission line (TL) technique,reflection loss of the $SrAl_xFe_{12-x}O_{19}$/LLDPE composite is additionally addressed, for conductorbacked single layer structure.Thickness optimization carried out for the same sample from 1 mm to 4 mm.

2. MATERIAL SYNTHESIS AND STRUCTURAL CHARACTERIZATION

Nanosized Al-substituted barium ferrite particles are preparedusing co-precipitation technique. According to the stoichiometriccomposition of $SrAl_xFe_{12-x}O_{19}$ (x=1, 2, 3 and 4), strontium nitrate (≥98%), aluminium nitrate nonahydrate (≥98%) and iron (III) nitrate nonahydrate (≥98%) are taken as precursors and dissolved indeionised water. Sodium hydroxide is addeddropwise in the solution to control the size of the particles. Oleicacid is used as surfactants in order to reduce inter-particle interaction and prevent agglomeration. The precipitate powder is dried in the hot-air oven and then, annealed atthree different temperatures 800˚C, 900˚C and 1000˚Cfor two hours to get the powder.XRD studies of the synthesized powder is carried out at different temperatures as shown in fig.1.

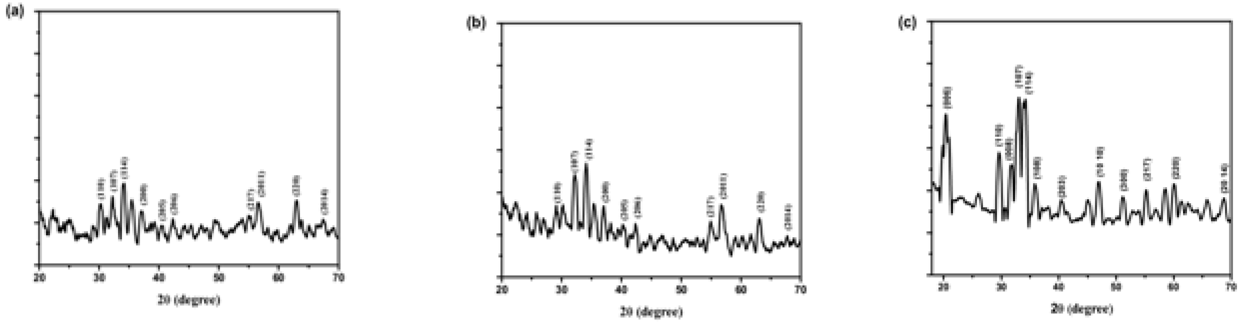

Fig 1: XRD pattern of $SrFe_{12}O_{19}$ annealed at (a) 800˚C, (b) 900˚C and (c) 1000˚C.

Formation of single phase M-type strontium ferrite is confirmedfrom X-ray diffraction (XRD) pattern, using Rigaku, Miniflex 200X-ray diffractometer with Cu Kαline of wavelength λ=1.541841 Å,recorded at 2θvalues from 10˚ to 70˚. Fig. 1(a–c) shows XRDpattern at different annealing temperature. 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 structureof the aluminium substituted strontium ferrites. The planes aredetermined from JCPDS card number 33-1340. No characteristicplane of Al^{3+}ions is observed confirming that the Al^{3+}ions enterthe 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)[10]. Crystallinity and size of the particles are calculated using Debye–Scherrer formula [11] and are found to be in nano size given in Table 1 along with the lattice constants. The lattice constantsaand cincrease with increasingannealing temperature.

Table 1 Calculated crystalline size (D) and lattice parameter of$SrAl_xFe_{12-x}O_{19}$ (x=1).

Ferrite	Annealing temperatures	Average crystalline size, D (nm)	Lattice parameter 'a' (Å)	Lattice parameter 'c' (Å)
$SrAl_xFe_{12-x}O_{19}$ (x=1)	800˚C	19.69	5.861	23.022
	900˚C	24.27	5.875	23.039
	1000˚C	28.8	5.887	23.047

It is noteworthy at lower sintering temperatures additional diffraction peaks are seen which s are absent at 1000˚C, showing complete annealing of the precursors. Subsequently, all the Al-doped ferrite powders (for x=2,3 and 4) are annealed at 1000˚C.

Composite samples with 60wt. % of nanosized $SrAl_xFe_{12-x}O_{19}$ (for x=1,2,3 and 4) in LLDPE matrix is developed using Al- doped strontium ferrite annealed at 1000˚C.LLDPE is used as the polymer matrix.The composite is prepared in situ by mixing mechanically 60 wt. % of the filler material into LLDPE powder and heating it for 2 hours. Then, the evaluation of complex permittivity and complex permeability of the samples in X- band (8–12.4 GHz) was determined by a vector network analyzer.

3. MICROWAVE CHARACTERIZATION

Complex permittivity($\varepsilon_r = \varepsilon_r^{'} - j\varepsilon_r^{''}$) and complex permeability($\mu_r = \mu_r^{'} - j\mu_r^{''}$)for 60 wt. % of $SrAl_xFe_{12-x}O_{19}$-LLDPE nano-composites over the X-band is measuredusing Nicolson–Ross methodusing AgilentWR-90x 11644A rectangular waveguide line compatible with Agilent8722ES vector network analyzer. For microwave characterization,pellets of dimensions 10.38 mmx22.94 mm x 4 mm are prepared.The frequency plots of ε_r', dielectric loss tangent, μ_r' and magnetic loss tangent are given in figures Fig. 2 (a-d).With increase in Al^{3+} ion substitution, ε_r'increases. The maximum $\varepsilon_r'$is observed for the $SrAl_xFe_{12-x}O_{19}$-LLDPE composite with x=4. But the dielectric and magnetic loss is more for x=3. With increase in the Al^{3+} ions, the exchange phenomenon between Fe^{3+} to Fe^{2+} reduces but the number of free Fe^{2+}ions increases in comparison to Fe^{3+}ions, thus leading to increase in complex permittivity values.

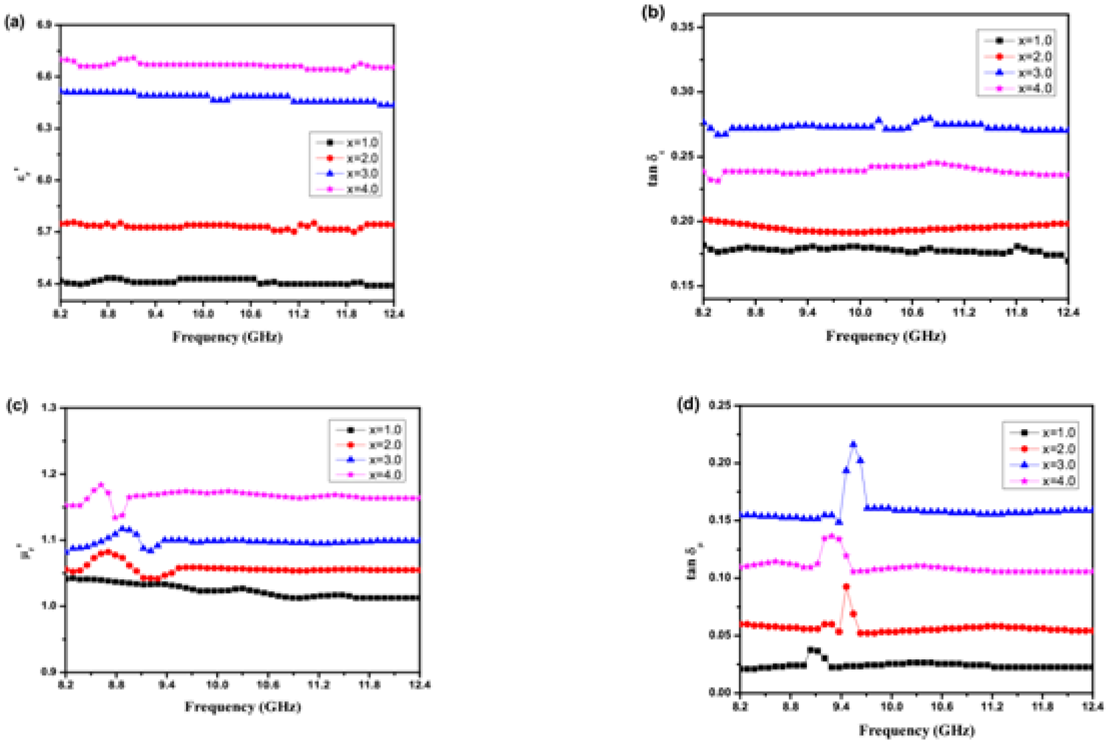

Fig 2: (a) Real part of complex permittivity (ε'_r), (b) Dielectric loss tangent ($tan\ \delta_\varepsilon$), (c) Real part of complex permeability (μ_r'), (d) Magnetic loss tangent ($tan\ \delta_\mu$) of $SrAl_xFe_{12-x}O_{19}$-LLDPE (x=1, 2, 3 and 4) composite.

The $SrAl_xFe_{12-x}O_{19}$-LLDPE nanocomposites are studiedfor its microwave absorption properties. Thickness optimization istheoretically carried out using TLM.

4. REFLECTION LOSS COMPUTED USING TRANSMISSION LINE METHOD (TLM)

The reflection loss of the designed singlelayer absorber based on TLM [12], is optimized with thickness for all the stoichiometric $SrAl_xFe_{12-x}O_{19}$-LLDPE nanocomposites using Eq.(1).

$$RL = 20\log\left|\frac{Z_{in} - Z_0}{Z_{in} + Z_0}\right| \qquad (1)$$

$$Z_{in} = Z_0 \tanh\left[j\frac{2\pi fd}{c}\sqrt{\mu_r \varepsilon_r}\right] \qquad (2)$$

where, Z_{in} is the input impedance at the air-absorber interphase, $Z_0$is the impedance of free space, f is the frequency of operation and d is the thickness of the composite. Fig. 3 shows the schematic of a single layer absorber material.

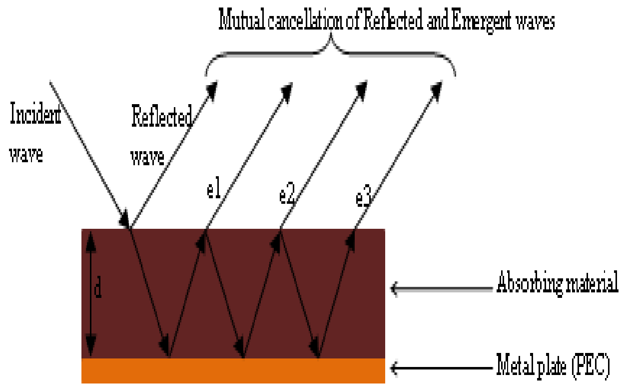

Fig 3: Schematic of a metal backed single layer absorber material.

The thickness of the absorber sample is varied from 1 mm to 4 mmin step of 0.5 mm. All the four $SrAl_xFe_{12-x}O_{19}$-LLDPE composites (x=1, 2, 3, 4)show -10 dB reflection loss bandwidth over the X-band for$d$=3 mm, as shown inFigs. 4(a-d).

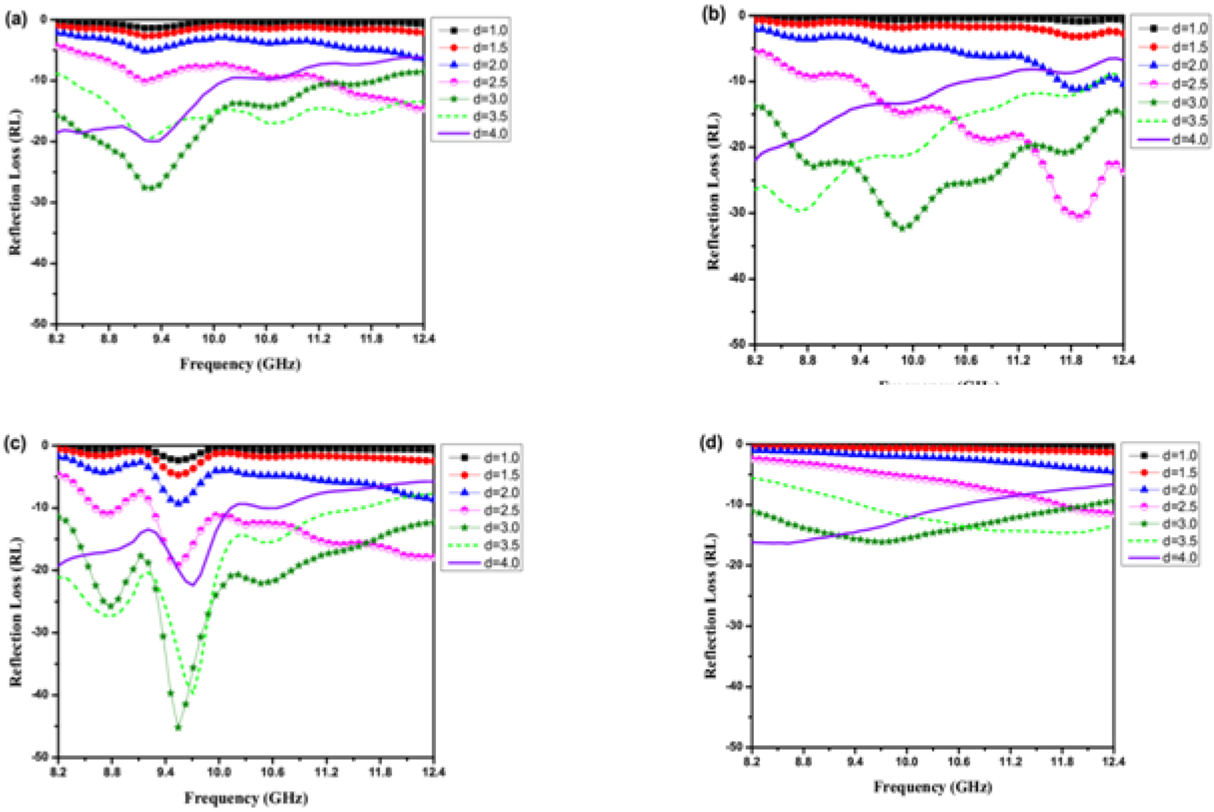

Fig 4: *RL*of $SrAl_xFe_{12-x}O_{19}$-LLDPEcomposite with thickness varying from 1mm to 4mm for (a) x=1 (b) x=2 (c) x=3 & (d) x=4 in X-band frequency range.

It can be seen that for $SrAl_xFe_{12-x}O_{19}$-LLDPE, x=3, the calculated reflection loss is highest among all the cases at*d*=3. Thus, *d*=3mmthickness sheets for all the compositions can be fabricated formeasurement of absorption.

5. CONCLUSION

Thickness optimization carried out for the same sample from 1 mm to 4 mm, showed maximum reflection loss for 3 mm thickness and -10 dB bandwidth (i.e. 90% absorption) over the whole band for $SrAl_xFe_{12-x}O_{19}$/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 developing thin, light weighed and broadband microwave absorbers using nanosized $SrAl_xFe_{12-x}O_{19}$ in LLDPE.

REFERENCES

[1] L.Q. You, J. Zheng, The magnetic properties of strontium hexaferrites with La–Cu substitution prepared by SHS method,Journal of Magnetism and Magnetic Materials, 318, 2007, pp. 74-78.

[2] S. Ounnunkada, P. Winotai, Properties of Cr-substituted M-type barium ferrites prepared by nitrate-citrate gel-autocombustion process, Journal of Magnetism and Magnetic Materials, 301, 2006, pp. 292-300.

[3] T.M. Clark, B.J. Evans, G.K. Thomson, 57 Fe Mössbauer spectroscopic investigation of complex magnetic structures in Ga, Sc, and In substituted *M*-type hexagonal ferrites,Journal of Applied Physics, 85, 1999, pp. 5229-5230.

[4] P.A.M. Castellanos, J.A. Rivera, A.C. Fuentes, R.L. Serrano, Magnetic and microstructural properties of the Ti^{4+}-doped Barium hexaferrite, Journal of Magnetismand Magnetic Materials 280, 2004, pp. 214-220.

[5] G. Albanese, M. Carbucicchio, L. Pareti, S. Rinaldi, E. Lucchini, G. Slokar, Low temperature specific heat of dilute iron-carbon martensite, Journal of Magnetism and Magnetic Materials, 15-18, 1980, 1453-1454.

[6] Vinod N. Dhage, M.L. Mane, A.P. Keche, C.T. Birajdar, K.M. Jadhav, Structural and magnetic behaviour of aluminium doped barium hexaferrite nanoparticles synthesized by solution combustion technique, Physica B, 406, 2011, pp. 789-793.

[7] M.R. Eraky, A.A. Beslepkin, S.P. Kuntsevich, Magnetic properties and NMR studies of the SrAlM hexagonal ferrite system, Materials Letters, 57, 2003, pp. 3427-3430.

[8] V. B. Bregar, Advantages of ferromagnetic nanoparticle composites in microwave absorbers, IEEE Trans. on Magnetics, 40, 2004, pp. 1679-1684.

[9] S.Motojima, Y. Noda, S. Hoshiya and Y. Hishikawa, Electromagnetic wave absorption property of carbon microcoils in 12–110 GHz region,Journal of Applied Physics, 94, 2003, pp. 2325-2330.
[10] J. Qiu, M. Gu, H. Shen, Microwave absorption properties of Al- and Crsubstituted M-type barium hexaferrite, Journal of Magnetism and Magnetic Materials, 295, 2005, pp. 263-268.
[11] H.P. Klug, L.E. Alexander, X-ray Diffraction Procedures for Poly Crystalline and Amorphous Materials, Wiley Interscience, New York, 1974.
[12] Y. B.Feng, T. Qiu and C. Y. Shen, Absorbing properties and structural design of microwave absorbers based on carbonyl iron and barium ferrite, Journal of Magnetism and Magnetic Materials,318, 2007, pp. 8–13.