# Magnetic and dielectric studies on Zn doped Mg ferrites (MgZn$_x$Fe$_{2-x}$O$_4$) nanofabricated using self-combustion method

**Anu**

*Department of Chemistry, BUC College, Batala (Gurdaspur)-143505, India.*

## Abstract

Herein, magnetic nanoparticles of Zn doped Mg ferrite have been obtained employing citric acid mediated sol-gel self-combustion route. Microstructural analysis using various spectroscopic studies e.g. X-ray diffraction (XRD), infrared study (IR), scanning electron, transmission electron microscopy (SEM, TEM) and magnetic study (VSM) reveal monophasic, chemically pure, nanocrystalline and magnetically ordered particles with mixed electro-magnetic properties. Dielectric study indicates that increase in Zn concentrations result in an increase of resistivity of doped Mg ferrites.

**Keywords:** Nanoparticles, self-combustion, electron microscopy, electro-magnetic properties.
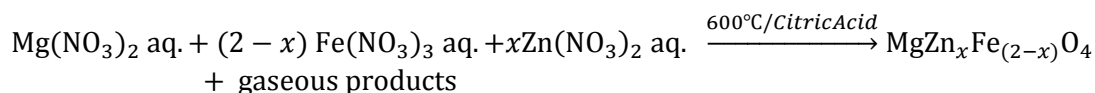
## 1. INTRODUCTION

Ferrite nanoparticles show unusual magnetic properties which are not observed in bulk material such as single domain behaviour and superparamagnetism [1,2]. Due to ease of preparation, low cost and high resistivity, ferrites have been regarded as better magnetic materials than pure metals and play an important role in the advancement of microwave materials technology over the past five decades. They find an ever increasing application in microwave devices, memory core of computers, radars, satellite communication and as permanent magnets [3-12].

Ferrites are essentially a dilute system of metal ions interspaced between oxygen ions. The oxygen ions play a role of mediating the magnetic interaction between the metal ions. The oxygen ions also insulate metal ions from each other which are the basis of much increased resistivity of ferrites. The magnetism in ferrites originates from the magnetic moments at the metal ions. The interaction of these magnetic moments through the intervening O$^{2-}$ ions is responsible for the observed magnetic order. This interaction is called superexchange interaction. The properties of the ferrite materials,

which decide the application areas are generally governed by the chemical compositions and procedures followed for their preparation. In the present study, solution combustion method has been undertaken to prepare powder ferrites with varying composition in pure phase. The method involves exothermic decomposition of the precursor that provides heat to the solid-state reaction, thus, decreasing the external temperature required for the obtention of ferrite. Therefore, stoichiometrically pure and single-phase ferrites are formed at lower temperature and in shorter time with greater surface area [3-5].

## 2.    MATERIALS AND METHOD

Stoichiometric quantities of the reactants i.e. $Mg(NO_3)_2$, $Zn(NO_3)_2$, $Fe(NO_3)_2.9H_2O$ and citric acid were stirred at a temperature of 80 ºC. After half an hour, temperature was increased to 130 ºC, so that the solution changes to the gel form and finally undergoes self-combustion to form fine powdered material. As-obtained mixture was then ignited for 3 hours in a muffle furnace maintained at 600ºC.

$$Mg(NO_3)_2 \text{ aq.} + (2-x)\,Fe(NO_3)_3 \text{ aq.} + x Zn(NO_3)_2 \text{ aq.} \xrightarrow{600°C/\textit{CitricAcid}} MgZn_x Fe_{(2-x)}O_4$$
$$+ \text{ gaseous products}$$

## 3.    RESULTS AND DISCUSSION:
### 3.1.    XRD Study

Figure 1a shows the X-ray diffraction patterns for different compositions of mixed Mg ferrites. The diffraction peaks reveal the existence of single phase cubic spinel ferrites and are comparable to those reported for reported ferrites [5-8]. Figure 3 shows the variation of lattice constant (Table 1) as a function of 'x' which is attributed to the substitution of larger $Zn^{2+}$cation (83 pm) for smaller $Fe^{3+}$cation (67 pm).
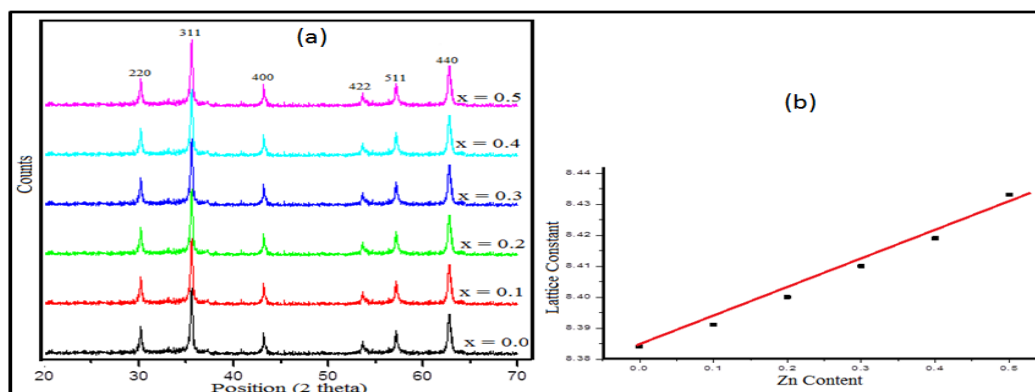


**Fig.1 (a):** X-ray powder diffraction patterns, (b) Variation of Lattice constant 'a' with Zn content for $MgZn_xFe_{2-x}O_4$;(x= 0 to 0.5)

Magnesium ferrite have all the $Mg^{2+}$ ions in octahedral position along with half of the $Fe^{3+}$ ions and remaining $Fe^{3+}$ ions occupy tetrahedral site. On addition of $Zn^{2+}$ ions which have strong affinity for tetrahedral site, only $Fe^{3+}$ ions present at tetrahedral site get replaced resulting in an increase in lattice parameter. The average particle size of the ferrite product calculated from the XRD pattern comes out to be 25-35 nm.

**Table-4:** Variation of physico-chemical parameters with composition 'x'.

| Composition (x) | Lattice parameter (a) | *Ms* (emu/g) | Resistivity (Ohm cm) |
|---|---|---|---|
| 0.0 | 8.384 | 24 | $7.02 \times 10^5$ |
| 0.1 | 8.390 | 35 | $8.5 \times 10^5$ |
| 0.2 | 8.399 | 46 | $1.3 \times 10^6$ |
| 0.3 | 8.409 | 53 | $1.5 \times 10^6$ |
| 0.4 | 8.418 | 38 | $1.7 \times 10^6$ |
| 0.5 | 8.433 | 26 | $1.8 \times 10^6$ |

### 3.2. MicrostructuralAnalysis

The size and shape of ferrite particles synthesized by the solution combustion route were also analyzed by scanning and transmission electron micrographs (SEM,TEM). An average particle size of 25-35 nm has been estimated by TEM for the nanocrystalline powders as shown in Figures 2 [12]. The smaller particle size may be attributed to the combustion synthesis involving molecular level heating without thermal gradient and requiring much smaller time than the conventional sintering ceramic technique.
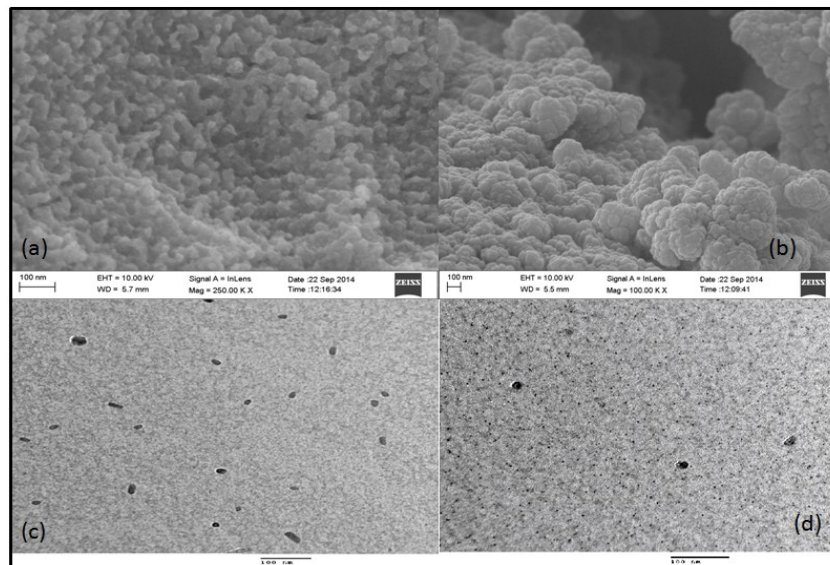


**Fig.2:** SEM micrograph for (a) x = 0.1, (b) x = 0.5, (c) TEM micrograph for x = 0.1, (d) x = 0.5

### 3.3.  Magnetic Study

Magnetic measurements reveal that all the samples show hysteresis loop, a typical ferromagnetic behavior of nanosized magnetic materials. Figures 3 show the variation of saturation magnetizationas a function of Zn content. The magnetic parameters (Table 1) for mixed magnesium ferrites show a similar trend to that observed for reported ferrites [8-10].
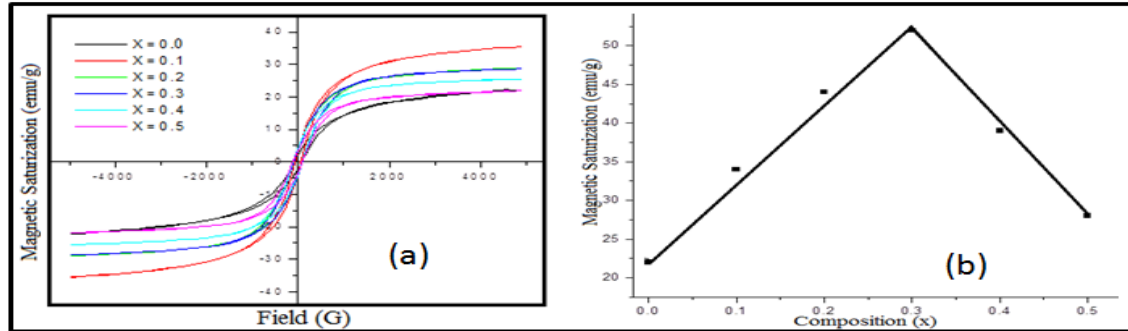


**Fig.3 (a), (b):** Variation of saturation magnetization with applied magnetic field.

### 3.4.  Dielectric Study

Dielectric properties for different ferrite samples were studied in the frequency range $10^2$ – $10^7$ Hz, (Fig. 4a). The frequency dependence of the dielectric constant ($\varepsilon'$) shows a continuous decrease with increase in frequency with pronounced dispersion at lower frequency and it remains almost independent of applied external field at high frequency domain. The existence of dielectric dispersion can be explained on the basis of Koop's two-layer model and Maxwell–Wagner polarization theory [6-9].
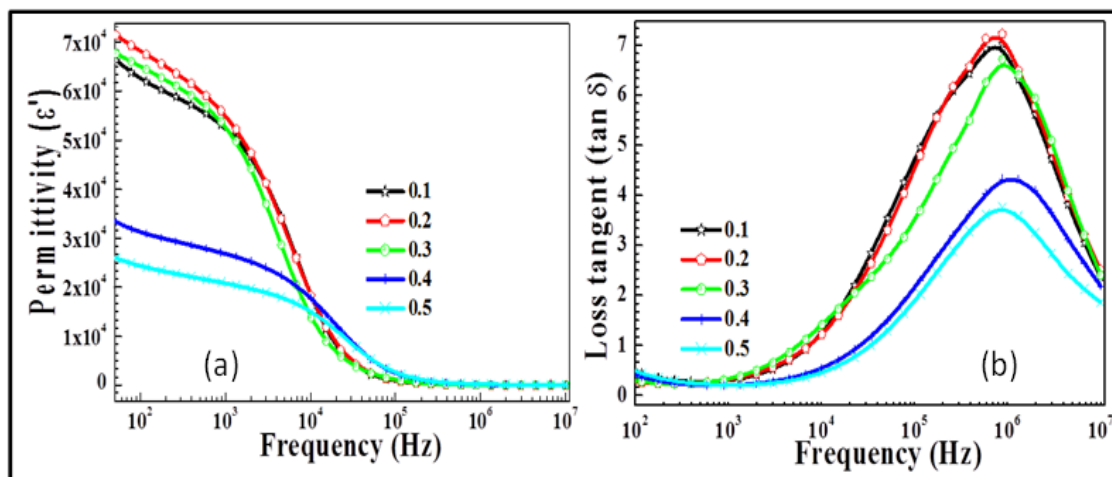


**Fig. 4:** Variation of (a) Dielectric constant (b)Tangent loss ($\delta$) with frequency.

Figure 4b shows an initial increase in the value of tangent loss ($\delta$) to attain a maxima followed by a regular decrease with frequency. It can be noted that the height of the peak increases with $Zn^{2+}$ ions substitution at x=0.1, and then it shows a subsequent decrease with increase of $Zn^{2+}$ ion concentration. The decrease of the height of the peak of tan $\delta$ may be attributed to the substitution of diamagnetic $Zn^{2+}$ions in place of $Fe^{3+}$ ions that limits the degree of conductivity by blocking hopping conduction mechanism, thus, resulting in an increase of resistivity.

## 4. CONCLUSIONS:

Citric acid mediated solution combustion method has been used for the synthesis of pure and doped Mg ferrite nanoparticles at shorter time and in lower temperature than the conventional ceramic method. X-ray powder diffraction studies reveal the formation of single phase spinel ferrites, which ensures thehigh purity of materials. The lattice parameter 'a' has been found to increase with increasing Zn content (x) which is attributed to the larger cationic radii of substituent $Zn^{2+}$ ions than $Fe^{3+}$ ions being replaced. An increase in the magnitude of saturation magnetization with increasing Zn content up to x = 0.3 has been explained on the basis of Neel's two sublattices model, while a decrease in the saturation magnetization with increasing Zn content for x > 0.3 has been described on the basis of Yafet and Kittel spin canted structure. The high dc-resistivity values obtained for the ferrites make them suitable for high-frequency applications and hence, play a key role as magnetic/engineering materials in the modern industry.

## ACKNOWLEDGEMENT

## REFERENCES

[1]  Adams, J.D., David, L.E., Dionne G.F., Schloemann E.F., and Stizer S., 2002, "Ferrite devices and materials", IEEE Trans. Microw. Theory Technol., 50, pp. 721–737.

[2]  Horwath, M.P., 2000, "Microwave applications of soft ferrites", J. Magn. Magn. Mater., 215, pp. 171–183.

[3]  Willard, M.A., Nakamura, Y., Laughlin, E.D., and Mc Henry, M., 1999, "Magnetic properties of ordered and disordered spinel phase ferrimagnets", J. Am. Ceram. Soc., 82, pp. 3342–3346.

[4]  Kotnala, R.K., Shah, J., Mathpal, M.C., Gupta, D., Purohit, L.P., and Kishan, H., 2009, "Role of modified active surface sites of magnesium ferrite for humidity sensing", J. Optoelect. Adv. Mater., 11, pp. 296–301.

[5] Busca, G., Finocchio, E., Lorenzelli, V., Trombetta, M., and Rossini, S.A., 1996, "IR study of alkene allylic activation on magnesium ferrite and alumina catalysts", J. Chem. Soc., 92, pp. 4687–4693.

[6] Gadkari, A.B., Shinde, T.J., and Vasambekar P.N., 2010, "Structural and magnetic properties of nanocrystalline Mg–Cd ferrites prepared by oxalate coprecipitation method", J. Mater. Sci. – Mater. Electron., 20, pp. 96–103.

[7] Patil, K.C., Aruna, S.T., and Mimani, T., 2002, "Combustion synthesis: an update", Curr. Opin. Solid State Chem., 6, pp. 507–512.

[8] Randhawa, B.S., Dosanjh, H.S., and Kaur M., 2009, "Preparation of spinel ferrites by citrate precursor route – a comparative study", Ceram. Int., 35, pp. 1045–1049.

[9] Singh, J., Kaur, H., Kaur, M., and Randhawa, B.S., 2010, "Preparation of copper ferrites from thermolysis of copper tris(malonato)ferrate", J. Appl. Phys., 107, pp. 520-522.

[10] Yang, J.M., and Yen, F.S., 2008, "Evolution of intermediate phases in the synthesis of zinc ferrite nanopowders prepared by tartarate precursor method", J. Alloys Compd., 450, pp. 387–394.

[11] Yahya, N., Arpin, M.N., Aziz, A.A., Daud, H., Zaid, H.M., PAL, L.K., and Mauf N., 2008, "Synthesis and characterization of magnesium zinc ferrite as electromagnetic source," Am. J. Eng. Appl. Sci., 1, pp. 53–58.

[12] Bakshi, M.S., 2009, "A simple method of superlattice formation step by step evaluation of crystal growth of gold nanoparticles through seed growth method," Langmuir, 25, pp. 12697–12705.