Study of Dielectric Properties of Nanocrystalline Zinc Dopped Magnesium Ferrite up to Microwave Frequencies

***Jyoti**

Assistant Professor

Department of Physics, Government College Bundi, Bundi, Rajasthan, PIN- 323001

Abstract. Zinc doped magnesium ferrites with the chemical composition $Zn_xMg_{1-x}Fe_2O_4$, where x = 0.2, 0.4 have been prepared by sol-gel auto combustion technique. X-ray diffraction (XRD) was used to confirm the formation of spinel phase. Room temperature dielectric constant measurements at lower frequencies (100 Hz to 120 MHz), show usual dielectric dispersion. Variation of AC conductivity as a function of frequency is studied and the results are explained by using Maxwell–Wagner two- layer model. The effect of frequency on dielectric constant (ε'), dielectric loss tangent (tan δ) and conductivity (σ_{AC}) has been discussed in terms of hopping of charge carriers between Fe^{2+} and Fe^{3+} ions. There is a strong co-relation between the conduction mechanism and the dielectric behaviour of ferrites. The dielectric constant at high frequencies is also measured using vector network analyzer (VNA) over a frequency range from 3.95 GHz to 5.85 GHz.

Keywords: Spinel, Nanocrystalline ferrites, conduction mechanism, dipolar polarization.

Introduction

Spinel ferrites have great importance in technological field because of their good magnetic properties like high permeability, high values of anisotropy and coercivity, as well as their extensive electrical properties like high resistivity, low eddy current losses and low dielectric losses [1, 2]. Ferrites containing Mg and Zn have higher resistivity which is more effective at high frequencies [3]. There are several synthesis techniques for ferrites like wet chemical co-precipitation, microwave refluxing, ceramic method, sol-gel auto combustion etc. [4]. In the present work the $Zn_1Mg_{1-x}Fe_2O_4$ (x = 0.2, 0.4) nanocrystalline ferrites are prepared by sol-gel auto combustion method. Study of their structural characterization and the dielectric properties upto microwave frequencies are main discussion of this paper.

Experimental

Synthesis Technique. $Zn_xMg_{1-x}Fe_2O_4$ (x=0.2, 0.4) nanocrystalline ferrites are synthesized by sol-gel auto combustion method. The analytical grade of metal nitrates $Zn(NO_3)_2.6H_2O$, $Mg(NO_3)_2.6H_2O$, $Fe(NO_3)_3.9H_2O$ and $C_6H_8O_7$ (citric acid) in 1:1 molar ratio are first dissolved in distilled water. Liquid ammonia is used to maintain the pH of the solution at about 7 by continuous stirring then the mixed solution is poured into a dish and heated at 70˚ C with constant stirring to transform it into a viscous gel. The resulting gel was heated upto 150˚ C to initiate combustion. As a result the dried gel burnt out completely to form loose powder sample.

Characterization Studies. For identification of phase, the samples were characterized by powder X-ray diffraction (XRD) technique using panalytical make X'Pert PRO MPD PW 3040 diffractometer using CuK_α (1.5413 Å) radiation in 2θ range of 25˚ to 70˚. This confirms the formation of single phase spinel ferrites. The average grain size (D) is determined from line broadening of (311) reflection peak using Debye Sherrer formula

$$D = 0.9\lambda/\beta\cos\theta \qquad (1)$$

Where θ is the Bragg angle for the highly intense peak (311) and β is full width at half maxima of that peak.

Dielectric Studies. Dielectric measurements at lower frequencies were performed on precision impedence analyzer over a frequency range from 100 Hz to 120 MHz. The dielectric constant (ε') was calculated using the formula (2)

$$\varepsilon' = Cd/\varepsilon_0 A \qquad (2)$$

Here 'C' is the capacitance in farad,'d' is the thickness of the pellet in meters, 'A' is the cross-sectional area of the flat surface of the pellet and ε_o is the permittivity of free space.
The high frequency dielectric measurements were carried out using vector network analyzer (VNA) over a frequency range 3.95 GHz to 5.85 GHz.

Results and Discussion

XRD analysis. The X-ray diffraction patterns of the samples $Zn_xMg_{1-x}Fe_2O_4$ (x = 0.2, 0.4) are shown in figure 1. The peaks in the x-ray spectrum confirm the single phase cubic spinel structure with no impurity phase. The values of lattice constants obtained from XRD data for the samples $Zn_{0.2}Mg_{0.8}Fe_2O_4$ and $Zn_{0.4}Mg_{0.6}Fe_2O_4$ are 8.4050 Å and 8.4239 Å respectively. The crystallite size of samples $Zn_{0.2}Mg_{0.8}Fe_2O_4$ and $Zn_{0.4}Mg_{0.6}Fe_2O_4$ are 28.137 nm and 30.156 nm respectively which are calculated by Scherrer equation.

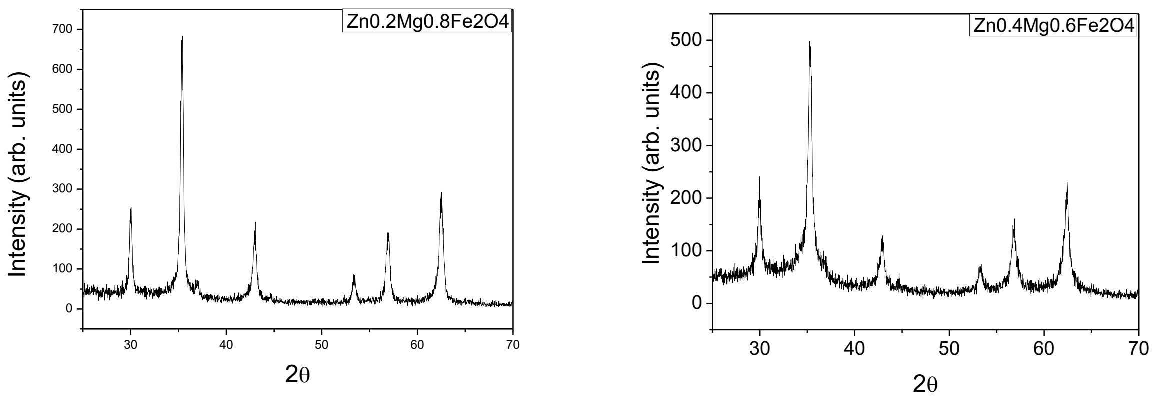
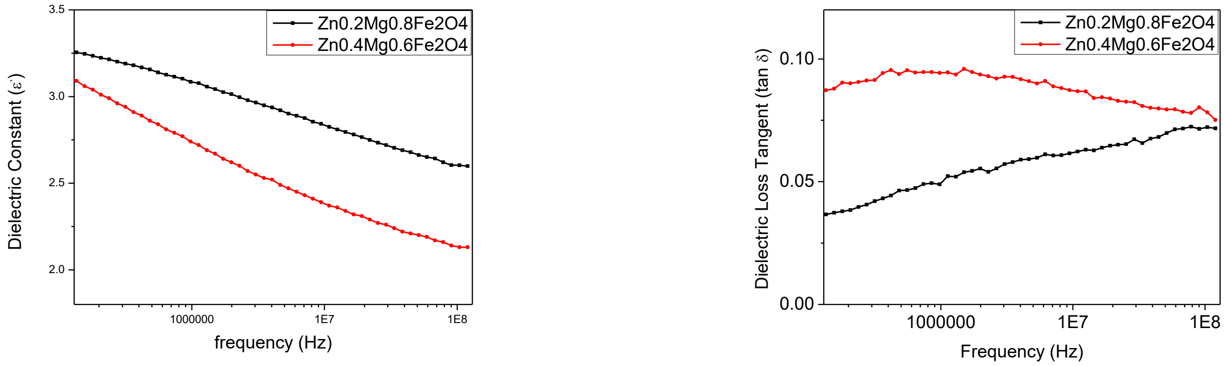


Fig. 1: *X-ray diffraction of nanocrystalline $Zn_xMg_{1-x}Fe_2O_4$ (x =0.2, 0.4) ferrite samples.*

Low frequency dielectric behaviour. The variation of dielectric constant (ε′) and dielectric loss tangent (tan δ) as a function of frequency at room temperature from 100 Hz to 120 MHz is represented by Figure 2(a) and (b) respectively. It is observed from figure 2(a) that dielectric constant decreases with increasing frequency exhibiting normal dielectric behavior. Dielectric dispersion is more at lower frequencies. The variation in dielectric constant decreases as frequency increases because after a certain frequency the electronic exchange between ferrous and ferric ions $Fe^{2+} \leftrightarrow Fe^{3+}$ cannot follow the external applied field. As Zn ions increases the dielectric constant decreases. This conduction like behavior can be explained on the basis of the mechanism of polarization in ferrites [1,5]. The addition of Zn ions reduces the Fe^{2+} ions on B-sites, which are easily polarizable ions. Therefore as Zn ions increases the polarization decreases, so the dielectric constant ε′ also decreases [6].
Figure 2(b) shows the variation of tan δ with frequency. A qualitative explanation can be given for the loss tangent tan δ. The conduction is considered as due to the hopping of electrons between Fe^{2+} and Fe^{+3} over the octahedral site. When this hopping frequency is nearly equal to that of the externally applied field frequency, a maximum of loss tangent may be observed.



(a) (b)

Fig. 2 : *Variation of (a) dielectric constant (ε') and (b)dielectric loss tangent (tan δ) with frequency at room temperature.*

AC conductivity. The AC conductivity of the ferrite samples was calculated by using the following relation,

$$\sigma_{AC} = 2\pi f \varepsilon_0 \varepsilon' \tan\delta \quad (3)$$

where, f is the frequency of the applied field, ε_0 is the absolute permittivity of air, ε' is the relative dielectric permittivity of the samples and tanδ is the loss factor.
Figure 3 represents the variation of AC conductivity as a function of frequency. Austin and Mott [7] discussed the electrical conduction mechanism in terms of the electron and polaron hopping. In the present work the conduction is due to small polarons. As the frequency of the external field increases, the conductive grains become more activate and this promotes the electron hopping between two adjacent octahedral sites (B-sites) and a transition between Fe^{2+} and Fe^{3+}, thereby increasing the hopping conduction. Therefore, a gradual increase in conductivity was observed with increasing frequency.

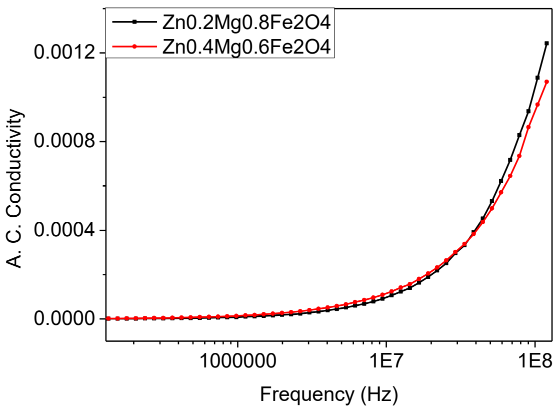
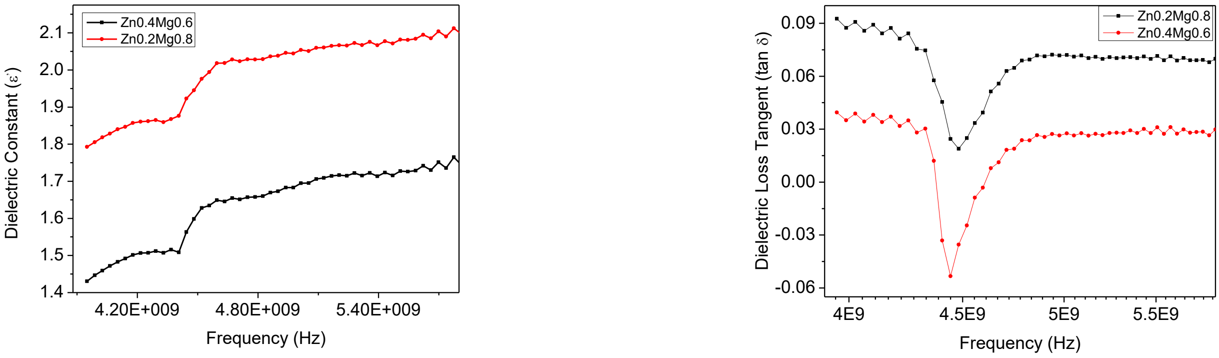


Fig. 3: *Variation of a. c. electrical conductivity of $Zn_xMg_{1-x}Fe_2O_4$ (x =0.2, 0.4) ferrite samples with frequency.*

High frequency dielectric behavior. figure 4 (a) shows the variation of dielectric constant as a function of frequency. There is a gradual increase in dielectric constant with frequency. The most probable mechanism is orientational or dipolar polarization in this frequency range. Figure 4 (b) shows the variation of dielectric loss tangent with frequency. It is observed that tanδ shows a minima for $Zn_{0.2}Mg_{0.8}Fe_2O_4$ and $Zn_{0.4}Mg_{0.6}Fe_2O_4$ ferrite at frequencies 4.48 GHz and 4.44 GHz samples respectively, which is a peculiar behaviour in ferrites.



(a) (b)

Fig. 4: *Variation of (a) dielectric constant (ε') and (b) dielectric loss tangent (tan δ) at microwave frequency at room temperature.*

Conclusions

The $Zn_{1-x}Mg_xFe_2O_4$ ferrites were synthesized by sol-gel auto combustion method. The XRD study confirms the formation of cubic spinel structure with all the characteristic reflections. The lattice constant *a* found to be increased with Zn content. Dielectric constant decreases rapidly with increasing frequency. Due to reduction of the Fe^{2+} ions on octahedral site, the addition of Zn ions increases the polarization and hence increases the dielectric constant ε'. A qualitative explanation is given for the loss tangent (tan δ). There is a strong correlation between the conduction mechanism and dielectric behavior A gradual increase is obtained in conductivity with frequency due to the more hopping conduction of electrons between Fe^{2+} and Fe^{3+}. A gradual increment in dielectric constant with frequency is observed even at microwave frequencies.

References

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