

# AC ELECTRIC TRANSPORT PROPERTIES OF Zn SUBSTITUTED Li FERRITES

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## Abstract

The electrical transport properties of  $Zn_xLi_{0.45-x/2}Fe_{2.55-x/2}O_4$  (where,  $x=0.0, 0.3, 0.5$ ) synthesized by conventional ceramic method have been investigated as a function of Zn content. The X-ray analysis confirms the formation of single phase cubic spinel structure. The ac resistivity and dielectric constant show opposite trend with increase in Zn content. The ac resistivity and dielectric constant of the samples are found to decrease with increase in frequency, exhibiting normal ferromagnetic behaviour. Dielectric constant of the samples decreases rapidly at lower frequencies and slower at higher frequencies which may be due to the Maxwell-Wagner interfacial polarization. The electrical properties can be explained on the basis of  $Fe^{2+}/Fe^{3+}$  ionic concentration.

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**Keywords:** XRD, Microstructure, Dielectric constant, AC resistivity

## Introduction

The polycrystalline ferrites have good dielectric properties that are dependent upon several factors such as the method of preparation, substitution of different cations, etc. The crystal structure, microstructure and cation distribution have a remarkable effect on ferrite properties and its applied aspect (Mazen et al., 1996; Liu et al., 1990). Several studies have been reported (Slama et al., 2010) on the effect of addition of divalent, trivalent and tetravalent ions on the magnetic properties of the lithium ferrites. Lithium and substituted lithium ferrites have been found to be very good substitutes for garnets in microwave devices due to their low cost, high resistivity and low eddy current losses. In the present investigation a systematic study of the structural, microstructure and electrical transport properties of Zn substituted Li ferrite is undertaken.

### Experimental Method:

Various composition of  $Zn_xLi_{0.45-x/2}Fe_{2.55-x/2}O_4$  was prepared by conventional ceramic method. High purity powders of  $Li_2CO_3$  (99%),  $ZnO$  (99.5%) and  $Fe_2O_3$  (99.5%) were used as raw materials. Stoichiometric amount of required powders were mixed thoroughly and then calcined at  $675^\circ C$  for 5 h in air.

The calcined powder were then pressed (8.5KPa) uniaxially into disc shaped (about 10 mm diameter, 2-3 mm thickness) and toroid shaped (about 10 mm outer diameter, 5mm inner diameter and 3 mm thickness) samples. The samples were sintered at  $1000^\circ C$  for 5h in air. The temperature ramps were  $10^\circ C/min$  for heating and  $5^\circ C/min$  for cooling. X-ray diffraction (XRD) measurements were performed [PHILIPS-PW-3040] with  $CuK_\alpha$  radiation ( $\lambda = 1.5418 \text{ \AA}$ ). The morphology and microstructural behavior of the samples were investigated with a Scanning Electron Microscope (SEM) [Inspect F50, EFI Company, Netherlands] from the mirror polished samples. Average grain sizes (grain diameter) of the samples were determined from SEM micrographs by linear intercept technique (Mendelson, 1969). The frequency characteristics of  $Zn_xLi_{0.45-x/2}Fe_{2.55-x/2}O_4$  ferrite sample i.e. ac resistivity and ac dielectric constant were investigated using a impedance analyzer [Wayne Kerr precision impedance analyzer, model no. 6500B] at room temperature in the frequency range 1 KHz to 120 MHz.

### Results and Discussion:

The XRD patterns for the various samples of  $Zn_xLi_{0.45-x/2}Fe_{2.55-x/2}O_4$  are presented in Figure 1. The XRD patterns for these compositions confirm the formation of single phase spinel ferrite with trace of impurity phase probably hematite in sample  $x=0$ , as depicted in Figure 1 between the peak (111) and (220). Analyzing the XRD patterns, we notice that the position of the peaks comply with those of the reported values (Brockman 1950). The observed peaks (111), (220), (311), (222), (400), (422), (511) and (440) which is either odd or even confirmed the spinel structure of the sample and also in agreement with other workers (Akther and Hakim, 2010; Maria et al., 2010).

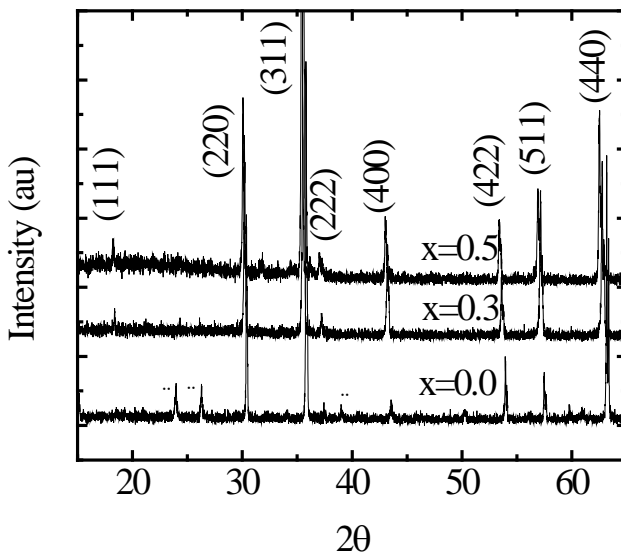


Figure 1. The X-ray diffraction pattern of Zn<sub>x</sub>Li<sub>0.45-x/2</sub>O<sub>4</sub> ferrites.

The electrical properties of soft ferrite are strongly influenced by its microstructures. The SEM micrograph for Zn<sub>x</sub>Li<sub>0.45-x/2</sub>Fe<sub>2.55-x/2</sub>O<sub>4</sub> compositions is shown in Figure 2. From analysis of micrographs it is found that the average grain size of the various samples of Zn-Li ferrite increases with increase in Zn<sup>2+</sup> content up to x = 0.3. A sudden decrease in average grain is observed beyond x = 0.3, which may be due to the fact that ions like Zn<sup>2+</sup>, Cd<sup>2+</sup> and Ti<sup>4+</sup> do not favour grain growth when present in excess quantities (Patil et al., 1991; Bellad et al., 2000).

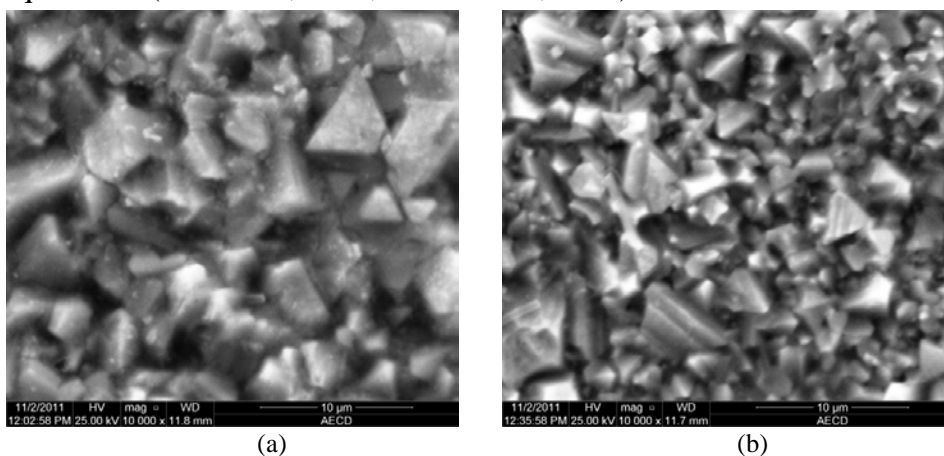


Figure 2. The SEM micrographs of various Zn<sub>x</sub>Li<sub>0.45-x/2</sub>Fe<sub>2.55-x/2</sub>O<sub>4</sub> sample (a) x=0.3 (b) x=0.5.

Figure 3 shows the variation of dielectric constant  $\epsilon'$  with frequency at room temperature for different compositions of  $Zn_xLi_{0.45-x/2}Fe_{2.55-x/2}O_4$  ferrites. It can be seen from the figure that the dielectric constant is found to decrease continuously with increasing frequency up to 1 MHz exhibiting the normal dielectric behavior of ferrites. The dielectric dispersion is rapid in the lower frequency region and remains almost independent at high frequency (1 MHz to 10 MHz). With further increase of frequency, it is observed that the value of  $\epsilon'$  increases with an increase of frequency and shows a resonance above 10 MHz. This behavior can be understood with the help of Maxwell-Wagner type interfacial polarization in accordance with Koop's phenomenological theory (Hankare et al., 2010), which is similar to that of the conduction mechanism. According to this theory, the conductivity of grain boundaries contributes more to the dielectric value at lower frequencies.

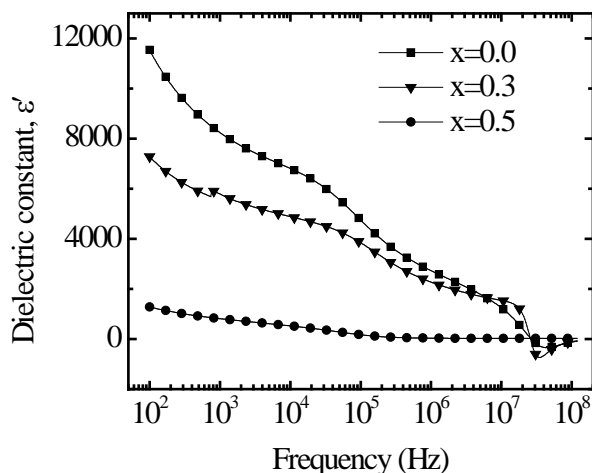


Figure 3. Variation of dielectric constant of various  $Zn_xLi_{0.45-x/2}Fe_{2.55-x/2}O_4$  with frequency.

In this model, the dielectric structure is assumed to consist of well conducting grain, which are separated by poorly conducting grain boundaries. The grains have higher values of conductivity and permittivity while the grain boundaries have lower values of these. At lower frequencies the grain boundaries are more effective than the grain controlling the electric conduction. Therefore, the permittivity is higher at lower frequencies and decreases with frequency. The electronic conduction in ferrites is mainly due to hopping of electron between ions of the same element existing in more than one valence state and distributed randomly over crystallographically equivalent lattice sites. A number of such ions formed during the sintering of ferrites.  $Fe^{2+}$  ions concentration depends upon several factors such as sintering temperature/time, environment and the grain structure. Creation of  $Fe^{2+}$  gives rise to electron hopping between Fe ions in +2 and +3 valance

states. The electronic exchange between  $\text{Fe}^{2+}$  and  $\text{Fe}^{3+}$  results in local displacement of charges in the direction of applied electric field, which is responsible for polarization in ferrites. The dielectric constant decreases with increasing frequency and then reaches a constant value due to the fact that beyond a certain frequency of external AC field, the electron exchange between  $\text{Fe}^{2+}$  and  $\text{Fe}^{3+}$  cannot follow the alternating field. The dispersion is large for  $x=0.0$ ,  $0.3$  while sample  $x=0.5$  showed a least frequency dependence. The presence of  $\text{Fe}^{2+}$  ions in excess amount favors the more dispersion. Similarly the weak dependence of dielectric constant on the frequency can be due to lack of  $\text{Fe}^{2+}/\text{Fe}^{3+}$  ions concentration (Maria et al., 2010).

The variation of dielectric loss tangent ( $\tan\delta$ ) with frequency is shown in Figure 4. The loss tangent remains almost constant at low frequency and a maximum loss tangent is observed at high frequency. A maximum can be observed when the jumping or hopping frequency of electrons between  $\text{Fe}^{2+}$  and  $\text{Fe}^{3+}$  becomes approximately equal to the frequency of the applied AC field and the phenomena is termed as ferromagnetic resonance (Rabinkin and Novikova, 1960). Similar results have been reported by others (Kumer et al., 1988) in case of Li-Ti ferrites and (Mahmud et al., 1991) in case of Ni-ferrites.

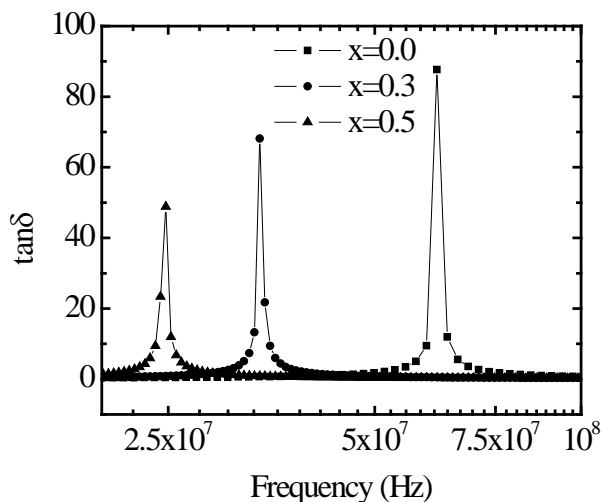


Figure 4. Variation of dielectric loss of various  $\text{Zn}_x\text{Li}_{0.45-x/2}\text{Fe}_{2.55-x/2}\text{O}_4$  ferrites with frequency.

Figure 5 shows the variation of ac resistivity with log frequency at room temperature. All the samples show a decrease in  $\rho$  with increase in frequency, which is also a normal behavior of ferrites. With increasing  $\text{Zn}^{2+}$  content, the frequency dispersion is enlarged significantly. The conduction mechanism in ferrites is explained on the basis of hopping of charge carriers

between the  $\text{Fe}^{2+}$  and  $\text{Fe}^{3+}$  ions on the octahedral site. The increase in frequency enhances the hopping frequency of charge carriers, resulting in an increase in the conduction process thereby decreasing the resistivity.

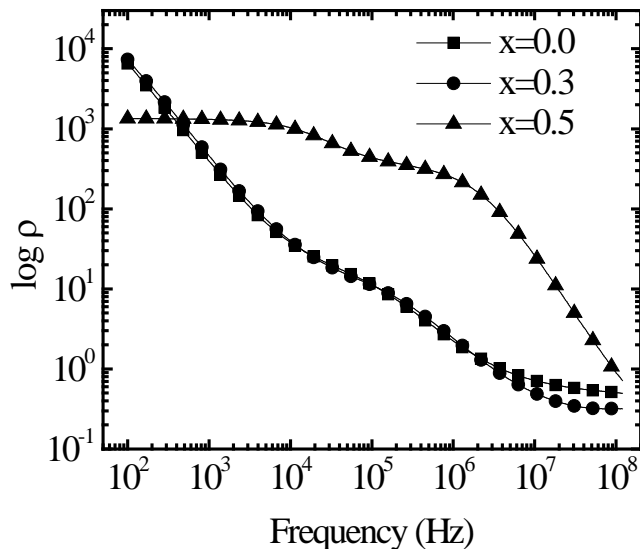


Figure 5. Variation of resistivity various of  $\text{Zn}_x\text{Li}_{0.45-x/2}\text{Fe}_{2.55-x/2}\text{O}_4$  ferrites with frequency.

Ferrites are low mobility materials and the increase in conductivity does not mean that the number of charge carriers increases, but only the mobility of charge carriers increases (Hauque et al., 2008). From the Figure 3 and Figure 5, it is also observed that the dielectric constant and resistivity behaves in an opposite manner.

### Conclusion:

The Zn substituted polycrystalline Li ferrite samples were synthesized by conventional ceramic method. The XRD analysis confirms the formation of single phase spinel ferrite except  $x = 0$ . The ac resistivity and dielectric constant of the samples are found to decrease with increase in frequency, exhibiting normal ferromagnetic behavior. The ac dielectric constant and resistivity dispersion with frequency has been explained on the basis of hopping of charge carriers between the  $\text{Fe}^{2+}$  and  $\text{Fe}^{3+}$  ions on the octahedral site.

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## References:

- Akther S. and Hakim M. A. Magnetic properties of cadmium substituted lithium ferrites. *Mater. Chem. Phys.*, 2010, Vol. 120, pp. 399.
- Bellad S. S., Watawe S. C., Shaikh A. M. and Chougule B. K. Cadmium substituted high permeability lithim ferrite. *Bull. Mater. Sci.*, 2000, Vol. 23, No. 2, pp. 83.
- Brockman F. G., Dowling P. H. and Steneck W. G. Dimensional Effects Resulting from a High Dielectric Constant Found in a Ferromagnetic Ferrite. *Phys. Rev.*, 1950, Vol. 77, pp. 85.
- Hankare P. P., Patil R. P., Sankpal U. B., Garadkar K. M., Sasikala R., Tripathi A. K. and Mulla L. S. Magnetic dielectric and complex impedance spectroscopic studies of nano crystalline Cr substituted Li-ferrite. *J. Magn. Magn. Mater.*, 2010, Vol. 322, pp. 2629.
- Haque M. M., Huq M. and Hakim M. A. Thermal hysteresis of permeability and transport properties of Mn substituted Mg-Cu-Zn ferrites. *J. Phys. D: Appl. Phys.*, 2008, Vol. 41, pp. 055007.
- Kumar B. K., Srivastava G. P. and Kishen P. in: *Proceedings of ICF5, Bombay, 1988*, pp. 233.
- Liu C. S., Shu W. B., Tung M. J. and Ke M. Y. Effect of partial substitution of  $\text{Ge}^{4+}$  for  $\text{Zn}^{2+}$  on the microstructure and temperature stability of Mn-Zn ferrites. *J. Appl. Phys.*, 1990, Vol. 67, pp. 5506.
- Mahmud M. A., El Nimar M. K., Tawfik A. A. and El Hasab M. Dielectric behaviour in Ni-Al ferrites at low frequencies. *J. Magn. Magn. Mater.*, 1991, Vol. 98, pp. 33.
- Maria K. H., Choudhury S. and Hakim M. A. Complex permeability and transport properties of Zn substituted Cu ferrites. *J. Bang. Acad. Sci.*, 2010, Vol. 34, No. 1, pp. 1.
- Mazen S. A., Eltalaky A., Mohamed A. Z. and Hashem H. A. Effect of  $\text{Ti}^{4+}$  substitution in copper ferrite: an analogous study. *Mater. Chem. phys.*, 1996, Vol. 44, pp. 293.
- Mendelson M.I. Average grain size in polycrystalline ceramics. *J. Am. Ceram. Soc.*, 1969, Vol. 52, No. 8, pp. 443.
- Patil R. S., Kakatjar V. S., Patil S. A., Sankpla A. M. and Sawant S. R. X-ray and bulk magnetic studies on  $\text{Li}_{0.50}\text{Zn}_x\text{Ti}_x\text{Fe}_{2.5-2x}\text{O}_4$ . *Mater. Chem. Phys.*, 1991, Vol. 28, No. 4. pp. 355.
- Rabinkin L. I. and Novikova Z. I. *Ferrites*. Minsk: Izv, Acad. Nauk USSR, 1960, 146.
- Slama J., Gruskova A., Soka M., Usakova M. and Jancarik V. Analysis of Magnetic Properties of Substituted Li Ferrites. *IEEE Trans. Mag.*, 2010, Vol. 46, No. 2. pp. 455.