

Structural and Electrical Characterization of Li-Zn Ferrites

M. Shahjahan, N. A. Ahmed, S. N. Rahman, S. Islam, N. Khatun, M. S. Hossain

Abstract — Four ferrite samples of $Li_{0.5x}Fe_{0.5x}Zn_{1-x}Fe_2O_4$, where $x=0.6, 0.7, 0.8, 0.9$ were prepared by conventional ceramic method. The dc electrical resistivity as a function of temperature has been studied and found to decrease with the increase in temperature. The Curie temperature (T_c) has been found to increase as the Zn content decreases from $x=0.6$ to $x=0.8$. But for the sample where $x=0.9$ the Curie temperature is less than that of the sample where $x=0.8$. The ac electrical conductivity (σ_{ac}), dielectric constant (ϵ), the dielectric loss tangent ($\tan\delta$) and quality control factor (Q -factor) as a function of frequency have also been studied. The experimental results indicate that for the first three samples the ac electrical conductivity (σ_{ac}) decreases with the increase in frequency up to 200 KHz and afterwards it increases with the increase in frequency. But for the sample where $x=0.9$ the ac conductivity (σ_{ac}) increases with the increase in frequency. It has been found that with the increase in frequency the dielectric constant (ϵ) and dielectric loss tangent ($\tan\delta$) decrease while the quality factor (Q -factor) and ac conductivity increases.

Keywords: Conventional Ceramic, Electrical Properties, Lithium Zinc, Surface Morphology.

I. INTRODUCTION

Li-Zn ferrites are very attractive for high frequency applications due to their excellent electrical and magnetic properties. Since their properties can be engineered during synthesis and processing steps, ferrite materials appear to have great technological potential.

Ferrites have a high electrical resistivity, small eddy current losses in them at high frequency and can therefore be used at high frequencies. Extensive research works have been made on ferrites by different researchers and found that the dielectric properties change with temperature and frequency of the applied field [1]. The study of ac conductivity at different temperatures and frequencies gives valuable information on conduction in materials based on localized electric charge carriers [2]. Li-Zn ferrites are promising for microwave devices [3] had synthesized Cu-Zn-Li ferrites using oxalate precursors with the intention to increase the Curie temperature (T_c) [4] has observed that with increase of Li^{1+} concentration the values of Curie temperature increase, whereas initial permeability decreases. In this communication, they reported their studies on permeability variation with respect to changes in composition, temperature and frequency for Li^{1+} substituted

Cu-Zn ferrites [5] prepared the high frequency Li-Zn ferrite by self-combination method with the rare earth substitutions. The oxides such as CaO, Na_2O , Sb_2O_3 , and ZrO_2 have some influence on the physical properties (grain growth, density, hardness) and on the crystallographic (lattice constant) or on the magnetic properties (saturation magnetization, initial permeability, curie temperature) of the Li-Zn ferrite. Six compounds with formula $Li_{0.3}Zn_{0.4}Fe_{2.26}R_{0.04}O_4$ where $R \equiv Yb, Er, Dy, Tb, Gd$ and Sm was studied. The results obtained reveal that by introducing a relatively small amount of R_2O_3 instead of Fe_2O_3 , important modifications of properties can be obtained. For example, R_2O_3 shifts the Curie point of Li-Zn ferrite to lower temperature and increases the electrical resistivity with over one order of magnitude. In 2002 Phanjouban and Prakash [6] have studied on the structural properties and dc conduction behavior of Li-Zn-Ti ferrites. The samples having the compositional formula $Li_{0.45+0.5t}Zn_{0.1}Ti_tFe_{2.45-1.5t}O_4$ with t varying from 0 to 1.2 in steps of 0.2 were prepared by the conventional dry ceramic method. XRD analysis showed all the samples to be single phase spinel and the lattice parameter was found to increase with increasing t . dc resistivity increases with Ti concentration. The normal dielectric behavior, such as decrease in dielectric constant with increasing frequency was observed by Reddy and Rao [7]. The present study has been undertaken to synthesize Li-Zn ferrites and study on their intrinsic properties (such as dc electrical resistivity, Curie temperature, ac electrical conductivity, dielectric constant, dielectric loss tangent and quality control factor as a function of frequency) which can be tailored to the requirement in electronic industries and information and communication technology.

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II. MATERIALS AND METHODS

In the Present study Li-Zn ferrites of the series, $\text{Li}_{0.5x}\text{Fe}_{0.5x}\text{Zn}_{1-x}\text{Fe}_2\text{O}_4$, where $x=0.6, 0.7, 0.8, 0.9$ for samples 1, 2, 3 & 4 respectively have been prepared using raw oxide collected from local market by conventional ceramic method at 1200°C for 3 hours. Samples were mixed with a mortar and pestle for fine particle size of the powders and their various intrinsic properties in relation to composition and sintering temperature have been studied. In the present measurement two-probe method has been used to measure the dc electrical resistivity. Activation energy and Curie temperature have been obtained from the resistivity vs. temperature curve. The frequency dependent ac conductivity, dielectric constant, dielectric loss tangent, quality factor (Q-factor) have been obtained by using a precision LCR meter. The instrument gave automatically the values of capacitance (C_p in pF), the resistance (R_p in $\text{K}\Omega$), the dielectric loss D ($\tan\delta$) and Q-factor by changing the frequencies.

III. RESULTS AND DISCUSSIONS

The effects of Lithium (Li) substitution V_2O_5 addition to Li-Zn ferrites compared in the SEM micrographs shown in figure 1. The micro structure of Li-Zn ferrites exhibits homogeneous grain distributions. The grain size and transition temperature (T_c) increase with the decreases of the Zn content while the grain size and Transition temperature (T_c) decrease with decreasing Li content in Li-Zn ferrites.

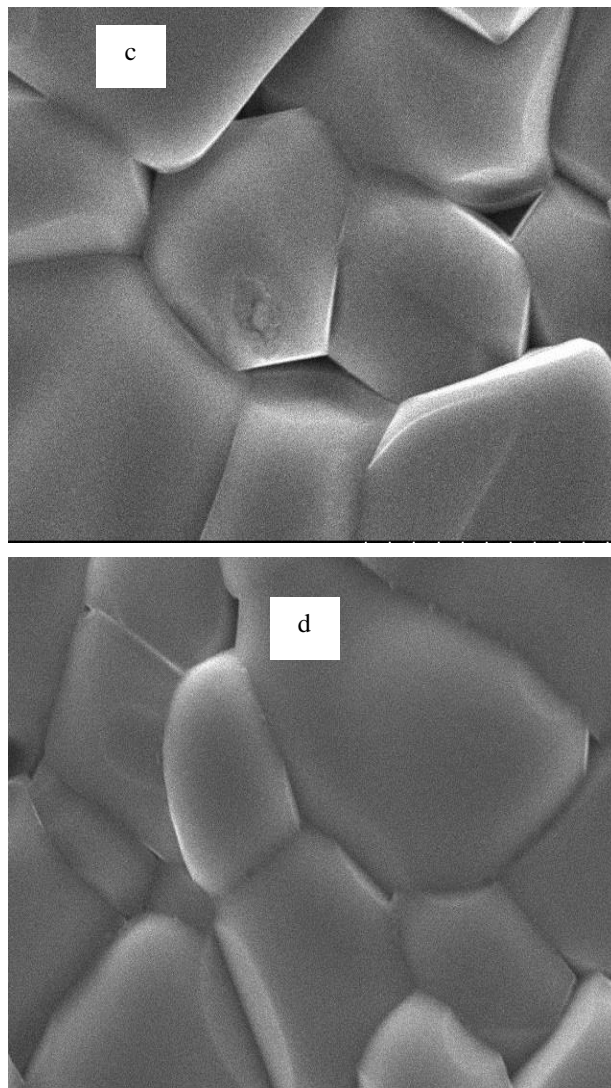
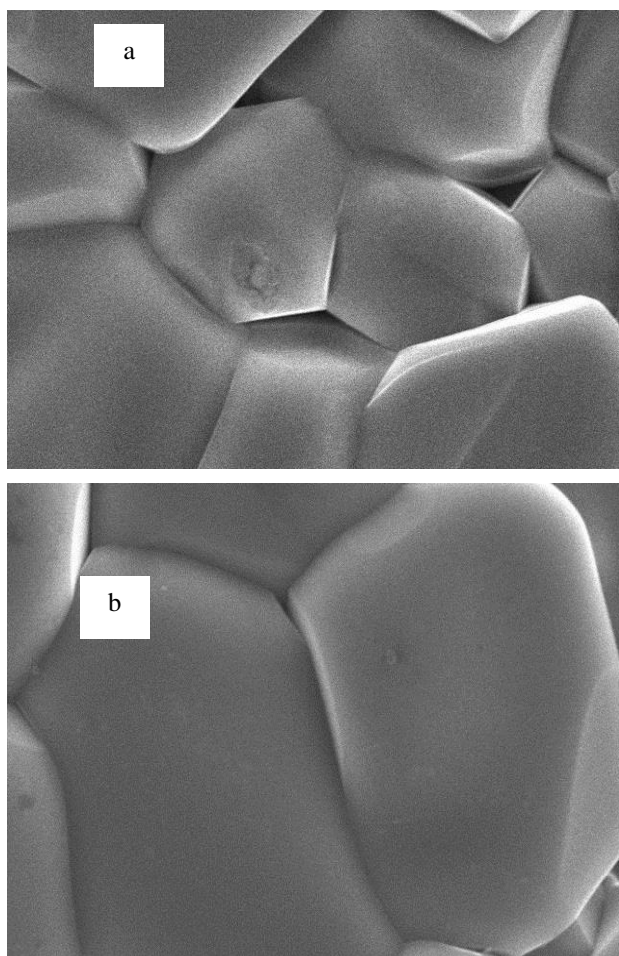


Fig.1. Scanning electron micrographs taken on the surface at $10\mu\text{m}$ scale of $\text{Li}_{0.5x}\text{Fe}_{0.5x}\text{Zn}_{1-x}\text{Fe}_2\text{O}_4$ samples sintered at 1200°C during 18 h with (a) $x=0.6$, (b) $x=0.7$, (c) $x=0.8$, (d) $x=0.9$

A. Composition Dependent dc Electrical Properties

The temperature dependence of dc electrical resistivity of four Li-Zn ferrite samples of the system $\text{Li}_{0.5x}\text{Fe}_{0.5x}\text{Zn}_{1-x}\text{Fe}_2\text{O}_4$ have been studied in the present research work and is shown in figures 2 to 5 respectively. From figures 2 to 5 it is observed that the dc resistivity of four samples increases slowly with the increase in temperature up to Curie temperature T_c and then decreases sharply beyond Curie temperature. It may be according to the chemical formula of our investigated series, $\text{Li}_{0.5x}\text{Fe}_{0.5x}\text{Zn}_{1-x}\text{Fe}_2\text{O}_4$, as Zn concentration increases, the Fe^{2+} concentration decreases for their occupation on octahedral sites. Thus the chance of formation of Fe^{2+} ions minimizes and the probability of hopping between Fe^{2+} and Fe^{3+} increases and the resistivity increases, which demands further study. This result is in good agreement with Sattar [14]. It is found that Curie temperatures for four samples are 270°C , 310°C , 395°C and 385°C respectively.

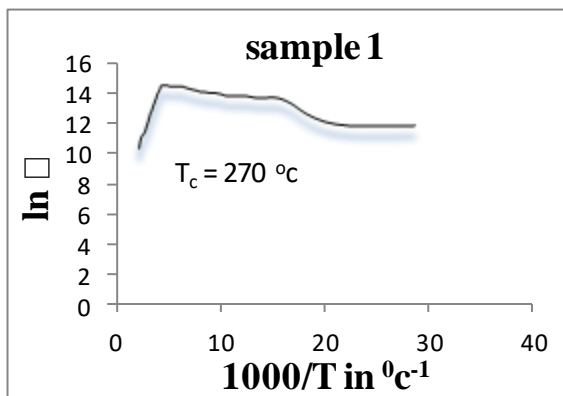


Fig. 2. Resistivity (lnρ) Vs. Temperature (1000/T)

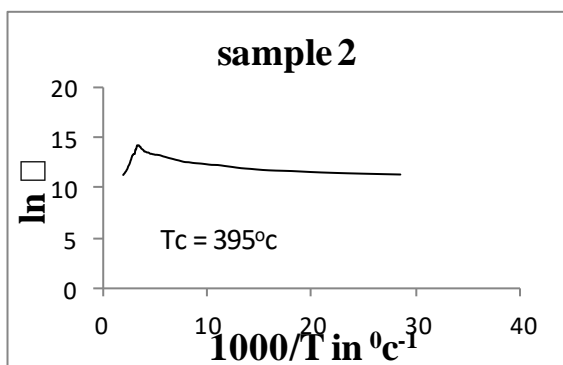


Fig.3. Resistivity (lnρ) Vs. Temperature (1000/T)

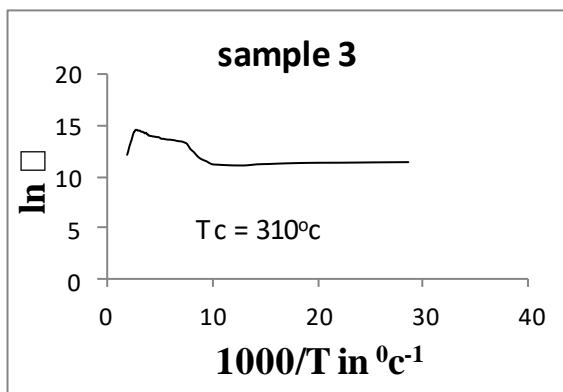


Fig. 4. Resistivity (lnρ) Vs. Temperature (1000/T)

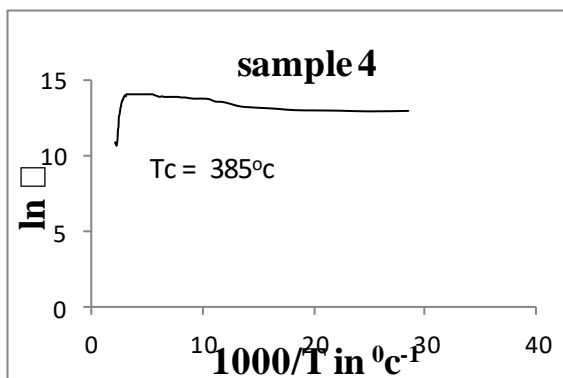


Fig. 5. Resistivity (lnρ) Vs. Temperature (1000/T)

It is also observed that the Curie temperatures and the activation energies increased for increased in Li concentrations from $x=0.6$ to 0.8 . But for Li concentration, $x=0.9$ Curie temperature and activation energy decreases slightly. It reveals that resistivity increases with the increase in zinc content. E. Rezlescu and Rezlescu [9] introduced a considerable change in chemical composition and hence corresponding change in properties.

Table-I: Curie temperature and Activation energy.

Sample No. & Chemical formula	Curie Temperature in °C	Resistivity, ρ in $\Omega\cdot m$ at room temp.	Resistivity, ρ in $\Omega\cdot m$ at T_c	Activation Energy, E_p in eV at T_c
Sample-1 ($x=0.6$)	270	1.548×10^3	8.76×10^3	0.0536
Sample-2 ($x=0.7$)	310	0.922×10^3	10.476×10^3	0.0609
Sample-3 ($x=0.8$)	395	0.911×10^3	10.212×10^3	0.0697
Sample-4 ($x=0.9$)	385	4.24×10^3	7.505×10^3	0.059

Due to evaporation of Zn^{2+} ions it leaves behind the voids in the lattice during the quenching period from very high temperature. They also observed that the diffusion coefficient is not sufficient to fill these pores. Therefore evaporation of zinc may be the interpretation of increasing electrical resistivity at higher sintering temperature. In present case zinc concentration also shifts the Curie point of Li-Zn ferrite to higher temperature. For samples where $x=0.6, 0.7,$ and 0.8 the Curie temperature increases with decrease in zinc concentration. But in the case of sample 4, the Curie temperature is $385^\circ C$ and activation energy is $0.0592 eV$ which shows lower values than that of the sample-3. This may be due to the influence of Li^{1+} as an effect of their ionic radius. The results obtained reveal that by changing lithium concentration, important modifications of properties can be obtained which demands further investigation. The sharp decrease in resistivity above the Curie temperature (T_c) can be explained on the basis of clusters of Zn^{2+} ions [8]-[10]. These clusters effectively reduce the concentration of stable bonds of $Fe^{2+} + Zn^{2+}$ and conduction may be due to more number of zinc ions at octahedral sites. The values of Curie temperature and Activation energy are presented in table-I.

B. Frequency Dependent Electrical Properties

The room temperature values of frequency dependent ac conductivity (σ_{ac}), dielectric constant (ϵ), dielectric loss tangent ($\tan\delta$) and Q-factor of mixed Li-Zn ferrites samples are shown in figures 6 to 9 respectively. The variation of ac conductivity as a function of frequency for mixed Li-Zn ferrites with different compositions is shown in figure 6. The room temperature values of ac conductivity (σ_{ac}) of four samples with the variation of frequency show that ac conductivity is dependent on frequency.



It can be seen from the figure that the value of ac conductivity decreases up to 200KHz with increasing frequency and then increases continuously with increasing frequency in the case of samples where, $x=0.6, 0.7$ and 0.8 . But this behavior deviates in the case of the sample where, $x=0.9$. D.Ravinder and K.Vijay Kumar [11] reported citing other references that a strong correlation between the conduction mechanism and the dielectric behavior of the ferrites starting with the conjecture that the mechanism of the polarization process in ferrites is similar to that of the electronic exchange between $Fe^{2+} \rightarrow Fe^{3+}$ results in local displacements which determine the polarization behavior of ferrites. A similar explanation is proposed for the composition and frequency dependence of the ac conductivity of the ferrites under this investigation. It may be stated that ferrite samples have the maximum divalent iron ion concentration. It is pertinent to mention that the number of ferrous ions on the octahedral site that play a dominant role in the process of conduction. It may be explained on the basis of the fact that the maximum number of ferrous ions which involve in the phenomenon of exchange $Fe^{2+} \rightarrow Fe^{3+}$ giving rise to increase in ac conductivity with the variation of frequency. Explanation of the behavior of sample for $x=0.9$ demands further investigation. Perhaps this sample is not stable with this composition. Zinc concentration may give rise to such anomalies.

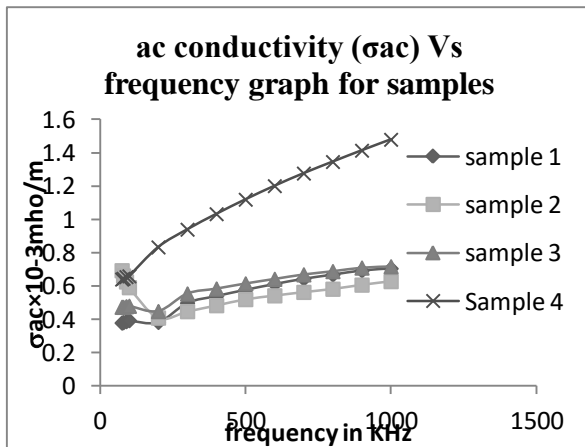


Fig.6. ac Conductivity (σ_{ac}) Vs. Frequency graph for samples.

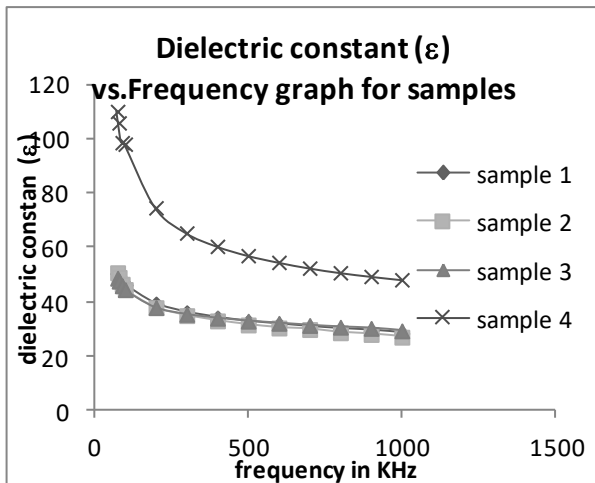


Fig.7. Dielectric constant (ϵ) Vs. Frequency graph for samples.

Dielectric constant (ϵ) Vs. frequency graphs of four samples are represented in figure 7. From the graph it is observed that the value of dielectric constant decreases continuously with increasing frequency. The dispersion of dielectric constant value for sample-4 is maximum. The decrease of dielectric constant with the increase of frequency as in the case of mixed ferrites is a normal dielectric behavior [11]. This behavior may be explained on the basis that beyond a certain frequency of the externally applied electric field, the electronic exchange between $Fe^{2+} \rightarrow Fe^{3+}$ cannot follow the varying field resulting in a decrease of dielectric constant. A similar explanation was given by Radha and Ravinder [12]. Figure 8 shows the variation of $\tan\delta$ with frequency of mixed Li-Zn ferrite samples. It can be seen from the figure that $\tan\delta$ shows a maximum value at 75KHz and minimum value at 1MHz. Dielectric loss tangent ($\tan\delta$) decreases with the increase in frequency.

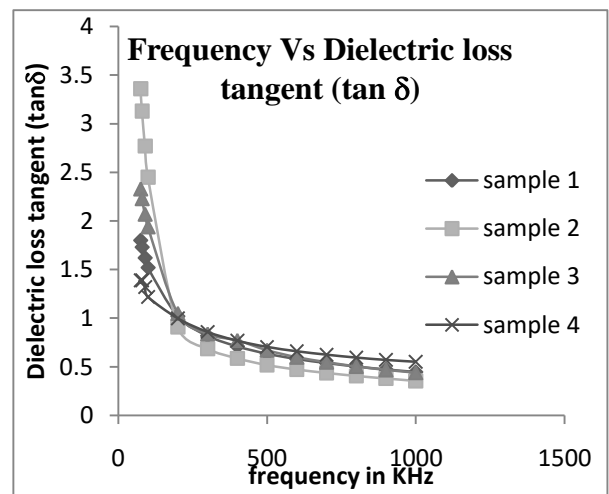


Fig.8. Dielectric Loss tangent ($\tan\delta$) Vs. Frequency graph for samples.

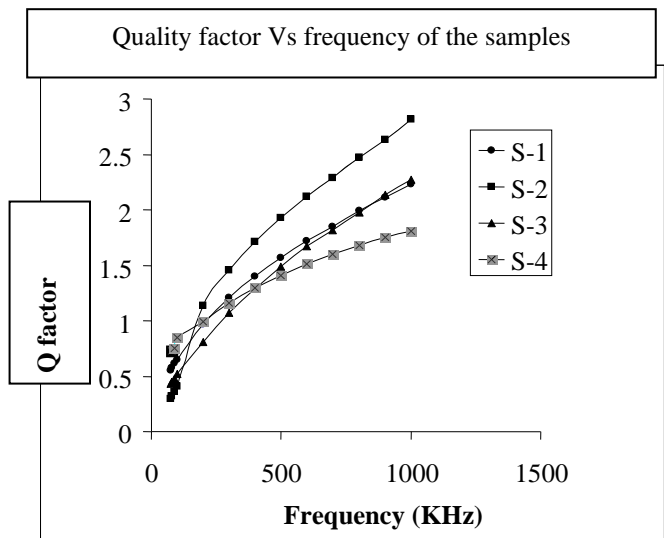


Fig. 9. Quality factor (Q-factor) Vs. Frequency for samples.

As reported by Iwauchi [13], there is a strong correlation between the conduction mechanism and the dielectric behavior of ferrites. The conduction mechanism is considered as due to hopping of electrons between Fe^{2+} and Fe^{3+} . As such when the hopping frequency is nearly equal to that of the frequency of externally applied electric field, a maximum of loss tangent observed. In case of the samples under investigation the hopping frequencies are of the appropriate magnitude to observe a loss of maximum at 75KHz. Q-factor Vs frequency graphs of four samples are represented in figure 8. From the graph it is observed that the Q-factor increases with the increase in frequency. Since Q-factor is a ratio of permeability to resistivity, this value gives an idea of good quality of ferrite samples. The ferrites having high Q-factor are very useful. In the case of the samples under investigation Q-factor is being increased with the rise in frequency. So these samples may be useful at microwave frequencies.

IV. CONCLUSIONS

The intrinsic properties of Li-Zn ferrite system is strongly dependent on chemical compositions, sintering temperatures and site locations of various cations. The dc resistivity of all four samples we investigated increases slowly with the increase in temperature up to Curie temperature, T_c and then decreases sharply beyond Curie temperature. Dielectric loss tangent and dielectric constant decrease rapidly with increasing frequency and reached almost at a constant value for higher frequency showing that they are independent of higher frequency. The ac conductivity and Q-factor were found to be higher for higher frequency. Comparing the electrical properties of four compositions, we can say that mixed $\text{Li}_{0.6}\text{Zn}_{0.4}\text{Fe}_2\text{O}_4$ ferrite shows highest Q-factor and therefore more elaborate investigation should be needed with this composition. In order to understand the exact nature, magnetization behavior of cations and their distribution in Li-Zn ferrite should need more investigation for better understanding of the quality of the synthesized materials.

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