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International Journal of Current Research Vol. 9, Issue, 10, pp.59176-59179, October, 2017

## INTERNATIONAL JOURNAL OF CURRENT RESEARCH

# **RESEARCH ARTICLE**

# SYNTHESIS AND AC PROPERTIES OF MIXED LI-ZN FERRITES

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## **ARTICLE INFO**

### ABSTRACT

*Article History:* Received 28<sup>th</sup> July, 2017 Received in revised form 15<sup>th</sup> August, 2017 Accepted 25<sup>th</sup> September, 2017 Published online 31<sup>st</sup> October, 2017

#### Key words:

Li-Zn spinel ferrite, Dielectric properties, AC electrical conductivity. The mixed polycrystalline ferrites  $Li_{0.5(1-x)}Zn_xFe_{2.5-0.5x}O_4$  were prepared by the conventional solid state method using high purity metal oxides  $ZnO_Fe_2O_3$  and  $LiCO_3$  for different concentration of  $Zn^{2+}$  ions. Double probe electrode method was used to study the AC properties: the AC electrical conductivity ( $\sigma$ ), the dielectric constant: real( $\varepsilon'$ ), imaginary ( $\varepsilon''$ ) and complex ( $\varepsilon^*$ ), and the dielectric loss tangent (tan  $\delta$ ) over therange of the applied frequency ( $f = 1 \ KHz - 5 \ MHz$ ) at room temperature.  $\sigma$  increased exponentially with the increasing f where the maximum value was for the sample  $Li_{0.3}Zn_{0.4}Fe_{2.3}O_4$ . The obtained results of the dielectric parameter ( $\varepsilon', \varepsilon'', \varepsilon^*$  and tan  $\delta$ ), also, decreased exponentially with increasing f, which confirms the normal spinel ferrite behavior. The behavior of AC properties can be elucidated on the basis of the exchanged electrons between  $Fe^{3+} + e^- \Leftrightarrow Fe^{2+}$ . The obtained results reveal that the AC properties change by substitution of the  $Zn^{2+}$  ions in Li-spinel ferrite matrix.

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Citation: Hussein Dawoud, Zana Abu Mosa and Samy Shaat, 2017. "Synthesis and ac properties of mixed li-zn ferrites", *International Journal of Current Research*, 9, (10), 59176-59179.

# **INTRODUCTION**

The significant electromagnetic of ferrites enable them to have wide variety of technological applications, aiming at a better material with excellent chemical stability, law eddy current, high permeability, high electrical resistivity, microwave components, biomedical potential applications in magnetic resonance imaging. The exploration of the electric properties of ferrite materialsoffers valuable information about the behavior of localized electric charge carriers and can understand the mechanism of electrical conduction and dielectric polarization. The properties of spinel ferrites with the chemical formula  $D_1^{2+}T_2^{3+}O_4^{2-}$  has a cubic structure packed of oxygen ( $O^{2-}$ ) ions (Shaath, 2012; Dawoudet al., 2016; Dawoud et al., 2017; Dawond, 1997; Dawoud and Shaat, 2006; Mazen and Dawoud, 1999), have tetrahedral  $(T_d)$  and octahedral  $(O_h)$  sites. These twositesthat occupies by metal ions depending on their radii and the valancey to form the cation distribution. It is worthy, to mention that the cations distribution among  $T_d$  and  $O_h$  sites plays an important role in controlling the electromagnetic and dielectric properties of spinel ferrite. Among of ferrite materials, Li-Zn spinel ferrite is, usually, used as magnetic material for different technological applications. So, Li-Znferrites have been developed for electronic applications. Even though *Li-Zn* ferritesystem exhibit excellent properties and can

be used for a variety of applications. In spite of, poordetailed studied has been reported in the literature on dielectric and AC conductivity properties of Li-Zn ferrite. Therefore, it was interesting in the present article to investigate the dielectric and AC conductivity properties of Li ferrite when added Zn content.

#### **Experimental Techniques**

The conventional solid state method was used to synthesize 25 grams of  $Li_{0.5(1-x)}Zn_xFe_{2.5-0.5x}O_4$  ferrites, where x = 0.0, 0.2,0.4, 0.6, 0.8 and 1.0, using high pure metal oxides  $ZnO_{1}Fe_{2}O_{3}$ and LiCO<sub>3</sub>. A stoichiometric quantities of metal oxides were grounded for 5 hr's, then was pre-sintered at750 °Cfor 3 hr'sin a crucible using a laboratory Furnace (BIFATHERM model AC62). After that, the prefired powder was well grounded for 3 hr's. The grounded powder was pressed under constant pressure of  $3 \times 10^8$  pa, by adding a small quantity of butyl alcohol as a binding material. The pressed powderwas formed into a disc shape with a radius 0.495 cm and thickness (0.4-0.7) cm. Then, all pressed discs were sintered at 1150 °C for 5hr's. After sintering process, the pressed discswere cooled down, gradually, to room temperature. Finally, the pressed discswere polished to obtain uniform parallel surfaces to study the AC and the dielectric properties. Double probe electrode method was used to examine the AC electrical conductivity, dielectric parameters over а variable range of frequency 1 KHz - 5 MHzat room temperature. The AC electrical conductivity ( $\sigma$ ) was calculated by the relation (Shaath, 2012).

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 $\sigma = t / aZ \tag{2}$ 

where *Z* the impedance of the sample, *t* is the thickness, and *a* is the cross-sectional area of a flat surface of the pressed disc.

The permittivity (dielectric constant) of the material such as the imaginary ( $\varepsilon$ "), the real ( $\varepsilon$ '), the complex ( $\varepsilon$ \*) can be calculated from (Abdul Khader *et al.*, 2016)

$$\varepsilon'' = \sigma / 2\pi f \varepsilon_o \tag{3}$$

where f is the applied frequency and  $\varepsilon_o$  is the permittivity of free space.

$$\varepsilon' = C/C_o \tag{4}$$

where C and  $C_o$  are the capacitance of the filled and unfilled of the pressed disc.

$$\varepsilon^* = \varepsilon - j\varepsilon'' \tag{5}$$

The dielectric loss tangent (tan  $\delta$ ) can be calculated by the following formula (Shaath, 2012)

$$\tan \delta = \varepsilon'' / \varepsilon' \tag{6}$$

# **RESULTS AND DISCUSSION**

#### AC conductivity

Fig. (1) shows the variation of  $\sigma$  over the range 1 KHz -5 MHz for the pressed discsamples. The points in this Fig. indicates to the calculated  $\sigma$ , where thered line is the fitted curve for the presented points. It is noticed that, o increases exponentially with increasing of f. All samples exhibit normal behavior with the variation of f. This is because of the applied force driven by f, which helps in transferring the charge carriers between the different conduction states. In line with, it may be attributed to the electron hopping or the electron exchange, i.e.  $Fe^{3+} + e^- \Leftrightarrow Fe^{2+}$  which occurs by the electron transform between the adjacentO<sub>h</sub> sites in the spinel lattice (Shaath, 2012). Based on the fitted curve, it can be deduced a mathematical expression for  $\sigma$ .

$$\sigma = B + Af^{S} \tag{7}$$

where *B* is y-axis offset, *A* has electrical conductivity unit and *S* is the universal exponential factor. Eq. (7) is similar to the equationthat is introduced in Refs. (Zaki*et al.*, 2013; Batoo and Ansari, 2012; Pervaiza and Gula, 2012).

$$\sigma = A f^{S} \tag{8}$$

The only difference between Equations (7) and (8) is the constant *B*. As a result, we suggest that Eq. (7) may be given a good description for the behavior of the  $\sigma$ in such material. The values of the fitted parameters *B*, *A* and *S* for all samples are seen in the Table (1).

However,  $\sigma$  is increased for the *Li-Zn* pinel ferrite is explained based on Verwey mechanism (Shaath, 2012). That is, the electron hopping may be occurred between the ions of the same element that present in more than one valence state and

distributed randomly over crystallographically inequivalent lattice sites (Dawoud *et al.*, 2010).





Table 1. Thevalues of *B*, *A* and *S* derived from the fitted curve for the Fig. 1. The ratio of the increment in the  $\sigma$  for each sample

Х	В	А	S	%
0.0	6.9523	0.02515	0.9571	93.27
0.2	5.6522	0.04290	0.9425	95.92
0.4	41.641	0.28868	0.7672	83.57
0.6	5.3923	0.00190	1.1628	87.78
0.8	15.001	0.32180	0.6964	88.18
1.0	0.5772	0.01259	1.0024	99.25

Depending upon the sintering conditions, the number of such ions may be produced during the preparation of the ferrite samples. It is known that, the partial reduction of the electron hopping,  $Fe^{3+} \Leftrightarrow Fe^{2+}$ , can take place at an elevated firing temperature (Shaath, 2012). Thus; the hopping of electron,  $Fe^{3+} \Leftrightarrow Fe^{2+}$ , occurs only by electron transform between the adjacentO<sub>h</sub> sites in the spinel lattice formed in the Li-Znspinel ferrite. This causes the increasing in electric conductance (Dawoud et al., 2010). Further comparison, at the highest frequency (5 MHz) for all samples, it was found that, the sample x = 0.4 (Fig. (2)) showed a rapidly increasing of  $\sigma$ which is equal to  $243(\Omega .m)^{-1}$ . This indicates that the sample with x = 0.4 has the high concentration of  $Fe^{2+}$  ions in the spinelstructure lattice (Shaath, 2012). In the same time, the lowest value of  $\sigma$  was registered for sample x = 0.6 which is 42  $(\Omega,m)^{-1}$ . The ratio of the increment for each of the given samples are shown in Table 1. Such ratio may be attributed to the concentration of  $Fe^{2+}$  ions the mixed *Li-Zn* spinel ferrite. Asimilar behavior was observed in various ferrite systems by several investigators (Dawoud et al., 2010; Alwash et al., 2016). The relaxation time  $(\tau_{\sigma})$  is a characteristic time constant of ferrimagnetic materials and the angular frequency( $\omega$ ). $\tau_{\sigma}$  can be described in terms of  $\sigma$  as below (Shaath, 2012)

$$\sigma = \sigma_h + \frac{\sigma_l - \sigma_h}{1 + (\omega \tau_{\sigma})^2} \tag{9}$$

where,  $\sigma_l$  and  $\sigma_h$  are  $\sigma_a$  low and high applied frequency, respectively. The estimated values of  $\tau_{\sigma}$  at f = 1 MHz for the samples, x = 0.0, 0.2, 0.4, 0.6, 0.8 and 1.0 are, respectively,

 $2.27 \times 10^{-5}s$ ,  $1.79 \times 10^{-5}s$ ,  $2.85 \times 10^{-5}s$ ,  $4.3 \times 10^{-5}s$ ,  $2.58 \times 10^{-5}s$  and  $1.48 \times 10^{-5}s$ .



Fig. 2. Variation of owith the concentration of Zn content

#### **Dielectric Properties**

Fig. 3(a-c) depicts the variation of the dielectric constants  $\varepsilon'$ ,  $\varepsilon''$ and  $\varepsilon^*$  with f. From this Fig. $\varepsilon'$ ,  $\varepsilon''$  and  $\varepsilon^*$  for all samples decreases with increasing of f. This decrease is more rapid in the low frequency region, butultimately this decrease becomes shiftlessat higher f. This behavior is subjected to dielectric polarization under the application of AC field. The decrease of dielectric constants with an increase of f observed in the case of mixed Li-Zn ferrites is a normal dielectric behavior of spinel ferrites, which are reported by several investigators (Azadmanjiri, 2008; Yadoji, 2003; Kumar et al., 2012; Krishna et al., 2012; Chavanet al., 2013; Devmunde et al., 2016; Soibam, 2016). As shown in Fig.3 the maximum value of the dispersion of the dielectric constants is for the sample with x =0.4. Herein, the concentration of  $Fe^{2+}$  ions is expected to be higher than in other compositions of mixed Li-Zn ferrites. Consequently, it is possible for these ions to be polarized to the maximum possible extent. Further, as the f of the externally applied field increases gradually, though the number of  $Fe^{2+}$ ions is present in the ferrite material. The value of  $\varepsilon^*$  decreases from 71732 at 1KHz to 756.45 at 5MHz. The reduction occurs because beyond a certain f of the externally applied electric field, the electronic exchange between  $Fe^{2+}$  and  $Fe^{3+}$  ions, cannot follow the alternating field. The variation of the dispersion of dielectric parameters with composition for other mixed Li-Zn ferrites explained by the fact that the electron exchange between  $Fe^{2+}$  and  $Fe^{3+}$  ions in an *n*-type semiconducting ferrite cannot follow the frequency of the applied alternating field beyond a critical value of the f. Such type of dielectric dispersion in ferrite system was analogous to Maxwell-Wagner model and Koops phenomenological theory, which suggested that ferrite system consist of a combination of highly conducting grains separated by poorly conducting grain boundaries (Zaki et al., 2013). As dielectric polarization in ferrites magnitude depends upon the percentage  $ofFe^{2+}$  and  $Fe^{3+}$  ion pairs at  $T_d$  and  $O_h$  sites.  $Fe^{2+}$  ions concentration largely affects the conduction phenomenon and depends upon type of cation substituting, synthesis route, sintering time and sintering temperatures. In the present case  $\varepsilon'$  and  $\varepsilon''$  both decreases, which can be explained by low  $Fe^{2+}$  ion concentration at  $O_h$  site causing a low value of resistivity and

hence a high value of dielectric parameter. As conductivity and relative permittivity has similar behavior.



Fig. 3. Plot of (a)  $\varepsilon^*$  (b)  $\varepsilon''$  and (c)  $\varepsilon'$  against *f* for all the samples at room temperature



Fig. 4. A Plot of  $\tan \delta$  against the *f* for all the samples at room temperature, the inset shows the peaks of  $\tan \delta$ 

Fig. (4) shows the variation of dielectric loss tangent (  $\tan \delta$  ) with the same range of f. It can be seen that; it has the same trend as dielectric parameters. It decreases exponentially with the increase in f and becomes constant up to f = 5 MHz due to the decreased polarization at high AC fields. As seen in Fig. (4),  $\tan \delta$  has a maximum value (peak values) at specific f. The peak values are found to shift towards higher fwith the increasing of  $Zn^{2+}$  ions. The peak values for the samples of  $Li_{0.5}Fe_{2.5}O_4$ ,  $Li_{0.4}Zn_{0.2}Fe_{2.4}O_4$ ,  $Li_{0.3}Zn_{0.4}Fe_{2.3}O_4$ ,  $Fe_{2.2}O_4, Li_{0.1}Zn_{0.8}Fe_{2.1}O_4$ , and  $ZnFe_2O_4$  were found at  $f = 20 \times 10^4$  $15.3 \times 10^4$ ,  $12 \times 10^4$ ,  $10 \times 10^4$ ,  $8 \times 10^{4}$  $2 \times 10^{4}$ and Hz, respectively. An qualitative explanation can be given for occurrence of the maximum in the  $\tan \delta$  versus f in the case of mixed Li-Zn spinel ferrite. As pointed by Iwauchi(Iwauchi, 1971) there is a strong correlation between the conduction mechanism and dielectric behavior of ferrites. The conduction mechanism in n-type ferrite 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 externally applied electric field, a maximum of  $\tan \delta$  may be observed. The condition for observing a maximumin the losses of a dielectric material is given by

$$\omega'\tau = 1 \tag{10}$$

where  $\omega' = 2\pi f_{max}$  is defined as the jumping probability per unit time, i.e. hopping probability(*P*). This means that  $f_{max}$  is directly proportional to *P* ( $f_{max} \propto P$ ).  $\tau$  is the relaxation time which is related to *P* by the relation  $\tau = 1/2P$ . Now a decrease of  $f_{max}$  with increasing of  $Zn^{2+}$  ions indicates that the *P* is decrease continuously. This behavior was, also, observed in various ferrite systems (Krishna *et al.*, 2012; Xavier *et al.*, 2015).

#### Conclusion

Substitution of the non-magnetic  $Zn^{2+}$  ions in *Li* spinel ferritewere successfully prepared by conventional solid statetechnique. AC electrical conductivity showed increasing with increasing of the appliedfrequency. The dielectric constant (real, imaginary and complex) and dielectric loss tangent were found to decrease for all samples with increasing of the applied frequency. The substitution of  $Zn^{2+}$  ionsin *Li* spinel ferrite shows remarkable influences in the AC properties.

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