International Journal of Current Engineering and Technology

E-ISSN 2277 – 4106, P-ISSN 2347 – 5161
Available at http://inpressco.com/category/ijcet

Research Article

Structural and Dielectric Characterization of $Co_{0.5}Zn_{0.5}Fe_2O_4$ Synthesis by Sol-Gel Auto Combustion Method

Inaam M. Abdulmajeed† and Lana Salar Khalil†*

†Baghdad University, College of Science, Physics Department, Iraq

Accepted 20 Jan 2016, Available online 31 Jan 2016, **Vol.6, No.1 (Feb 2016)**

Abstract

Zink cobalt ferrite ($Co_{0.5}Zn_{0.5}Fe_2O_4$) were synthesized using sol-gel auto combustion method. The synthesized particles were characterized using X-ray diffraction, the XRD patterns corresponding to spinel crystalline ($Co_{0.5}Zn_{0.5}Fe_2O_4$). Dielectric permittivity (ε') of $Co_{0.5}$ $Zn_{0.5}$ Fe_2O_4 decreases while conductivity (σ) increases with increasing frequency. The samples have high values of ε' at low frequencies. The D.C electrical resistivity was investigated from room temperature to 423 K, the variation of dc conductivity decreases by increasing temperature hence resistivity increases.

Keywords: *Co-Zn ferrites; sol gel auto combustion; X-ray Diffraction; dielectric behavior; dc conductivity*

Introduction

Over the last few decades the interest towards soft magnetic materials has substantially increased due to increasing demand for improved performances of electromagnetic gadgets, memory or data storage devices, sensors etc. Magnetic materials which have combined electrical and magnetic properties are known as ferrites. Iron oxide and metal oxides are the main constituents of the ferrites (Kittle, 2005).

The electrical conductivity of ferrites at room temperature varies from10^3 ($\Omega^{-1}.cm^{-1}$) to 10^{-11} ($\Omega^{-1}.cm^{-1}$) depending on the chemical composition and nature of constituent ions. The low electrical conductivity in comparison with that of magnetic metals has been the main aspect of ferrites. The frequency dependent dielectric behavior creates considerable interest in understanding many physical and chemical properties. These high and low resistivities of ferrites are mainly explained on the basis of actual location of cations in the spinel structure and hopping mechanism (S. B. Patila.et.al, 2014; E. C. Snelling, 1969). The principle application of dielectric and the ac electrical properties is the capacitive element in electronic circuits and as electrical insulators. Therefore, the dielectric constant, dielectric loss factor and conductivity are the important parameters. Most of the high frequency applications related to electric properties are concerned with dielectrics (J. Smite and H.P.J.Wijin, 1959). The advantages of ferrites, which are ceramic in nature, over the other available dielectric materials are elastic properties, temperature performance and greater resistance to environmental changes particularly at high temperature and their ability to form gas tight seals with metals. The dielectric properties of ferrites basically depend on several factors such as chemical composition, method of preparation and additives. These properties are sensitive to microstructure, grain size, grain boundaries and porosity. The study of composition and frequency dependence of dielectric behavior gives valuable information regarding localized charge carriers and dielectric polarization (R. Rani.*et.al*, 2013; G. Sathishkumar, 2010).

Experimental

Preparation of Co-Zn ferrite by sol-gel method

Ferrite powders were prepared by sol-gel auto combustion method. In this method; Cobalt nitrate, Iron nitrate, Zinc acetate, Citric Acid and sodium hydroxide were used as a starting raw materials, Each one of the component ($Fe(NO_3)_3.9H_2O$), ($Co(NO_3)_2.6H_2O$) and ($(CH_3COO)_2.Zn.2H_2O$) was dissolved using stoichiometric ratios to prepare $Co_{0.5}$ $Zn_{0.5}$ Fe_2O_4 ferrite, The iron solution, cobalt solution and zinc solution were mixed using magnetic stirrer and so the cobalt zinc ferrite solution was prepared, While the Co-Zn ferrite solution was still on the magnetic stirrer, drops of 1N for NaOH solution was added slightly to the solution until the gel was formed, The PH of the solution was measured for $Co_{0.5}$ $Zn_{0.5}$ Fe_2O_4, where the gel formation begins at PH (8), Addition of citric acid that leads to hear notifying the combustion which helps in reducing the particle size of produced gel.

*Corresponding author: **Lana Salar Khalil**

Filtrate the solution with filter papers to get out the gel then washing it with deionised distilled water for at least three times.

After filtering the gel product, dry it at 80^0 for one day in a programmed electrical oven to get the ferrite powder. The gel was then crushed and fired at temperature 200°C. The powders were finally cooled by switching off the furnace to room temperature.

For the purpose of molding samples and processing; the prepared samples were compact in mold having10 mm diameter with thickness of 2mm under pressing load 3 tons to get a pellets. All compact samples were sintered in electric furnace, The sintering temperature was 500°C for two hours, with a heating rate of 5°C /min, then the samples were cooled down to the room temperature.

Dielectric Measurements

LCR meter model 4194A made in Japan is used to measure the dielectric properties (dielectric constant, loss tangent and the conductivity).

Result and discussion

The polycrystalline $Co_{0.5}$ $Zn_{0.5}$ Fe_2O_4 ferrite have been investigated byx-ray diffraction technique which is shown in Figure (1). The X-ray diffraction pattern of the sample $Co_{0.5}$ $Zn_{0.5}$ Fe_2O_4 shows that the absence of extra lines in the pattern which confirms the single phase cubic spinel structure formation of samples. Also, the x-ray spectrum showed all characteristic planes of Co-Zn spinel ferrites (220, 311, 222, 400, 422, and 511), the main diffraction peak of spinel ferrite that is (311) peak is the more intense one that was considered as a measure of its degree of crystallinity. The lattice parameter for prepared $Co_{0.5}$ $Zn_{0.5}$ Fe_2O_4 was 8.3935 A°.

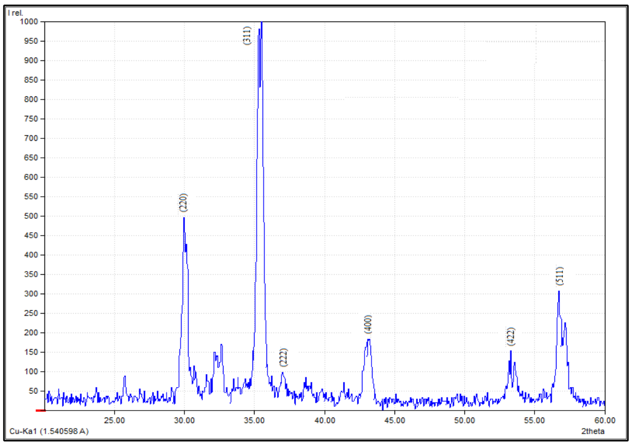

Fig 1 XRD patterns for $Co_{0.5}Zn_{0.5}Fe_2O_4$samples prepared by sol-gel

Electrical properties of Co-Zn ferrite

The electrical properties of Co substituted Zn ferrite ($Co_{0.5}Zn_{0.5}Fe_2O_4$) in this study includes the a.c measurement, d.c measurement and dielectric constant, the a.c measurement were measured under maximum frequency at about 5MHz.

Dielectric constant

The dielectric constant, **ε'**, was calculated from the measured value of capacitance (**$\varepsilon' = C/C_o$** , where C_o is the capacitance of free space.

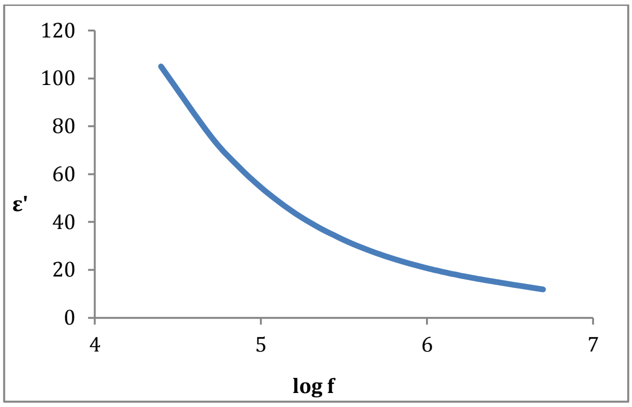

Fig 2 The variation of dielectric constant with frequency logarithm (logf)for ($Co_{0.5}Zn_{0.5}Fe_2O_4$)

Figure (2) shows the variation of dielectric constant with frequency logarithm at room temperature for ($Co_{0.5}Zn_{0.5}Fe_2O_4$). It can be seen from this figure that the dielectric constant is found to decrease continuously with increasing of frequency for the specimen exhibiting normal dielectric behavior of ferrites. The dielectric dispersion is rapid at lower frequency region and remains almost independent at the high frequency region. The incorporation of Zn into the ($Co_{0.5}Zn_{0.5}Fe_2O_4$.) has no pronounced effect on the dielectric constant at high frequency, but significantly decreases the dielectric constant in the low frequency range. This type of behavior was observed in a number of ferrites such as Mg-Zn ferrites (B. K. Bammannavar. *et al*, 2007). The dielectric behavior of ferrites may be explained on the basis of the mechanism of the dielectric polarization process and is similar to that of the conduction process. The electronic exchange $Fe^{2+} \leftrightarrow Fe^{3+}$ gives the local displacement of electrons in the direction of the applied electric field, which induces the polarization in ferrites (K. A. Hossain. *et al*,2013). The magnitude of exchange depends on the concentration of Fe^{2+}/ Fe^{3+} ion pairs present in the B site for the present ferrite. The presence of Fe^{2+} ions in excess amount favors the polarization effects. The observed decrease in dielectric constant with increase in frequency is due to the fact that above certain frequencies the electronic exchange between Fe^{2+} and Fe^{3+} ions does not follow the frequency of the applied ac field. The samples have high values of ε' in the order of 10^3-10^8 at low frequencies. This could be explained using Koop's phenomenological theory (Koops), which was based on the Maxwell-Wagner model (Maxwell and Wagner) for the inhomogeneous double layer dielectric structure. The dielectric structure was supposed to be composed of the fairly well-conducting ferrite grains. These are separated by the second thin layer of grain boundaries which are poorly conducting substances. These grain boundaries could be formed during the sintering process due to the superficial reduction or oxidation of crystallites in the porous materials as a result of their direct contact with the firing atmosphere (M. T. Farid. *et. al,* 2015). The grain boundaries of lower conductivity were found to be effective at lower frequencies while ferrite grains of high conductivity are effective at high frequencies (Koops 1951, Kumar and Srivastava 1994). This explains the higher values of ε′ at lower frequencies and the decrease in ε′ as the frequency increases.

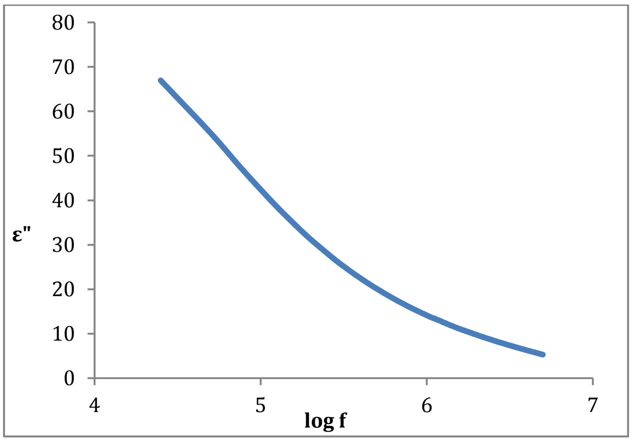

Fig 3 The variation of the imaginary part of dielectric constant with angular frequency logarithm (logf)for ($Co_{0.5}Zn_{0.5}Fe_2O_4$)

The real ε' and imaginary ε parts of the dielectric permittivity are given by the following Debye equation (Kingery*et al.* 1976) as

$$\varepsilon' = \varepsilon\infty + \frac{\varepsilon^{o} - \varepsilon\infty}{1 + \omega^2\tau^2} c_{\circ}^{\infty}$$

Where, τ is the relaxation time, $\epsilon_\circ$ the dielectric permittivity at a very low frequency and ϵ_∞ the dielectric permittivity at a very high frequency, respectively. This Debye equation indicates that ε′ decrease as the frequency increases. This is in agreement with the behaviour of ε′ in Figures 2 and 3 for the present studied samples(K. A. Hossain. *et al*,2013).

Electrical conductivity

The dependence of the measured total conductivity (σ_{tot}) on the frequency for different frequencies in the range of (50Hz to 5M Hz) for Co-Zn ferrite ($Co_{0.5}Zn_{0.5}Fe_2O_4$) is shown infigure 4. It was observed that σ_{tot} increased with the increase of frequency.

In general, the conductivity of semiconductor consists of free or weakly bound electronic and ionic charges and trapped ionic charges in the semiconductor.

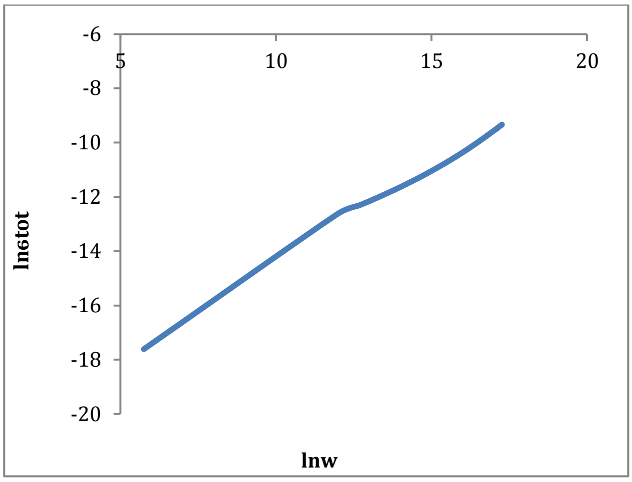

Fig 4 Variation of total conductivity with angular frequency for ($Co_{0.5}Zn_{0.5}Fe_2O_4$) ferrite

The free charges are free to move in electrical field independent on frequency, and contribute to the direct current conductivity. While charge carriers that are trapped in the semiconductor require alternating electrical filed at certain frequency to liberate the ions from one site to another in succession by hopping mechanism and contribute to the alternating current conductivity.

It was observed that the electrical conductivity increases with increase in frequency. These result shows that the conductivity was behaving frequency dependence for the ferrite compositions. The electron hopping between Fe^{+2} and Fe^{+3} ions on the octahedral sites in addition to the hole hopping between Zn^{+2} ions on B site will contribute to electric conduction in ferrites(D. B. Fahad ,2014).

The dielectric loss

Figure 5 shows that tanδ decreases with increasing frequency. When the frequency of the applied ac electric field is much smaller than hopping frequency of electrons between Fe^{2+} and Fe^{3+} ions at adjacent octahedral sites, the electrons follow the field and hence the loss is maximum.

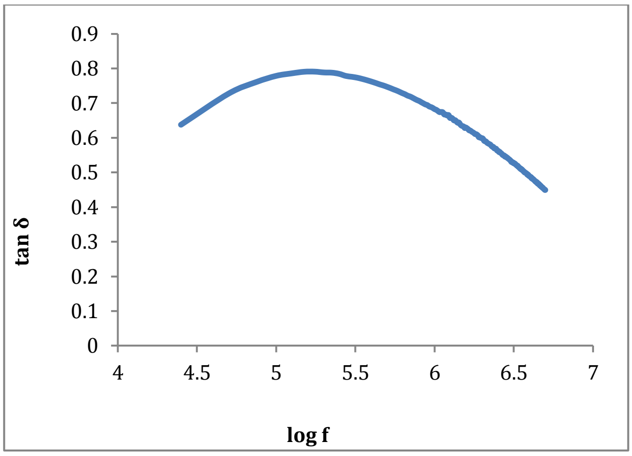

Fig 5 Variation of dielectric loss with the frequency for ($Co_{0.5}Zn_{0.5}Fe_2O_4$) ferrite

At higher frequencies of the applied electric field, the hopping frequency of the electron exchange between Fe^{2+} and Fe^{3+} ions cannot follow the applied field beyond certain critical frequency and the loss is minimum.At low frequency (tanδ) is high and decreases rapidly at high frequency in accordance with Koops phenomenological theory. Therefore, it is expected that energy loss is high at low frequency region while it is low at high frequency region (M. T. Farid, 2015).

Dc electrical conductivity

In order to study conductivity mechanisms, it is convenient to plot logarithm of the conductivity (log σ) as a function of 1000/T for prepared ferrite samples in the range 303–423K for ($Co_{0.5}Zn_{0.5}Fe_2O_4$), as shown in fig(6). This figure shows that by increasing temperature, conductivity decreases hence resistivity increases. This confirms that the ferrite under investigation has semiconductor behavior. The increase in conductivity may be due to the presence of Fe^{2+} ions as zinc is added, which produced during sintering. Another reason for increase in conductivity on increasing zinc composition is due to that Zn^{2+} ions prefer the occupation of tetrahedral sites (A) and Co ions prefers the occupation of octahedral sites (B). While Fe ions partially occupies A and B sites. On increasing Zn^{2+} concentration at A sites, Co ions concentration at B sites will decrease. This leads to migration of some Fe ions from A site to B site to balance the reduction in Co ions concentration at B sites. As a result the number of ferric and ferrous ions at B sites which are responsible for electrical conductivity in ferrites increases consequently resistivity decreases by increasing Zn^{2+} ion concentration. This same trend of conductivity has been reported (N. Ferrite *et. al,*2012).

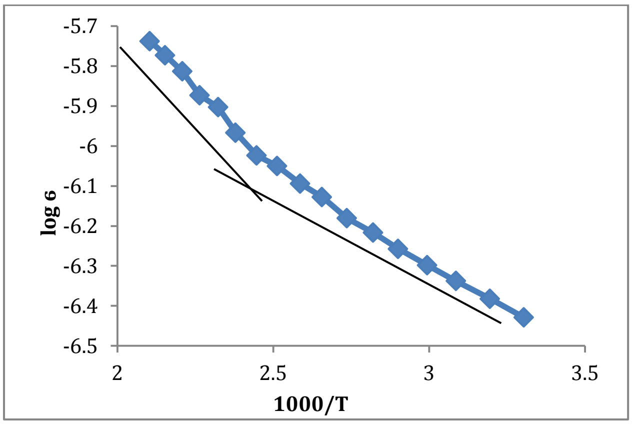

Fig 6 The variation of log(б)with 1000/T for ($Co_{0.5}Zn_{0.5}Fe_2O_4$)

Conclusions

The polycrystalline $Co_{0.5}$ $Zn_{0.5}$ Fe_2O_4 ferrite with spinel structure can be synthesis by sol-gel auto combustion method.

Dielectric permittivity (ε′) of $Co_{0.5}$ $Zn_{0.5}$ Fe_2O_4 decreases while conductivity (σ) increases with increasing frequency. The samples have high values of ε' at low frequencies. This could be explained using Koop's phenomenological theory which was based on the Maxwell-Wagner model. The behavior of electrical conductivity is explained by hopping mechanism between Fe^{+2} and Fe^{+3} ions at octahedral sites.

The maximum of the loss factor (tan δ) is noticed in the range of frequency at about (1-6) KHz. This maximal value is observed when the hopping frequency is equal to the frequency of the applied field.

The dc conductivity variation with temperature shows that the increasing in temperature gives conductivity decreases hence resistivity increases. This confirms that the ferrite under investigation has semiconductor behavior.

References

Kittel, Charles - Introduction To Solid State Physics, 8th edition.

S. B. Patil, R. P. Patil, J. S. Ghodake, and B. K. Chougule (2014), Journal of Magnetism and Magnetic Materials, Temperature and frequency dependent dielectric properties of Ni – Mg – Zn – Co ferrites, J. Magn.Magn.Mater., vol. 350, pp. 179–182

E.C. Snelling (1969), Soft-Ferrites, 2nd edition, Iliffe Books Ltd, London

J. Smit and H. P. J. Wijn, Ferrites (1959), Philips Technical Library, Eindhoven.

R. Rani, G. Kumar, K. M. Batoo, and M. Singh (2013), Electric and Dielectric Study of Zinc Substituted Cobalt Nanoferrites Prepared by Solution Combustion Method, Am. J. Nanomater., vol. 1, no. 1, pp. 9–12.

G. Sathishkumar, Synthesis (2010.), Structural and Dielectric Studies of Nickel Substituted Cobalt-Zinc Ferrite, Mater. Sci. Appl., vol. 01, no. 01, pp. 19–24.

B. K. Bammannavar, L. R. Naik, R. B. Pujar, and B. K. Chougule (Oct 2007), Preparation , characterization and physical properties of Mg-Zn ferrites, vol. 14, pp. 381–385.

K. A. Hossain, S. Akther, and D. K. Saha (2013), Dielectric and transport properties of zn-substituted cobalt ferrites, vol. 37, no. 1, pp. 73–82.

K. R. Reddy, P. S. C. Rao, and R. E. Jessup (1982), The Effect of Carbon Mineralization on Denitrification Kinetics in Mineral and Organic Soils, Soil Sci. Soc. Am. J., vol. 46, no. 1, p. NP.

D. B. Fahad (2014), Structural, Electrical and Optical Properties of CuLa y Fe 2-y Ferrite System.

M. T. Farid, I. Ahmad, S. Aman, M. Kanwal, G. Murtaza, I. Ahmad, and M. Ishfaq, Sem (2015), Ftir and Dielectric Properties of Cobalt Substituted, vol. 11, no. 1, pp. 1–10.

N. Ferrite and E. T. Al (2012), Structural and Electrical Conductivity Studies in, vol. 2012, no. September, pp. 185–191.