AC Conductivity and Dielectric Properties of Cu–Zn ferrites

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***Abstract**: In this work, we have studied the effects of the Zn^{2+} ions on the electric and the dielectric properties of the Cu spinel ferrite. The mixed Cu-Zn spinel ferrite, of chemical formula $Cu_{1-s}Zn_sFe_2O_4$, where s stepped by 0.2 according to ($0.0 \le s \le 1.0$), were prepared from purity metal oxides using the standard ceramic preparation. The AC conductivity was determined for the ferrite samples in the applied frequency range $(10^4 - 10^6)Hz$. In this range of frequency, the AC conductivity increases rapidly as a function of the applied frequency.*

The dielectric properties for the ferrite samples were also determined at room temperature. The general trend for all samples was found to decrease continuously with increasing of the applied frequency. The measurements of the electric and the dielectric properties show that, the behavior of the ferrite samples is similar to that of the semiconductor materials. The results of the electric and dielectric properties are inadequate to previous studies for various ferrite compounds. The electric conductivity for the samples was explained using the electron hopping model.

Introduction

Spinel ferrites and their properties have been the subject of many recent [1-5] investigations. High electrical resistivity and low eddy current losses at high frequency make them widely useable for the cores of high frequency electronic devises [2]. It is therefore important to study electric and dielectric properties of spinel ferrites. Various investigators have studied the electric properties of the spinel ferrites [3].The dielectric behavior of Co-Zn [6], Li-Zn [7] and Mn-Zn[8] ferrite systems as a function of frequency was also investigated. They attributed the dielectric behavior in above systems to Maxwell – Wegner polarization. A strong correlation between the conduction mechanism and the dielectric behavior for ferrites has been reported by Lwauchi [9]. The dielectric behavior of copper- containing ferrites was investigated by Reslescu et al. [10]. They studied the dielectric parameter as function of composition, and frequency for Cu-Ni and Cu-Mn ferrites. In fact there is no cut study of electric and dielectric behavior of Cu- Zn ferrites, particularly at low frequency. Therefore, a detailed investigation of frequency and composition dependence of the dielectric constant and dielectric loss tangent are carried out in the present work and the results are presented in this paper.

Experimental details

Zn-Cu ferrites of different compositions, with formula $Cu_{1-s}Zn_sFe_2O_4$ where s ranges from 0 to 1 in steps of 0.2, were prepared by usual double sintering ceramic technique, using high purity oxides, CuO, ZnO, and Fe_2O_3. The ferrite powders were presintered at 750 ˚C for 3h and cooled to room temperature. The reacted materials were wet-milled and a small quantity of polyvinyl alcohol was added as a binder. The ferrite granules were pressed by hydraulic press at a pressure of 3×10^3kg/cm^2 to form pellets of 5mm radius and 3-5mm thickness. The pellets were sintered at 1100˚C for 5h. The samples then were cooled at a rate of 1K/ min. The pellet samples were polished and smoothed to have uniform plane surfaces. Silver was pasted on sides to ensure good electrical contacts.

The AC conductivity measurements were carried out using the complex impedance technique. In this technique, a Function generator Model GFG 8050 is connected across the sample and a standard non-conductive resistance (R). A dual channel oscilloscope (CRO) Model GBS 620 is used to measure the total input voltage (V_T). Also, the voltage (V_{R}) across the non – inductive resistance is determined. Thus, the AC conductivity σ can be calculated as follows;

$$\sigma = Z^{-1} L/ S \ (\Omega^{-1} m^{-1})$$

where, Z , L and S are the impedance ,thickness and the cross – sectional area of the sample respectively.

The dielectric loss tangent was obtained from Lissajous figure displayed along-persistence cathode ray oscilloscope according to Collett et al.[11]. V_T is connected to X-plate of the oscilloscope and V_R to Y- plate .Hence the loss tangent tan δ is equal to cot φ where $\varphi = \sin^{-1}(V_\theta/V_T)$. The dielectric constant **ε** was also calculated using AC conductivity σ_{AC} and the dielectric loss tangent tan δ according to the following formula [12].

$$\varepsilon = \frac{\sigma_{AC}}{\nu \tan\delta} 1.8\times10^{10} \qquad (1)$$

where ν is the frequency of the applied field.

Results and Discussion

1. Conductivity

The variation of the AC conductivity with the applied frequency that ranges from $10^4\,Hz$ up to $10^6\,Hz$ was studied at room temperature for the samples of the ferrite system $Cu_{1-s}Zn_sFe_2O_4$.

Figure (1-a,b), displayed the AC conductivity σ_{AC} for the given samples showed a continuous increasing with the increasing of the applied frequency. All samples exhibit normal behavior with the variation of the applied frequency. It is also clear that, the high value of the AC conductivity is obtained for the sample with s = 1.0. This indicates that, the AC conductivity depends strongly on the increase in the Zn^{2+} ions in the samples. The dispersion in σ_{AC} with the applied frequency that explained by Koop's theorem [13],is shown in the figures (1a,b) which supposed the ferrites compact acts as a multilayer capacitor. In this model, the ferrite grain and grain boundaries have different properties. This gives rise that the effect of the multilayer capacitor rises with frequency and leads to the increase of the conductivity.

The simplest theoretical expression for the AC conductivity σ_{AC} can be described in the form [14]

$$\sigma_{AC} = \nu \frac{\varepsilon \tan\delta}{1.8X10^{10}} \Omega^{-1} m^{-1} \qquad (2)$$

where ν is the applied frequency, ε is the dielectric constant and tan δ is the dielectric loss tangent.

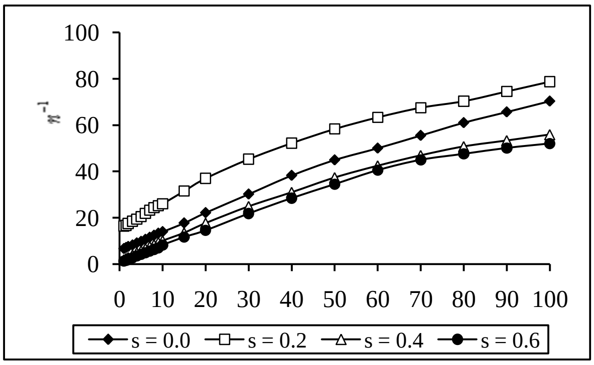

Fig. (1a): A Plot of σ versus the applied frequency for the samples with s = 0.0, 0.2, 0.4 **and** 0.6 **at room temperature.**

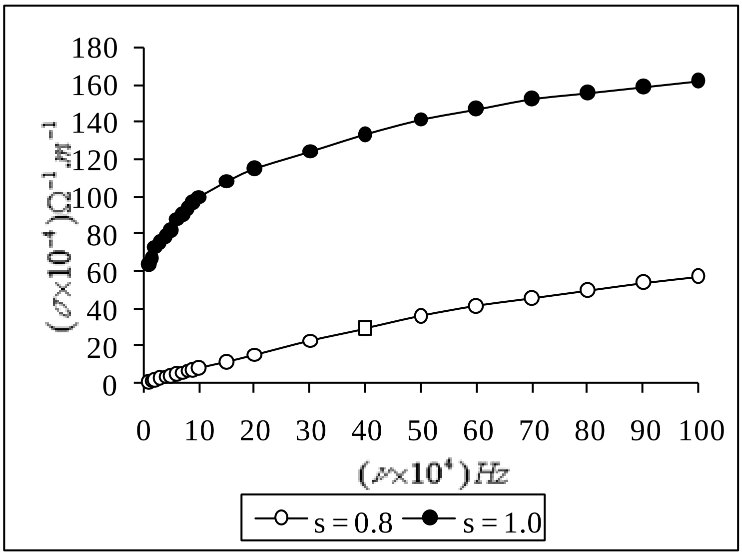

Fig. (1b): A Plot of σ versus the applied frequency for the samples with s = 0.8 **and** 1.0 **at room temperature.**

From this equation, it is clear that, σ_{AC} is directly proportional to the applied frequency, therefore, as shown in the figure (1a,b), σ_{AC} increases with the increasing of the applied frequency. It was also found that, σ_{AC} for the Cu spinel ferrite increased by 90.48 %,where the applied frequency increased. This may be attributed to the electron hopping or the electron exchange, i.e. $Fe^{3+} \Leftrightarrow Fe^{2+}$ or $Cu^{1+} \Leftrightarrow Cu^{2+}$, which occurs by the electron transform between the adjacent O_h sites in the spinel lattice [15], or by the electron exchange according to the formula $Fe^{3+} + Cu^{1+} \rightarrow Fe^{2+} + Cu^{2+}$.

However, σ_{AC} is increased for the *Zn* spinel ferrite by 42%. This is explained on the basis of **Verwey** mechanism [16]. 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 in equivalent lattice sites. Depending upon the sintering conditions, number of such ions may be produced during the preparation of the ferrite samples.

It is known that, partial reduction of the electron hopping, $Fe^{3+} \Leftrightarrow Fe^{2+}$, can take place at elevated firing temperature [17]. Thus; the hopping of electron, $Fe^{3+} \Leftrightarrow Fe^{2+}$, occurs only by electron transform between the adjacent O_h sites in the spinel lattice formed in the *Zn* spinel ferrite. This causes the reduction in electric resistance [18,19]. In general, the average percentage of the increasing of the conductivity for all samples of the given system is equal to 83%, due to the higher concentration of Fe^{2+} ions in the spinel structure lattice which leads to higher conductivity [20].

Further comparison, at the frequency $\nu = 10^6 Hz$ for all samples, it was found that, the sample with s = 1.0 showed a rapid increase in σ_{AC} which is equal to $162 \times 10^{-4} \Omega^{-1}.m^{-1}$. This is due to the existence of a maximum value of the divalent iron Fe^{2+} ions among all the mixed *Cu*-*Zn* spinel ferrite, as discussed **in** the *IR* spectra analysis [21]. A similar behavior was observed in various ferrite systems by several investigators [22-25]. The relaxation in σ_{AC} can be described in terms of relaxation [26]

$$\sigma_{AC} = \sigma_{h\nu} + \frac{\sigma_{l\nu} - \sigma_{h\nu}}{1 + (\omega\tau_{\sigma})^2} \qquad (3)$$

where $\sigma_{l\nu}$ is the value of σ_{AC} at low frequency and $\sigma_{h\nu}$ the value of σ_{AC} at high applied frequency.

The relaxation time τ_{σ} is a characteristic time constant of ferrimagnetic materials and $\omega = 2\pi\nu$ is the applied angular frequency .The relaxation time τ_{σ} was also calculated at different values of σ_{AC} for all samples. It was found that, the average value of τ_{σ} is approximately given by $\tau_{\sigma} = 2.9X10^{-6}$ sec.. This agreed well with the value obtained for the mixed *Li-Cu* ferrite [27].

2. Dielectric Properties

For the given mixed *Cu-Zn* spinel ferrite the variation of the dielectric constant ε is represented in figure (2-a,b), where the frequency ranges from $10^4 Hz$ up to $10^6 Hz$ at room temperature The variation of the dielectric loss tangent $\tan(\delta)$ is also represented in figure (3a,b).

Theoretically, the relation of **ε** and $\tan(\delta)$ with the applied frequency ν is obtained as follows;

$$\varepsilon \tan(\delta) = \frac{\sigma_{AC}}{\nu} 1.8 \times 10^{10} \qquad (4)$$

The values of dielectric constants **ε** that presented in the previous results are calculated by using equation (4) . The general trends for all compositions is that, **ε**

and $\tan(\delta)$ were found to decrease continuously with increasing the applied frequency and showed normal dielectric behavior of the spinel ferrite. This behavior of dielectric may be explained qualitatively by considering that the mechanism of polarization process in ferrite is similar to that the conduction process [28]. The electron hopping occurs by electron transfer between adjacent octahedral sites (B –site) in the sup lattice. Thus by electronic exchange (Fe^{2+} + Cu^{2+} ↔ Fe^{3+} + Cu^{+}) one obtained local displacements of electrons in the direction of the applied electric field, these displacements determine the polarization of the ferrite. The effect of polarization is to reduce the field inside the medium [15].

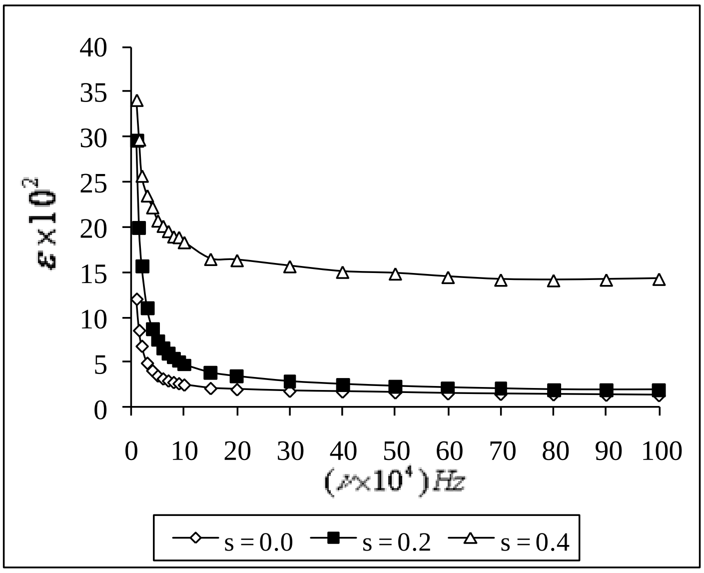

Fig. (2a): A Plot of ε against the applied frequency for the samples with s = 0.0, 0.2 and 0.4 at room temperature.

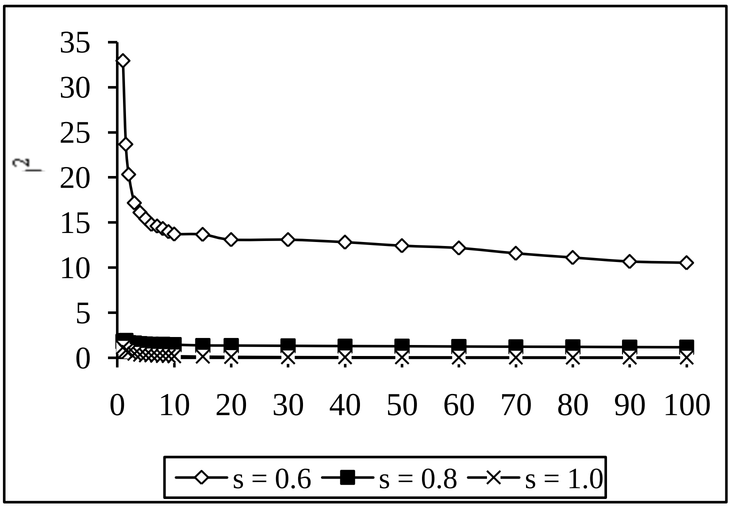

Fig. (2b): A Plot of ε against the applied frequency for the samples with s = 0.6, 0.8 and 1.0 at room temperature.

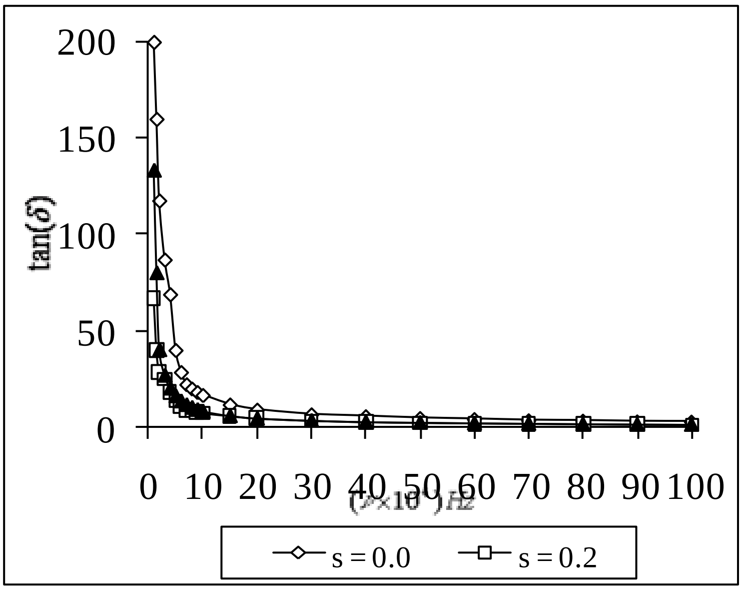

Fig. (3a): A Plot of $\tan(\delta)$ against the applied frequency for the samples with s = 0.0, 0.2 and 0.4 at room temperature.

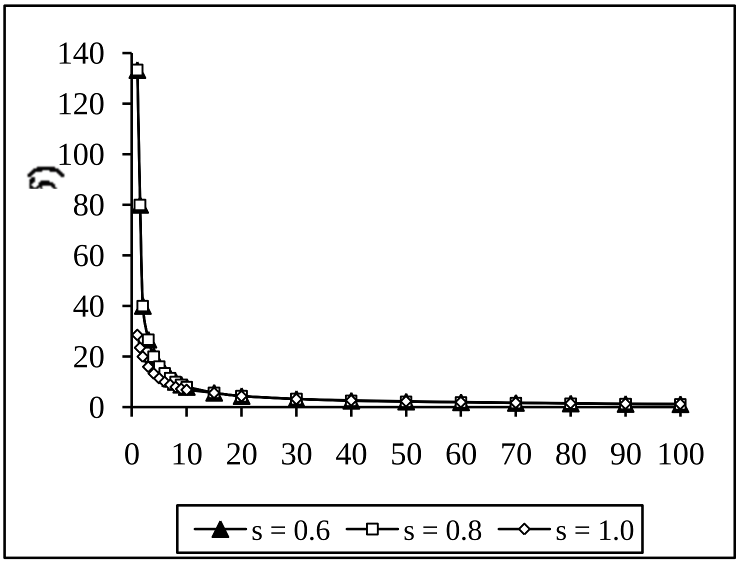

Fig. (3b): A Plot of $\tan(\delta)$ against the applied frequency for the samples with s = 0.6, 0.8 and 1.0 at room temperature.

Therefore, the dielectric constant of a ferrite substance decrease substantially as the frequency is increased. This behavior was, also, observed in various ferrite systems [29 -31].

The obtained results of the dielectric constant indicated that, the *Zn* spinel ferrite was given a maximum dielectric dispersion among all the mixed *Cu-Zn* spinel ferrite which can be explained on the basis of the available Fe^{2+} ions on the octahedral O_h sites of the crystal. **. Lwauchi** [9] has pointed out that there is a strong correlation between the conduction mechanism (hopping mechanism) and the dielectric behavior for ferrites. By the electric exchange $Fe^{2+} \leftrightarrow Fe3^{+} + e^{-1}$ one obtained local displacements of electrons in the direction of the applied electric field. When the applied frequency is increased the dielectric constant **ε** and $\tan(\delta)$ decreased due to the fact that beyond a certain frequency of the electric field, the electronic exchange between ferrous Fe^{2+} and ferric Fe^{3+} ions, cannot follow the alternating field Thus, the dielectric constant **ε** and $\tan(\delta)$ may decrease substantially as the frequency is increased.

Conclusions

1. Measurements showed that, the Cu-Zn samples have the same behavior of semiconductor materials.
2. The *AC* conductivity for the ferrite samples shows a continuous increasing with increasing of the applied frequency. All samples exhibited normal behavior with the variation of the applied frequency.
3. Dielectric constant and dielectric loss tangent were found to decrease for all samples with increasing the applied frequency. This behavior was similar to the normal dielectric behavior of spinel ferrites.
4. A strong correlation between the conduction mechanism and the dielectric behavior of Cu-Zn ferrites was also found.

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