



Characteristics of AC. Conductivity and Dielectric Behavior of $\text{Cu}_{0.5}\text{Ti}_{0.5}\text{Ho}_x\text{Fe}_{2-x}\text{O}_4$ Ferrites

Muthafar F. Al-Hilli*

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

Abstract

Ferrites with the formula $\text{Cu}_{0.5}\text{Ti}_{0.5}\text{Ho}_x\text{Fe}_{2-x}\text{O}_4$ ($x=0$ and 0.09) were prepared by standard ceramic method. The powder mixtures were presintered at 900°C for 5h. The final sintering of the pellets was performed at 1100°C for 2 hrs. The dielectric properties and AC conductivity were measured at different temperatures over the frequency range 100Hz - 10MHz. The variation in dielectric constant with frequency revealed that dispersion is due to the Maxwell–Wagner type of interfacial polarization in accordance with Koop’s phenomenological theory. This ferrite showed high value of dielectric constant. At low frequencies the dielectric constant and dielectric loss factor was found to decrease with the increase in frequency and Ho addition. The dielectric loss decreased with temperature rise. The frequency dependence of dielectric loss tangent is found to be abnormal at various temperatures, the abnormal behavior of dielectric relaxation processes was observed. The prepared ferrite showed low range of AC conductivity. The AC conductivity was noticed to increase with frequency and temperature. While the conductivity decreased with Ho addition.

Keywords: ferrites, Ac conductivity, dielectrics

خصائص التوصيلية الترددية والسلوك العزلي لفيراييت $\text{Cu}_{0.5}\text{Ti}_{0.5}\text{Ho}_x\text{Fe}_{2-x}\text{O}_4$

مظفر فؤاد جميل الحلبي*

قسم الفيزياء، كلية العلوم، جامعة بغداد، بغداد، العراق

الخلاصة

تم تحضير الفيراييت بالصيغة $\text{Cu}_{0.5}\text{Ti}_{0.5}\text{Ho}_x\text{Fe}_{2-x}\text{O}_4$ حيث ان ($x=0$ and 0.09) بطريقة تحضير المساحيق السيراميكية. تم اجراء الحرق الاولي لمخلوط المساحيق عند درجة حرارة 900°C لمدة خمس ساعات اما الحرق النهائي للعينات القرصية فقد تم جرى عند درجة حرارة 1100°C لمدة ساعتين. تم قياس الخصائص العزلية والتوصيلية الترددية. عند درجات حرارية مختلفة على المدى الترددي 100Hz - 10MHz. تبين ان علاقة تغيير ثابت العزل مع التردد هي من نوع ماكويل- واغنر لاستقطابية بين السطحية تبعا لنظرية كوب. اظهرت عينات الفيراييت المحضرة قيم عالية لثابت العزل الكهربائي. عند الترددات الواطئة كانت قيمة ثابت العزل وقيمة الفقد العزلي تقل مع زيادة التردد وكذلك نسبة الاضافة للهولوميوم. وجد ان قيمة الفقد العزلي تقل مع زيادة درجة الحرارة. وان علاقة ظل زاوية الفقد العزلي للفيراييت اعتمادا على التردد عند مختلف درجات الحرارة كانت من نوع السلوك غير الاعتيادي لعملية الاسترخاء العزلي. اظهرت نتائج الفيراييت المحضر قيم واطئة للتوصيلية الكهربائية. كانت التوصيلية الكهربائية تزداد مع درجة الحرارة، بينما تقل مع نسبة اضافة الهولوميوم.

*Email: mfj972@yahoo.com

Introduction

Ferrites are essential materials that are employed in the fabrication of magnetic, electronic and micro-wave devices. Ferrites gained their technological importance from their high resistivity and low eddy current losses. The Copper content was found to have a significant influence on the electromagnetic properties [1]. It was reported that the dielectric parameters are greatly influenced by the presence of Cu^{1+} ions which appears during the sintering process of these ferrites by the reduction of Cu^{2+} . This results in abnormal behavior dielectric properties as a function of frequency and temperature [2]. Rare earth elements (R) are well known to have large magnetic moments, large magneto crystalline anisotropy and very large magnetostriction at low temperatures due to the localized nature of 4f electrons. Rare earth iron based compounds (RFe_2) possess large magnetostriction and these compounds are used as active elements in several magnetostrictive transducers. However, the high conductivity of these compounds limits the usage of the transducers to low frequencies (few kHz). Substitution of rare earth ion into the ferrites spinel structure was reported to cause structural distortions which induce strains in the material. This significantly modifies its electrical and magnetic properties [3]. However, to the best of my knowledge, the effects of Ho doping on the dielectric properties of Cu-Ti ferrites are not well described in literature. This work reports the dielectric properties of Ho doped Cu-Ti ferrite as a function of composition, frequency and temperature.

Experimental

Ferrite samples with the chemical formula $\text{Cu}_{0.5}\text{Ti}_{0.5}\text{Ho}_x\text{Fe}_{2-x}\text{O}_4$, where $x=0$ and 0.09 were prepared by standard ceramic method. Analytical grade oxides supplied by BDH chemicals Ltd. with purity of 99.9 % of Fe_2O_3 , Ho_2O_3 , CuO and TiO were mixed in the proper molar ratios. The mixtures were ground to a very fine powder and then sintered for 5h at temperature of 900°C . The sintered mixtures were grinded and mixed again, and then pressed using pressure of 4 tons into pellet shape with diameter of 2cm. Then the samples were presintered at 1100°C for 2 hrs and then naturally cooled to room temperature. Both sides of the pellet were polished and coated with Al by thermal evaporation as contact electrodes for electrical measurements. The dielectric constant, Dielectric loss factor and loss tangent (δ), were measured over the frequency range 100Hz - 10MHz using an HP4284A LCR meter. The variation of capacity and dielectric properties with temperature was measured from $28-200^\circ\text{C}$.

Results and discussion

Figures-1 and 2 show the relationship of the dielectric constant of the undoped and Ho ($x=0.09$) doped samples with \ln frequency at different temperatures, respectively. The dielectric constant decreased with frequency increase. It can be seen that the dielectric constant decreases rapidly with increasing frequency and then reaches almost a constant value. The components of polarizability are $\alpha_e + \alpha_i + \alpha_o + \alpha_s$, representing susceptibilities associated with, electronic, ionic, orientation, and space charge polarization, respectively. Electronic polarization is due to shifts or displacement of electron clouds in the dielectric field away from their equilibrium positions, resulting in a net dipolar response.

It occurs in all solids up to optical frequencies $\sim 10^{16}$ Hz. Ionic polarization results from similar ionic displacement in the field and occurs up to the infrared region of $10^{10}-10^{13}$ Hz. In contrast, orientation polarization is both frequency (time) dependent and temperature dependent, since it represents dipole orientation and ion jump polarization. The remaining process, the space charge or interfacial polarization, is produced by traveling charge carriers. However, they exhibit different frequency responses and can be separated. At low frequencies, all four sources of polarization are important, whereas at optical frequencies, i.e., above optical phonon frequencies, only the electronic polarization comes into play. In the absence of dipolar polarization, ϵ^* should be nondispersive for frequencies up to microwave frequencies. Thus, dispersion below microwave frequencies results from dipolar polarization effects and space charge or interfacial polarization. Therefore, they play a main role in our dielectric measurement.

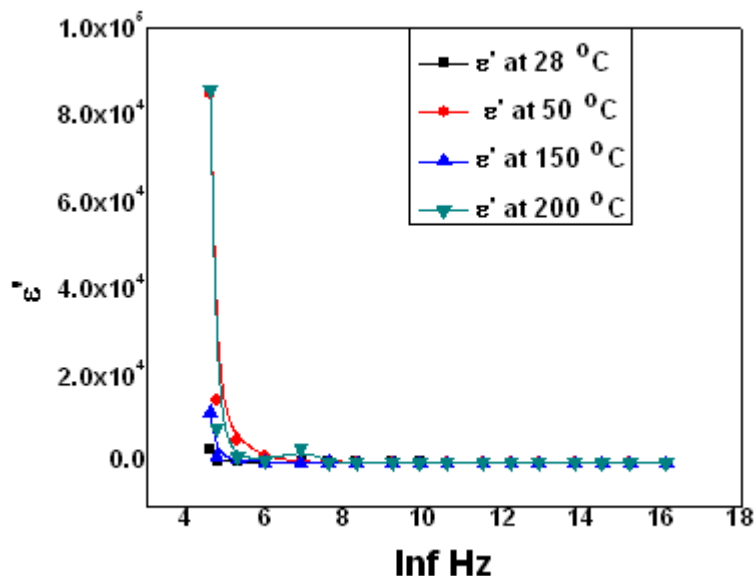


Figure 1- dielectric constant versus ln frequency for the sample $\text{Cu}_{0.5}\text{Ti}_{0.5}\text{Fe}_2\text{O}_4$ measured at different temperatures.

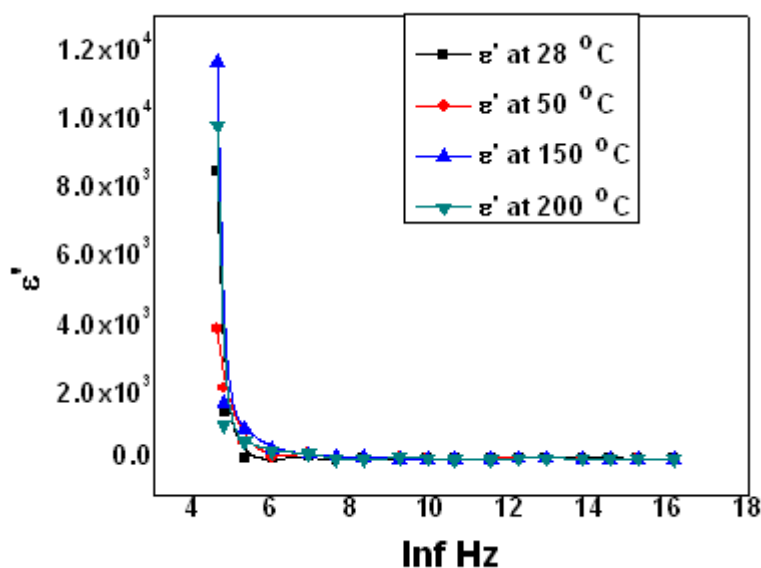


Figure 2- dielectric constant versus ln frequency for the sample $\text{Cu}_{0.5}\text{Ti}_{0.5}\text{Ho}_{0.09}\text{Fe}_2\text{O}_4$ measured at different temperatures.

The imaginary part of the dielectric constant versus ($\ln f$) at different temperatures, for the undoped and doped samples is shown in Figures-3 and 4, respectively. Both real dielectric constant ϵ' and imaginary dielectric constant ϵ'' are temperature and frequency dependent over the temperature range of room temperature to 200 °C. A thermally activated process (that is dipole relaxation process), shown as a drop of the real dielectric constant, gradually shifts to lower frequency as the temperature decreases [4]. The variation of dielectric constant with frequency reveals the dispersion due to the Maxwell–Wagner type of interfacial polarization in accordance with Koop's phenomenological theory. The polarization in ferrites is through a mechanism similar to the conduction process. By electron exchange between $\text{Fe}^{2+} \leftrightarrow \text{Fe}^{3+}$ the local displacement of electrons in the direction of the applied field occurs and these electrons determine the polarization. The polarization decreases with increasing frequency and then reaches a constant value due to fact that beyond a certain frequency of external field, the electron exchange $\text{Fe}^{2+} \leftrightarrow \text{Fe}^{3+}$ cannot follow the alternating field. That's why the

dielectric constant decreases rapidly with increasing frequency and then reaches a constant value. The dielectric constant was found to decrease with Ho addition. This may be because the Ho enhanced the grain growth leading to decrease in the interfacial polarization which plays a significant role in polycrystalline samples.

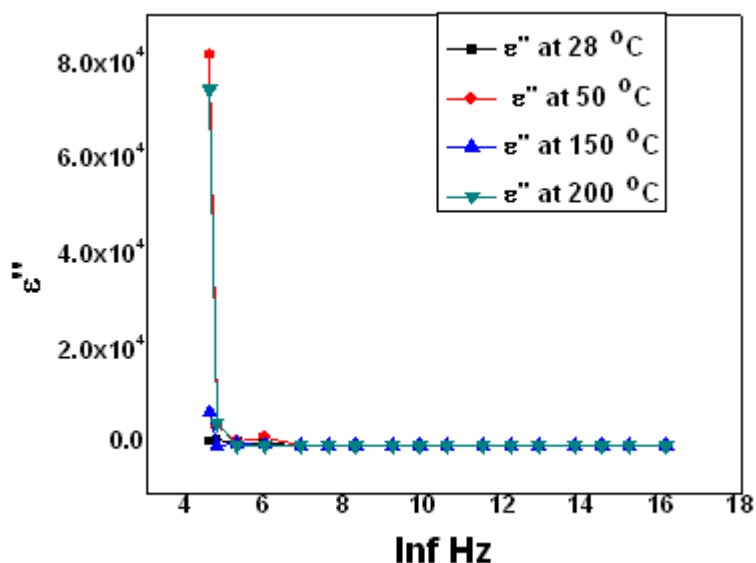


Figure 3- dielectric loss factor versus ln frequency for the sample $\text{Cu}_{0.5}\text{Ti}_{0.5}\text{Fe}_2\text{O}_4$ measured at different temperatures.

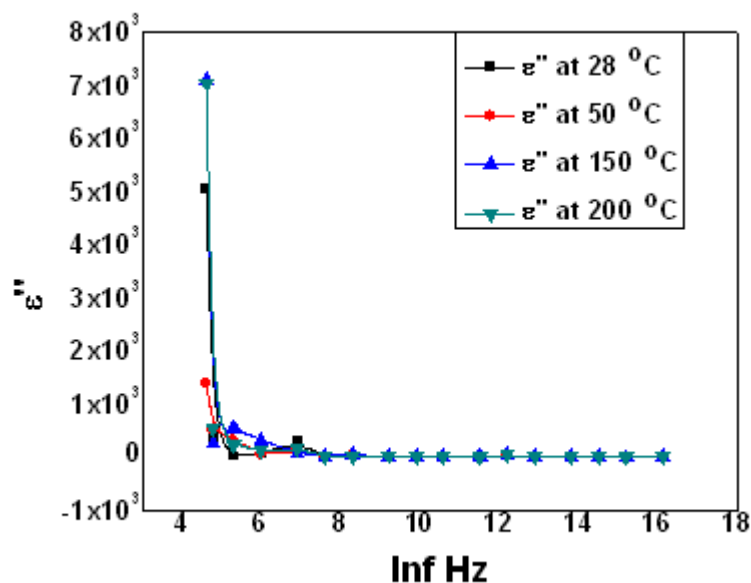


Figure 4- Dielectric loss factor versus ln frequency for the sample $\text{Cu}_{0.5}\text{Ti}_{0.5}\text{Ho}_{0.09}\text{Fe}_2\text{O}_4$ measured at different temperatures.

The variation of loss tangent ($\tan \delta$) with frequency is shown in Figures-5 and 6. It can be seen that for all of the samples it decreases continuously with increasing frequency and none of the samples exhibit the loss peak. The peaking behavior occurs when the jump frequency of electrons between Fe^{2+} and Fe^{3+} is equal to the frequency of the applied field. If the resistivity is very high, as in most microwave ferrites, it can be shown that

$$\tan \delta = 1/\omega \epsilon_0 \rho$$

where ω_0 is the angular frequency corresponding to the maximum value of $\tan \delta$. Thus the dielectric loss tangent of such ferrites is expected to decrease approximately inversely with frequency [5].

The dielectric loss was also noticed to decrease with ho addition. May be due to the rare earths oxides are known as low dielectric loss materials.

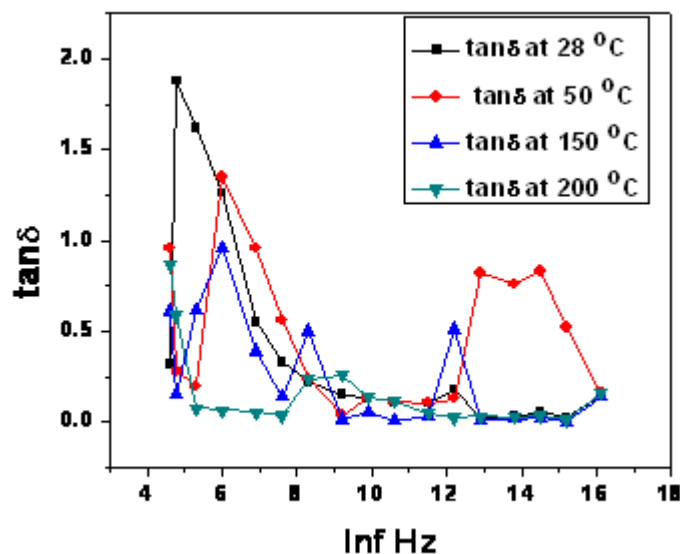


Figure 5- dielectric loss tangent versus ln frequency for the sample $\text{Cu}_{0.5}\text{Ti}_{0.5}\text{Fe}_2\text{O}_4$ measured at different temperatures.

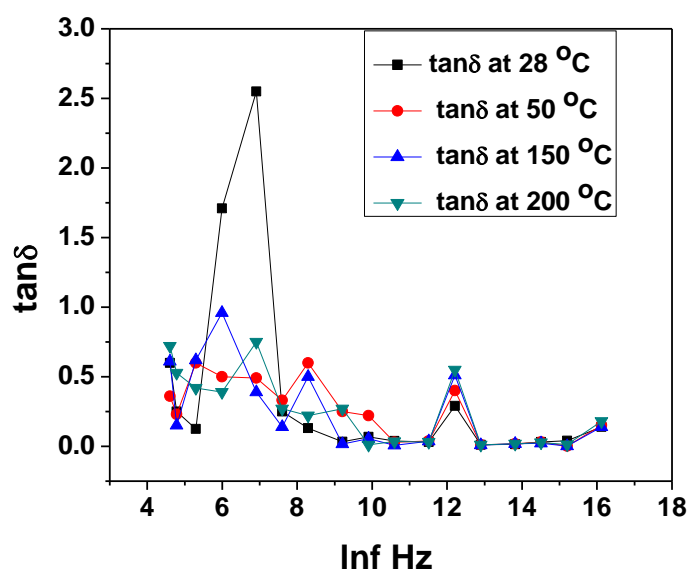


Figure 6- dielectric loss tangent versus ln frequency for the sample $\text{Cu}_{0.5}\text{Ti}_{0.5}\text{Ho}_{0.09}\text{Fe}_2\text{O}_4$ measured at different temperatures.

The AC conductivity dependence upon frequency is illustrated in Figures-7 and 8. The conductivity found to increase with frequency, which agrees with the relation:

$$\sigma_{AC} = \omega \epsilon_0 \epsilon'' = \omega \epsilon_0 \epsilon' \tan \delta$$

where ϵ_0 is the vacuum permittivity, ϵ' is the dielectric constant, ϵ'' is the dielectric loss factor and $\tan \delta$ is the tangent of dielectric loss angle. This behavior with frequency can be explained by Koop's theorem, which supposed that the ferrite compact acts as a multilayer capacitor. In this model, the ferrite grain and grain boundaries have different properties. The effect of the multilayer capacitor increases with frequency; as a result the conductivity increases [6].

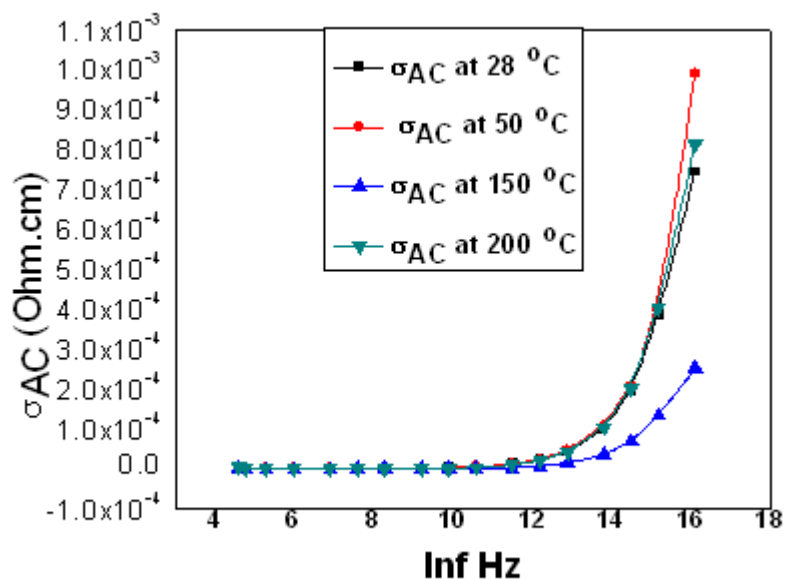


Figure 7- AC conductivity versus ln frequency for the sample $\text{Cu}_{0.5}\text{Ti}_{0.5}\text{Fe}_2\text{O}_4$ measured at different temperatures.

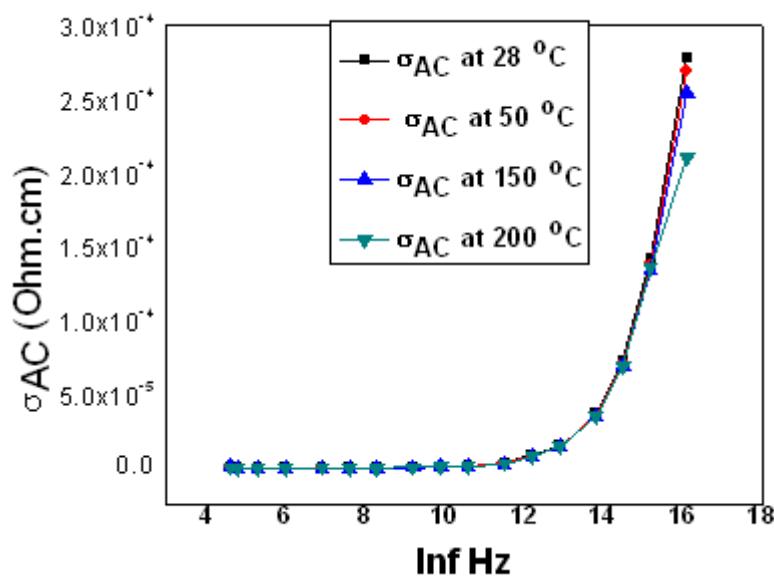


Figure 8- AC conductivity versus ln frequency for the sample $\text{Cu}_{0.5}\text{Ti}_{0.5}\text{Ho}_{0.09}\text{Fe}_2\text{O}_4$ measured at different temperatures.

Conclusion

The dielectric constant decreases rapidly with increasing frequency and then reaches a constant value. The dielectric constant was found to decrease with Ho addition. The change of loss tangent ($\tan \delta$) with frequency showed abnormal behavior. The conductivity was found to increase with frequency.

References

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