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Electromagnetic properties of polycrystalline $Co_{0.5}Zn_{0.5}Fe_2O_4$ at various sintering temperature

M. H. R. Khan[1] and A. K. M. Akther Hossain[2]

Abstract: Polycrystalline $Co_{0.5}Zn_{0.5}Fe_2O_4$ ferrite has been synthesized by solid state reaction method. The variation of dielectric constant, AC resistivity and dielectric loss have been studied as a function of frequency and sintering temperature. The dielectric constant, AC electrical resistivity, and dielectric loss decrease at low frequency region and remain constant at high frequency region. Effect of sintering temperature on dielectric constant and AC resistivity has been discussed. It is found that the dielectric constant for this mixed ferrite increases with increasing sintering temperature. At the same time AC resistivity decreases. Initial permeability of this ferrite was measured at two different temperatures-one at room temperature and another at liquid nitrogen temperature. It is observed that initial permeability of liquid nitrogen temperature is less than that of room temperature. It increases with increasing sintering temperature. DC electrical resistivity has been observed to decrease with increasing temperature. The plot of ln ρ_{DC} is observed to be linear, indicating that the sample is thermally activated. The temperature dependence of DC resistivity shows the semiconductor nature.

1. Introduction:

The polycrystalline ferrites have electrical and dielectric properties that are dependent upon several factors such as the methods of preparation, sintering temperature and substitution of different cations etc. The excellent electrical and magnetic properties make them suitable for high frequency applications in the field of telecommunication. They can also be used as an electromagnetic wave absorber; multilayer chip indicator and transformer core [1-5]. They have the spinel structure AB_2O_4 in which A and B are tetrahedral and octahedral sites, respectively [6]. Among spinel ferrites, cobalt ferrites are very important because they have been proven to be useful in many magnetic applications [7-8]. Cobalt based ferrites have high dielectric constants which make them very useful for microwave applications [9]. Dielectric, electrical and magnetic properties of these ferrites depend on temperature, frequency of applied electric field and its structure [10]. Several investigations on the cobalt based ferrite have been carried out to improve the properties of these ferrites [11-12]. The complex permeability spectra were also examined from the view point of the microstructure, as well as the chemical composition [1]. However, the magnetic properties such as permeability measured at liquid nitrogen temperature of $Co_{0.5}Zn_{0.5}Fe_2O_4$ ferrites at high frequencies, have not been intensively investigated yet. Also no reports have been found in the literature on electrical and dielectric properties of the $Co_{0.5}Zn_{0.5}Fe_2O_4$ ferrites. The aim of the present work is to investigate the effect of sintering temperature and frequency on the dielectric and AC electrical properties and temperature dependent DC electrical resistivity of the $Co_{0.5}Zn_{0.5}Fe_2O_4$ ferrite.

2. Materials and Methods

The spinel ferrites with nominal composition $Co_{0.5}Zn_{0.5}Fe_2O_4$ was prepared by the standard solid state reaction method. Details sample preparation procedure has been explained elsewhere [1]. The dielectric properties measurements and AC resistivity have been carried out by using an impedance analyzer (model no. 6500B) in the frequency range 20Hz - 120MHz. The surface of the disks was coated on adjacent faces with silver paste, thereby forming parallel plate capacitor geometry. The real ($\varepsilon^{/}$) and imaginary ($\varepsilon^{//}$) parts of dielectric constant were calculated using the following relations:

$$\varepsilon^{/} = \frac{Ct}{\varepsilon_0 A} \qquad \text{and} \qquad \varepsilon^{//} = \varepsilon^{/} \tan\delta \ , \qquad [1]$$

where C is the capacitance, t is the thickness of the sample, A is the cross-sectional area, tan δ is loss factor and ε_0 is the free space permittivity. The DC electrical resistivity for the sample was measured by two probes method in a temperature range 25˚ - 200˚C. The relationship between resistivity and temperature may be expressed as

$$\rho = \rho_0 e^{\frac{E_a}{k_B T}} \qquad [2]$$

where k_B is the Boltzmann constant and E_a is the activation energy corresponding to the electrical process.

[1] *Associate Professor, Department of Arts & Sciences, Ahsanullah University of Science and Technology, Dhaka*
[2] *Professor, Department of Physics, Bangladesh University of Engineering and Technology, Dhaka*

3. Results and Discussion

Fig. 1 shows the variation of $\varepsilon^{/}$ as a function of frequency for the samples sintered at different temperatures.

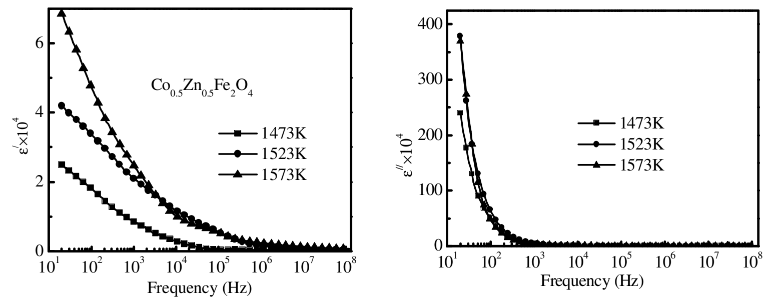

Fig. 1. The variation of $\varepsilon^{/}$ and $\varepsilon^{//}$ as a function of frequency of $Co_{0.5}Zn_{0.5}Fe_2O_4$ sintered at 1473, 1523 and 1573K.

The values of $\varepsilon^{/}$ approximately in the order of 10^4 at low frequencies are observed for $Co_{0.5}Zn_{0.5}Fe_2O_4$ ferrite sintered at different temperatures. Hence, $Co_{0.5}Zn_{0.5}Fe_2O_4$ is a good dielectric material. The complex dielectric constant decreases with increase in frequency for the sample at different sintering temperatures. The value of real part of dielectric constant is much higher at lower frequencies. At very high frequency region, its value becomes so small that it becomes independent of frequency. These types of dielectric behavior are also observed by other investigations [11–12].

The $\varepsilon^{/}$ as function of frequency may be explained on the basis of Maxwell and Wagner's two layer model [13-14]. According to this model, it is the conductivity of grain boundaries that contributes more to the dielectric value at lower frequencies [15]. This causes the localized accumulation of charges under the influence of electric field, which results in interfacial polarization. At higher frequencies the electron exchange between ferrous and ferric ions ($Fe^{2+} \leftrightarrow Fe^{3}$) can not follow the alternating field, which causes a decrease in the contribution of interfacial polarization in dielectric constant. As a result we observe a decrease in $\varepsilon^{/}$.

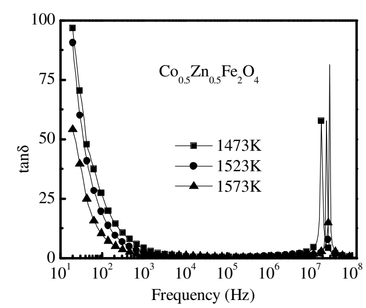

Fig. 2. The variation of tan δ as a function of frequency of $Co_{0.5}Zn_{0.5}Fe_2O_4$ sintered at 1473, 1523 and 1573K.

The variation in tanδ with frequency is shown in Fig. 2. It follows the similar trend as that of $\varepsilon^{/}$ with frequency. The tan δ peaks of the samples have been observed in the higher frequency region. The dielectric loss is one of the most important properties of the ferrites, which depends on the processing and sintering conditions, chemical composition, and the type of additives. It is reported that the dielectric behaviors of ferrites strongly depend on the conduction mechanism [16-17]. The tanδ peak is expected when the hopping frequency of the electron between Fe^{2+} and Fe^{3+} ions is approximately equal to that of the external applied electric field. These peaks are the result of resonance due to the matching of the time period of the applied electric field with those of the corresponding relaxation phenomena. However, the peaks are not observed for the prepared samples clearly because they appear at frequencies greater than 120MHz.

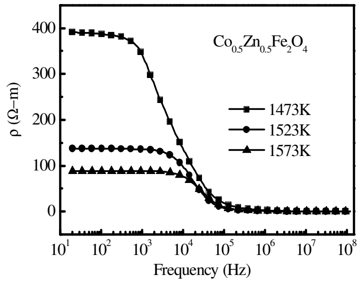

Fig. 3. AC resistivity as function of frequency for $Co_{0.5}Zn_{0.5}Fe_2O_4$ ferrites sintered at 1473, 1523 and 1573K.

Fig. 3 shows the variation of AC resistivity (ρ_{AC}) with frequency measured at room temperature. All the samples show decrease in ρ_{AC} with the increase in frequency up to 10^5 Hz and after that ρ_{AC} remains constant and becomes negligibly small at higher frequencies. It is also clear from this figure that the dispersion is more at low-frequency region which might be due to the difference in the concentration of Fe^{3+}/Fe^{2+}. The conductivity of $Co_{0.5}Zn_{0.5}Fe_2O_4$ can be explained on the basis of the electron hopping between Fe^{2+} and Fe^{3+} ions on octahedral B-sites [18]. During the sintering process Fe^{2+} ions in the lattice are created due to zinc loss. Higher the sintering temperature, greater is the possibility of Fe^{2+} formation. Creation of Fe^{2+} ions gives rise to electron hopping between the Fe ions in Fe^{+2} and Fe^{+3} valence states [18]. Consequently, resistivity decreases with increasing sintering temperature. It is observed from the Figs. 2 and 3 that the AC resistivity and dielectric constant show an inverse trend. It is seen from the Fig. 4a that the DC electrical resistivity (ρ_{DC}) shows a linear decrease with temperature.

The variation of ρ_{DC} with temperature is explained on the basis of actual location of cation in the spinel structure and increasing drift mobility of the charge carriers.

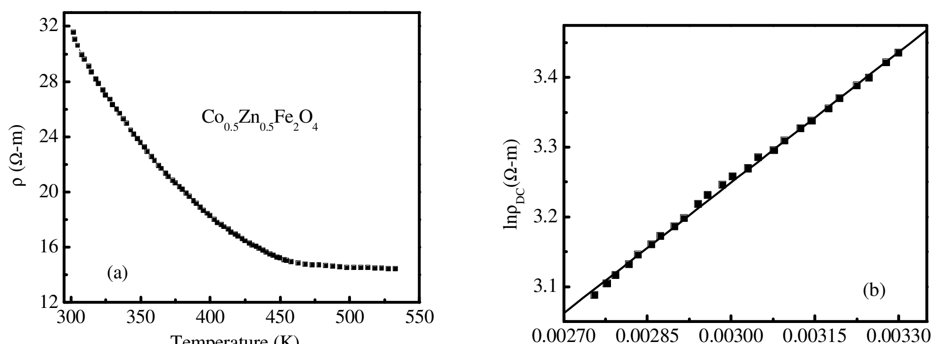

Fig. 4. (a) Temperature dependent ρ_{DC} (b) the variation of $\ln \rho_{DC}$ as a function of inverse temperature of $Co_{0.5}Zn_{0.5}Fe_2O_4$ sintered at 1523K.

The decreasing ρ_{DC} with increasing temperature of this ferrite confirms semi-conducting behavior. The activation energy was calculated using equation 2 from the slope of $\ln \rho_{DC}$ vs. inverse temperature plot as shown in Fig.4b. The plot of $\ln \rho_{DC}$ is observed to be linear, indicating that the samples are thermally activated.

Fig. 5a shows the initial permeability ($\mu_i^/$) of polycrystalline $Co_{0.5}Zn_{0.5}Fe_2O_4$ measured at liquid nitrogen temperature sintered at 1473, 1523 and 1573K as well as Fig.5b shows the effect of temperature on $\mu_i^/$ measured at both room temperature and liquid nitrogen temperature. For both of them, the frequency response $\mu_i^/$ shows the common feature. The $\mu_i^/$ was almost independent of the frequency. The Fig.5 shows that resonance frequency has not been observed in the frequency range 100Hz to 120MHz. It may be found at higher frequency than that of 120MHz. It is observed that $\mu_i^/$ at liquid nitrogen temperature is less than that of room temperature due to increasing anisotropy constant.

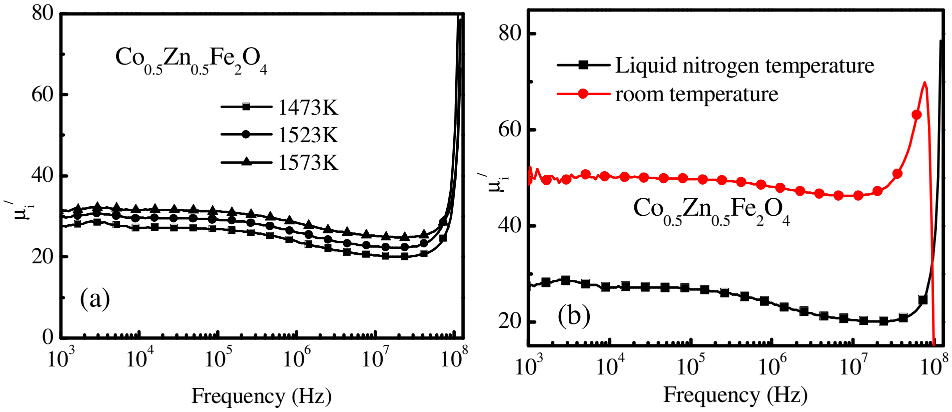

Fig. 5. (a) The μ_i' measured at liquid nitrogen temperature as a function of frequency for $Co_{0.5}Zn_{0.5}Fe_2O_4$ sintered 1473, 1523 and 1573K (b) the variation of μ_i' measured at both room temperature and liquid nitrogen temperature of $Co_{0.5}Zn_{0.5}Fe_2O_4$ sintered at 1473K

It is reported that the anisotropy constant increases with decreasing temperature [3]. Effect of anisotropy constant on μ_i' already discussed elsewhere [1]. The increasing μ_i' with increasing sintering temperature may be explained on the basis of continuous grain growth.

4. Conclusions

The dielectric constant, AC resistivity, dielectric loss, DC electrical resistivity measured at room temperature and initial permeability measured both at liquid nitrogen temperature and room temperature of $Co_{0.5}Zn_{0.5}Fe_2O_4$ ferrites have been investigated. The experimental results indicate that with increasing sintering temperature both dielectric constant and dielectric loss increase and AC electrical resistivity decreases. DC electrical resistivity decreases as the temperature increases indicating that the samples have semiconductor-like behavior. The plot of $\ln \rho_{DC}$ vs. inverse temperature confirms that the conduction is thermally activated. The electric conduction is similar to the dielectric polarization and is related to the electron exchange interaction between the ferrous and ferric ions on the octahedral sites. The initial permeability of liquid nitrogen temperature is less than that of room temperature. Low values of loss factor exhibited by this ferrite suggest its utility in inductor and transformer applications.

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References

[1] M.H.R. Khan, A.K.M. Akther Hossain, J. Magn. Magn. Mater. **324** 550–558 (2012).
[2] J. Azadmanjiri, H. K. Salehani, M. R. Barati, F. Farzan, Mater. Lett. **61**, 84(2007).
[3] Z.Yue, L. Li, J. Zhou, H. Zhang, Z. Gui, Mater. Sci. Eng. B **64**, 68 (1999).
[4] J. Mürbe, J. Töpfer, J. Magn. Magn. Mater. **324**, 578–583(2012).
[5] M.Goodarz Naseri, E. Bin Saion, H. Abbastabar Ahangar, M. Hashima, A. H. Shaari, J. Magn. Magn. Mater. **323**, 1745–1749(2011).
[6] Valenzuela, R., Magnetic Ceramics, Cambridge University Press, Cambridge, 1994.
[7] Lucas, A., Lebourgeois, R., Mazaleyrat, F., Laboure, E., J. Magn. Magn. Mater., **323**, 735–739, (2011).
[8] Surendra S. More, Ram H. Kadam, Ankush B. Kadam, Dhanraj R. Mane, Govind K. Bichile, Cent. Eur. J. Chem. **8(2)**, 419–425, (2010).
[9] M.A. Ahmed and M.A. El Hiti, J. Phyh. III **5** , 775(1995).
[10] R. V. Magalaraja, S. Ananth a kumar, P. Manohar, F. D.Gnanam, J. Magn. Magn. Mater. **253,** 56(2002).
[11] Muhammad Javed Iqbal , Mah Rukh Siddiquah, J. Alloys Compd. **453**, 513–518(2008).

[12] Mohd. Hashim, Alimuddin, Shalendra Kumar, B. H. Koob, Sagar E. Shirsath, E. M. Mohammed, Jyoti Shah, R. K. Kotnala, H. K. Choi, H. Chung, Ravi Kumar J. Alloys Compd. (Article in Press)
[13] J.C. Maxwell, Electricity and Magnetism, vol. 1, Oxford University Press, Oxford, 1929 (Section 328).
[14] K.W. Wagner, Ann. Phys. **40**, 817 (1913).
[15] I. T. Rabinkin, Z. I. Novikova, Ferrites, Izv Acad. Nauk USSR Minsk, 1960.
[16] A. Thakura, P. Mathura, M. Singh, J. Phys. Chem. Solids **68**, 378–381(2007).
[17] M.L.S. Teo, L.B. Kong, Z.W. Li, G.Q. Lin, Y.B. Gan, J. Alloys Compd. **459**, 567–575 (2008).
[18] E. J. W. Verway, P. W. Haayman, Physica **8,** 979(1941).