Dielectric Properties and Magnetic Susceptibility of Mn-Zn Ferrites/Sio$_2$ Composites

D. El Kony and S. A. Saafan

Physics Department, Faculty of Science, Tanta University, Tanta, Egypt.
samiasaafan@yahoo.com

Abstract: Dielectric properties and magnetic susceptibility were measured for four compositions of Mn Zn ferrites ($Mn_{1-x} Zn_x Fe_2O_4$; x = 0.8, 0.6, 0.4 and 0.2).Then composites of these ferrites with SiO_2 were prepared by mixing 25% weight of SiO_2 powder with 75% weight of each composition of the ferrite powders. The dielectric properties and the magnetic susceptibility were measured again for the composite samples. The results are displayed as functions of temperature and frequency for the dielectric constant, loss tangent and AC conductivity, whereas for the magnetic susceptibility only results at room temperature are reported. The results show that adding silicon dioxide has significantly reduced the conductivity or in other words increased the resistivity while the magnetic susceptibility still have high values in the same order of magnitude. These findings may be useful in many applications that require the reduction of eddy currents while keeping the magnetic susceptibility in the same range.

[D. El Kony and S. A. Saafan. **Dielectric Properties and Magnetic Susceptibility of Mn-Zn Ferrites/Sio$_2$ Composites.** *J Am Sci* 2012;8(10):51-57]. (ISSN: 1545-1003). http://www.jofamericanscience.org. 9

Keywords: Dielectric Properties; Soft ferrites; magnetic susceptibility

1. Introduction

Nowadays, ferrites have a primary position of economic and engineering importance within the family of magnetic materials because of their excellent physical properties. Practically all TV sets, portable radios, long distance carrier telephone circuits are employing ferrite cores in filters, antennas and transformers [1]. Soft ferrites, in particular, are among the most widely used magnetic materials having low cost, high performance for high-frequency applications. Mn-Zn ferrites have many important applications in this context [2].

Composites of ferrites and polymers may have very promising properties and represent a very diverse field of research. Silicon dioxide- sometimes known as silica - is one of the most abundant compounds in nature as sand or quartz and also it is manufactured in several forms. Previously published researches on ferrite/ SiO_2 composites had included investigations of the effect of silica and/or ferrite content on their magnetic properties [3-6], or investigation of some biological aspects such as their absorption behavior of bovine serum albumin by using UV [7]. The effect of varying complexing agent in the preparation of $Ni_{0.5}Zn_{0.5}Fe_2O_4$/20wt% SiO_2 nano-composites by auto-combustion on the structural and magnetic properties of the composites was also studied concluding that the complexing agent in the starting solution influenced the magnetic interaction between NiZn ferrite and silica, and then determined on the particle size [8]

The present work is devoted to investigate the effect of adding 25% weight of SiO_2 to four samples of Mn-Zn ferrites ($Mn_{1-x} Zn_x Fe_2O_4$; x = 0.8, 0.6, 0.4 and 0.2) on some of their dielectric and magnetic properties. Such an investigation has not been published before in literature as far as the authors know. The experimental findings of such a research may be of basic importance for materials designers in different applications.

2. Experimental

Four samples of Mn-Zn ferrites ($Mn_{1-x} Zn_x Fe_2O_4$; x = 0.8, 0.6, 0.4 and 0.2) were previously prepared by the conventional ceramic method. The details of preparation and characterization by X-ray diffraction, IR and Mossbauer spectroscopy had been previously published [9]. Part of the powders of different compositions was pressed to disc-shaped pellets. This group of pure ferrites represents the first category of the investigated samples. Some other part of the ferrites was mixed with 25% weight of $Si\ O_2$ and pressed to pellets also. These composite samples represent the second category. Some dielectric and magnetic properties were investigated for the two categories of samples.

The ac conductivity, dielectric constant, and loss tangent tan δ are studied as functions of frequency at different temperatures by using the circuit shown in reference [10]. The surfaces of each disc-shaped pellet have been coated with silver paste to improve the contact with two electrodes inside an evacuated silica tube cell surrounded by a home-made heater. In this circuit, the sample is connected in series with a resistor R that must be adjusted such that the value of V_R is small enough ($\leq 1\%$) compared to the applied voltage, V, (r.m.s = 1 Volt). The lock in amplifier (Stanford Research Systems SR 510) reads the voltage drop, V_R, on the resistance, R, and the phase angle, ϕ, between the applied voltage, V, and the current I passing through the resistor R. The current through the sample can be considered as formed of two parts, the displacement current, I_C, and the conduction current I_r which lags behind I_C by an angle of 90°. V_R and ϕ were measured as a function of temperature and frequency for all samples of the two categories. AC conductivity σ_{ac}, dielectric constant ε' and dielectric loss tanδ were calculated as functions of frequency and/or temperature and composition (from the experimental values of V, V_R, ϕ, $\omega = 2\pi f$, R, d, A; where d is the thickness of the sample (cm) and A is its surface area (cm^2)) by using the following equations:

$$\sigma'_{ac} = \frac{d}{rA} = \frac{dV_R \cos\phi}{AVR} \quad (1)$$

$$\varepsilon' = \frac{C}{C_o} = \frac{11.3 dV_R \sin\phi}{RA\omega} \times 10^{12} \quad (2)$$

$$\tan\delta = \frac{1}{\tan\phi} = \frac{I_r}{I_C} \quad (3)$$

Finally, mass susceptibility of all samples was measured at room temperature by an MS2B magnetic susceptibility meter (Bartinglon Instrument Ltd.). These values of the mass susceptibility were displayed as function of composition represented by the Zn content (x) in the two investigated categories.

3. Results and discussion

3.1 The dielectric constant behavior

The variation of the dielectric constant, with the frequency in logarithmic scale at different temperatures for the samples of the two categories under investigation is shown in Fig. (1:a-d) for the ferrites samples and in Fig. (4:a-d) for the composites samples. In either category, the dielectric constant, ε', is found to decrease as the frequency increases. It can be seen that this decrease is relatively sharp at lower frequencies, while it is slower at higher frequencies. The high values of dielectric constant – especially in the first category of pure ferrites - are often explained in literature on the basis of Koops phenomenological theory [11]. That is, in ferrites the system may be thought of as consisting of well-conducting grains separated by highly resistive grain boundaries [12]. The charge carriers upon reaching the grain boundary by hopping encounter different resistance so that accumulation of charges at the separating boundaries occurs causing what is called interfacial polarization and hence the dielectric constant value is highly raised [13-15]. As we can see a hopping electron in the direction of the external applied field increases the dielectric polarization, therefore, there is a strong correlation between the conduction mechanism and the dielectric behavior in ferrites or in other words the mechanism of the polarization process in ferrites is similar to that of the conduction process [16]. This assumption is enhanced by comparing the results of the first category of pure ferrites to the second one of composites containing silicon dioxide. Where, it is obviously seen that the values of the dielectric constant of the composites are relatively lower than those of the pure ferrite samples in consistency with the conductivity measurements displayed in Fig. (3; a-d) and Fig. (6:a-d) below.

However, as the frequency of the applied field is increased, the dielectric constant decreases until reaching a constant value, because beyond a certain frequency of the applied electric field the polarization cannot follow its alternations [17].

The variation of the dielectric constant with temperature for the samples of the two categories under investigation is also obviously shown in Fig. (1:a-d) and Fig. (4:a-d). Generally, an increase in the dielectric constant is observed with increasing temperature, the increase being quite significant at lower frequencies. The increase in temperature thermally activates the charge carriers, increasing the electron hopping, thereby enhancing the dielectric constant.

It is worth mentioning that there are four main contributions to the polarization in ferrites viz. electronic, atomic, dipolar and interfacial polarization. Electronic and atomic polarization are important at high frequencies and are temperature independent, while the latter two play dominant role at lower frequencies and they are both temperature dependent. Also, interfacial polarization is known to increase with temperature whereas dipolar polarization decreases with temperature. The observed increase in dielectric constant with temperature at low frequency is supportive of the fact that interfacial polarization is the main contributor to the dielectric constant in the present samples.

The composition dependence (Zn concentration (x) and SiO_2 addition) of the dielectric constant of the two categories of samples at room temperature and at applied frequency of 100 KHz is shown in Fig. (7). It can be noticed that the variation of ε' with Zn content has the same trend in the pure samples and in the silicon dioxide containing samples, with a significant decrease in value in the composites samples as mentioned above. This similarity in variation with the Zn content in the two categories reinforces the validity of the measurements and also it can be interpreted according to the cationic distribution in the pure ferrite samples previously published [9]. This cationic distribution had been proposed according to Mossbauer spectroscopy analysis. At first when x = 0.2, Mn had a fraction of 0.12 in the A-site and 0.68 in the B-site while the fraction of Fe ions in the A-site had been 0.68 and 1.32 in the B-site. Hen at x = 04, the fractions had become for Mn 0.132 and 0.468 and for iron 0.468 and 1.532 respectively. Upon further increase of Zn content – with the well known A-site preference of the Zn ions at x = 0.6 and x= 0.8, all Mn ions had occupied the B- sites only, along with an increase in the fraction of Fe ions in the B- sites. Bearing in mind that the existence of cations with different valencies of the same element in the same site is the origin of hopping conduction mechanism in ferrites, we can obviously see that the slight increase of the dielectric constant from the sample of x = 0.2 to the sample of x= 0.4 is quite logic. The decrease upon changing x = 0.4 to x = 0.6 may be attributed to the decrease of the fraction of Mn^{+2} - Mn^{+3} pairs on the B-sites associated with a very small and the insignificant increase of Mn_{1-x} Zn_x pairs. At x = 0.8 the decrease in Mn fraction on the B-site (0.2) had been well compensated by a significant increase of the Mn_{1-x} Zn_x pairs causing an increase in hopping of charge carriers and consequently in the dielectric constant.

3.2 The dielectric loss behavior

The variation of dielectric loss tangent (tan δ) as a function of frequency (f) at different temperatures for the two categories of samples is shown in Fig. (2: a-d) and (4: a-d). The figures show the normal expected behavior with frequency, i.e., tan δ decreases with increasing frequency. The dielectric loss decreases rapidly at low frequency region, while rate of decrease is slow in high frequency region and then becomes almost frequency independent. Such a behavior can be explained on the basis that in low frequency region, which corresponds to high resistivity (due to the grain boundary), more energy is required for electron exchange between Fe^{2+} - Fe^{3+} pairs in the octahedral sites, as a result the loss is high. Moreover this is in consistency with the mathematical definition of tan δ which is equal to ε''/ ε'; where ε'' represents a measure of the resistive current and ε' represents a measure of the capacitive current in the electrical equivalent circuit of the sample. In high frequency region, which corresponds to low resistivity (due to grains), a small energy is required for electron transfer between Fe ion pairs at the octahedral site. Moreover, the dielectric loss tangent also depends on number of factors, such as stoichiometry, Fe^{2+} and Mn^{2+} contents, and structural homogeneity, which in turn depends upon the composition of the samples, this may interpret the slight increase of loss tangent in the second category of composite samples than the corresponding pure ferrite samples of the first category [13] .

Again, it can be seen that the dielectric loss of all the samples increases with an increase of temperature at the same frequency, which is expected as the temperature increases, the resistivity of the samples decreases [13].

3.3 The ac conductivity behavior

The complex ac conductivity is given by

$$\sigma^*(\omega) = \sigma'_{ac} + i\sigma''_{ac} \qquad (4)$$

Where, the real part of this function consists of two components:

$$\sigma'_{ac} = \sigma_1(T) + \sigma_2(T,\omega) \qquad (5)$$

The first term, $\sigma_1(T)$ is actually the dc electrical conductivity $\sigma_{dc}(T)$, it is temperature dependent and frequency independent. It is related to the drift mobility of electric charge carriers and obeys an Arrhenius relation [18]. The second term $\sigma_2(T,\omega)$ is frequency and temperature dependent, it is attributed to the dielectric relaxation caused by the localized electric charge carriers and obeys the power law form:

$$\sigma_2(T,\omega) = A\omega^s \qquad (6)$$

Where, A and s are composition and temperature dependent parameters [19-20]

Fig. (3:a-d) and Fig. (6:a-d) display the frequency dependence of σ'_{ac} of the samples of both categories; pure ferrites and composites respectively. As obviously seen the measurements taken at different temperatures reveal the semiconducting nature of all samples. And since it is well established that the conduction process has a strong correlation with the interfacial polarization in ferrites , therefore all the argument above considering the variation of dielectric constant with frequency and composition is applicable to the ac conductivity. Fig. (8) enhances this assumption as well.

3.4 Magnetic susceptibility measurements

The mass susceptibility values of all samples measured at room temperature by an MS2B magnetic susceptibility meter are displayed in table 1 and is graphically presented as function of composition in Fig. (9) for the two investigated categories. The behavior with the Zn content (x) is the same in the two categories enhancing the accuracy of the measurements and has almost the same order of magnitude in both categories. His may be useful in applications that require increasing resistivity while keeping the magnetic susceptibility in the same order of magnitude.

Table (1)

***Mass susceptibility at room temperature measured for the composition $Mn_{1-x}Zn_xFe_2O_4$ with and without* sio₂**

Zinc content	χ_m (emu/g) x 10^{-6} (measured by a susceptibility meter)	
X	Pure ferrites	Composites of ferrites + 25% SiO_2
0.2	3.0410	1.4005
0.4	3.6270	2.0259
0.6	2.9465	1.6714
0.8	2.2551	1.4698

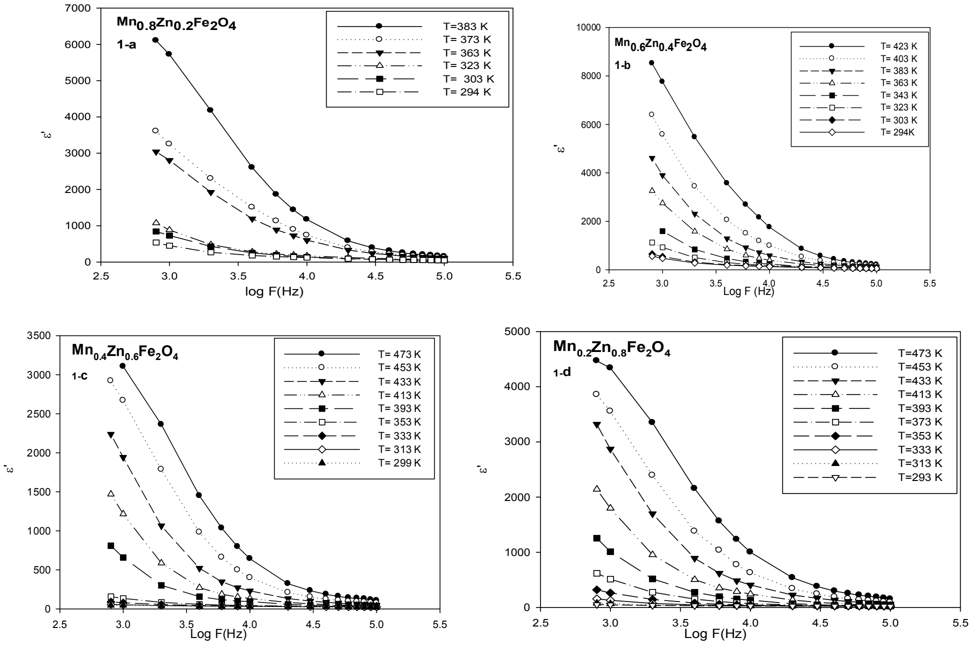

Fig.(1) the relation between έ vs. log F for the samples $Mn_{1-x}Zn_xFe_2O_4$; (x= 0.2, 0.4, 0.6, 0.8)

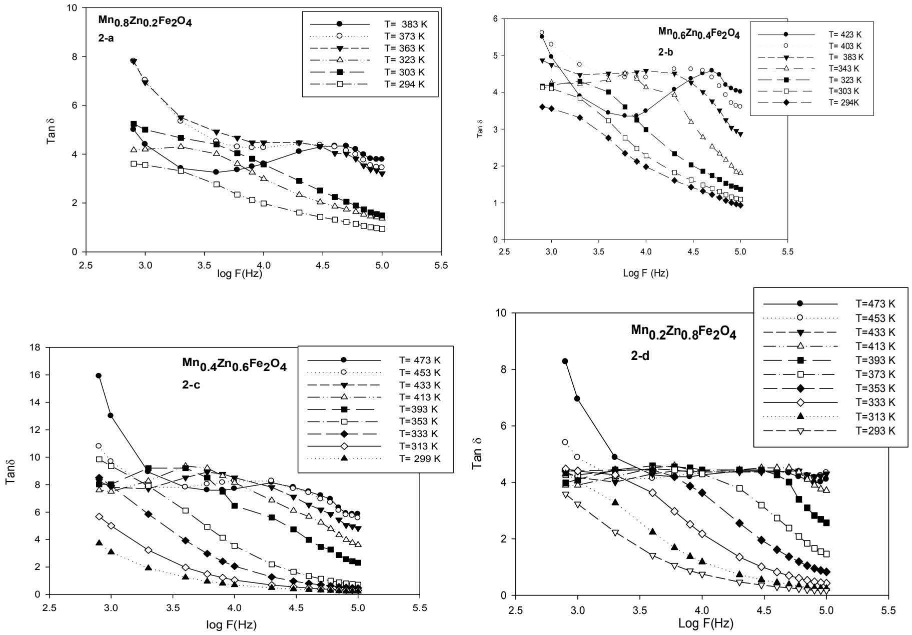

Fig.(2) the relation between tanδ vs. log F for the samples $Mn_{1-x}Zn_xFe_2O_4$; (x= 0.2, 0.4, 0.6, 0.8)

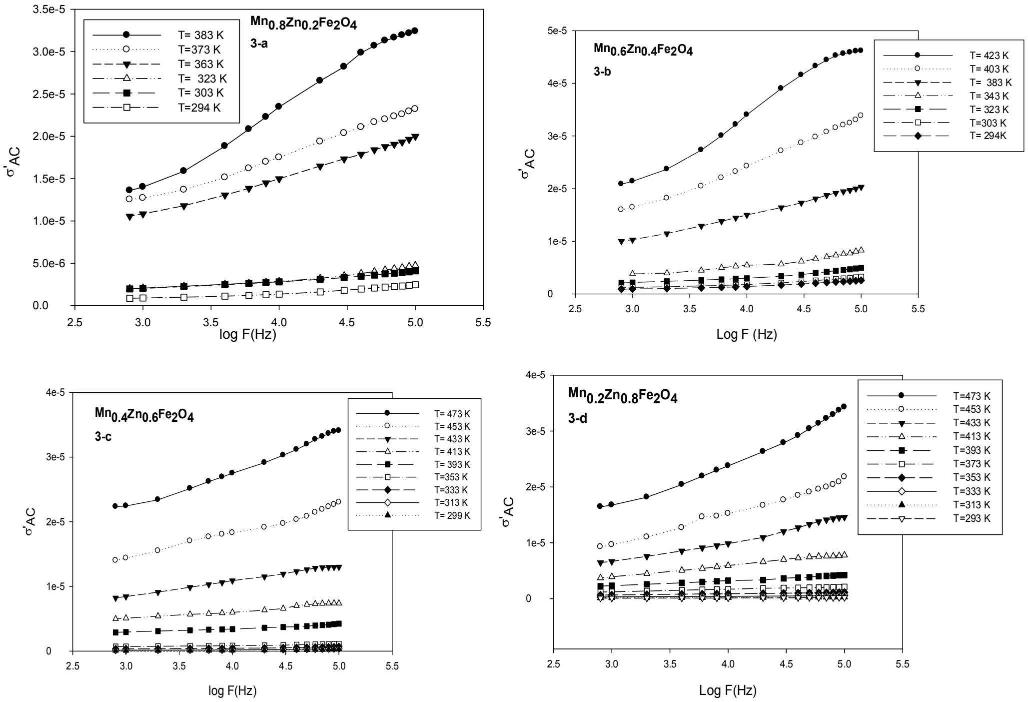

Fig.(3) the relation between σ'$_{Ac}$ vs. log F for the samples $Mn_{1-x}Zn_xFe_2O_4$; (x= 0.2, 0.4, 0.6, 0.8)

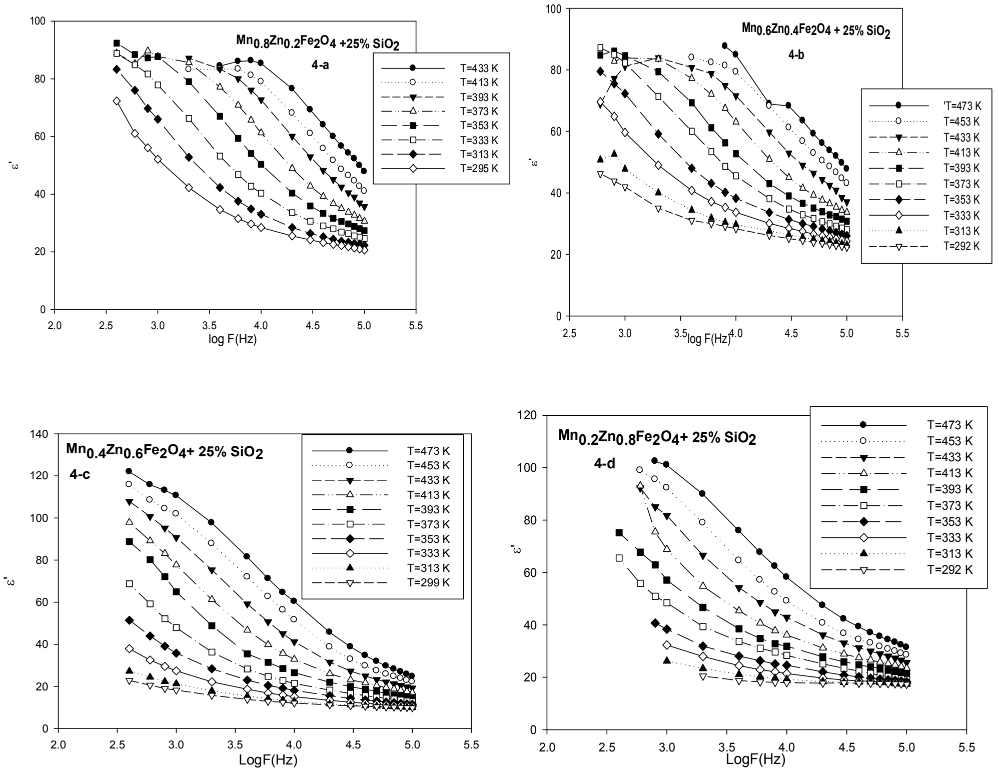

Fig.(4) the relation between έ vs. log F for the samples $Mn_{1-x}Zn_xFe_2O_4$; (x= 0.2, 0.4, 0.6, 0.8)with 25%w. of SiO_2

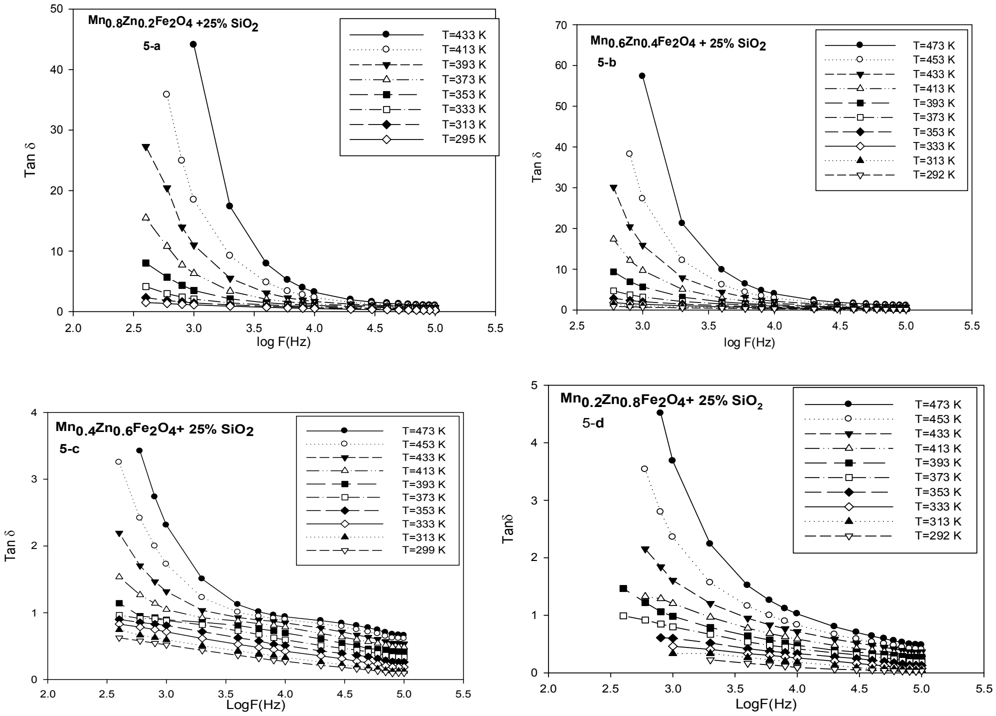

Fig.(5) the relation between tanδ vs. log F for the samples $Mn_{1-x}Zn_xFe_2O_4$; (x= 0.2, 0.4, 0.6, 0.8) with 25%w. of SiO_2

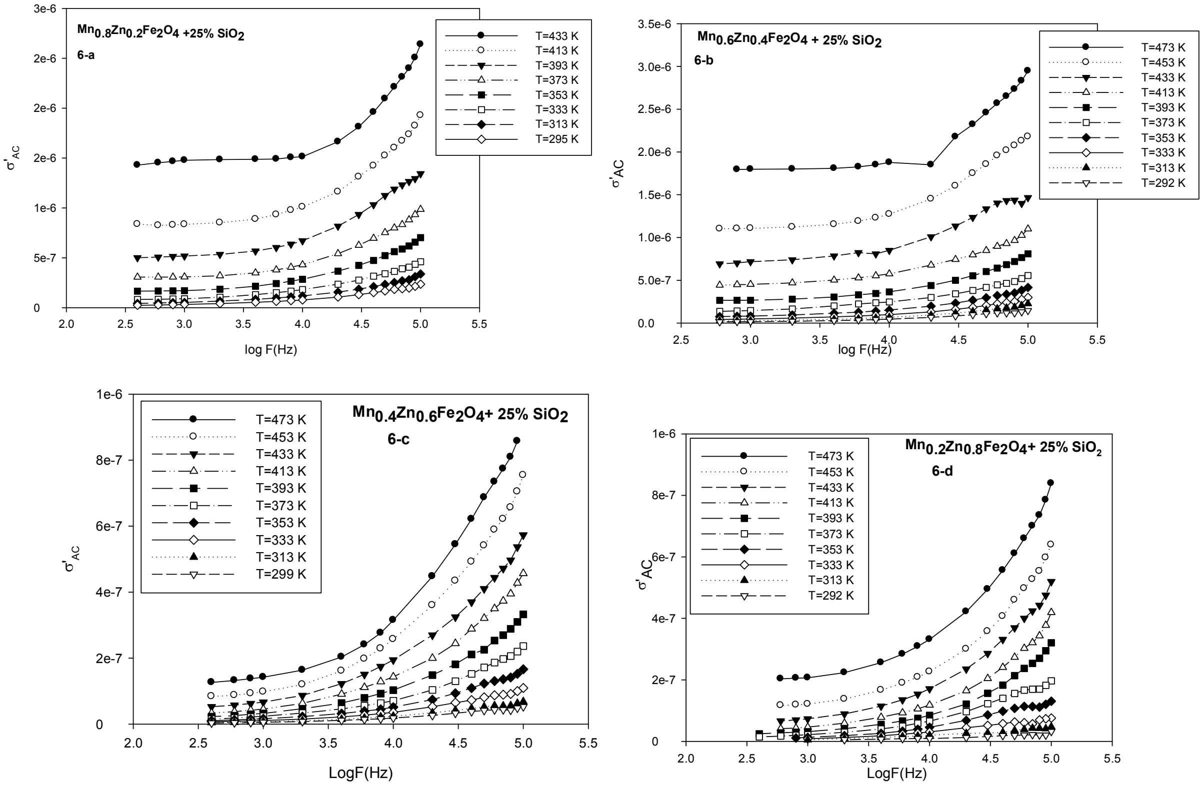

Fig.(6) The relation between σ'_{Ac} vs. log F for the samples $Mn_{1-x}Zn_xFe_2O_4$; (x= 0.2, 0.4, 0.6, 0.8) with 25%w. of SiO_2

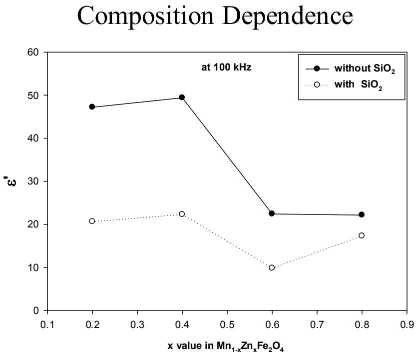

Fig.(7) the relation between έ vs. x values of the samples $Mn_{1-x}Zn_xFe_2O_4$; (x= 0.2, 0.4, 0.6, 0.8) without and with 25%w. of SiO_2

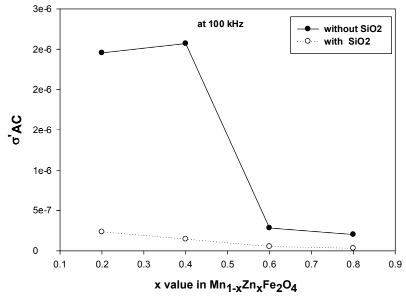

Fig.(8) the relation between σ'_{Ac} vs. x values of the samples $Mn_{1-x}Zn_xFe_2O_4$; (x= 0.2, 0.4, 0.6, 0.8) without and with 25%w. of SiO_2

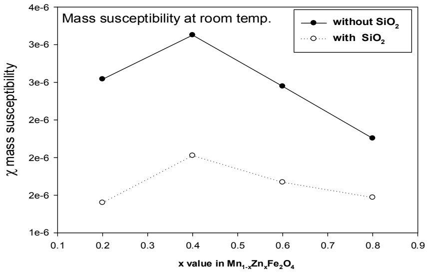

Fig.(9) the relation between Mass susceptibility vs. x values of the samples $Mn_{1-x}Zn_xFe_2O_4$; (x= 0.2, 0.4, 0.6, 0.8) at room temperature without and with 25%w. of SiO_2

Conclusion

The strong well known correlation between conduction mechanism and interfacial polarization mechanism in ferrites is also observed in composites of ferrites and 25% of SiO_2. Therefore, the addition of SiO_2 to the investigated samples of ferrites has decreased both of the conductivity and the dielectric constant. Whereas, for the magnetic susceptibility, the pure ferrite samples and the composite samples have the same trnd of behavior with composition and almost the same order of magnitude.

Acknowledgement

The authors are very grateful to Prof. Dr. M. Amer for providing them by the pure ferrite samples used in the present investigation.

Corresponding authors

D. El Kony
Physics Department, Faculty of Science, Tanta University, Tanta, Egypt.
samiasaafan@yahoo.com

References

[1] Hashim Alimuddin M., , Shalendra Kumar, Sikander Ali , Koo B.H., Chung H., Kumar R., J. Alloys Compd. 511 (2012) 107– 114.

[2] Thakur A., Mathur P. and Singh M., J. Phys. & Chem. of solids 68 (2007) 378-381.

[3] Praveena K., Sadhana K., Ramana Murthy S., Journal of Magnetism and Magnetic Materials 323 (2011) 2122–2128.

[4] Sunil Rohilla, SushilKumar, Aghamkar P., Sunder b S., Agarwal A., Journal of Magnetism and Magnetic Materials 323 (2011) 897–902.

[5] Wua K.H., Changa Y.C., Changa T.C., Chiub Y.S., Wub T.R., Journal of Magnetism and Magnetic Materials 283 (2004) 380–384.

[6] Shuping Zhang , Dawei Donga, Yu Suia, Zhiguo Liu , Hongxia Wang , Zhengnan Qian , Wenhui Sua, Journal of Alloys and Compounds 415 (2006) 257–260.

[7] Ruijiang Liua, Xiangqian Shena, Chengtao Jianga, Fuzhan Songa, Hongxia Li, Journal of Alloys and Compounds 511 (2012) 163– 168.

[8] Wua K.H., Tinga T.H., Lia M.C., Ho W.D., Journal of Magnetism and Magnetic Materials 298 (2006) 25–32.

[9] Amer M.A., phys. Stat. sol. (a) 151, (1995) 205.

[10] Elkestawy M.A., Journal of Alloys and Compounds 492 (2010) 616–620.

[11] Koops C. G., Phys. Rev. 83 (1) (1951) 121.

[12] Wang H.B., Liu J.-H., Li W.F., Wang J.-B., Wang L., Song L.-J., Yuan S.-J., Li F.-S., J. Alloy. Comp., 461 (2008) 373–377.

[13] Batoo K. M., Kumar S., Lee C. G., Alimuddin, J. Curr. Appl. Phys. 9 (2009) 1072-1078.

[14] Soibam I., Phanjoubam S., Sharma H.B., Sarma H.N.K., Laishram R., Prakash C., Solid State Comm. 148 (2008) 399-402.

[15] Thakur A., Mathur P.. Singh M., J. Phy. Chem. Sol., 68 (2007) 378-381.

[16] Ravinder D. , Latha K., J. Mater. Lett. 41(1999) 247-253.

[17] Zaky A.A. and Hawley R., "Dielectric Solids", New York: Dover Publications Inc., 1988.

[18] El-Nim M.K. r, Moharram B.M., Saafan S.A., Assar S.T., J. of Magn. Magn. Mater. 322 (2010) 2108–2112.

[19] Abdeen A.M., J. Magn. Magn. Mater. 185 (1998) 199–206.

[20] M.A. El Hiti, J. Magn. Magn. Mater. 164(1996) 187-196.

9/5/2012