AC Conductivity and Dielectric Properties of Mn Mg Zn-ferrite /polystyrene composite

D. El Kony & M. A. Elkestawy *

Physics Department, Faculty of Science, Tanta University, Tanta, Egypt
**Physics Department, Faculty of Science, Suez Canal University, Suez, Egypt.*

The ac conductivity and dielectric properties of four composites prepared by mixing $Mn_{0.8-x}Mg_xZn_{0.2}Fe_2O_4$ (x = 0, 0.4) with polystyrene (30% wt. and 45% wt.), were investigated as a function of frequency over a limited range of temperature – to avoid melting of polystyrene - by using a complex impedance technique. The ac conductivity shows the expected increase with increasing frequency. The conductivity shows an interesting change in behavior from an NTC thermistor behavior to a semiconductor like behavior. This suggests further studies to explore new applications. Moreover, the dielectric constant and the dielectric loss tangent (tan δ) have low values suggesting that the studied composites may be good candidates for some practical applications that require minimization of eddy currents.

1. Introduction:

Spinel ferrites have been investigated intensively in recent years due to their potential applications in non-resonant devices, radio frequency circuits, high quality filters, rod antennas, transformer cores, read/write heads for high-speed digital tapes and operating devices [1- 7] Most commercially important ferrites are consisted of mixed ferrites which actually consist of solid solutions of the various simple ferrites. The effect of each simple ferrite can be used to balance or optimize a specific property of the final mixed ferrite. For example, manganese zinc ferrites have higher permeability at low or medium frequencies while magnesium ferrite, $MgFe_2O_4$, which is a soft magnetic n-type semiconducting material, is considered as a cheaper constituent and may be added to other ferrites to enhance their resistivity [1]. Moreover, for polymer composites to be used in electric packaging, they must have a good combination of thermal and dielectric properties .The aim of the present research is to study the effect of introducing both Mg and polystyrene on the dielectric properties of the composition $Mn_{0.8}Zn_{0.2}Fe_2O_4$ at different frequencies over a limited range of temperature - because the glass transition temperature of polystyrene is known to be 100.9^{o}C [9].

2. Experimental Techniques:

$Mn_{0.8-x}Mg_xZn_{0.2}Fe_2O_4$ (x = 0, 0.4) was prepared by the conventional ceramic method and the X-ray diffraction results (Table.1) confirms the formation of the required spinel ferrite as identified by the ICDD cards No. [74-2397 and 88-1942 and 89-0599] . Polystyrene was mixed with the two ferrite samples by two ratios (30% wt. and 45% wt.) at room temperature to obtain four composite powder samples, these two ratios are chosen so that the polystyrene properties don't dominate, causing the loss of ferrite known benefits in various applications . The samples were thoroughly ground for about 6-8 hours to reach homogeneity then pressed into pellets. Dielectric analysis (DEA) was performed for the four samples. DEA measures the electrical properties of a material as a function of temperature, and frequency; where two fundamental electrical characteristics of materials are investigated; the capacitive (insulating) nature, which represents its ability to store electrical charge; and the conductive nature, which represents its ability to transfer electrical charge [10]. The surfaces of each pellet were coated with a thin layer of silver paste to provide good Ohmic contact to the external electrodes. The measurements were carried out inside an evacuated silica tube by using a complex impedance technique [11]. The circuit is illustrated in Figure (1); where the sample is labeled "S" and the thermocouple is not shown in figure for inconvenience but in fact it is touching the surface of the sample. Ac voltage V with frequency f is applied to the sample. A small resistor R is connected in series with the sample. The current is given by ($I=V_R/R$) where V_R is the drop voltage on R. A lock-in amplifier (lock-in amplifier Stanford SR 510 type) is used in the circuit to measure simultaneously the voltage V_R, the frequency of the applied voltage f and the phase difference between the applied ac voltage V and V_R. From those readings the ac conductivity (σ'_{ac}), the dielectric constant (ε') and the dielectric loss tangent ($\tan \delta$) can be calculated.

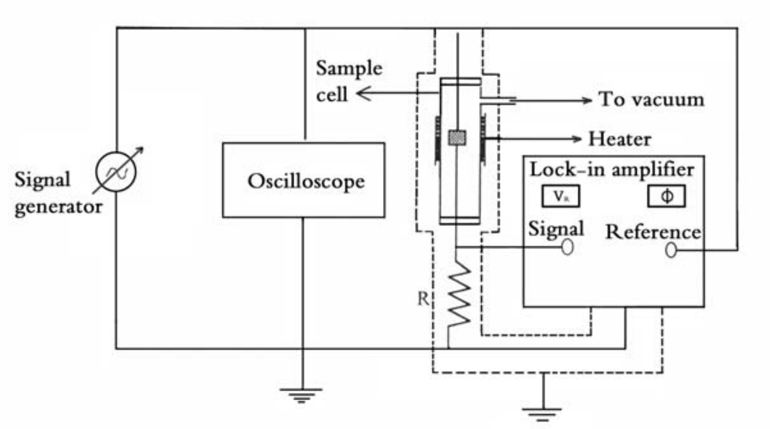

Fig. (1): Block diagram of the circuit used for ac measurements.

Table(1):d-spacing, relative intensities and (hkl) plans of the system Mg0.4Mn0.4Ze0.2Fe2O4 and Mg0.8Zn0.2Fe2O4

$Mg_{0.8}Ze_{0.2}Fe_2o_4$			$Mg_{0.4}Mn_{0.4}Zn_{0.2}Fe_2O_4$			
Angle 2-Theta	d-value Angstrom	Intensity%	Angle 2-Theta	d-value Angstrom	Intensity%	hKL
30.08	2.97	38.3	29.9	2.97	27.1	220
35.42	2.54	100	35.3	2.53	100	311
37.05	2.44	6.9	36.87	2.42	7.8	222
43	2.11	17.2	42.85	2.1	31.5	400
53.3	1.72	11.5	53.15	1.72	20.6	422
56.9	1.62	37.3	56.63	1.62	38.1	511
62.49	1.488	29.2	62.4	1.49	35.8	440
73.9	1.287	5.1	73.52	1.28	5	533

3. Results and Discussion:

3.1. AC Conductivity and Dielectric Properties

Figure (2: a-d) illustrates the frequency dependence of the real part of the complex ac conductivity σ'_{ac} for the four investigated composites: $Mn_{0.8-x}Mg_xZn_{0.2}Fe_2O_4$ (x = 0, 0.4) with polystyrene content (30% wt. and 45% wt.), at different values of temperature. By checking out Figure (2: a-d) where (a) $Mn_{0.8}Zn_{0.2}Fe_2O_4$ with polystyrene content (30% wt.), (b) $Mn_{0.8}Zn_{0.2}Fe_2O_4$ with polystyrene content (45% wt.), (c) $Mn_{0.4}Mg_{0.4}Zn_{0.2}Fe_2O_4$ with polystyrene content (30% wt.) and (d) $Mn_{0.4}Mg_{0.4}Zn_{0.2}Fe_2O_4$ with polystyrene content (45% wt.), we can find that the conductivity has low values ranging from (1.57×10^{-10}

to 2.212×10^{-7} $\Omega^{-1}cm^{-1}$) and increases with increasing frequency as expected in heterogeneous dielectrics where it is well known that any solid consisting of phases with different conductivity has an overall conductivity which increases with frequency. This is because at high frequencies localized charge motion makes it possible to take maximum advantage of well conducting regions, while at lower frequencies charge transport must extend over longer distances and is limited by poorly conducting regions [12] in other words at high frequencies when 1/ωc is minimum, a maximum conduction in the highly conducting regions is achieved- giving the observed increasing conductivity with increasing frequency. It is also observed that by increasing temperature, the conductivity slightly decreases until the temperature becomes 343 K, after which the conductivity slightly increases. This behavior is equivalent to a reverse in the direction of slope in a σ'_{ac} vs. T graph which has been reported before by others in some Mn Zn ferrite samples and Ni Cu Zn ferrite samples too [13, 14]. Such a change in the slope is sometimes attributed to a change in the activation energy of the conduction process [15].

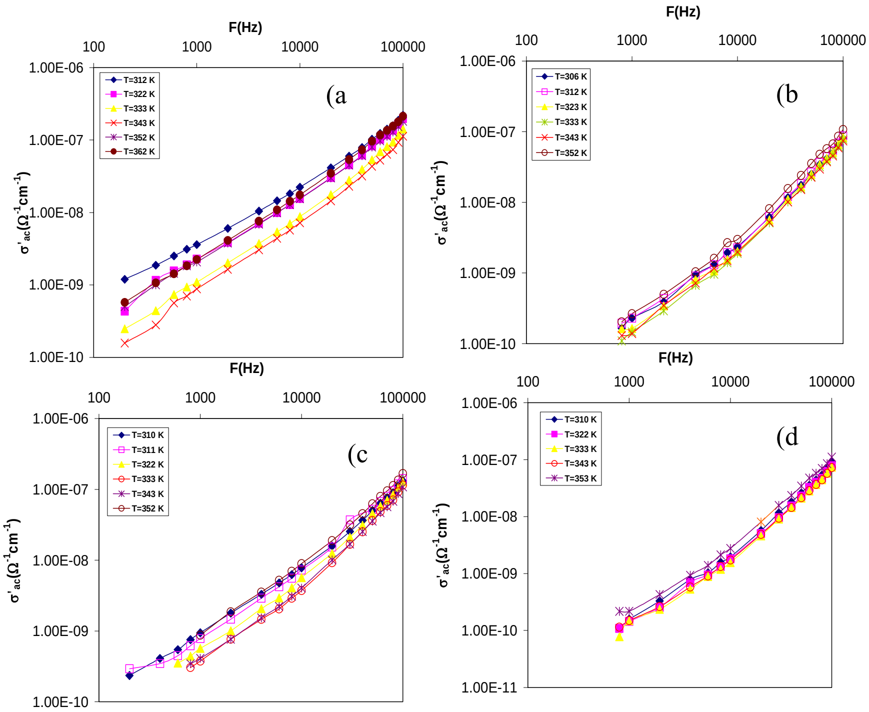

Fig. (2): σ'_{ac} ($\Omega^{-1}cm^{-1}$) versus F (Hz) at selected temperatures for the studied samples where (a) $Mn_{0.8}Zn_{0.2}Fe_2O_4$ with polystyrene content (30% wt.), (b) $Mn_{0.8}Zn_{0.2}Fe_2O_4$ with polystyrene content (45% wt.), (c) $Mn_{0.4}Mg_{0.4}Zn_{0.2}Fe_2O_4$ with polystyrene content (30% wt.) and (d) $Mn_{0.4}Mg_{0.4}Zn_{0.2}Fe_2O_4$ with polystyrene content (45% wt.).

Moreover, the behavior of the conductivity in the first range of temperature (up to 343K) is similar to the behavior of a thermistor having a negative thermal coefficient (NTC), and it is repeated and confirmed in the four samples so that it may be recommended to further study this point carefully to explore some potential applications of these composites.

Figure (3: a-d) shows that the qualitative behavior of the dielectric constant ε' as a function of frequency F (at some selected temperatures) is almost similar in the four investigated samples whereas for the quantitative behavior, it is observed that the dielectric constant ε' of the samples with polystyrene percentage 45% wt. are less than of those having polystyrene percentage 30% wt., this is in agreement with expectation since it is known that the pure polystyrene has a low value of dielectric constant ranging from 2 to 3 [16]. Also as expected a dispersion appears with frequency where the polarization cannot continue to follow the variation of the field as the frequency increases so that the dielectric constant decreases. In addition, in ferrites, there is a strong correlation between the conduction mechanism and the dielectric behavior. In other words the mechanism of the polarization process in ferrites is similar to that of the conduction process where it is observed that the electronic exchange between Fe^{2+} and Fe^{3+} ions results in local displacements which determine the polarization [17] and simultaneously the conduction. The results of the present work also show that the dielectric constant έ and electrical conductivity σ_{ac} follow the same trend; where, as the temperature increases the dielectric constant ε' decreases until 343 K after which the dielectric constant begin to increase with increasing temperature. This behavior is in consistency and confirms the preceding discussion of the conductivity.

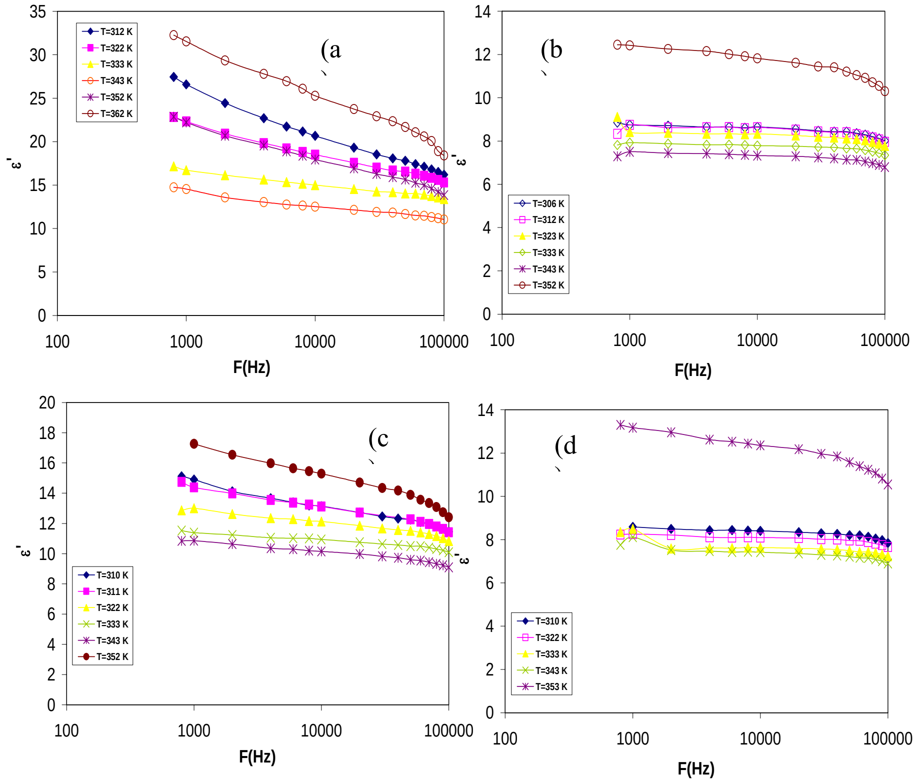

Fig. (3): Dielectric constant ε' versus F (Hz) at selected temperatures for the studied samples where (a) $Mn_{0.8}Zn_{0.2}Fe_2O_4$ with polystyrene content (30% wt.), (b) $Mn_{0.8}Zn_{0.2}Fe_2O_4$ with polystyrene content (45% wt.), (c) $Mn_{0.4}Mg_{0.4}Zn_{0.2}Fe_2O_4$ with polystyrene content (30% wt.) and (d) $Mn_{0.4}Mg_{0.4}Zn_{0.2}Fe_2O_4$ with polystyrene content (45% wt.).

Figure (4: a-d) shows the behavior of the loss factor (tanδ) as a function of frequency F (at some selected temperatures). At most of the selected temperatures as the frequency increases the loss factor (tanδ) increases at first slightly then considerably with increasing frequency as if it going to display a maximum but out of the range of measurements .It may be noticed that at the temperature 312K in the first sample Fig(4-a),there is at first a decrease in tanδ with increasing frequency ,which may be an indication of a previous maximum of tanδ at just a lower frequency (out of the range of the measurements) suggesting that the rate of hopping of charge carriers at this frequency may coincide with the applied frequency[18,19].

The increase in tanδ with frequency may be attributed to the increase in ε'' – which represents the resistive current and is proportional with the conductivity – with increasing frequency by a faster rate than that of decreasing ε' in the defining relation of (tanδ): tanδ = ε''/ε' [20].

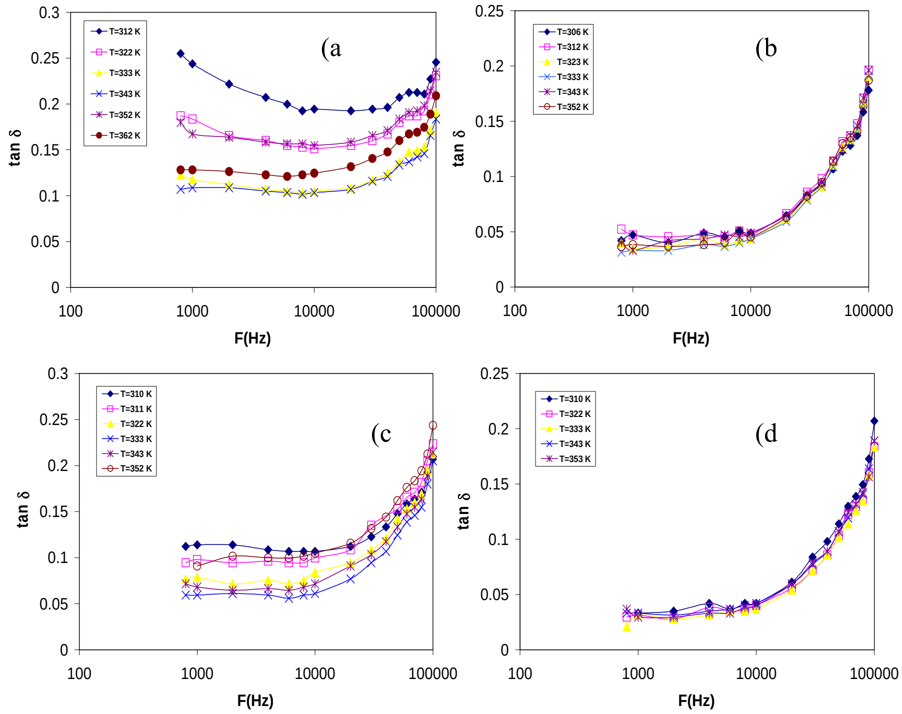

Fig. (4): tanδ versus F (Hz) at selected temperatures for the studied samples where (a) $Mn_{0.8}Zn_{0.2}Fe_2O_4$ with polystyrene content (30% wt.), (b) $Mn_{0.8}Zn_{0.2}Fe_2O_4$ with polystyrene content (45% wt.), (c) $Mn_{0.4}Mg_{0.4}Zn_{0.2}Fe_2O_4$ with polystyrene content (30% wt.) and (d) $Mn_{0.4}Mg_{0.4}Zn_{0.2}Fe_2O_4$ with polystyrene content (45% wt.).

4. Conclusion:

1- The samples show an interesting behavior of conductivity similar to an NTC themistor until the temperature becomes 343 K where by increasing temperature, the conductivity slightly decreases.
2- The dielectric constant as a function of temperature confirms this interesting behavior and also confirms the well-known correlation between the conduction mechanism and the polarization mechanism in ferrites.
3- This behavior is repeated in the four samples so that it may be recommended to further study this point carefully to explore some potential applications of these composites
4- The dielectric constant and the dielectric loss tangent (tan δ) have low values suggesting that the studied composites may be good candidates for some practical applications that require minimization of the eddy currents.

5. References:

2. Goldman, Modern Ferrite Technology, Marcel Dekker, Inc., New York, (1993).
3. S.K Prandhan, S. Bid, M. Gateshki, V. Petkov, *Materials Chemistry and Physics*, **93**, 224–230 (2005).
4. P. Ravindernathan, K.C. Patil, *J. Mater. Sci*. **22**, 3262 (1987).
5. H. Igarash, K. Ohazaki, *J. Am. Ceram. Soc*.**60**, 51 (1997).
6. A.Goldman, *Am. Ceram. Soc. Bull*. **63**, 582 (1984).
7. D. Stoppels, *J. Magn. Magn. Mater*. **160**, 323 (1996).
8. K.H. Lee, D.H. Cho, S.S Jeung, *J. Mater. Sci. Lett*. **16**, 83 (1997).
9. C.S. Kim,Y.S. Yi, K.T. Park, H. Namgung, J. G. Lee, *J. Appl. Phys*. 85, 5223 (1999).
10. Suzhu Yu, Peter Hing, and Xiao Hu, *J. Appl. Phys*., **86**, No. 1, 1July (2000).
11. Yu, Hig, and Hu, *J. Appl. Phys*., Vol. **88**, No. 1, 1 July (2000).
12. A.M. Ahmed, M.A. El Hiti, M.M. Mosaad and S. M. Attia, *Journal of Magnetism and Magnetic Materials* **146**, 84 (1995).
13. Jeppe C. Dyre and Thomas B. Schroder, "*Universality of ac conduction in disordered solids*". Reviews of Modern Physics, Vol. **72**, No. 3, 873 (2000).
14. E. Ateia, M.A. Ahmed, A.K. El-Aziz, *Journal of Magnetism and Magnetic Materials*, **311**, 545 (2007).

15. M. Kaiser, S. S. Ata-Allah, *Materials Research Bulletin* **44**, 1249 (2009).

16. J. Smitt, H.P.J. Wijn, Ferrites, John Wiley & Sons, New York, (1959).

17. Suzhu Yu, Peter Hing and Xiao Hu, *J. Appl. Phys*., **88**, No. 1, 1 July (2000).

18. D.Ravinder and K.V. Kumar, "*Dielectric behaviour of erbium substituted Mn–Zn ferrites*". Bull. Mater. Sci., **24**, 505 (2001) .

19. S. S. Ata-Allah, M.K. Fayek, *Phys. Stat. Sol*.,(a)**175**, 725 (1999).

20. M. A. Elkestawy, S. Abdelkader, M. A. Amer, *Physica* **B405**, 619 (2010).

21. K. Jonscher, Universal Relaxation Law, Chelsea dielectric spress, London, (1996).