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Infrared Absorption and Dielectric Properties of Mg-Zn Ferrite

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Absract

The mixed ferrites Mg_{1-x} Zn_x Fe_2O_4 (where x=0.0,0.2,0.3,0.4,0.5and 0.6) were prepared by the ceramic method. Infrared absorption (IR) and dielectric properties were used to investigate this ferrite. The IR spectra in the range from 200 to 1000 cm^{-1} were reported and only, two bands were observed. The band υ_A around 560 cm^{-1} and the band υ_B around 400 cm^{-1} were assigned to the tetrahedral and octahedral complexes, respectively. The a.c. conductivity of the above compositions was studied in a wide range of temperature from 300 up to 720K. Over this range of temperature, the a.c. conductivity seems to be independent of frequency at high temperatures for all values of x. In the relatively low temperature regions, the frequency dissipation is attributed to the hopping conduction of the localized carriers. The dielectric constant έ, dielectric loss ε'' and loss tangent tan δ increase with increasing temperature. The dielectric behaviour is explained by using the mechanism of polarization process, which is correlated to that of electron exchange interaction.

Introduction

The ferromagnetic oxide known as ferrites have the general formula $M^{2+}Fe^{3+}O^{+4}$ where M is a divalent metallic ion such as Fe^{2+}, $Ni_2^{+,}$ Mg^{2+},Cu^{2+} and Zn^{2+} or mixture of these ions. Ferrites and the ferromagnetic oxides, are suitable materials for high frequency application [1]. It is well known that the intrinsic properties of ferrites largely depend on chemical composition and preparation conditions. By introducing relatively small amount of foreign ions, the structural and electrical properties can be modified [2,3]. The electrical conductivity in Ni containing Zn-Mg ferrites [4], Mg - ferrites [5] and Zn-Mg ferrite [6] were studied.

In this work the dielectric properties of Mg_{1-x} Zn_x Fe_2 O_4 (x = 0, 0.1, 0.3, 0.4, 0.5, and 0.6) are reported at four constant frequency 10^2, $10^{3,}$ 10^4, and 10^5 Hz in the temperature range of (300- 700K), and the conduction mechanism was discussed.

Experimental

Polycrystalline samples of mixed ferrites $Mg_{1-x}Zn_xFe_2O_4$ (with x = 0.0,0.1,0.2,0.3,0.4,0.5 and 0.6) were prepared by a standard ceramic method. The details of the method of preparation were given in [7]. The IR spectra in the range 200-1000 cm^{-1} were recorded at room temperature using a Perking Elmer infrared spectrometer model 1430. The ac conductivity was measured at four constant frequencies (10^2, $10^{3,}$ 10^4, and 10^5 Hz) over a wide range of temperature (300- 720K) using Stanford – research system (model SR720) LCR meter) with a personal computer which has a basic accuracy of 0.05%. The drive voltage of LCR meter used in the present work is $0.5V_{rms}$

Results and Discussion

IR Absorption Spectra

The IR spectra of the investigated composition $Mg_{1-x}Zn_xFe_2O_4$ recorded in the range of 200-1000 cm^{-1} are shown in Fig. (1) and the absorption bands are listed in Table (1)**.**

Table 1: The IR absorption bands of $Mg_{1-X}Zn_xFe_2O_4$

A- Site				B- Site							
X	υ_A cm^{-1}	I_A%	Γ_A cm^{-1}	υ_B cm^{-1}	I_B%	Γ_B cm^{-1}	I_A / I_B	Γ_A / Γ_B	F_1 X 10^6 dyne cm^{-1}	F_2 X 10^6 dyne cm^{-1}	$\theta_{D,}$ K
0.0	565	42.5	160	420	22	75	1.93	2.13	2.927	1.579	704
0.1	560	32	130	415	14.5	70	2.20	1.86	2.876	1.579	701
0.2	560	32	135	405	12.5	95	2.56	1.42	2.876	1.467	690
0.3	560	35.5	130	410	14.5	100	2.45	1.3	2.876	1.542	697
0.4	555	31	160	405	19	145	1.63	1.1	2.835	1.467	687
0.5	450	31	125	405	14	115	2.21	1.09	2.77	1.542	690
0.6	550	25.5	135	405	10	110	2.55	1.23	2.77	1.467	683

According to Waldron [8] the ferrites can be considered continuously bonded crystals, meaning that the atoms are bonded to all nearest neighbourse by equivalent forces. In ferrites the metal ions are situate in two different sublattices designated tetrahedral (A-site) and octahedral (B-site) according to the geometrical configuration of the oxygen nearest neighbours. Waldron [8] and Hafner [9] have attributed the band around 600 cm^{-1} to stretching vibrations of the tetrahedral groups (υ_A) and that around 400 cm^{-1} to the octahedral groups (υ_B). In the present study the absorption bands for Mg-Zn ferrites are found to be in the expected range of the high frequency band υ_A is in the range 550 – 565 cm^{-1} and the lower frequency band υ_B is in the range of 405-420 cm^{-1}.

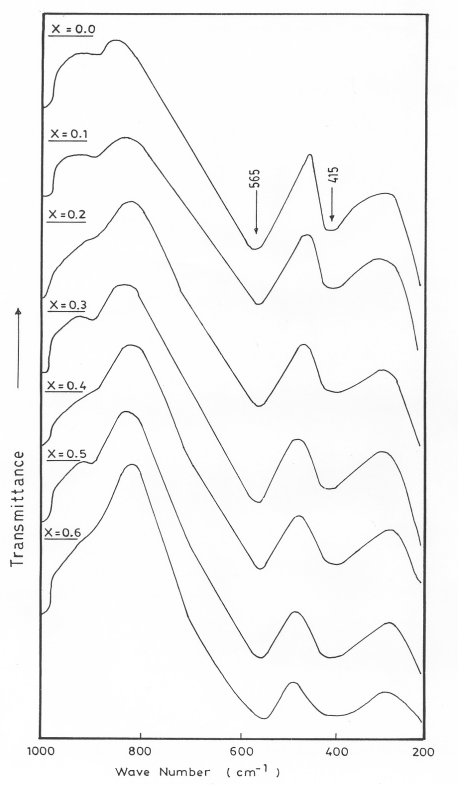

Figure 1: The IR absorption spectra of $Mg_{1-x}\ Zn_x\ Fe_2O_4$

The force constant of stretching vibration was calculated [10] for the A and B- sites and tabulated in Table 1. It was found to be in the ordinary value. Also, the Debye temperature of the investigated composition was calculated using the relation [8];

$$\theta_D = \lambda c.\upsilon_{av} = 1.438\ \upsilon_{av}, \quad (1)$$

where υ_{av} represents the average value between υ_A and υ_B frequencies. Fig.(2a) shows the relation between θ_D and composition. It is clear that θ_D decreases with increasing Zn content. From the IR spectra of Mg-Zn ferriteza There is a small decreasing about 15 cm in the wave number for each site. From IR spectra, decreases from 565 to 550 cm^{-1} and from 420 to 405 cm with increasing Zn content in the composition.

In our previous study [11-13], it was found a good relation between the cation distribution and the intensity ratio and broadening of each site (A&B-sites). Fig. (2b) demonstrates the relation between composition and the intensity ratio (I_A/I_B) and broadening ratio(i.e half band width Γ_A/Γ_B). It is clear that the two ratios depend on the statistical distribution of the cation on each site. The behavior of Fig.(2b) is divided into two regions according to the previous suggestion of cation distribution [7];

$$(Zn^{2+}_{x}Mg^{2+}_{y}Fe^{3+}_{1-x-y})^{A}[Mg^{2+}_{1-x-y}\,Fe^{3+}_{1+x+y}]^{B}\,O_4,\ x \leq 0.3$$

and $$(Zn^{2+}_{x-y}Fe^{3+}_{1+y-x})^{A}[Mg^{2+}_{1-x}\,Zn^{2+}_{y}Fe^{3+}_{1+x-y}]^{B}\,O_4,\ x \geq 0.4$$

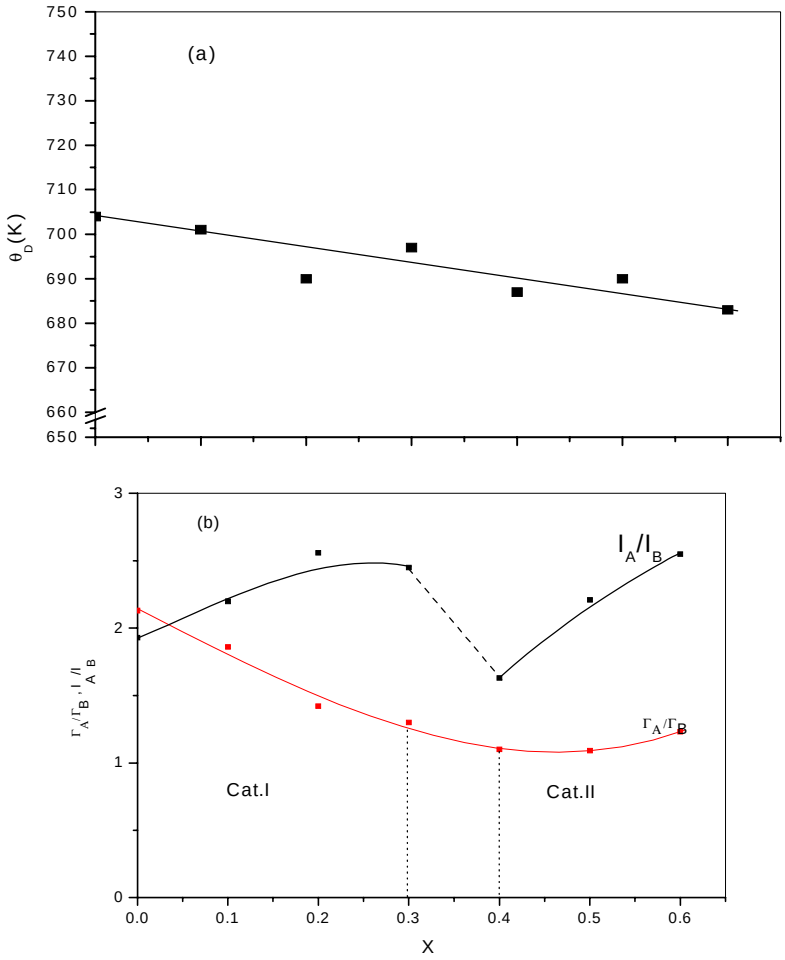

Figure 2: The dependence of a) Debye temperature, b) the ratio of half band width of site –A to that of site-B (Γ_A/Γ_B) and the ratio of the intensity of the absorption band of site-A to that of site-B(I_A/I_B), on x.

According to this distribution , the Zn^{+2} ions can exist on the A-site only with small ratio of Mg^{+2} ions for $x \leq 0.3$, but for $x \geq 0.4$, a ratio of Zn^{+2} ions migrate to B-site and prevent the Mg^{+2} ions to exist in A - site. This cation distribution is reflected on the behavior of Fig.(2b).

AC Conductivity

Over a wide range of temperature from 300K up to 700K, the ac conductivity was measured at four different frequencies 10^2, $10^{3,}$ 10^4, and 10^5 Hz for the investigated compositions as shown in Fig.(3). It can be seen that the Mg-Zn ferrite shows a semiconducting trend, as commonly seen in most ferrites. From Fig.(3) the conductivity at higher temperatures seems to be frequency independent while a little dispersion was observed at relatively low temperatures.

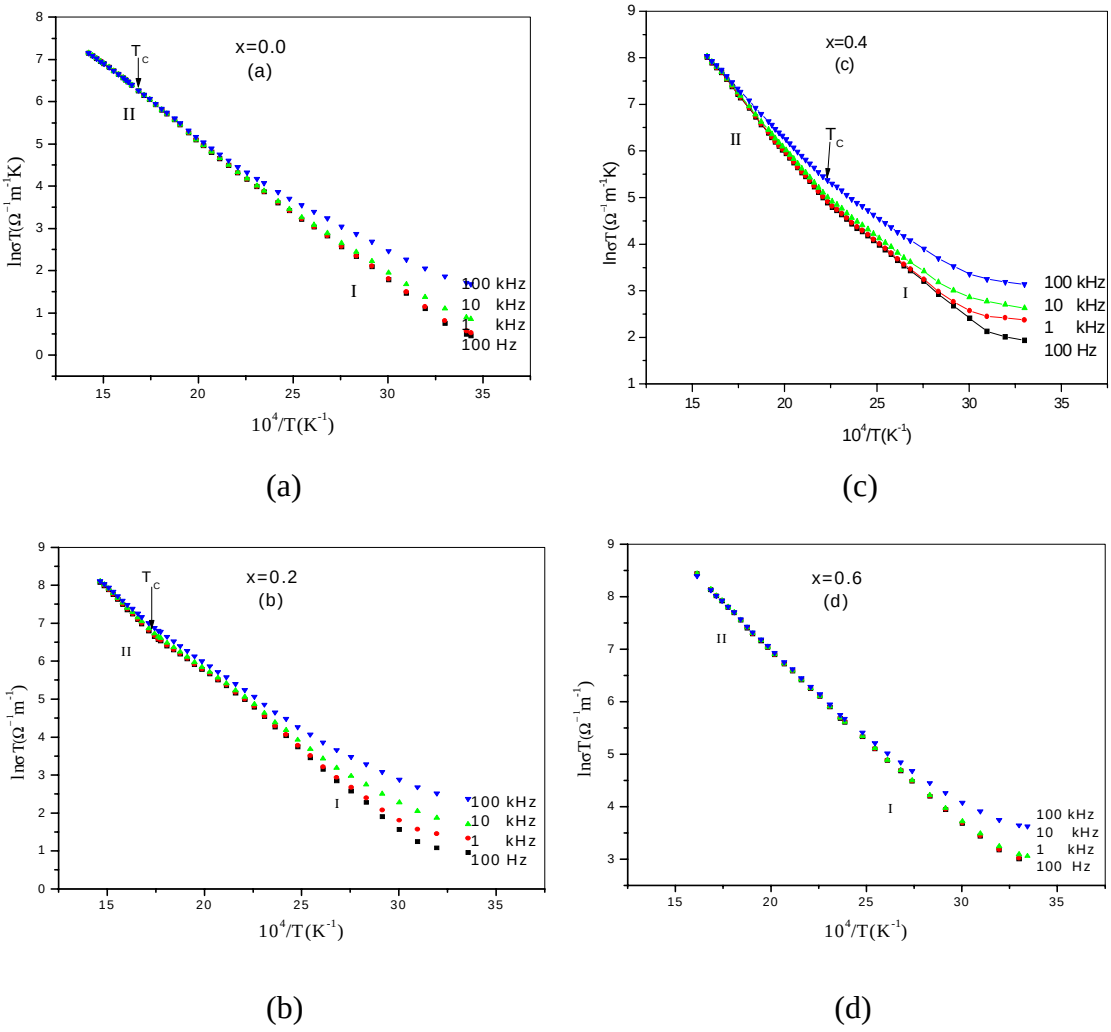

Fig.(3) Temperature dependence of the ac conductivity for x=0,0.2,0.4 and 0.6 In the dispersion region (region I), the ac conductivity increases with increasing frequency. The variation of the conductivity in this region is called the ac part, where the applied frequency gives clear variation in the values of conductivity. In general, this region is being below T_C, i.e. in the region of ferromagnetic range. In the second region (region II), where the conductivity is frequency independent and temperature dependent, is called the dc part. The last region exists in the paramagnetic region i.e. above T_C. The experimental data of the ac conductivity of Mg -Zn ferrite successfully comply with the following equation [14]:

$$\sigma = \frac{const}{T} \exp(-E_\sigma / KT) \qquad (2)$$

where E_σ represents the activation energy of the conduction mechanism and the other parameters have their usual meaning. The values of E_σ (I) for each composition were calculated in the temperature range between 350-430K at four different frequencies.

The obtained values of E_σ (I) are tabulated in Table(2). It can be seen that the activation energy decreases with increasing frequency and also the activation energy in region II was determined and it has one value corresponding to the change in frequency.

Table 2: Values of the activation energy for different values of x in regions I and II as a function of frequency.

E_σ (I)					**E_σ (II)**
X	10^2 Hz	10^3 Hz	10^4 Hz	10^5 Hz	Frequency independent part
0.0	0.28	0.27	0.25	0.20	0.30
0.2	0.35	0.32	0.27	0.27	0.44
0.4	0.32	0.32	0.29	0.26	0.48
0.6	0.27	0.26	0.26	0.21	0.33

From Fig (3,a-d)**,** it was observed for samples of $x \leq 0.4$ that at low temperature the frequency dispersion of ac conductivity is more pronounced than the other samples x = 0.5 and 0.6 . It is suggested that the dispersion at room temperature in the relation of $\ln\sigma T$ versus T^{-1} can be manifested by the percentage of change in $\ln T\sigma$, $(\Delta \ln\sigma T)$ %, corresponding to the change of frequency from 10^2 to10^5 Hz can also be taken as an indication for the dispersion as shown in Fig (4)**.** It is clear that the dispersion increased with increasing Zn concentration for $x < 0.3$ and then decreases when Zn migrates from A-site to B-site. The frequency dispersion of the ac conductivity in region I, can be attributed to the ac hopping conduction of the localized carriers [15]. Therefore, the hopping conduction is expected to be responsible for the conduction mechanism in Mg-Zn ferrite.

From the Fig (5,a-d) in the range of temperature below 400 K, it is clear that for some composition ε decreases (such as: x =0.4 and 0.6) with increasing temperature (at lower frequencies) and also decreases with increasing frequency. For other compositions there is a slight variation in ε with both temperature and frequency. In region T < 400K, thermal energy given to the sample is not sufficient to free the localized frozen dipoles to be oriented in the field direction. So the electron polarizability plays a significant role in the dielectric behavior in the temperature range of T < 400 K, where the electron exchange between $Fe^{2+} \rightleftharpoons Fe^{3+}$ takes place.

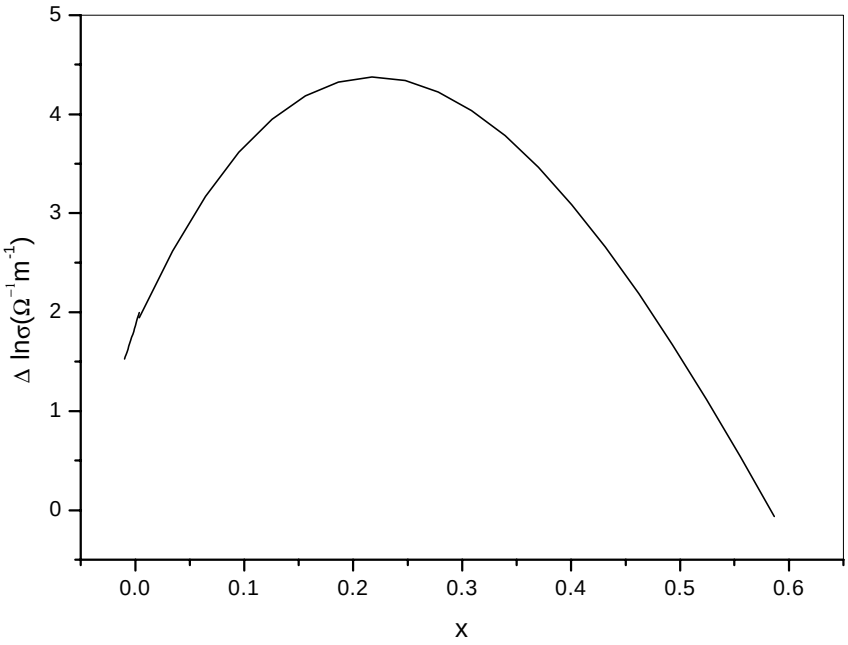

Figure 4: The variation of (ΔlnσT)% with x.

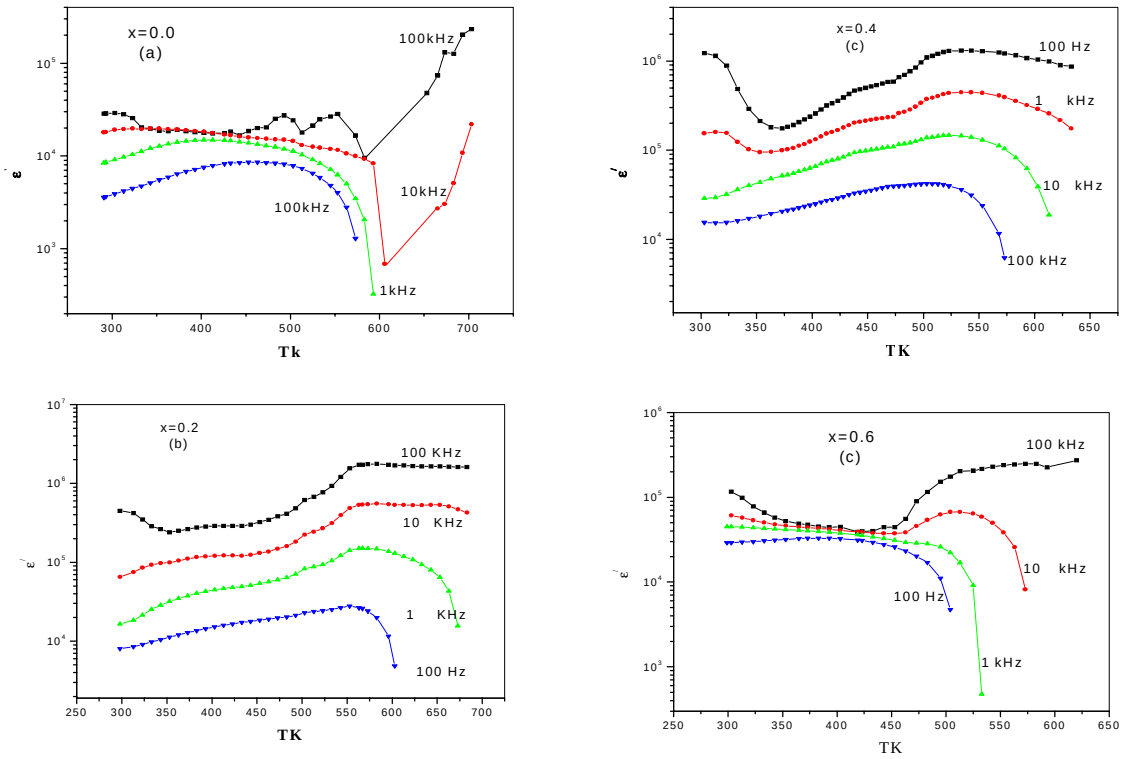

Figure 5: Variation of dielectric constant (έ) at different frequencies for x = 0, 0.2, 0.4 and 0.6.

In the range of temperature between 400 and 550 K, there are increasing in the values of dielectric constant έ with increasing temperature. In this range of temperature, the thermal energy is sufficient to liberate the frozen dipoles and the applied electric field aligned these dipoles in its direction. In this range of temperature, thermal energy given to the sample decreases the internal viscosity, increasing the randomness (entropy) and consequently έ increases. For T > 550 K, it is clear that έ decreases with increasing the temperature especially at higher frequencies. Several authors [16,17,18] observed this behavior. The decrease in έ takes place when the jumping frequency of the charge carriers cannot follow the applied electric field and consequently the friction between them will be increased.

The changes in the dielectric loss tangent (tan δ) with temperature are represented in Fig (6). By increasing temperature, tan δ begins to increase until it attains a maximum value. Similar behaviour was observed in zinc substituted magnesium rich manganese ferrites [19] and also in rare earth substituted Cu-Zn ferrite [20]. Moreover, it was mentioned that the dielectric material is a mixture of two or more polar substances. The dependence of tan δ vs. T may have two or more maxima produced by the effect of the separate components of the mixture [21]. According to this fact and taking Koop's model into consideration, where the material is assumed to be composed of grains and grain boundaries with different parameters, one may expect that each of the grains and the grain boundaries have their characteristic maximum in tan δ.

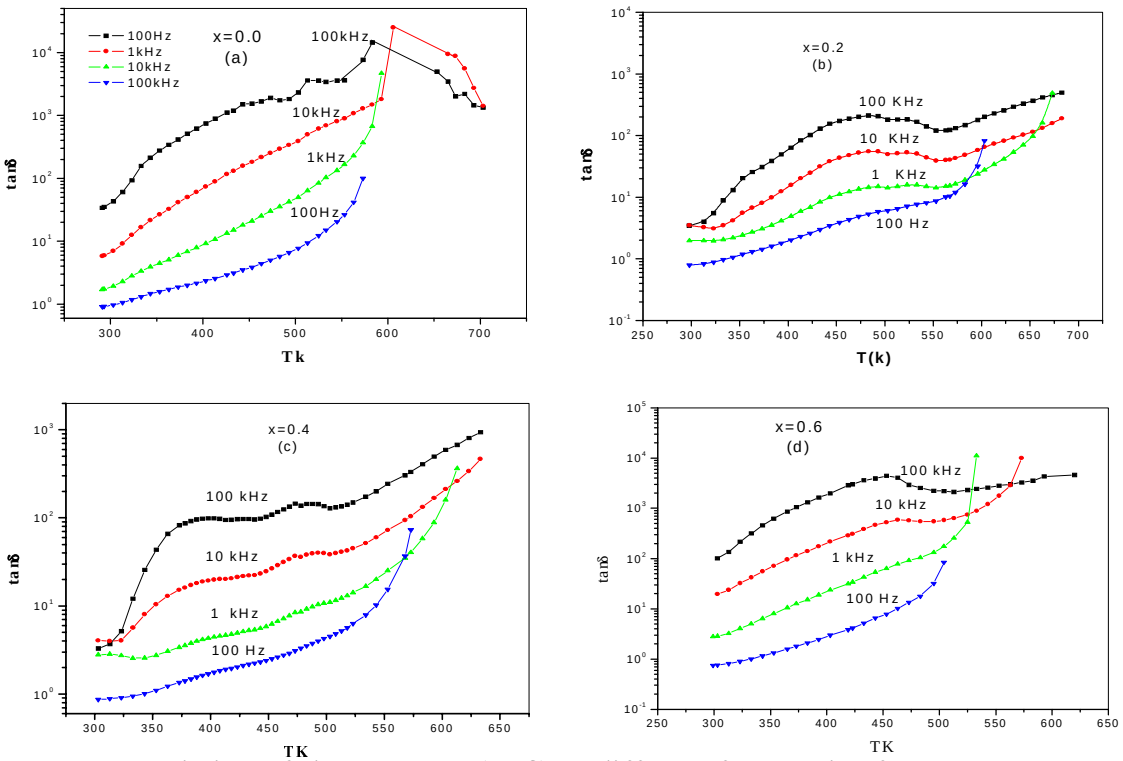

Figure 6: variation of loss tangent (tanδ) at different frequencies for x = 0, 0.2, 0.4 and 0.6.

Conclusions

The IR spectra confirmed the formation of spinel structure and gave information about the distribution of ions between the two sites (A and B)

The temperature dependence of ac conductivity of Mg-Zn ferrites in the temperature range from 300K up to700K showed that there is a little dispersion at relative low temperatures and also two region in ac conductivity, where, region I is called the ac part, in which the applied frequency gives clear variation in the conductivity. Region II is called the dc part, where the conductivity is frequency independent and temperature dependent.

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