Journal of Biomedical and Bioengineering, ISSN: 0976 – 8084 & E-ISSN: 0976–8092, Vol. 1, Issue 1, 2010, PP-21-29

Dielectric behaviour study of nanocrystalline Co-Zn ferrite

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Abstract- Dielectric properties are studied as a function of electric field frequency for $Co_{0.5}Zn_{0.5}Fe_2O_4$ prepared by wet chemical co-precipitation method. The composition is characterized by X-ray diffraction technique (XRD). XRD study shows formation of single-phase homogeneous compound with cubic structure. The crystal size is calculated from XRD data by using Scherrer equation and is confirmed by SEM, which reveals the formation of nanocrystalline ferrite. Dielectric constant (ε'), complex dielectric constant (ε'') and dielectric loss tangent (tan δ) are measured in the frequency range up to 10KHz, at different temperatures (300K to 900K) and they show dispersion with decrease in frequency and increase in temperature. Thermal variation of ε', ε'' and tan δ has been studied at four different frequencies 100Hz, 120Hz, 1 KHz, and 10 KHz. The variation of these parameters with frequency is explained qualitatively with the aid of Koops phenomenological theory. The observed results can be explained on the basis of an electron exchange between Fe^{2+} and Fe^{3+} ions. Improved dielectric properties are with nanocrystallinity of the prepared samples.

Keywords- Nanocrystalline Co-Zn ferrite, dielectric constant, dielectric loss

Introduction

Ferrimagnetic cubic spinels namely ferrites possess properties of both magnetic materials and electric insulator. These properties make ferrites as an important material in many technological applications. These are preferred because of their high permeability in the radio frequency region, high electrical resistivity, mechanical hardness, chemical stability, etc. [1, 2]. There is a growing interest of researchers from last few decades in nanocrystalline ferrites. The properties of these nanocrystalline ferrite particles are considerably different from those of their bulk counterparts.

The nanocrystalline ferrites can be prepared by different techniques. Number of chemical methods was developed to prepare nanocrystalline spinel ferrites. The methods include wet chemical co-precipitation [3], microwave refluxing [4], sol-gel [5], hydrothermal [6], glass crystallization [7], salt melt technique [8] etc. Processing of ferrites has gained tremendous importance in recent times to meet the high performance demands on ferrites in keeping pace with the vast emerging technologies. Thus the quality of ferrite powders has strong influence on the performance of final device.

Ferrite formation in the wet process has attracted significant interest in a ferrite coating process of electric devices [9] and in many other devices. These important applications of ferrites are constituents of their unique structural, chemical and physical properties. These properties are improved when ferrites are prepared by wet-chemical method [10, 11].

Among spinel ferrites, cobalt ferrite, $CoFe_2O_4$ is especially interesting because of its high cubic magnetocrystalline anisotropy, high coercivity and moderate saturation magnetization. Recently, cobalt ferrite nano particles were also known to be a photo-magnetic material, which shows interesting light-induced coercivity change [12, 13]. Also Co-Zn ferrite found to possess good elastic properties [14].

Many researchers have studied the magnetic properties of nano-size cobalt and mixed cobalt ferrites. The properties studied are the magnetic properties like saturation magnetization [15], Bohr magnetron number and AC susceptibility [16], coercivity and Mössbauer study [17], etc. This literature survey is not showing any report on study the dielectric properties of nanocrystalline cobalt-zinc ferrite prepared by wet chemical method.

In our previous study we found that the substitution of non-magnetic zinc ions has great impact on structural and dielectric properties of nanocrystalline Ni-Zn ferrites [10]. So by considering the technological importance of cobalt ferrite, impact of non-magnetic zinc substitution on its properties and with the above literature survey, we decided to study the dielectric properties of the ferrite $Co_{0.5}Zn_{0.5}Fe_2O_4$ prepared by wet chemical co-precipitation method.

Experiments

Preparation of the sample

The Zn substituted cobalt ferrite $Co_{0.5}Zn_{0.5}Fe_2O_4$ was prepared by air oxidation of an aqueous suspension containing Co^{2+}, Zn^{2+}, and Fe^{3+} cations in proper proportions. The starting solution was prepared by mixing appropriate amounts of $FeSO_4.7H_2O$, $CoSO_4.7H_2O$ and $ZnSO_4.7H_2O$ (all 99.9% pure supplied by s.d. fine, India). A two molar NaOH solution is used as precipitating agent and H_2O_2 was used as an oxidant, which helps to convert Fe^{2+} to Fe^{3+} by the following reaction [18].

$Fe^{2+} + H_2O_2 \rightarrow Fe^{3+} + OH^{-} + OH^{\cdot}$.

The details of method of preparation were reported in our previous work [3]. The sample was filtered, and washed several times by distilled water. The wet sample of the ferrite was annealed at 725^0C for 16 h. The powder sample was palletized at the pressure of 4 tons to yield a pellet of 10mm diameter and 2mm thickness.

Measurement of the parameters

The X-ray powder diffraction pattern was recorded on a Philips X-ray diffractometer (Model Joel-DX-8030) at room temperature in a 2□ scanning range from 20^0 to 80^0. The particle morphology was studied by scanning electron microscopy (SEM). The dielectric constant (ε′), dielectric loss or complex dielectric constant (ε″) and dielectric loss tangent (tanδ) were measured as a function of frequency by using LCR-Q meter (Model HP4284). The dielectric measurements were made using two-probe method at the frequencies 100Hz; 120Hz; 1KHz and 10KHz in temperature range 300K to 900K.

Results and discussion

XRD and SEM

XRD pattern of the ferrite $Co_{0.5}Zn_{0.5}Fe_2O_4$ annealed at 725K show all the peaks of the spinel structure. The diffraction pattern is shown in Fig. 1. The value of lattice constant was obtained from XRD data with an accuracy of ±0.002Å and is 8.4Å. The particle size of the ferrite sample was obtained from broadening of XRD peak by using the Scherrer equation [19], and is 56nm. It is evident from the particle size that wet chemical co-precipitation method yields the particles of the order of nanometer dimension. The particle size is confirmed by SEM data (Fig. 2).

Dielectric properties

The real and imaginary part of the dielectric constant (ε'), dielectric loss (ε") and loss tangent (tanδ) of $Co_{0.5}Zn_{0.5}Fe_2O_4$ was computed according to Smit and Wijn [20] as a function of frequency and temperature.

Fig. 3, 4 and 5 show the variation of the dielectric constant (ε′), dielectric loss (ε″) and dielectric loss tangent (tanδ) respectively as a function of frequency at different temperatures of the ferrite. It is observed from the Fig. 3 and 4 that dielectric constant as well as dielectric loss decreases with increasing frequency exhibiting normal ferrimagnetic behaviour. A more dielectric dispersion is observed at lower frequency region and it remains almost independent of applied external field at high frequency. Similar behaviour of dielectric constant with frequency was observed by many researchers [21, 22]. The dielectric dispersion observed at lower frequency region is due to Maxwell-Wagner [23] interfacial type of polarization, which is in agreement with Koops phenomenological theory [24]. The decrease in dielectric constant with frequency can be explained by the supposition that the mechanism of polarization process in ferrite is similar to that of conductivity process. By the electronic exchange, $Fe^{2+} \rightarrow Fe^{3+} + e^{-1}$, one obtains local displacement of electrons in the direction of electric field. These displacements determine the polarization in ferrites. It is known that effect of polarization is to reduce the field inside the media. The decrease of polarization with increase of frequency may be due to the fact that, beyond a certain frequency of the electric field, the electronic exchange between ferrous and ferric ions cannot follow the alternating field. Hence dielectric constant may decrease substantially as frequency is increased.

Fig. 5 shows that the variation of dielectric loss tangent (tanδ) with frequency at 423 K to 923K for the ferrite where the parameter tanδ decreases exponentially with the increase of frequency. According to [25], there is a strong co-relation between the conduction mechanism and the dielectric behaviour of ferrites. A maximum value in tanδ can be observed when the hopping frequency is approximately equal to that of the externally applied electric field [26].

The plot of ε' versus temperature and ε'' versus temperature shows that, both ε' and ε'' increases with increasing temperature. Also the plot of dielectric loss tangent (tanδ) shows that tanδ increases with increasing temperature.

This behaviour of temperature dependence of ε' and ε'' is in good agreement with the results on other soft ferrites [27] for which ε' and ε'' increases with increasing temperature.

According to Rabkin and Novikova [28], the process of dielectric polarization in ferrites takes place through a mechanism similar to conduction process by electron exchange $Fe^{2+}\leftrightarrow Fe^{3+}$ and $Co^{2+}\leftrightarrow Co^{3+}$ one obtains local displacement of the electrons in the direction of electrical field; these displacements determines the polarization. Both types of carrier 'n' and 'p' contribute to polarization and they depend on the temperature. Since, the influence of temperature on the electronic exchange $Fe^{2+}\leftrightarrow Fe^{3+}$ and $Co^{2+}\leftrightarrow Co^{3+}$ is more pronounced than on the displacement of peak carriers, ε' and ε'' increase rapidly with increase in temperature. Also tanδ increases with increase in temperature.

Conclusion

The wet chemical co-precipitation method yields ferrite particles of nano meter in size. The dielectric constant, dielectric loss and dielectric tangent loss of the Co-Zn ferrite are frequency as well as temperature sensitive. The frequency and temperature dependence of the dielectric constant arises from the two types of charge carriers present in this ferrite.

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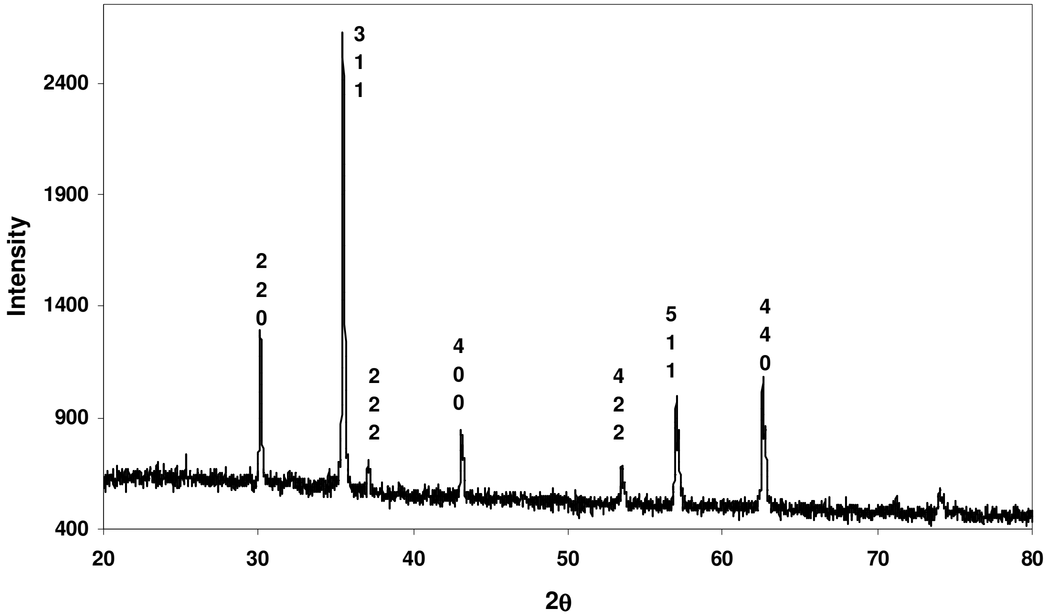

Fig. 1 - X-ray diffraction pattern of the ferrite $Co_{0.5}Zn_{0.5}Fe_2O_4$

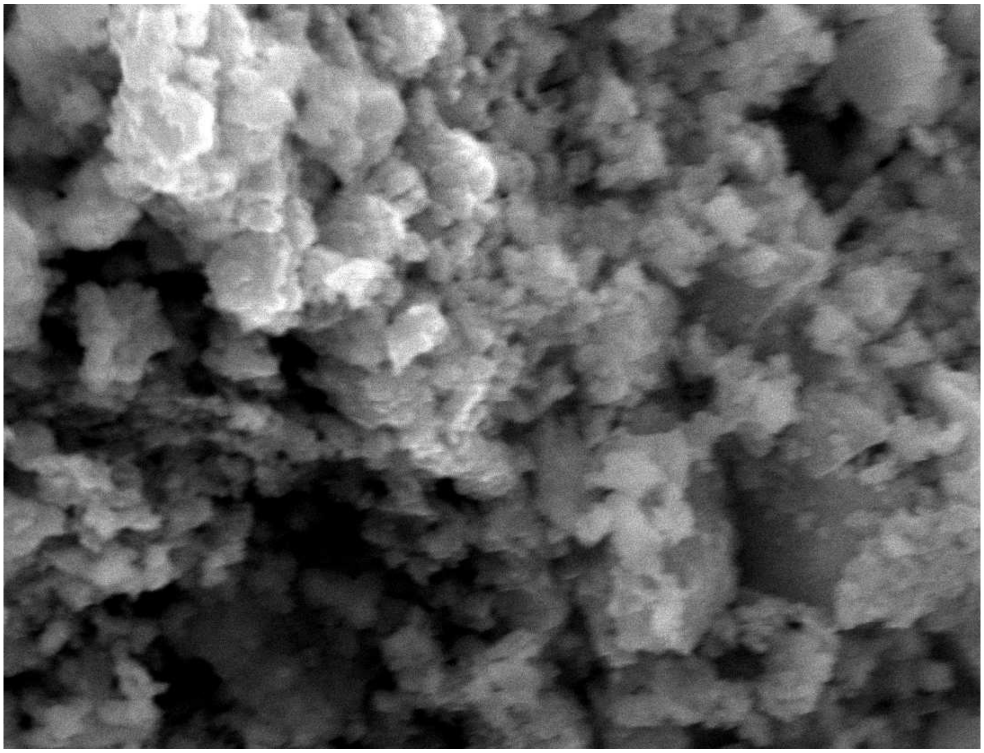

Fig. 2 - Scanning electron micrograph of the ferrite $Co_{0.5}Zn_{0.5}Fe_2O_4$

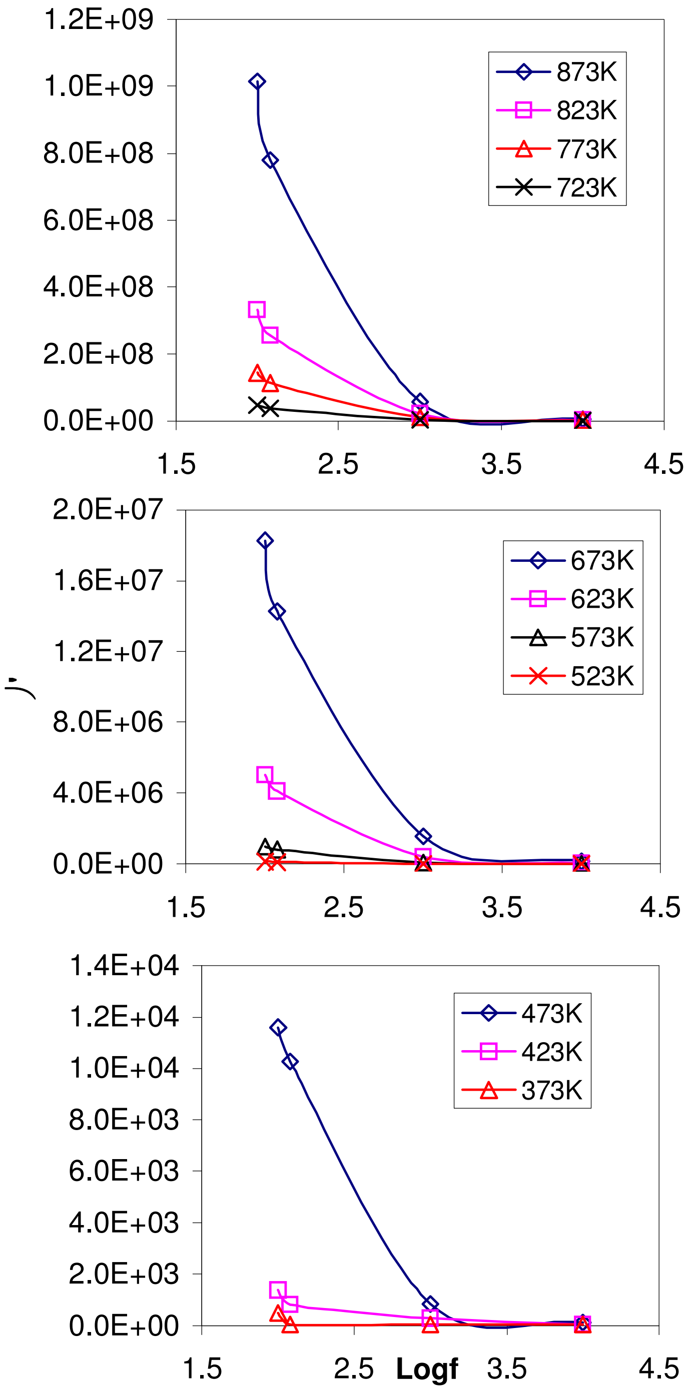

Fig. 3 - Variation of the dielectric constant (□') with logf of the ferrite $Co_{0.5}Zn_{0.5}Fe_2O_4$

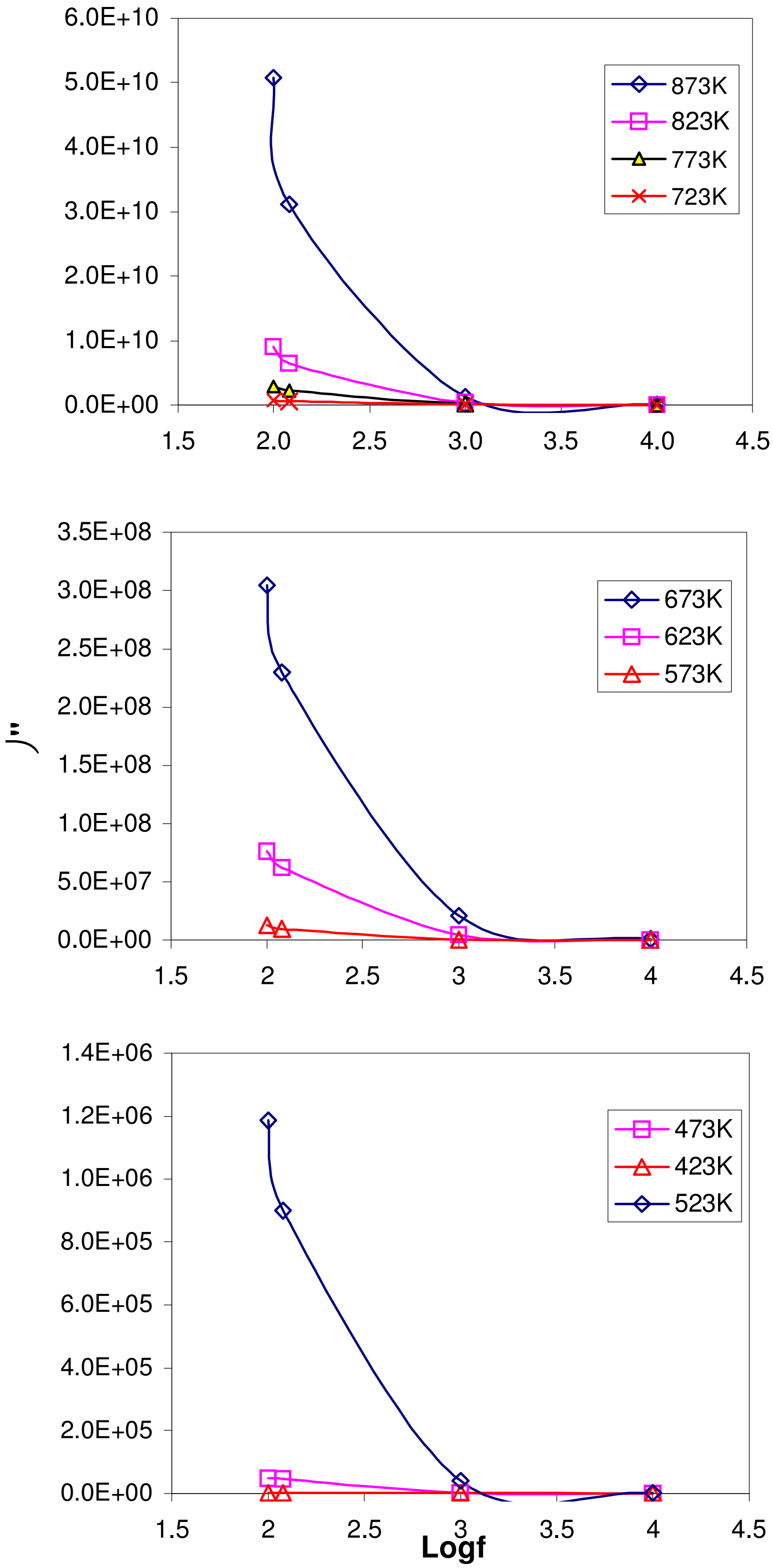

Fig. 4 - Variation of the dielectric loss (□") with logf of the ferrite $Co_{0.5}Zn_{0.5}Fe_2O_4$

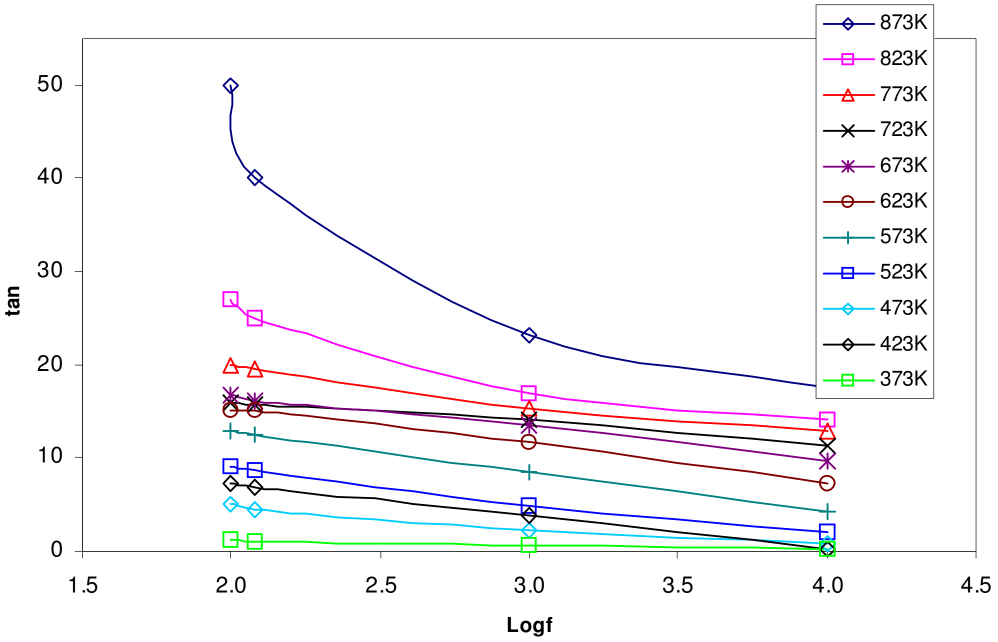

Fig. 5- Variation of the dielectric loss tangent (tan□) with logf of the ferrite $Co_{0.5}Zn_{0.5}Fe_2O_4$

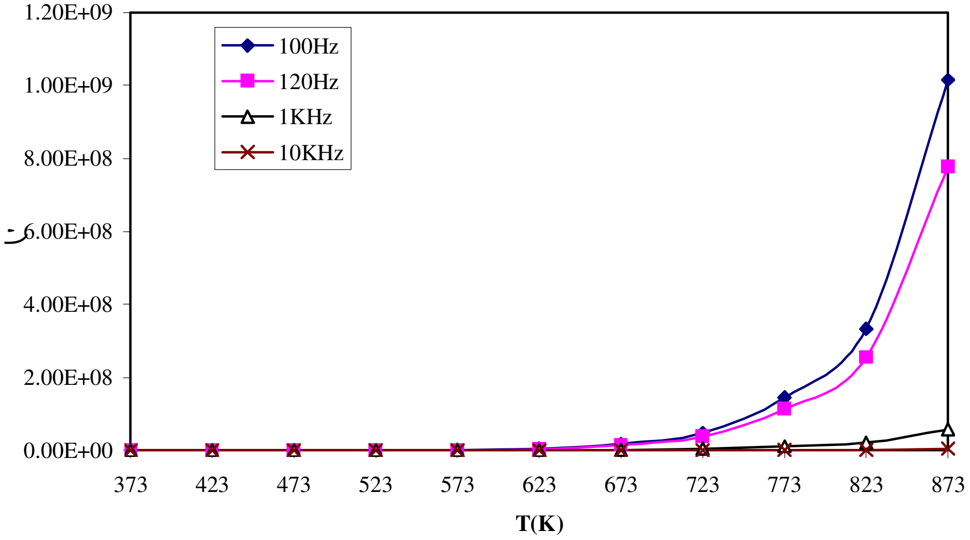

Fig. 6 - Variation of the dielectric constant (□') with temperature of the ferrite $Co_{0.5}Zn_{0.5}Fe_2O_4$

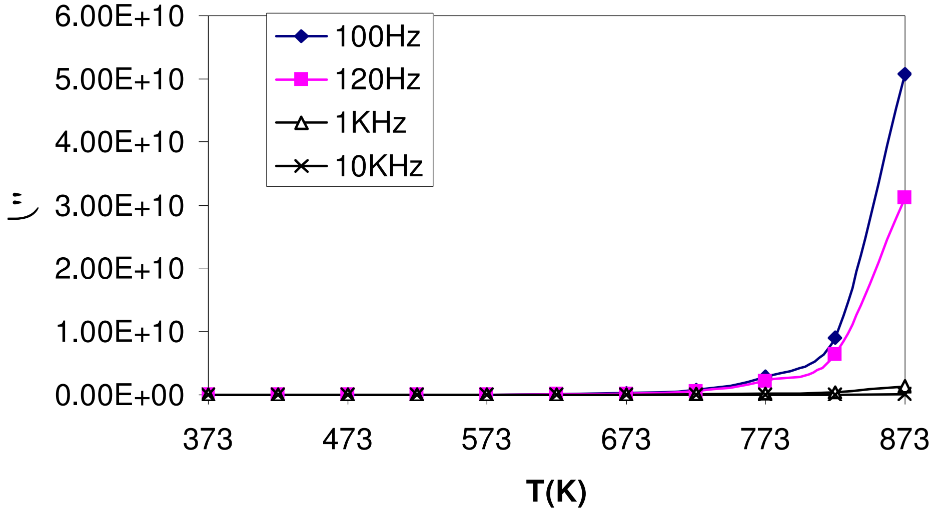

Fig. 7 - Variation of the dielectric constant (□") with temperature of the ferrite $Co_{0.5}Zn_{0.5}Fe_2O_4$

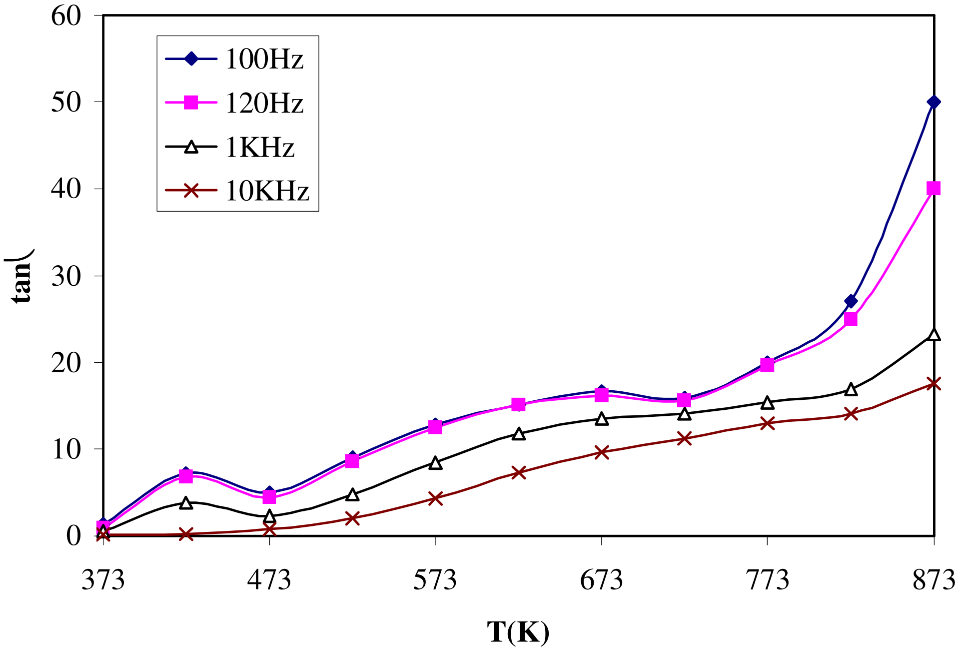

Fig. 8 - Variation of the dielectric loss tangent (tan□) with temperature of the ferrite $Co_{0.5}Zn_{0.5}Fe_2O_4$