Dielectric behavior in nanocrystalline Al doped Co-Zn ferrite

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A series of the sample of the system $Co_{1-x}Zn_xFe_{2-x}Al_xO_4$ (x = 0.0 - 0.6) were prepared by using wet chemical co-precipitation method. The dielectric constant (ε'), dielectric loss (ε'') and dielectric loss tangent (tanδ) were measured in the frequency range 100Hz to 1MHz at the room temperature and as a temperature with fixed frequency of 1kHz. All the samples shows the dielectric dispersion. The dielectric constant (ε'), dielectric loss (ε'') with frequency shows decreasing nature. This behaviour of a dielectric is explained qualitatively in terms of the supposition that the mechanism of the polarization process in ferrite is similar to that of the conduction process. The dielectric constant (ε'), dielectric loss (ε'') and dielectric loss tangent (tanδ) with temperature at fixed frequency shows decreasing nature.

1. Introduction

Ferrites forming a group of semiconducting materials are of great technological importance because of their twin property of magnetic conductor and electrical insulator. They can be used in memory cores, high frequency transformers, antenna, recording heads and radar absorbing paints [1]. Hence the study of magnetic and electric properties is important and useful. Studies on the effect of temperature, composition and frequency on the dielectric behaviour offer valuable information about conduction phenomenon in the ferrite based on localized electric charge carriers [2]. Ferrites also have vast application from microwave to radio frequencies. The low conductivity of ferrite causes them for its use in microwave applications. They exhibit high resistivity, and dielectric properties. The dielectric properties of ferrites are dependent upon several factors including the method of preparation, chemical composition and grain structure or size. When a ferrite is sintered under slightly reducing condition, the valence state changes, the individual cation found in the sample leads to high conductivity and when such a material is cooled in a oxygen atmosphere, it is possible to form films of high resistivity over the constituent grain. Such ferrites in which the individual grains are separated by either air gaps or low conducting layers behave as inhomogeneous dielectric material. This aroused considerable interest in the low frequency range (10^2 Hz - 10^5 Hz) dielectric behaviour of ferrite. Among the many workers involved in this type of studies, the prominent once are Koops [3] Moltgen [4], Kamiyoshi [5], Iwauchi et. al. [6,7], Rezlescu [8,9] and Josyolu [10].

The cobalt ferrite ($CoFe_2O_4$) possesses an inverse spinel structure and the observed degree of inversion depends upon heat treatment [11]. Many workers [12-14] have studied the structural, electrical and magnetic properties of cobalt ferrite and cobalt-substituted ferrite. It has been reported that the substitution of tetravalent ions in cobalt ferrite influences the structural, electrical and magnetic properties [15,16]. To our knowledge no one has reported the dielectric behaviour of $Co_{1-x}Zn_xFe_{2-x}Al_xO_4$. Therefore the paper reports the study of the effect of temperature, frequency and composition on the cobalt ferrite prepared by wet chemical co-precipitation method.

2. Experimental

The spinel ferrite system under investigation $Co_{1-x}Zn_xFe_{2-x}Al_xO_4$ with variable composition (x = 0.0 - 0.6) was prepared by air oxidation of an aqueous suspension containing Co^{2+}, Zn^{2+}, Al^{3+}, Fe^{3+} cations in proper proportion. The starting solutions were prepared by mixing 50 ml of aqueous solution of $FeSo_4 \times 7H_2O$, $CoSo_4 \times 7H_2O$, $ZnSo_4 \times 7H_2O$ and $Al_2(So_4)_3 \times 16H_2O$ in stoichiometric proportions. A two molar (2M) solution of NaOH was prepared as a precipitant. It has been suggested that the solubility product constant (K_{sp}) of all the constituents always exceeds when the starting solution is added into the precipitant. Therefore, in order to achieve simultaneous precipitation of all the hydroxide $Co(OH)_2$, $Zn(OH)_2$, $Al(OH)_3$ and $Fe(OH)_2$, the starting solution (pH ≈ 3) was added to the solution of NaOH and a suspension (pH = 11) containing dark intermediate

precipitation was found. Then the suspension was heated and kept at a temperature of 60^0C, while oxygen gas was bubbled uniformly into the suspension to stir it and to promote the oxidation reaction until all the intermediate precipitant changed into the dark brownish precipitate of the spinel ferrite. The samples were filtered, washed several times by distilled water. The wet samples of Co-Zn-Fe-Al system were annealed at 800^0C for 12 hr.

Dielectric measurement as a function of frequencies in the range 100 Hz-1 MHz at the room temperature and also as a function of temperature in the range 300-800 K for 1KHz frequency were carried out using a LCR meter (HP-4284A) in conjunction with a laboratory designed cell, the temperature controller ($\pm1^0c$) using two probe method. The dielectric constant is calculated by using the formula,

$$\varepsilon^{'} = \frac{Cd}{\varepsilon_0 A} \qquad (1)$$

where, C is the capacitance of the pallet in pf, d is the diameter of the pallet, A is the cross sectional area of the flat surface of the pellet and ε_0 is the constant of permitivity for free space.

The dielectric loss (ε'') was calculated using the relation

$$\varepsilon'' = \frac{1}{2\pi \cdot 1000 \cdot \varepsilon_0} \sigma \qquad (2)$$

where, σ is a.c. conductivity,

The dielectric loss tangent (tanδ) was calculated using the relation

$$\tan\delta = \frac{\varepsilon''}{\varepsilon'} \qquad (3)$$

The Aim of the present work is to study the dielectric constant (ε'), dielectric loss (ε") and dielectric loss tangent (tanδ) were measured in the frequency range 100Hz to 1MHz at the room temperature and as a temperature with fixed frequency i.e. 1KHz.

3. Results and discussion

3.1 Frequency dependent dielectric properties

The a.c. parameters such as dielectric constant (ε′), dielectric loss (ε″) and dielectric loss tangent (tan δ) and a.c. resistivity depend on the frequency. The measurements of a.c. resistance (R) and capacitance (C) were made with the help of LCR-Q meter (HP-4284A) in the frequency range 20 Hz to 1 MHz. The dielectric

constant (ε′) and dielectric loss tangent (tanδ) are calculated using the values of R and C.

Fig. 1 shows the variation of dielectric constant with frequency measured at room temperature for all the samples. It can be seen from Fig 1 that the dielectric constant ε′ decreases with increase in frequency. The observed behaviour of dielectric constant with frequency may be due to Maxwell-Wagner interfacial type of polarization [17, 18] which is in good agreement with Koops phenomenological theory [19], by the electronic exchange, $Fe^{3+} \rightarrow Fe^{2+} + e^{-1}$, one obtains local displacement of electrons in the direction of electric field. These displacements determine the polarization in ferrites. 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 Fe^{3+} and Fe^{2+} cannot follow the alternating field.
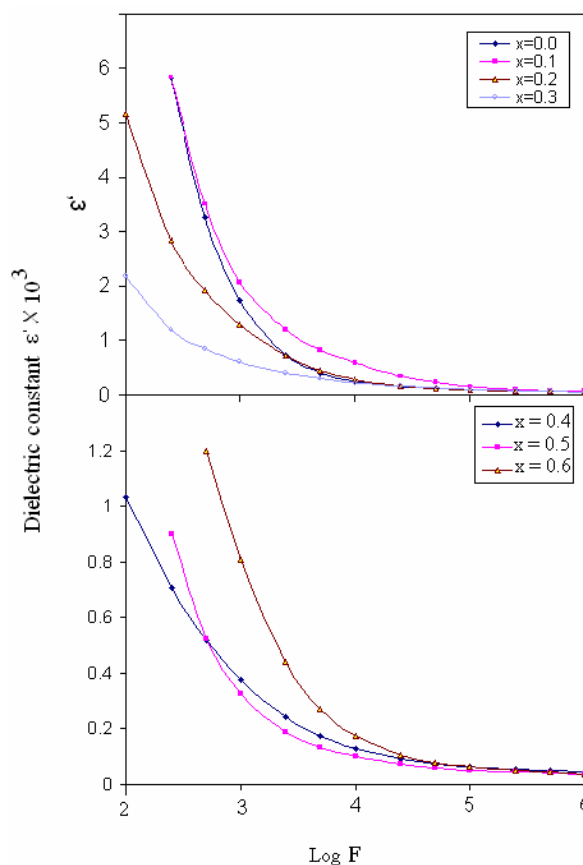

Fig. 1. The variation of dielectric constant (ε′) with log F of the system $Co_{1-x}Zn_xFe_{2-x}Al_xO_4$ (x = 0.1 to 0.6).

Therefore, the real part of dielectric constant decreases with increasing frequency. Similar behaviour of dielectric loss is observed and is shown Fig 2.
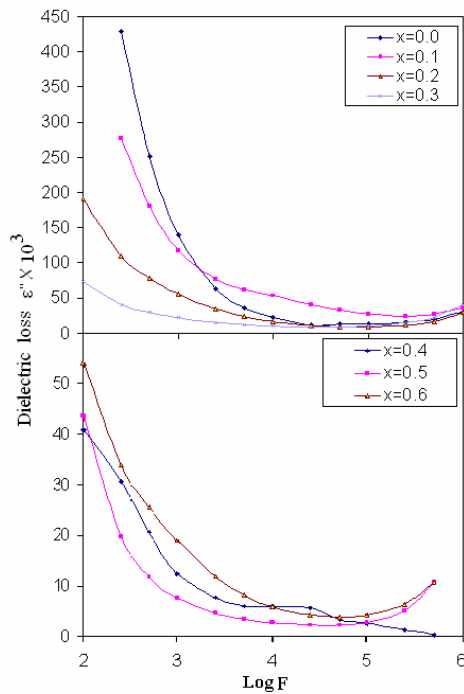
Fig. 2. The variation of dielectric loss (ε'') with log F for the system $Co_{1-x}Zn_xFe_{2-x}Al_xO_4$ (x = 0.1 to 0.6).

The variation of dielectric loss tangent against log F at room temperature is shown in Fig. 3. It is clear from Fig 3. that all the samples show a normal dielectric behaviour with frequency.
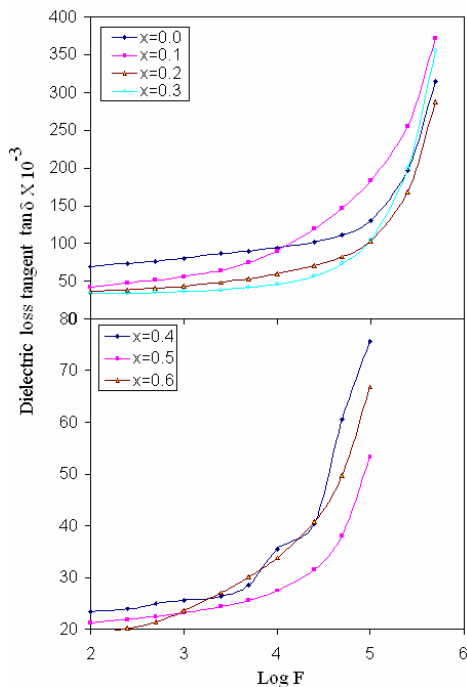

Fig. 3. The variation of dielectric loss tangent (tanδ) with log F for the system $Co_{1-x}Zn_xFe_{2-x}Al_xO_4$ (x = 0.1 to 0.6).

The parameter tanδ increases experimentally with the increasing frequency. The maximum in tanδ occur when the jump frequency of electron between Fe^{2+} and Fe^{3+} is equal to the applied frequency [20].

3.2 Temperature dependent dielectric properties

The real (ε') and imaginary part of the dielectric constant, loss tangent (tan δ) of $Co_{1-x}Zn_xFe_{2-x}Al_xO_4$ was computed according to Smit and Wijn [21] as a function of temperature. A.C. conductivity measurements were carried out in the temperature range 300-800 K. Fig. 4, 5 and 6 shows the variation of the dielectric constant (ε'), dielectric loss (ε'') and dielectric loss tangent (tan δ) respectively as a function of temperature at 1KHz.
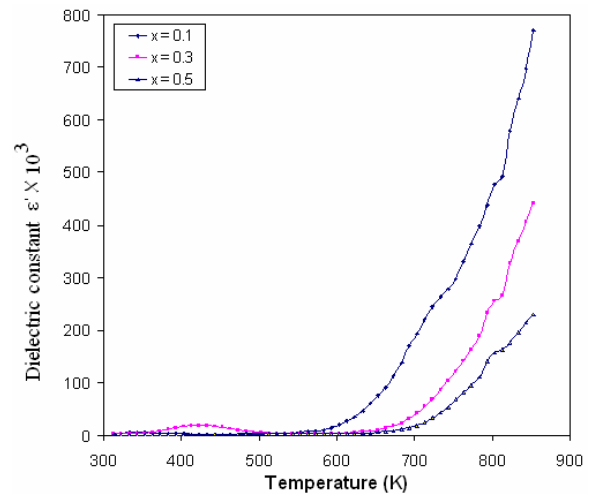

Fig. 4. The variation of dielectric constant (ε') as a function of temperature (K) at 1 kHz of the typical samples x = 0.1, 0.3 and 0.5 for the system $Co_{1-x}Zn_xFe_{2-x}Al_xO_4$.

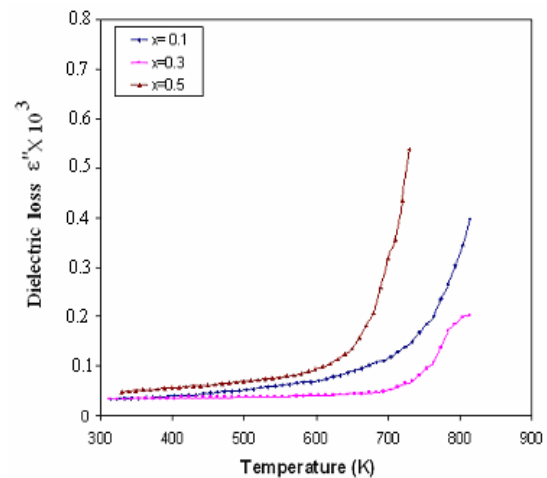*Fig. 5. The variation of dielectric loss (ε'') with temperature (K) of the typical samples x = 0.1, 0.3 and 0.5 for the system $Co_{1-x}Zn_xFe_{2-x}Al_xO_4$.*
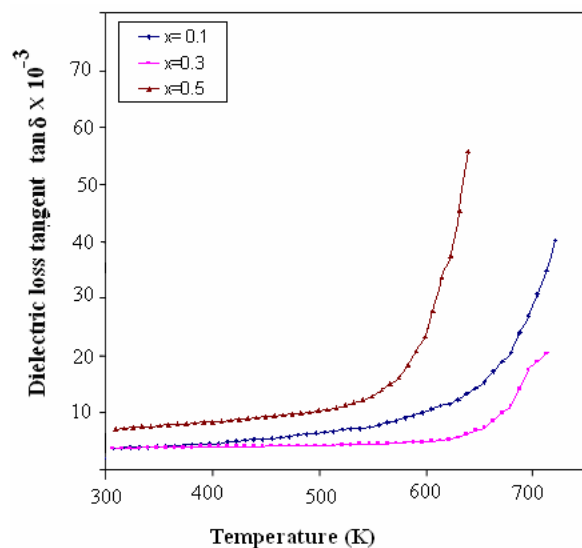
Fig. 6. The variation of dielectric loss tangent (tanδ) with temperature (K) of the typical samples x = 0.1, 0.3 and 0.5 for the system $Co_{1-x}Zn_xFe_{2-x}Al_xO_4$.

The temperature dependence of dielectric constant (ε′), dielectric loss (ε″) and dielectric loss tangent (tanδ) can be explained on the basis of polarization effect. The number of space charge carriers governs the space charge polarization. As temperature increases electrical conductivity increases due to the increase in thermally activated drift mobility of electric charge carriers according to hopping conduction mechanism. Hence, dielectric polarization increases which causes the increase in dielectric constant (ε′), dielectric loss (ε″) and dielectric loss tangent (tanδ). This behaviour of dielectric constant (ε′), dielectric loss (ε″) and dielectric loss tangent (tan δ) is in good agreement with those reported by other workers [22, 23].

4. Conclusions

The dielectric properties, the dielectric constant (ε′)and the dielectric loss (ε″) , decrease with increasing frequency for all Co-Zn-Fe-Al ferrite compositions. This behaviour of a dielectric is explained qualitatively in terms of the supposition that the mechanism of the polarization process in ferrite is similar to that of the conduction process (hopping). Temperature dependence of dielectric constant, dielectric loss and dielectric loss tangent at 1 kHz shows increasing behaviour.

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