Frequency and Composition Dependant Dielectric properties of Indium Substituted Cobalt ferrite

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Abstract: *The nanoparticles of indium substituted cobalt ferrite were prepared by sol-gel auto combustion method. The prepared spinel ferrite compositions were studied for their dielectric properties. The dielectric properties as a function of frequency and composition were studied by two probe method. The dielectric parameters show the maximum values at low frequencies and minimum values at high frequencies.*

Introduction:

Ferrite nanoparticles have attracted an increasing interest because of their wide area of applications as magnetic recording, storage, catalyst, biotechnology [1, 2] etc. In the recent years, the interest in the use of nanoparticles in biomedical application has greatly increased. The size and composition of nanoparticles influence the applications of magnetic nanoparticles.

Spinel ferrites are an important class of magnetic materials, composed of iron oxide and metal oxide, having large variety of electrical and magnetic properties. They possess high electrical resistivity, low eddy current and dielectric losses and good magnetic properties. Nano size spinel ferrites are very promising materials in technological applications in many important fields.

Cobalt ferrite is a unique spinel ferrite with multiple properties like high saturation magnetization, high magnetic permeability, high Curie temperature, positive magneto-crystalline anisotropy, high electrical resistivity, low eddy current and dielectric losses [3, 4]. These remarkable properties make cobalt ferrite useful in many electrical, electronic telecommunication etc. fields. The sol-gel auto combustion method is a unique and versatile wet chemical method used for synthesis of nano sized spinel ferrite materials. It is a simple process, significant in saving the time and energy consumption, low cost, produces high purity materials compared to other methods. The method is commonly used and employed to obtain improved powder characteristics, more homogeneity and nano sized particles, which results in modifying the dielectric, and other properties of spinel ferrite.

Results and discussion:

Indium substituted cobalt ferrite was prepared by using sol-gel auto combustion synthesis technique. The prepared samples were characterized by XRD for their structural studies as reported elsewhere [5].

Dielectric measurements

Frequency dependence

The dielectric properties were studied over the frequency range from 100 Hz to 1 MHz by using LCR-Q meter (HP 4248A). The values of dielectric constant (ε'), dielectric loss (ε'') and dielectric loss tangent (tan δ) were determined using the following relations

$$\varepsilon' = \frac{Ct}{\varepsilon_0 A} \quad (1)$$

$$\varepsilon'' = \varepsilon' \tan\delta \quad (2)$$

where, C is the capacitance of the pellet in farad, t is the thickness of the pellet in meter, A is the cross sectional area of the flat surface of the pellet and ε_0 is the permittivity constant of free space.

The effect of frequency f on the dielectric constant (ε') and loss (ε'') is shown in Figs. 1 and 2 for

$CoIn_xFe_{2-x}O_4$ (for x = 0.0, 0.1, 0.2, 0.3, 0.4 and 0.5) samples prepared by sol–gel technique.

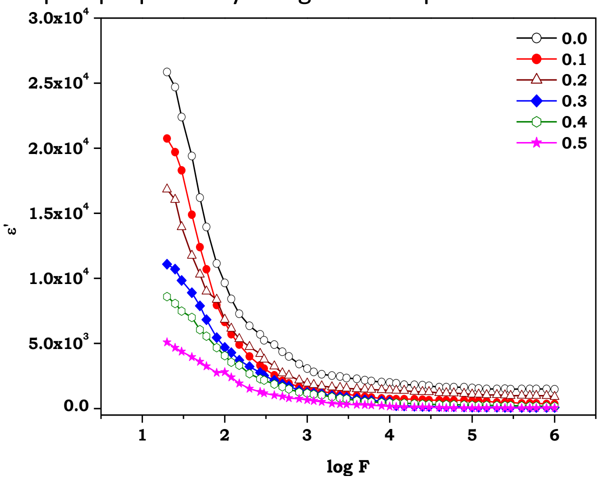

Figure 1: Variation of dielectric constant (ε') with logf for $CoIn_xFe_{2-x}O_4$

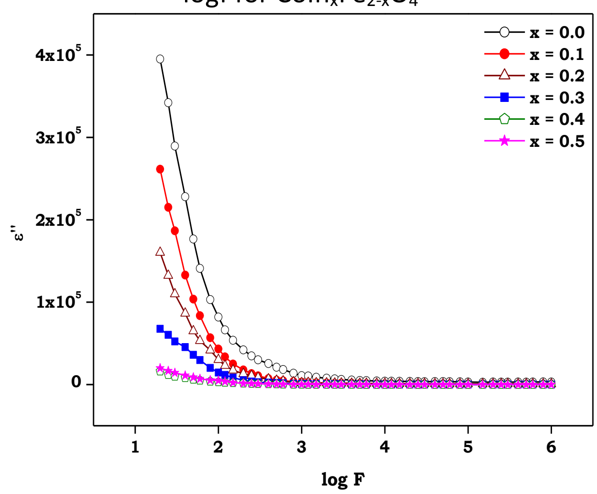

Figure 2: Variation of dielectric loss (ε'') with logf for $CoIn_xFe_{2-x}O_4$

It can be seen from Figs. 1 and 2 that both dielectric constant and loss, decrease with increase of frequency. Similar type of behavior was observed in the literature [6, 7]. The dispersion (exponential decay) in ε' and ε'' with increase in frequency is due to the fact that the polarization decreases with increasing frequency and then reaches a constant value. The observed variation in ε' and ε'' may be understood on the basis of space charge polarization, which is due to an inhomogeneous structure governed by the number of space charge carriers and the resistivity of the samples [8] it can be understood on the basis of Maxwell–Wagner type interfacial polarization and is in agreement with Koop's phenomenological theory [9, 10]. The dielectric structure of ferrites is made up of well conducting layer of grains followed by poorly conducting layer of grain boundaries and the high value of dielectric constant arises from the space charge polarization produced at the grain boundary. The polarization mechanism involves the exchange of electrons between the ions of the same element, which are present in more than one valence state and are distributed randomly over crystallographic equivalent sites. Here the exchange of electrons mainly takes place between Fe^{3+} and Fe^{2+} ions present at octahedral [B] sites. During this exchange mechanism, the electrons have to pass through the grains and grain boundary of the dielectric medium. Owing to high resistance of the grain boundary, the electrons accumulate at the grain boundary and produce space charge polarization [11].

The dielectric loss tangent for each sample was calculated using the values of dielectric constant and the dielectric loss. The variation of dielectric loss tangent (tanδ) with frequency is shown in Fig. 3. It is evident from Fig. 3 that, like dielectric constant, dielectric loss tangent also decreases exponentially with increasing frequency.

According to Iwauchi [12], there is a strong correlation between the conduction mechanism and the dielectric behavior of ferrites. The dielectric loss in ferrites is considered to originate from two mechanisms namely electron hopping and charge defect dipoles. The former contributes to the dielectric loss at low frequency region. At high frequency the dielectric loss manly results from defect dipoles to the field. These dipoles are formed due to the change in the cation state (Fe^{3+}/Fe^{2+}) during the sintering process. Relaxation of dipoles under an electric field is decreased with increase in frequency ultimately results in the dielectric loss at high frequency [13]. The observed variation of dielectric loss tangent with frequency is normal behaviour of ferrites at low frequency (corresponds to the high resistivity of grain boundaries) more energy is required for electron hoping and as result the loss is high. In the high frequency region (corresponds to high conductivity of grains) energy required for the hopping of electrons is less and hence the tanδ decreases [14].

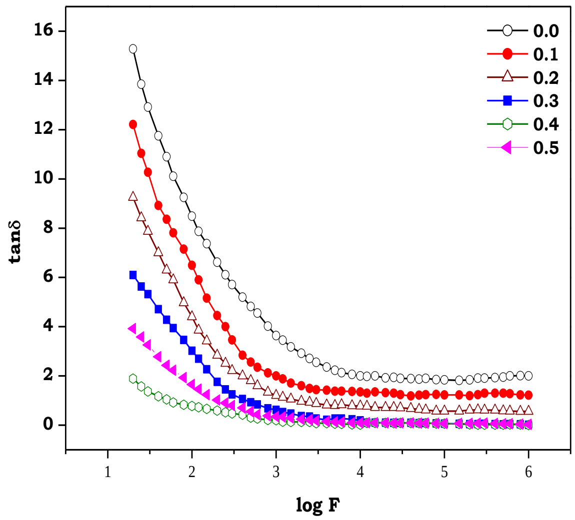

Figure 3: Variation of dielectric loss tangent (tanδ) with logf for $CoIn_xFe_{2-x}O_4$

Compositional Dependence

The values of dielectric constant, dielectric loss and dielectric loss tangent at 100 Hz and 1 MHz frequency are given in Table 1.

Table 1: Room temperature dielectric constant (ε'), dielectric loss (ε'') and dielectric loss tangent (tanδ) at 100 Hz and 1 MHz, for $CoIn_xFe_{2-x}O_4$ nanoparticles

Variation of dielectric constant (ε') determined at frequencies 100 Hz and 1 MHz as a function of In^{3+} concentration x is shown in Fig. 4.

Comp . x	f =100 Hz			f =1 MHz		
	ε'	ε"	tan δ	ε'	ε"	tan δ
0.0	965	8192	8.4	148	2960	2.0
0.1	664	4313	6.4	358	429	1.2
0.2	684	3009	4.4	900	414	0.46
0.3	470	1457	3.1	179	2.68	0.01
0.4	406	3162	0.7	77	0.30	0.00
0.5	280	4928	1.7	49	1.56	0.03

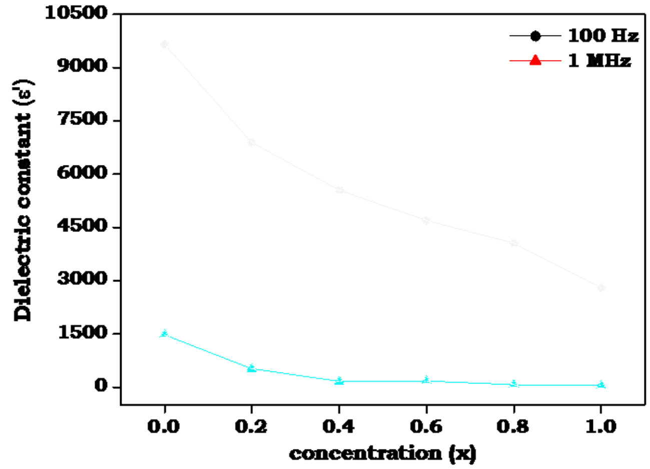

Figure 4: Variation of dielectric constant with indium concentration x at 100 Hz and 1 MHz for $CoIn_xFe_{2-x}O_4$

It can be seen from Fig. 4 that the dielectric constant (ε') of $CoIn_xFe_{2-x}O_4$ ferrite nanoparticles decreases with the increase of In^{3+} concentration. This decrease in dielectric constant (ε') can be explained as In^{3+} ions are substituted for Fe^{3+} ions, the concentration of Fe^{3+} ions is decreased and the rate of hopping process is reduced.

The mechanism for the electrical conduction is similar to that of the dielectric polarization. The compositional dependence of dielectric constant can be explained on the basis of this theory. In ferrites, the electronic exchange between $Fe^{2+} \rightarrow Fe^{3+}$ in octahedral sites results in local displacement of electrons in the direction of the applied electric field which determines electric polarization behaviour of the ferrites [15].

It is known that In^{3+} ions occupy octahedral sites [17] due to their larger ionic radius. The concentration of Fe^{3+} ions at B-sites decreases monotonously with increasing concentration of Indium. The reduction in the values of dielectric constant with increasing concentration of Indium is due to depleting concentration of iron ions at [B] sites which play a dominant role in dielectric polarization. The electron transfer between Fe^{2+} and Fe^{3+} ions ($Fe^{2+} \leftrightarrow Fe^{3+} + e^-$) will be hindered i.e. the polarization decreases. Consequently, dielectric constant decreases with Indium contents.

Conclusion:

The dielectric constant, dielectric loss and dielectric loss tangent all decreases exponentially with increase in frequency. The dielectric constant measured at low and high frequency also decreases with indium substitution x

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