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Dielectric Properties of Ni-Zn Ferrites Synthesized by Citrate Gel Method

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ABSTRACT

Ni-Zn ferrite with composition of $Ni_{1-x}Zn_xFe_2O_4$ (x = 0.0, 0.2, 0.4, 0.6, 0.8, 0.9, 1.0) were prepared by citrate gel method. The Dielectric Properties for all the samples were investigated at room temperature as a function of frequency. The dielectric constant shows dispersion in the lower frequency region and remains almost constant at higher frequencies. The frequency dependence of dielectric loss tangent ($\tan\delta$) is found to be abnormal, giving a peak at certain frequency for mixed Ni-Zn ferrites. A qualitative explanation is given for the composition and frequency dependence of the dielectric loss tangent.

Keywords: Ferrites; Citrate Method; Lattice Parameter; Dielectric Constant; Dielectric Loss

1. Introduction

Ni-Zn ferrite is a well-known spinel magnetic material. In the inverse spinel structure of $NiFe_2O_4$, the tetrahedral sites are occupied by ferric ions and octahedral sites by ferric and nickel ions. $Ni_{1-x}Zn_xFe_2O_4$ ferrites are ferrimagnetic materials with a large number of technological applications in telecommunications and entertainment electronics. Ni-Zn ferrites are among the most widely used soft magnetic materials because of high frequency applications as they posses high electrical resistivity and low eddy current losses [1-3]. It is known that magnetic properties of ferrites are sensitive to preparation technique and their microstructures [4]. The electrical and magnetic properties of such ferrites depend strongly on distribution of cations at the tetrahedral (A) and octahedral (B) sites in the lattice [5-7]. It is well known that zinc ions can be used to alter the saturation magnetization. It is believed that the addition of zinc ions also affects the lattice parameter and it would therefore be expected to change the Curie temperature of the material [8]. The substitution of divalent ions in pure ferrites leads to the modification of the structural, electrical and magnetic properties [9]. The conventional solid-state reaction route is widely used for the production of ferrite because of its low cost and suitability for large scale production.

The citrate method is used to speed up the synthesis of complex materials. It is a simple process, which offers a significant saving in time and energy consumption over the traditional methods. Several researchers have reported the synthesis of Ni-Zn ferrites using different techniques like, refluxing process [10], ceramic [11], hydrothermal [12], combustion [13], co-precipitation [14], reverse micelle process [15], spark plasma sintering [16], micro emulsion [17] and ball milling etc.

In this work, we present the results of systematic doping of non-magnetic Zn content on the dielectric properties of Ni-Zn ferrite synthesized by citrate method.

2. Experimental

The starting materials were nickel nitrate, zinc nitrate, iron nitrate, citric acid and ammonia all of analytical grade. The solution of nickel nitrate ($Ni(NO_3)_2 \cdot 6H_2O$, ferric nitrate ($Fe(NO_3)_3 \cdot 9H_2O$) and zinc nitrate ($Zn(NO_3)_2 \cdot 6H_2O$) in their stoichiometry were dissolved in a de ionized water. Citric acid was then added to the prepared aqueous solution to chelate Ni^{2+}, Zn^{2+} and Fe^{3+} in the solution. The molar ratio of citric acid to total moles of nitrate ions was adjusted at 1:3. The mixed solution was neutralized to pH 7 by adding ammonia (NH_3) solution. The neutralized solution was evaporated to dry ness by heating at 100°C on a hot plate with continuous stirring, until it becomes

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viscous and finally formed a very viscous gel. Increasing the temperature up to 200°C leads to ignition of gel. The dried gel burnt completely in a self propagating combustion manner to form a loose powder. Finally the burnt powder was calcined in air at temperature of 1000°C for one hour to obtain spinel phase. Afterwards the powder was pressed into pellets of thickness 3 mm and a diameter of 10 mm with press by applying a pressure of 2 tons/in^2. The final sintering was done at 1000°C, afterwards the pellets were coated with silver paint for better electrical contact to measure the dielectric properties.

The structural characterizations of all samples were carried out by X-ray diffraction (XRD) and conforms the well defined single phase spinel structure. XRD data were taken at room temperature using CuKα radiation. The dielectric data are measured by LCR meter at room temperature in the frequency range 2 Hz to 2 MHz.

3. Results and Discussions

All the zinc substitute nickel ferrites of the various compositions show the crystalline cubic spinel structure. The sharp peaks showed all-crystalline nature of single phase ferrite. The lattice parameter of individual composition was calculated by using the formula

$$a = d \left(h^2 + k^2 + 1^2 \right)^{1/2}$$

where, a = lattice constant;
       d = inter planar distance;
      $(h, k, 1)$ = the Miller indices.

The variation of lattice parameter with zinc composition is shown in **Figure 1**. The lattice parameter is found vary linearly with increasing zinc concentration, there by indicating that the Ni-Zn ferrite system obeys Vegard's law [18]. A similar behavior of lattice constant with dopant concentration was observed by several investigators in various ferrite systems [19-21]. The variation in

lattice constant with zinc content can be explained on the basis of the ionic radii of Zn^{2+} (0.82 Å) ions is higher than that of Ni^{2+} (0.78 Å) [22].

3.1. Dielecric Constant with Frequency

The effect of frequency on the real dielectric constant (ε') can be seen from **Figure 2** and that the value of dielectric constant decreases continuously with increasing frequency. The decrease in the values of dielectric constant as the frequency increases can be due to electron exchange interaction between Fe^{2+} and Fe^{3+} ions, which cannot follow the alternating electric field. The decrease of dielectric constant with increase of frequency as observed in the case of Ni-Zn ferrite. A similar behavior was also observed in [23-26] of various ferrite systems. The explanation for the decrease in the values of ε' as the frequency increases can be related to electron exchange interaction between Fe^{2+} and Fe^{3+} ions which cannot follow the alternation of the electric field beyond a certain frequency.

3.2. Dielectric Loss Tangent (tanδ) with Frequency

The variation of tanδ with frequency can be seen from **Figure 3**, it can be seen from the figure that in the case of $NiFe_2O_4$, $Ni_{0.8}Zn_{0.2}Fe_2O_4$, $Ni_{0.6}Zn_{0.4}Fe_2O_4$ and $Ni_{0.4}Zn_{0.6}Fe_2O_4$ tanδ shows maximum at a frequency of 21 Hz, in the case of $Ni_{0.1}Zn_{0.9}Fe_2O_4$, $ZnFe_2O_4$ tanδ shows maximum at a frequency of 22 Hz and $Ni_{0.2}Zn_{0.8}Fe_2O_4$ tanδ shows maximum at 25 Hz. A qualitative explanation can be given for the occurrence of the maxima in tanδ verses frequency curves in $NiFe_2O_4$, $Ni_{0.8}Zn_{0.2}Fe_2O_4$, $Ni_{0.6}Zn_{0.4}Fe_2O_4$, $Ni_{0.4}Zn_{0.6}Fe_2O_4$, $Ni_{0.1}Zn_{0.9}Fe_2O_4$, $ZnFe_2O_4$ and $Ni_{02}Zn_{0.8}Fe_2O_4$, as pointed out by Iwauchi [27], there is a strong correlation between the conduction mechanism and the dielectric behavior of ferrites. The conduction mechanism in n-type ferrites is considered as due
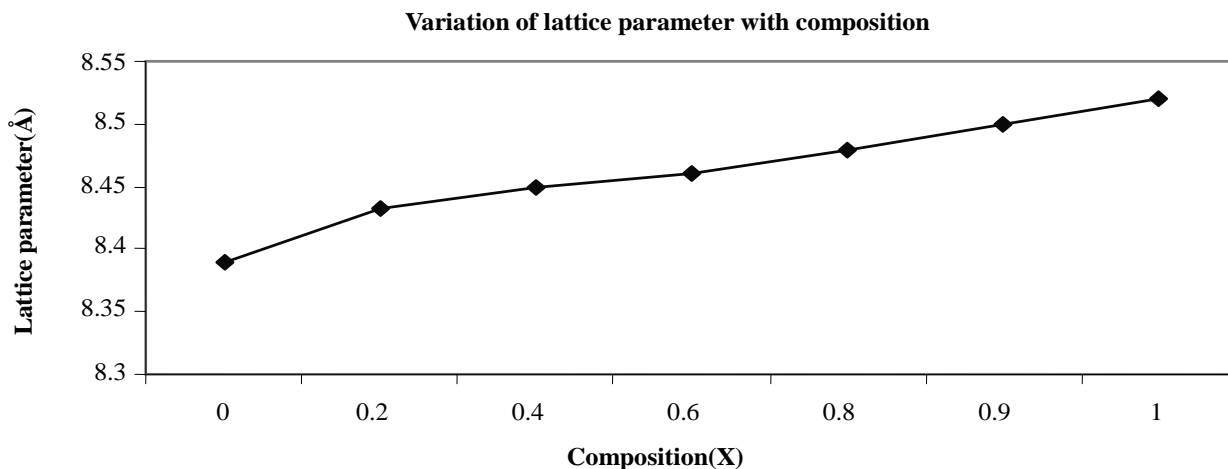

Variation of lattice parameter with composition

Figure 1. Variation of lattice parameter with composition.

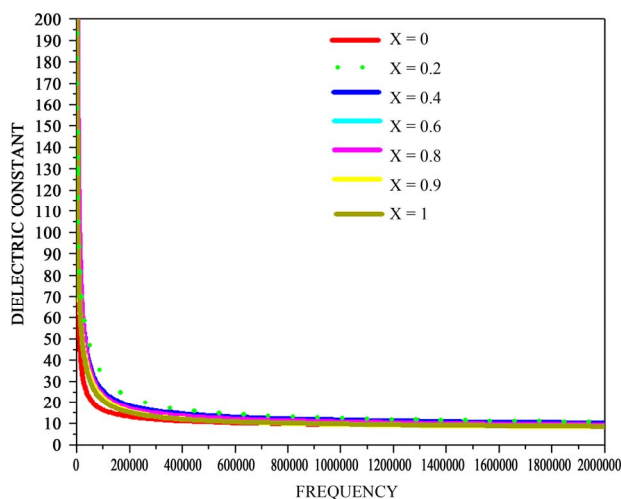   *WJCMP*

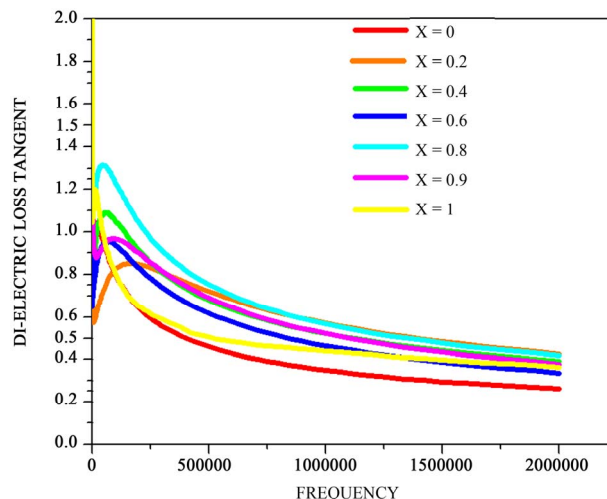**Figure 2. Variation of dielectric constant with composition.**

Figure 3. variation of dielectric loss tangent with composition.

to hopping of electrons between Fe^{2+} and Fe^{3+} situated on the octahedral sites. As seen, when the hopping frequency is nearly equal to that of external applied electric field a maximum of loss tangent may be observed [28]. As such it is possible that in the case of in $NiFe_2O_4$, $Ni_{0.8}Zn_{0.2}Fe_2O_4$, $Ni_{0.6}Zn_{0.4}Fe_2O_4$, $Ni_{0.4}Zn_{0.6}Fe_2O_4$, $Ni_{0.1}Zn_{0.9}Fe_2O_4$, $ZnFe_2O_4$ and $Ni_{02}Zn_{0.8}Fe_2O_4$, the hoping frequencies are of the approximate magnitude to observe a loss maximum at 21, 22, 25 Hz respectively.

3.3. Conclusion

It may be concluded that a series of Ni-Zn ferrite with compositional formula $Ni_{1-x}Zn_xFe_2O_4$ where x = 0.0, 0.2, 0.4, 0.6, 0.8, 0.9 & 1.0 are prepared by citrate gel method. The lattice constant was found to be increases with zinc composition. The variation of dielectric constant and dielectric loss tangent was explained on the basis of electronic exchange between the Fe^{2+} and Fe^{3+} ions.

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