

## INFLUENCE OF YTTRIUM SUBSTITUTION ON STRUCTURAL AND TRANSPORT PROPERTIES OF Ni-Zn ( $\text{Ni}_{0.25}\text{Zn}_{0.75}\text{Y}_x\text{Fe}_{2-x}\text{O}_4$ ) FERRITE

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### ABSTARCT

The Polycrystalline Ni-Zn ferrite of composition  $\text{Ni}_{0.25}\text{Zn}_{0.75}\text{Y}_x\text{Fe}_{2-x}\text{O}_4$  [ $x = 0.00, 0.02, 0.04, 0.06$  and  $0.08$ ] were prepared by double sintering solid state reaction method. The X-ray diffraction pattern ensures the formation of face centered cubic (fcc) type spinel crystal structure. With the increase of Yttrium (Y) content, the lattice parameter increases. The permeability of all the samples was studied extensively by observing the real part of permeability  $\mu'$ , the imaginary part of permeability  $\mu''$  and the relative quality factor ( $Q$ -factor) as a function of frequency. Here the real part of permeability remains unchanged until at a high frequency and then decreases. The  $Q$ -factor initially has a trend of increasing with frequency and after a certain frequency it decreases. The dielectric constants for all samples were inspected with frequency variation. With the increasing frequency, the dielectric constant has a trend of decreasing and it becomes frequency independent at high frequency region.

**Keywords:** Ferrites, XRD, Complex permeability, Frequency dependence, Dielectric constant

### 1. INTRODUCTION

Ferrites are the ferromagnetic oxide materials that are famous for their vast applications in human life as well as in research areas. Polycrystalline ferrites are a type of ferromagnetic materials that has been studied extensively for their importance in both fundamental and experimental aspects. Various effective magnetic and transport properties such as high resistivity, low magnetic loss, thermodynamic stability, high permeability, chemical stability, mechanical hardness, low cost etc. can be found in polycrystalline ferrites. These properties provide precise information to make them tremendously applicable for various electronic devices such as microwave devices, multilayer chip inductors, transformers, electric generator storage devices, etc (Nakamura, 1997; Shahida Akhter *et al.*, 2013).

The structural, magnetic and transport properties of polycrystalline ferrites depend on the synthesis technique, thermal operation, pH value, sintering temperature and relative distribution of cations of the specimen. In case of fcc type spinel ferrite, the metal ions are occupied by two sites naming A-site (tetrahedral) and B-site (octahedral). Various magnetic behaviors like paramagnetism, ferromagnetism, antiferromagnetism, ferrimagnetism, superparamagnetism etc. are deeply dependent on A- and B-site cation distribution (Gupta *et al.*, 1968; Lange, 1989; Kigery *et al.*, 1975; Ahmed *et al.*, 2003). Basically, the lattice of tetrahedral A-site ions magnetized spontaneously in one direction and in the opposite direction for the lattice in octahedral B-site ions. However, the value of magnetization are not equal for A and B sublattice. There is no cancellation occurs by two opposite magnetic and it results a net spontaneous magnetization (Shil *et al.*, 2013).

The present work deals with to prepare and synthesize Ni-Zn ferrite by conventional solid state reaction method and to study the results of Yttrium doping on structural and transport phenomenon's including XRD pattern, relative permeability and dielectrics measurement observation.

### 2. EXPERIMENTAL

To synthesize the samples using solid state reaction method, the powdered composition of  $\text{Ni}_{0.25}\text{Zn}_{0.75}\text{Y}_x\text{Fe}_{2-x}\text{O}_4$  [ $x = 0.00, 0.02, 0.04, 0.06$  and  $0.08$ ] were usually prepared from the analytical grade powder of raw oxides (NiO, ZnO,  $\text{Y}_2\text{O}_3$  and  $\text{Fe}_2\text{O}_3$ ) that were bought from market by crushing the materials, grinding as well as milling. Solid state reaction occurs between evidently regular crystal lattices, where the kinetic motion is very much confined. Moreover, it also depends on the lattice defects presented in crystals. In solid state reaction method, appropriate amounts of two or more components of chemical compounds are carefully grinded together and mixed thoroughly in a mortar with pestle with appropriate homogenization. Before heat treatment, the powdered samples were taken to make both pellet and ring shaped samples using die-punch assembly of Hydraulic press. The ground powders were then taken for presintering in air at a temperature above  $850^\circ\text{C}$ . The calcined powders were then further crushed to prepare fine powders. The pellets/disc and toroid shaped samples were made of these powders using uniaxial or iso-static pressure. Sintering was carried

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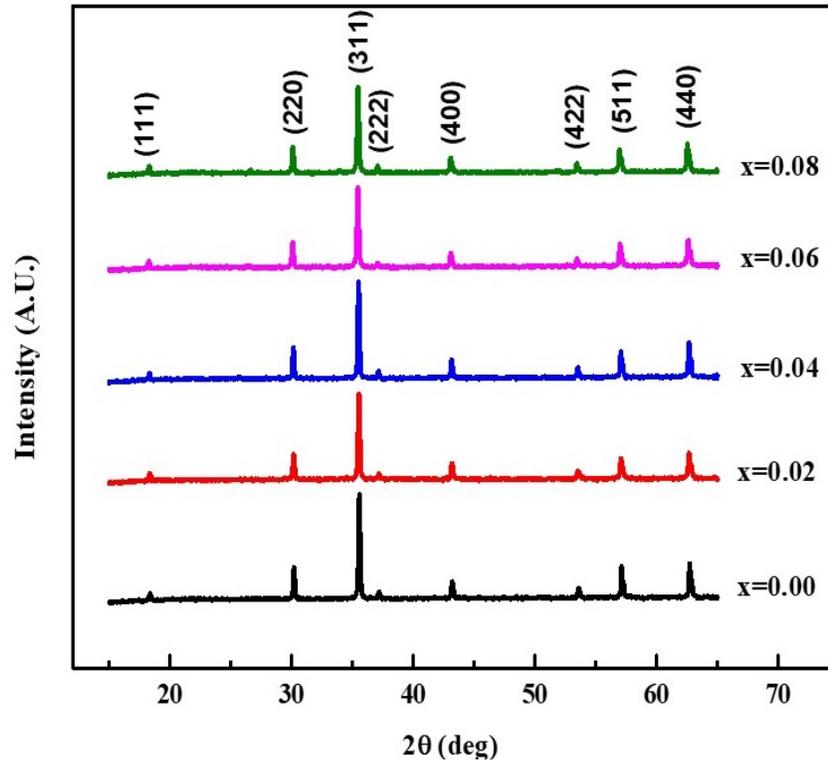
out in the solid state, at temperature 1150°C. For structural characterization of the specimens, X-ray diffraction pattern were carried out for both as-dried samples and for Y doped samples using Philips X'pert Pro powder X-ray diffractometer. The transport properties were observed by using Wayne Kerr Precision (6500B series, England) Impedance analyzer in the solid state physics laboratory of KUET.

### 3. RESULTS AND DISCUSSION

#### 3.1 Structural Investigation

The structural view or the various phases of Y doped Ni-Zn ferrites were confirmed from XRD analysis. Figure 1 shows the XRD patterns of  $\text{Ni}_{0.25}\text{Zn}_{0.75}\text{Y}_x\text{Fe}_{2-x}\text{O}_4$  [ $x = 0.00, 0.02, 0.04$  and  $0.06$ ] ferrites sintered at 1150 °C for 3 hours. The indexing for all samples that are shown in XRD (Figure 1) indicating the formation of fcc type spinel structure. The Figure 1 shows eight sharp peaks and the peaks can be indexed as (111), (220), (311), (222), (400), (422), (511), and (440). Here, all the miller indices of a peak are either all odd or even, which confirmed that the samples are spinel lattice with cubic structure. This also confirms the homogeneity of the studied samples (Shil *et al.*, 2013; Vucinic-Vasic *et al.*, 2013). The lattice parameter “a” of pure samples and the samples with different concentrations of Y were determined using the following formula,

$$a = d_{hkl} \times \sqrt{h^2 + k^2 + l^2} \quad (1)$$



**Figure 1:** XRD patterns for the  $\text{Ni}_{0.25}\text{Zn}_{0.75}\text{Y}_x\text{Fe}_{2-x}\text{O}_4$  [ $x = 0.00, 0.02, 0.04, 0.06$  and  $0.08$ ] sintered at 1150 °C.

**Table 1:** Data of the lattice parameter (a), bulk density ( $\rho_B$ ), X-ray density ( $\rho_x$ ), porosity (%) for  $\text{Ni}_{0.25}\text{Zn}_{0.75}\text{Y}_x\text{Fe}_{2-x}\text{O}_4$ , [ $x=0.00, 0.02, 0.04, 0.06$  and  $0.08$ ]

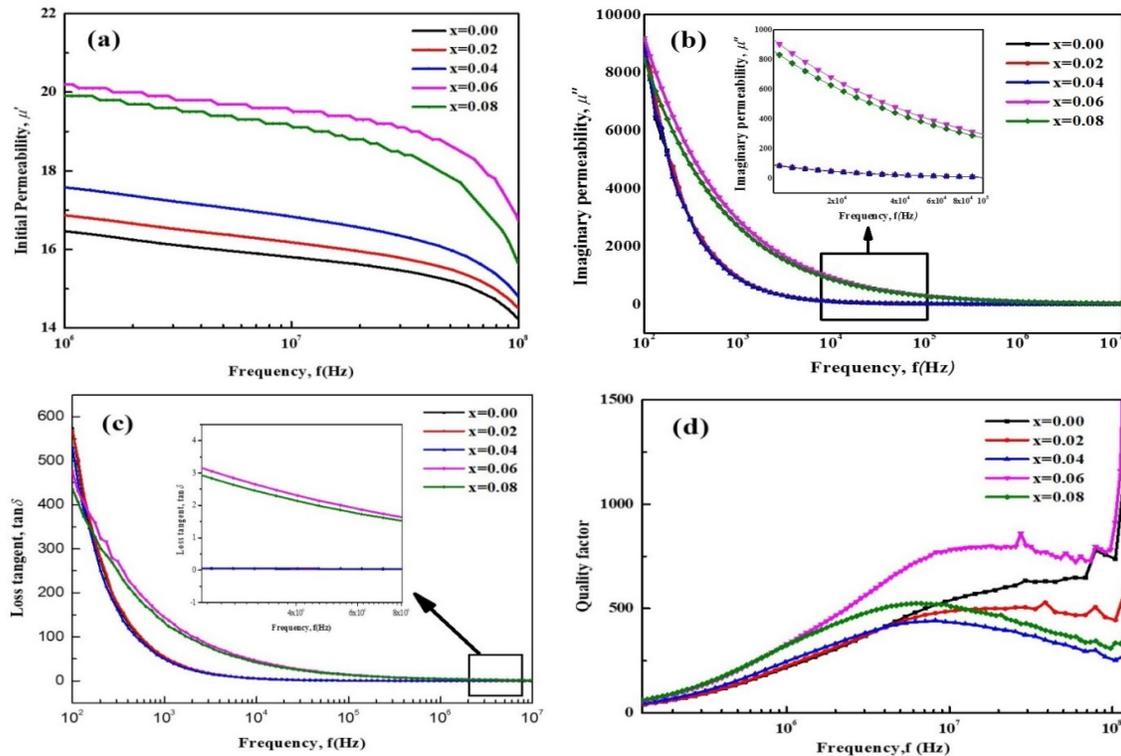
Content	a (Å)	$\rho_x$ (gm/cm <sup>3</sup> )	$\rho_B$ (gm/cm <sup>3</sup> )	Porosity (%)
X= 0.00	8.375	5.41	4.90	9.55
X= 0.02	8.384	5.41	4.75	12.20
X= 0.04	8.399	5.40	4.82	10.60
X= 0.06	8.403	5.40	4.91	9.10
X= 0.08	8.398	5.43	4.86	10.50

Various parameters such as Lattice parameter, density and porosity are shown in Table 1. It can be seen here that with the increase of Y content in  $\text{Ni}_{0.25}\text{Zn}_{0.75}\text{Y}_x\text{Fe}_{2-x}\text{O}_4$ , lattice parameter increases which results the

variation of surface to volume ratio. The increase in lattice parameter happens because of the replacement of comparatively larger  $Y^{3+}$  with  $Fe^{3+}$  ion in octahedral B-site. Here, with the increase of Y content, Bulk density and X-ray density increases. Moreover, the porosity has the trend of decreasing.

### 3.2 Frequency Dependent Permeability Observation

The expression for the complex permeability can be represented by  $\mu = \mu' - i\mu''$ , where  $\mu'$  and  $\mu''$  indicating the real part and imaginary part of the complex permeability respectively. (Haque *et al.*, 2008; Hu Jun *et al.*, 2005; Tsutaoka *et al.*, 1995; Wohlfarth, 1980; Smit *et al.*, 1959). For all the samples of  $Ni_{0.25}Zn_{0.75}Y_xFe_{2-x}O_4$ , ferrites, the complex permeability has been observed within the frequency range from 1 KHz to 120MHz that are shown in figure 2. It can be seen that  $\mu'$  remains almost persistent until very high frequency (around 60MHz) and then drops rapidly. The fairly constant  $\mu'$  values over a wide range of frequency region are known as the zone of utility because it demonstrates the stability of composition and quality of the prepared samples. Here the initial permeability remains stable for a long frequency range due to bouncing of electron between  $Fe^{2+}$  and  $Fe^{3+}$ . It could be observed that the significant rise of initial permeability occurs with substitution of Y content. A ferrite inductor exhibits the properties of a low loss constant self-inductor where  $\mu'$  becomes stable and the core becomes mostly inductive (Snelling, 1988; Khan *et al.*, 2013). From the Figure 2 the ranges of operating frequency up to 30MHz at constant frequency all the doped samples wider than the other ferrite sample and quality of the doped  $Y^{3+}$  in Ni-Zn ferrites. The substitution of ferromagnetic  $Fe^{3+}$  by diamagnetic  $Y^{3+}$  in the spinel is not significant for increasing permeability. In this present case  $Y^{3+}$  incorporated into the lattice is very small and hence slight enhanced permeability with increasing  $Y^{3+}$  ions due to incorporation. The imaginary part of permeability  $\mu''$  exhibits exponentially decreasing phenomenon with the increase of frequency and at comparatively high frequency, the value of  $\mu''$  becomes stable for all the concentrations.



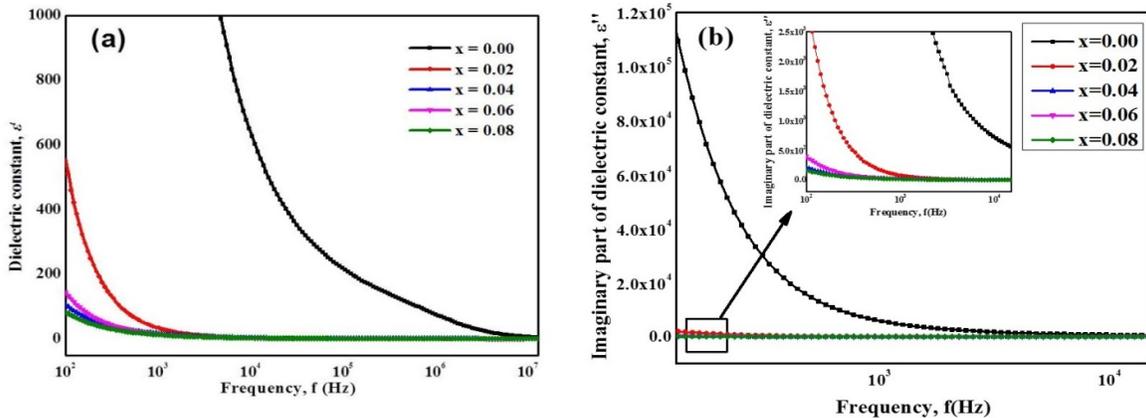
**Figure 2:** The frequency dependent (a) Real part of complex permeability, (b) Imaginary part of complex permeability, (c) Loss tangent and (d) Quality factor of  $Ni_{0.25}Zn_{0.75}Y_xFe_{2-x}O_4$  [ $x = 0.00, 0.02, 0.04, 0.06$  and  $0.08$ ].

Q-factor for all the samples has a trend of increasing with frequency and at a point it becomes maximum. After that point, deterioration occurs which is responsible for the dropping of domain wall motion with respect to the applied magnetic field. The loss tangent  $\tan \delta$  is related to Q-factor reciprocally. That's why, with the increase of frequency it decreases for all the samples that are presented in Figure 2(c).

### 3.3 Frequency Dependent Dielectric Properties Observation

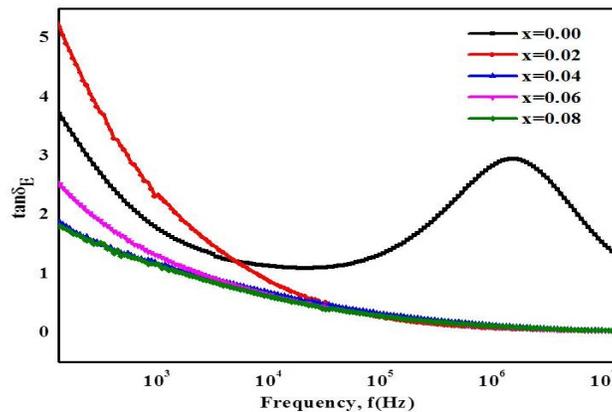
The variation of dielectric constant  $\epsilon'$  for all samples with respect to frequency (ranging from 1 KHz to 120 MHz) at room temperature are shown in Figure 3. It can be found here that, continuous decrease of dielectric constant occurs with the increase of frequency for all the specimens. The decreasing phenomenon of dielectric constant with increasing frequency is a conventional behavior of ferrites, which was observed before for various polycrystalline ferrites (Jadhav *et al.*, 1999; Kodama, 1999).

At high frequency, there is no significant effect of dielectric constant due to rare earth substitution like  $Y^{3+}$  into these ferrites. But at low frequency, a drastically decreasing dielectric constant can be observed. The electron distribution between  $Fe^{2+}$  and  $Fe^{3+}$  is responsible for the contained displacement of electrons in the direction of applied electric field, that creates the polarization (Zhenxing Yue *et al.*, 2001). Here the density of  $Fe^{2+}/Fe^{3+}$  on B site operates the magnitude of exchange of electron.



**Figure 3:** The frequency dependent dielectric of (a) real part and (b) Imaginary part of  $Ni_{0.25}Zn_{0.75}Y_xFe_{2-x}O_4$  [ $x = 0.00, 0.02, 0.04, 0.06$  and  $0.08$ ].

The imaginary part of complex dielectric constant,  $\epsilon''$  was also decreased with increasing frequency as shown in Figure 3(b). For very high frequency the value of  $\epsilon''$  is so small that it does not depend on applied frequency. The rapid decrease of  $\epsilon''$  at lower frequencies can also be explained by space charge polarization. We know that ferrites are made up of highly conducting layer of grains followed by less conducting layer of grain boundaries. During the exchange process between  $Fe^{3+}$ - $Fe^{2+}$  the electrons have to propagate through the grains and grain boundary of the dielectric medium. After that the electrons gather together at grain boundary that causes space charge polarization. At low frequency the grain boundaries become more effective where grains are effective at high frequency. For this reason, grain boundary effect is responsible for decreasing imaginary part of the dielectric constant rapidly at low frequency region. At high frequency the polarization decreased, thus resulting in a decrease in the imaginary part of the dielectric constant.



**Figure 4:** The loss tangent for the frequency dependent dielectric of  $Ni_{0.25}Zn_{0.75}Y_xFe_{2-x}O_4$  [ $x = 0.00, 0.02, 0.04, 0.06$  and  $0.08$ ].

Figure 4 shows the variation of  $\tan \delta_E$  with frequency for ferrites at room temperature. The values of loss tangent decreases with increasing Y content in  $Ni_{0.25}Zn_{0.75}Y_xFe_{2-x}O_4$  ferrites. The variation of  $\tan \delta_E$  with respect to

frequency can be explained by two mechanisms: electron hopping and charge defect dipoles. The formation of dipoles contributes to loss tangent in the low frequency range and defect dipoles are responsible at high frequency range in ferrites are formed due to exchange  $\text{Fe}^{3+}$ - $\text{Fe}^{2+}$  exchange, during sintering process. In the low frequency region ferrite contains high resistivity, then grain boundary requires more energy to exchange electron between  $\text{Fe}^{2+}$ - $\text{Fe}^{3+}$  ions. For this reason the energy loss factor,  $\tan \delta_E$  is high in the low frequency range (Slatineanu *et al.*, 2013). At high frequency region the resistivity is very low, that's why it requires very small amount of energy for electron interaction between  $\text{Fe}^{2+}$  and  $\text{Fe}^{3+}$ . But for  $x = 0.0$ , no Yttrium was doped and hence at high frequency region, the material exhibits slightly resistive behavior at a specific frequency (nearly  $10^6$  Hz) for which it is required noticeably higher energy for the exchange of electron which shows controversial behavior than other samples at that frequency region.

#### 4. CONCLUSIONS

The effects of  $\text{Y}^{3+}$  doping on  $\text{Ni}_{0.25}\text{Zn}_{0.75}\text{Y}_x\text{Fe}_{2-x}\text{O}_4$  were inspected and some key points can be summarized as follows:

- The X-ray diffraction pattern for all compositions confirms the origination of fcc type cubic spinel structure.
- The lattice parameters have the trend of increasing with the increase of  $\text{Y}^{3+}$  in the composition. Moreover, the bulk density and X-ray density also shows increasing phenomenon.
- The frequency dependent initial permeability ( $\mu'$ ) remains almost unchanged until very high frequency and then drops rapidly.
- With the increase of  $\text{Y}^{3+}$ , the rapid decrease of dielectric constant happens in the low frequency region and becomes independent of frequency at high frequency region.

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