

## Structural and Electric Properties of Titanium Substituted Ni-Cu-Zn Ferrite

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### Abstract

*This paper reports the preparation and characterization of  $Ti_x(Ni_{0.12}Cu_{0.28}Zn_{0.6}Fe_{2}O_{4+\delta})_{1-x}$  ferrite by solid state reaction. In present work, the impact of  $Ti^{4+}$  substituted  $Ti_x(Ni_{0.35}Cu_{0.05}Zn_{0.6}Fe_{1.98}O_{4+\delta})_{1-x}$  ferrite is investigated by varying the concentration of the  $Ti^{4+}$  with  $x = 0.003, 0.0765, 0.15$ . The compound is synthesized by solid state method. Structural and electrical properties have been explored using XRD, FTIR, and dielectric spectroscopy technique. X-ray diffraction (XRD) patterns reveal the formation of the cubic spinel phase in the samples after sintering the compound at  $1000^{\circ}C$ . The mean crystallite size  $D_{XRD}$  of the samples determined from XRD line broadening is 35.18–44.68 nm. The dielectric constant vary as a function of frequency and composition at room temperature. It is observed that with the increase in the concentration of  $Ti^{4+}$  ions dielectric constant increases.*

**Keywords:** Ferrite; X-Ray Diffractions; Spinel Structure; dielectric measurement

### Introduction

Nano crystalline spinel ferrites have been investigated in the recent years by many researchers. These ferrites are materials of interest because of their unique electric, dielectric and magnetic properties. These ferrites are very important group of magnetic materials due to their extensive use in a wide range of applications from low to high permeability devices including electronics, ferrofluid, magnetic drug delivery microwave devices, and high density information storage devices [1-5].

### Experimental Details

#### Sample preparation

Titanium substituted NiCuZn ferrites of the cubic spinel structure  $[Ti_x(Ni_{0.35}Cu_{0.05}Zn_{0.60}Fe_{1.98}O_{4+\delta})_{1-x}]$  was synthesized by conventional solid state technique. High purity 'AR' grade oxides of iron, titanium, nickel, copper and zinc were used to prepare  $[Ti_x(Ni_{0.35}Cu_{0.05}Zn_{0.60}Fe_{1.98}O_{4+\delta})_{1-x}]$  ( $0.003 \leq x \leq 0.15$ ) ferrite by using solid state reaction method. The stoichiometric proportions of these constituent oxides were weighed, intimately mixed and

the resulting powders were ball-milled using Agate mortar and pestle in aqueous medium. Ball milled compound is calcined at 750<sup>0</sup>C for six hours. Subsequently the compound obtained is put in the pellet form and the pellets were sintered at 1000<sup>0</sup>C for four hours.

X-ray diffraction measurement was carried using XPERT-PRO diffractometer system with CuK<sub>α</sub> radiation of wavelength 1.540598 nm. Dielectric measurement were performed using LCR HI- Tester (HIOKI) in the frequency range 20Hz to 20MHz at room temperature.

## Results and Discussion

### X-Ray Diffraction

Figure 1 shows the powder XRD pattern of Ti<sub>x</sub>(Ni<sub>0.12</sub>Cu<sub>0.28</sub>Zn<sub>0.6</sub>Fe<sub>2</sub>O<sub>4+δ</sub>)<sub>1-x</sub> (0.003 ≤ x ≤ 0.15) ferrite. The positions and relative intensities of all the peaks indicate that the crystalline structure of the products favors the formation of cubic spinel phase only, which is accordant to JCPDS card NO. 35-1373. From the analysis of the XRD data, it has been observed that all the samples exhibit single phase cubic spinel structure. Peak intensity for (311) plane goes on decreasing as the concentration of Ti is decreased. This gives the evidence of symmetrical distribution of the crystal structure on account of Titanium. The broad XRD lines indicate that the particles are of nanosize range.[6] The peaks of (2 2 0), (3 1 1), (2 2 2), (4 0 0), (4 2 2), (5 1 1) and (4 0 0) have been deconvoluted to Lorentzian curves for the determination of the crystallite size using full-width at half-maximum.

The crystallite size (D) of the particle of the powder was determined using Scherer formula

$$D(hkl) = 0.9 \lambda / \beta \cos \theta \quad \dots\dots\dots(1)$$

where, D(hkl) is the mean dimension of the crystallite perpendicular to the plane (hkl), β is the full width at half maximum in radians and θ is the Bragg angle for the actual peak.

The lattice parameter ‘a’ is calculated using the relation,

$$a = \sqrt{(h^2 + k^2 + l^2)} \quad \dots\dots\dots(2)$$

since each primitive unit cell of the spinel structure contains 8 molecules, the value of X-ray density (dx) is calculated according to the relation,

$$d_x = \frac{8M}{Na^3} \quad \dots\dots\dots(3)$$

Where, ‘N’ is the Avogadro number 6.022x10<sup>23</sup>, ‘M’ is the molecular weight of the sample and ‘a’ is the lattice parameter.

The average particle size of  $\text{Ti}_x(\text{Ni}_{0.12}\text{Cu}_{0.28}\text{Zn}_{0.6}\text{Fe}_2\text{O}_{4+\delta})_{1-x}$  with  $x = 0.003, 0.0765$  and  $0.15$  has been calculated using the Debye–Scherrer formula[7] having value 35.1825, 44.6861 and 37.8646 nm respectively. The lattice parameters[8] and x-ray density[8] have been calculated assuming the spinel cubic symmetry and taking the three most intense peak at (311), (440) and (220).

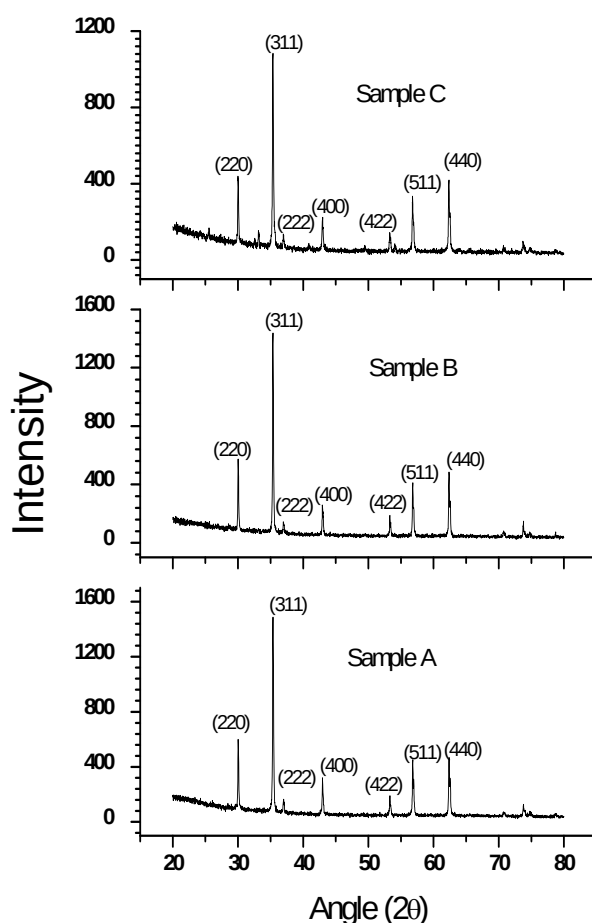


Fig. 1 XRD of  $\text{Ti}_x(\text{Ni}_{0.12}\text{Cu}_{0.28}\text{Zn}_{0.6}\text{Fe}_2\text{O}_{4+\delta})_{1-x}$  with  $x = 0.003$ (sample A),  $0.0765$ (sample B) and  $0.15$ (sample C) concentrations.

### Dielectric measurement

In figure - 2 the dielectric constant of  $\text{Ti}_x(\text{Ni}_{0.12}\text{Cu}_{0.28}\text{Zn}_{0.6}\text{Fe}_2\text{O}_{4+\delta})_{1-x}$  decreases rapidly as the frequency increases, indicating dispersive behavior in the low-frequency region and reaching the saturation limit at higher frequencies. The observed behavior may be caused by Maxwell–Wagner interfacial polarization, in agreement with Koops phenomenological theory. [9-11] The high value of the dielectric constant of ferroelectric compounds at low frequencies is commonly

explained by space charge polarization that is caused by the inhomogeneous dielectric structure. The inhomogeneities in the ferrite and ferroelectric composites normally arise from impurities, porosity, and grain structure.

The dielectric constant of ferrites declines as the frequency increases and remains constant at high frequencies, because above a certain frequency of electric field, the electric dipole does not follow the alternating field. However, the higher dielectric constant of sample C with  $x=0.15$  with respect to sample 'A' and 'B' in  $\text{Ti}_x(\text{Ni}_{0.12}\text{Cu}_{0.28}\text{Zn}_{0.6}\text{Fe}_2\text{O}_{4+\delta})_{1-x}$  compound is due to high polarization on account of increasing concentration of titanium in the compound.

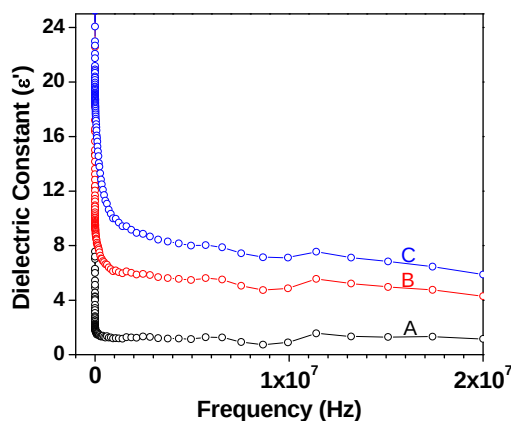


Fig. 2: Variation of dielectric constant with titanium concentration

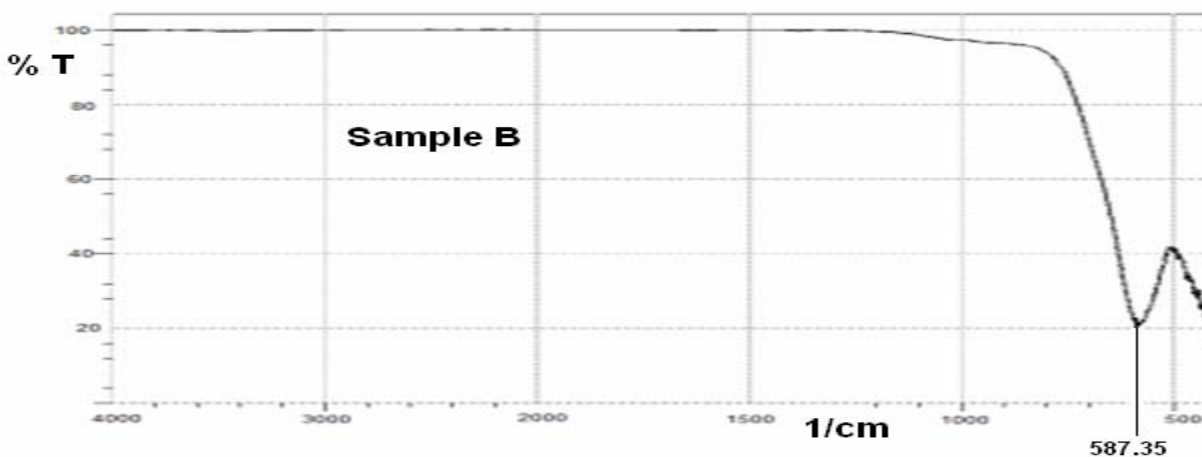
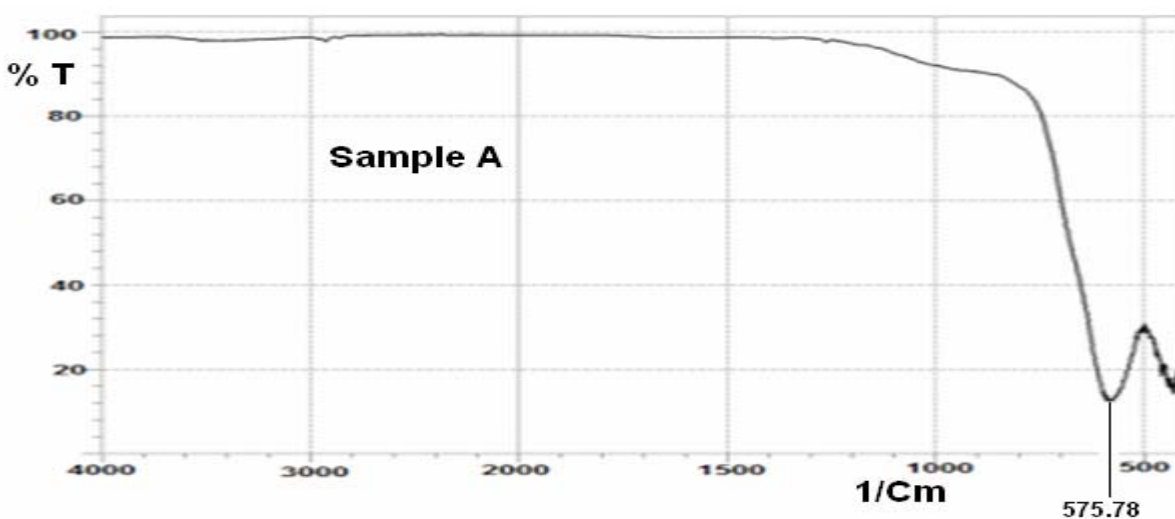
### FTIR measurement

FTIR characterization of the compound  $\text{Ti}_x(\text{Ni}_{0.12}\text{Cu}_{0.28}\text{Zn}_{0.6}\text{Fe}_2\text{O}_{4+\delta})_{1-x}$  with  $x = 0.003$  (sample A),  $0.0765$  (sample B) and  $0.15$  (sample C) concentrations were performed in the range of  $500\text{--}4,000\text{ cm}^{-1}$  (Figure 3).

The shift in the transmittance curve of FTIR results from  $575.78\text{ cm}^{-1}$  for sample A, where the  $\text{Ti}^{+4}$  concentration ( $x=0.003$ ) is least, to the  $587.35\text{ cm}^{-1}$  for sample C ( $x=0.15$ ) shows that the energy band gap is increasing which manifest the formation of the nanoparticle. The peak at  $587.35\text{ cm}^{-1}$  is owing to the presence of ferrite nano-particles.[12]

**TABLE 1.** Data on lattice constant (a), crystallite size (D), X-ray density ( $d_x$ ) NiZnCu ferrite

| X      | Molecular wt of<br>$Ti_x(Ni_{0.12}Cu_{0.28}Zn_{0.6}Fe_2O_{4+d})_{1-x}$<br>g/mole | a<br>Å | crystallite size<br>(D)<br>nm | X-ray density ( $d_x$ )<br>g/cm <sup>3</sup> |
|--------|----------------------------------------------------------------------------------|--------|-------------------------------|----------------------------------------------|
| 0.003  | 239.2743                                                                         | 7.2741 | 35.1825                       | 436.9110                                     |
| 0.0765 | 227.5226                                                                         | 7.2729 | 44.6861                       | 415.4526                                     |
| 0.15   | 215.7709                                                                         | 7.0369 | 37.8646                       | 393.9942                                     |



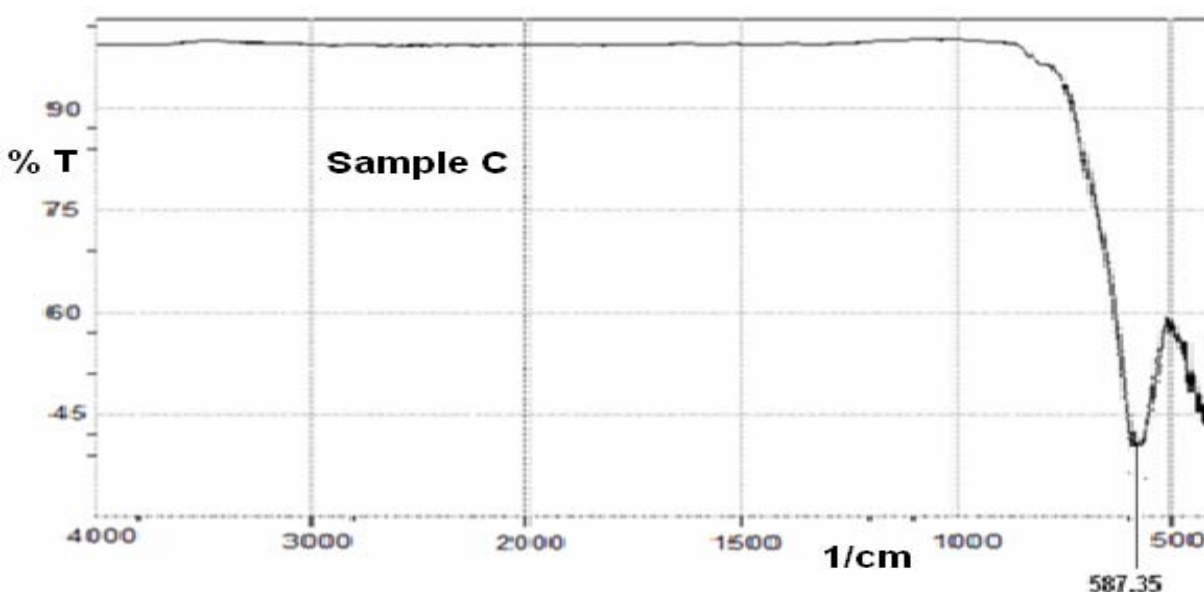


Fig.3 FTIR spectra for  $\text{Ti}_x(\text{Ni}_{0.12}\text{Cu}_{0.28}\text{Zn}_{0.6}\text{Fe}_2\text{O}_{4+\delta})_{1-x}$  with  $x = 0.003$ (Sample A),  $0.0765$ (Sample B) and  $0.15$  (Sample C) concentrations

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