Investigation of Dielectric Properties of (1-x) $Ni_{0.53}Cu_{0.12}Zn_{0.35}Fe_{1.88}O_4$ ferrite + (x) $Gd_{0.2}Ce_{0.8}O_3$ for Multilayer Chip Inductors

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***Abstract:* The composite of (1-x) $Ni_{0.53}Cu_{0.12}Zn_{0.35}Fe_{1.88}O_4$ + (x) $Gd_{0.2}Ce_{0.8}O_3$(x=0.10,0.20,0.30) were prepared by mixing nanocrystalline $Ni_{0.53}Cu_{0.12}Zn_{0.35}Fe_{1.88}O_4$ and $Gd_{0.2}Ce_{0.8}O_3$powders at different weights percents. The powders of NiCuZn ferrite were synthesized using sol gel method. The powders were densified using conventional sintering method at 1000°C/2 hrs. The phase and morphology of the composites was observed with X-ray diffraction (XRD) and Scanning Electron Microscope (SEM). The frequency dependence of real (ε′) and imaginary (ε″) parts of permittivity was measured in the range of 1MHz-1.8GHz.**

***Index Terms*—Composites, X-ray diffraction, Electrical properties.**

I. Introduction

Ferrites are usually as core materials in inductor which behaves as an impedance device and has a coil for inducing inductance to an electronic circuit. A ferrite core typically amplifies or multiplies the inductance of a coil by the permeability of the core. The chip inductor is fabricated by laminating ferrite layers and internal conductors and co-firing for monolithic structure [1]. Ferrite chip inductors are one of the important components for majority of electronic products such as cellular phones, computer notebooks, hard and floppy drives etc [2]. The decrease of sintering temperature along with increase of densification of the material are desirable conditions such that it can be co-fired with silver (Ag) internal electrode for MLCI application. The various concentration of zinc and copper and optimization of zinc are essential to achieve desirable electromagnetic properties in the ferrites those have been reported in the literature [3]. This makes Ni substituted Zinc ferrites dominant materials for multilayer chip inductor (MLCI) applications as they have good electromagnetic properties. With development of communication technology, the high performance MLCIs used in the frequency range from 500 MHz to 2 GHz. The MLCIs should have good density, high permeability and low dielectric constant assuring the high resonance frequency. The addition of ferroelectric material such as $BaTiO_3$ with NiCuZn ferrites resulted increase in the cutoff frequency [4]. The rare earth substituted materials are becoming the promising materials for different applications. Addition of small amount of rare earth ions to ferrite samples produces a change in electrical and structural properties depending on the type of substitution. Zhao et al. [5] investigated that the crystallite size of the Ni-Mn ferrites decreased as Gd ions were doped. Ahmed et al. [6] reported the electrical properties of Mg-Ti doped ferrites like doped with rare earth ions Er, Ce and Nd increased the resistivity. Sun et al. [7-8] investigated the effects of rare earth ions on the properties of $(Ni_{0.5}Zn_{0.5})Fe_{1.98}RE_{0.02}O_4$ (RE = Y, Eu or Gd) nominal compositions. The partial substitution of rare earth materials increased the density, higher electrical resistivity and relative loss factor. Hence in the present study, the NiCuZn ferrite particles were mixed with GdC in different weight percentages and sintered to form composite ferrite consisting of ferrite and rare earth materials. This paper reports the preparation, characterization and dielectric properties of rare earth doped ferrites composites.

II. Experimental

Analytical grade nickel nitrate, zinc nitrate, copper nitrate, iron nitrate and citric acid were used as raw materials to synthezied the $Ni_{0.53}Cu_{0.12}Zn_{0.35}Fe_{1.88}O_4$ ferrite material. The stoichometric proportions of molar ratio 1:1 of metal nitrates and citric acid is dissolved in the deionized water. During this process, the solution was continuously stirred using a magnetic agitator. Ammonia was added to the solution to adjust the pH value to about 9. The precipitate is then heated continuously until all the gel burnt out to form powder. The powder is then crushed using agate mortor and then heated at 65°C for 12 hours. The particle size was estimated to be around 35 nm for NiCuZn ferrite using XRD. The $Gd_{0.2}Ce_{0.8}O_3$ (GdC)is directly purchased from the Cottor international manufacture with specified grain size of 50 nm and 99.9% purity.

The ferrite powder and (GdC) material is mixed in different weight percentages of x=0.10, 0.20, 0.30. The powder was uniaxially pressed at a pressure of 1500 kg/cm^2 to form green pellets. The pellets were sintered at 1000°C and characterization is done using X-ray diffraction (XRD) and Scanning electron microscope (SEM). The phase identification of the sintered samples was performed using X-ray diffraction (XRD) with Cu-Ka radiation. The frequency

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dependence of complex permittivity was measured in the range of 1MHz to 1.8GHz using Agilent 4291B Impedance/Material Analyzer.

III. RESULTS AND DISCUSSION

X-ray diffraction patterns of composite ferrite of NiCuZn ferrite and (GdC) materials are shown in Figure 1., indicating that the sintered powders show the peaks for both the materials. Fig.1 also indicates the presence of two crystallite phases of NiCuZn ferrite and both (GdC) which coexist in all of three sintered composites. The lattice constants of composite material is estimated and tabulated in Table 1. from the table, it can be observed that the lattice constants decreases with increase in rare earth dopants. It was also found that the density of the present composites vary 90% that of theoretical density and hence the values of porosity vary around 10%.

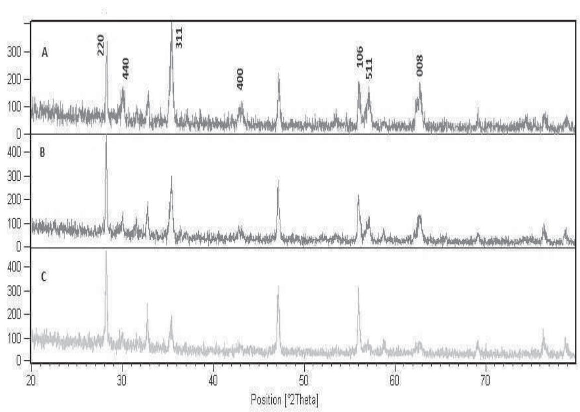

Figure 1. XRD of the composites

Sample Name	Lattice constant		Grain Size (SEM) nm	ε′ (1GHz)	ε″ (1GHz)
	a(A°) ±0.001	c(A°) ±0.001			
A	5.351	7.656	92	19.64	0.384
B	5.276	7.456	104	19.24	0.337
C	5.125	7.223	117	18.00	0.318

Table I. Data of lattice constant, grain size and electrical properties

Figure 2. shows the SEM pictures for conventional sintered samples. The average size of the composite ferrite, geometrically estimated from SEM photographs of composite sintered samples and results are presented in Table I. In the images, the black grains are ferrite grains and the white ones are ferroelectric grains. With increasing ferrite content, the ferrite grains increase and the ferroelectric grains decrease continually.

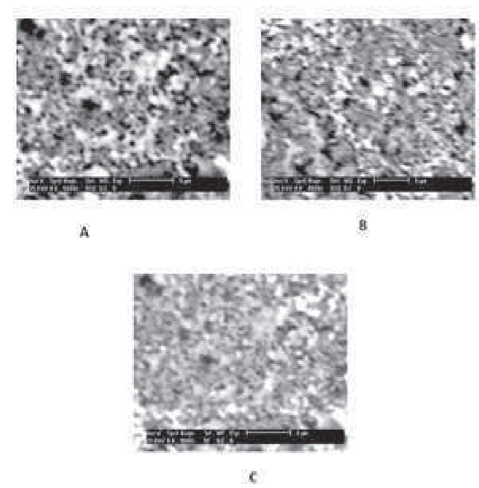

Figure 2. SEM for composites

The decrease of lattice constant and increase in grain size as the rare earth substance increases is due to the deformation of the structure. The difference in their ionic radii will lead to strains, which may result in domain wall motion resulting in deformation of the spinel structure [9]. The rare earth ions tendency to prefer the octahedral sites by replacing iron ions (Fe^{3+}) [10]. Ferrimagnetism is largely governed by Fe-Fe interaction (the spin coupling of the 3d electrons). If the rare earth ions enter the spinel lattice, the R_E-Fe interactions also appears (4f-3d coupling), which can lead to changes in the structural, electrical, magnetization and Curie temperature [11].

The frequency variation of real (ε′) and imaginary (ε″) parts of permittivity for all the samples under investigation was measured in the frequency range of 1 MHz to 1.8 GHz and obtained results are plotted in Fig.3 & Fig.4. It can be seen from the figures that the value of ε′ remains constant upto a frequency 600 MHz and increases further increase of frequency. In all the samples a resonance peak and anti resonance was observed above 1.49 GHz. This behaviour can be explained in the following way: The ε' remains constant in the frequency range from 1 MHz to 600 MHz due to the hoping electrons will not follow the external applied field. Where as the increase of ε' from 600 MHz to 1.49 GHz is may be due to the following of hopping electrons, the external field. When the hoping frequency of the electrons is equal to that of the external applied electric field, a dielectric resonance peak is obtained in the dielectric constant. The

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decrease of real part of permittivity (ε′) is attributed to the introduction of $Gd_{0.2}Ce_{0.8}O_3$which decreases the dipoles in the composite materials.

Frequency dispersion of permittivity for composites decreases with increasing rare earth dopants content due to the decrease in relaxor behavior caused by the incorporation of the ferrites [12]. The decrease in permittivity and broadness observed is due to incorporation of rare earth dopants which dilutes ferroelectric properties which can be observed from the figures. It can be observed from the figure that as the rare earth dopant increases in the composite materials, the ferroelectric transition is shifting towards the higher frequency side.

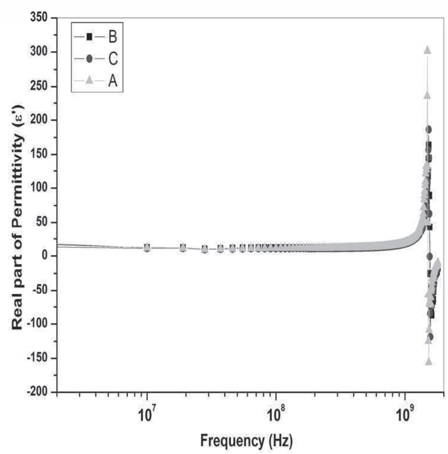

Figure 3. Variation of Real part of permittivity with frequency for composites

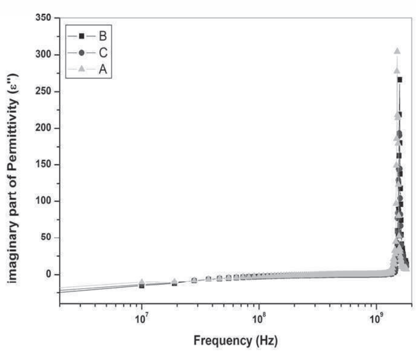

Figure 4. Variation of imaginary part of permittivity with frequency for composites

The frequency dependence of imaginary part of permittivity (ε″) has been measured on all the composites in the range of 1MHz to 1.8GHz and obtained results were plotted in Figure 4. A figure suggests that the value of ε″ is small and remains almost constant from 1 MHz to 600 MHz. The ε″ value increases with an increase of frequency from 600 MHz and finally shows a resonance peak above 1.4 GHz. The variation of ε″ with frequency may be explained similar to that of ε' variation with frequency. These relative permittivity and imaginary part of permittivity factors is very important for Multi-Layer Chip Inductor (MLCI) where ferrite thin films are separated by electrodes and the parasitic capacitance of this assembly is very important for multilayer devices. The capacitance and tan δ/ imaginary part of permittivity should be as low as possible for a good inductor material. All the composite ferrites shows very low capacitance and loss factor (tan δ) for wide frequency range.

CONCLUSION

High dense (90%), homogeneous and small grained (1-x) $Ni_{0.53}Cu_{0.12}Zn_{0.35}Fe_{1.88}O_4$ + (x) $Gd_{0.2}Ce_{0.8}O_3$(x=0.10,0.20,0.30) were prepared using conventional sintering method at 1000^oC/1200min. The average grain sizes of all the composite lies between 90nm and 120nm. The present composites show low dielectric constant and low losses at 1MHz and resonant frequency of all the sintered samples found to be greater than 1.4GHz. As the ferrite content increases in the composite materials, the ferroelectric transition temperature is shifting towards the lower frequency side.

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