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Magnetic and Electrical Properties of Gd Doped Ni-Cu-Zn-Fe_2O_4

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Abstract

$Ni_{0.5}Cu_{0.25}Zn_{0.25}Gd_xFe_{2-x}O_4$ (x = 0.0, 0.025, 0.05, 0.075, 0.1) ferrites were synthesized using an oxalic-based precursor method. The T_C for all the $Ni_{0.5}Cu_{0.25}Zn_{0.25}Gd_xFe_{2-x}O_4$ (x = 0.0, 0.025, 0.05, 0.075, 0.1) samples was measured by using one of the double coil susceptibility setup. In all the samples it is observed that, at a certain temperature, susceptibility falls to zero indicating the Curie temperature (T_C) and ferrimagnetic samples are converted into paramagnetic sample at that temperature. The electrical properties were investigated for these samples. Dielectric properties and ρ_{dc} properties were observed to decrease with the increase in the frequency for all the Gd doped ferrite samples.

Keywords

Ni-Cu-Zn-Gd Nanoferrites, Magnetic Properties, Electrical Properties

1. Introduction

Spinel Ni-Cu-Zn ferrites are one of the potential materials used in high frequency applications and in magnetic storage devices [1]. They are used as recording heads, inductors, deflection yokes, transformer cores, etc. [2] [3]. These ferrites with different chemical compositions in different forms like, thin films and nano powder have been investigated for their structural, electrical and magnetic properties in recent years. In these ferrites, if partial doping of +2, +3 ions are replaced in the place of Fe^{3+} ions, it may lead to the structural distortion thereby enhancing the magnetic properties. Rare earth doped Ni-Cu-Zn ferrites results in the improved magnetic and optical properties [4] [5] [6] [7] [8]. Higher percentage of rare earth doping in ferrites usually contributes for the formation

of rare earth secondary phases, and is observed for only few kind of rare earth elements [9]. It is well known that the magnetic properties of the ferrite materials depends on the type, ionic radius and concentration of the doping ions (magnetic/nomagnetic nature) [10], grain and morphology of the samples and methods of preparation [11] [12]. Doping these ferrites with various transition elements leads to important changes in their structural, electrical and magnetic properties.

Synthesis of Gd doped Ni-Cu-Zn ferrites is a challenging task because of co-existence of undesired phase like Fe_2O_3 along with the spinel. It is known that rare-earth ions play an important role in determining the magnetocrystalline anisotropy in 4f–3d intermetallic compounds [13]. The presence of Gd^{3+} ions influences mainly the magnetic anisotropy of the system. The magnetic properties of ferrites can be changed by the substitution of various kinds of divalent ions or by introducing a relatively small amount of rare-earth ions. Substitution of rare earth ion into the spinel structure has been reported to lead to structural distortion and to induce strains and to significantly modify the electrical and magnetic properties [14] [15]. It is found that all the rare earth ions favor in the occurrence of secondary phases resulting in the increase of bulk density and electrical resistivity [16] [17]. From our literature review we observed that till now no researcher have reported Gd doped $Ni_{0.5}Cu_{0.25}Zn_{0.25}Fe_2O_4$ ferrite. Therefore, in this present work, we made an attempt for systematic doping of Gd in Ni-Cu-Zn ferrite synthesized using oxalic acid-based precursor method [18] [19] [20] to investigate their magnetic and electrical properties.

2. Experimental Procedure

$Ni_{0.5}Cu_{0.25}Zn_{0.25}Gd_xFe_{2-x}O_4$ (x = 0.0, 0.025, 0.05, 0.075, 0.1) ferrite nanopowders were synthesized using an oxalate-based precursor method [18] [19] [20]. All the chemicals used were a.r. grade from Sigma–Aldrich and had purities ≥ 99%. In this synthesis process, nickel nitrate hydrate ($Ni(NO_3)_2{\cdot}6H_2O$), cupric nitrate hydrate ($Cu(NO_3)_2{\cdot}6H_2O$), zinc nitrate hydrate ($Zn(NO_3)_2{\cdot}6H_2O$), gadolinium oxide (Gd_2O_3) and ferric nitrate nonahydrate ($Fe(NO_3)_3{\cdot}9H_2O$) were used as the starting materials. The entire synthesis process is described elsewhere [19]. The resultant mixtures were evaporated on a hot plate at ~150˚C for 2 h. The obtained raw powders were thermally heat treated at 450˚C for 4 h. The T_C for all the $Ni_{0.5}Cu_{0.25}Zn_{0.25}Gd_xFe_{2-x}O_4$ (x = 0.0, 0.025, 0.05, 0.075, 0.1) samples was measured by using one of the double coil susceptibility setup. The dielectric properties were measured using a Nova Control, Alpha high performance frequency analyzer.

3. Results and Discussions

The Curie temperature (T_C) for all the samples was measured by using one of the double coil susceptibility models available. The graphs of the magnetization vs absolute temperature T are plotted which are shown in **Figure 1**. The plots of all

the samples show ferrimagnetic behavior. In all the cases it is found that at a certain temperature, susceptibility falls to zero indicating the Curie temperature (T_C) and ferrimagnetic sample is converted into paramagnetic sample.

In the present system $Ni_{0.5}Cu_{0.25}Zn_{0.25}Gd_xFe_{2-x}O_4$ (x = 0.0, 0.025, 0.05, 0.075, 0.1) we have obtained the Curie temperature as 426˚C for $Ni_{0.5}Cu_{0.25}Zn_{0.25}Gd_xFe_2O_4$ samples. From this observation we can say that due to non-magnetic Gd doping T_C is decreased. Curie temperatures T_C of all the samples are given in **Table 1** and the variation of T_C with x is shown in **Figure 2**. From **Figure 2**, it is observed that T_C keeps on decreasing due to increasing non-magnetic Gd^{3+} content *x*. This indicates that ferrimagnetic behavior reduces with addition of non-magnetic Gd^{3+} ions. This is accredited to decrease in magnetic (A-B) interactions and increase in (B-B) interaction due to the substitution of Fe ions Gd ions. The substitution of Gd^{3+} ions reduces the active magnetic moment therefore the T_C goes on decreasing with increase in non-magnetic Gd content *x* in this ferrite system. The results of A. C. susceptibility are in good agreement with those of magnetization.

Table 1. Curie temperature (T_C) of $Ni_{0.5}Cu_{0.25}Zn_{0.25}Gd_xFe_{2-x}O_4$ (x = 0.0, 0.025, 0.05, 0.075, 0.1) ferrites.

x	Tc (˚C)
0	426
0.025	385
0.05	319
0.075	252
0.1	213

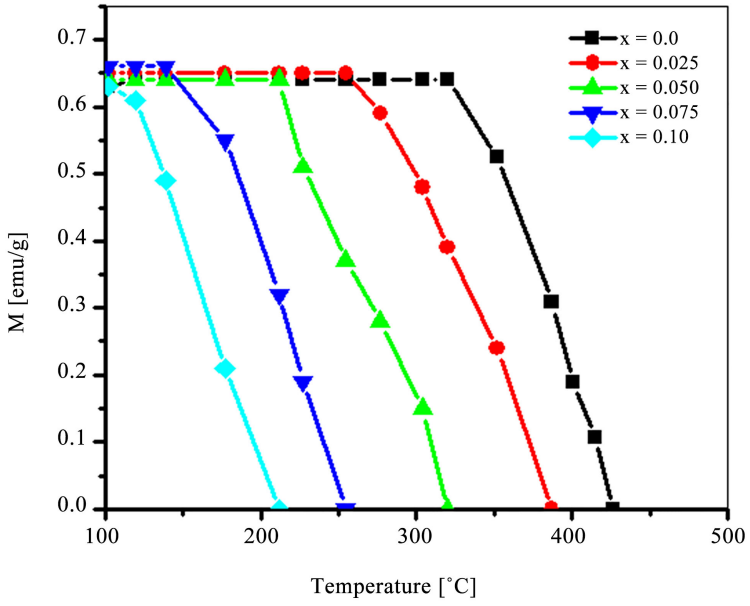

Figure 1. Curie temperature (T_C) measurements for $Ni_{0.5}Cu_{0.25}Zn_{0.25}Gd_xFe_{2-x}O_4$ (x = 0.0, 0.025, 0.05, 0.075, 0.1) ferrites samples.

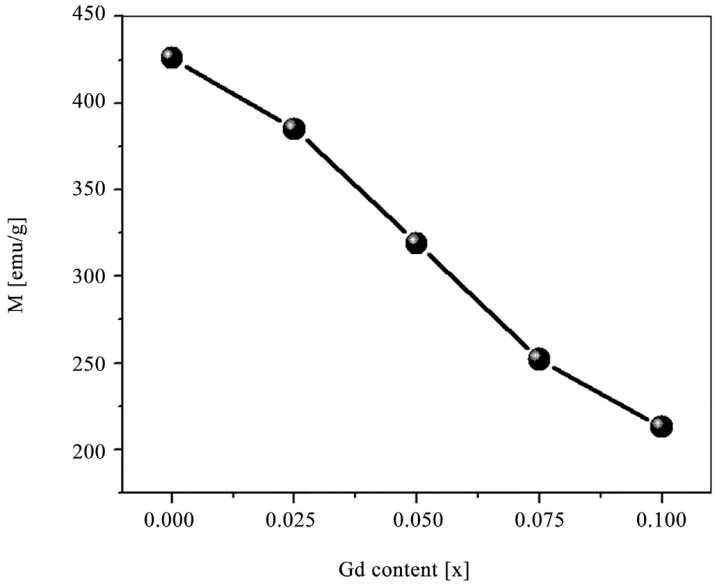

Figure 2. Curie temperature (T_C) with Gd content x for $Ni_{0.5}Cu_{0.25}Zn_{0.25}Gd_xFe_{2-x}O_4$ (x = 0.0, 0.025, 0.05, 0.075, 0.1) ferrites samples.

The dielectric behavior among ferrites is considered to be one of the most important electrical properties that predominantly based on the synthesis technique, annealing time, annealing temperature and type of dopant and its quantity. The variations in the dielectric constant (ε') with frequency for $Ni_{0.5}Cu_{0.25}Zn_{0.25}Gd_xFe_{2-x}O_4$ (x = 0.0, 0.025, 0.05, 0.075, 0.1) ferrites samples are shown in **Figure 3**. All the samples are observed to exhibit dielectric dispersion. The dielectric constant was observed to decrease initially with increasing frequency and then to become almost constant at higher frequencies. Different grain sizes show different dielectric constants but the behaviors are same. At a certain frequency, these samples show a frequency-independent characteristic behavior that can be explained using the well-known Maxwell-Wagner-type interfacial polarization, that is in accord with Koop's theory [21] [22] [23]. The dielectric polarization among ferrites is same as that of the conduction mechanism taking place with hopping of electronics. Hopping of electrons among the Fe^{3+} - Fe^{2+} in the applied field direction takes place to determine the polarization. With the increase in frequency the polarization decreases and attains a constant value. This is based on the fact, that after a particular applied frequency, the electron will exchange between $Fe^{2+} \leftrightarrow Fe^{3+}$ which may not follow the alternating electric field. Higher dielectric constant values at lower frequencies will be due to greater number of Fe^{2+} ions, interfacial dislocations, grain boundary defects, oxygen vacancies [21] [22] [23] [24]. The decrease in the dielectric constant (ε') with increasing frequency is a natural phenomenon due to the fact that any element contributing to polarization will show a lagging effect with the applied field at larger frequencies.

The observed dielectric constant values for our NiCuZnGd ferrite samples are little more than those reported for NiCuZn ferrite synthesized by using different

processes and different compositions. The conduction mechanism among the ferrites is majorly due to the hopping of electrons of the same element with different oxidation states. The lower dielectric constant values are observed among ferrites that are annealed at lower temperatures due to the low chance of ions existing with different valance states which reduces the probability of hopping electrons [24] [25] [26] [27] [28]. Also, the grain/particle size, density, stoichiometry and homogeneity of the ferrites are observed to affect the dielectric constant values [26]. Therefore, as our samples are annealed at higher temperatures, the dielectric constants are observed to be more.

The tanδ with the applied frequency is measured at room temperature. The dielectric loss tangent data plotted in **Figure 4** clearly shows that in all the samples the dielectric loss tangent slowly increased with the increase of frequency till a particular frequency, after which it slowly decreases. Different grain sizes show different dielectric loss tangent curves and different values. With the application of an external alternating field having the same frequency, the maximum electrical energy will be transfer to the oscillating ions; due to this, the power loss in the ferrites rises [24] [27]. The peaks in the dielectric loss tangent appear with the applied field time is in phase with the dielectric and when the condition, $\omega\tau = 1$ is satisfied, where $\omega = 2\pi f$, f is the applied field frequency, τ is the relaxation time, which is connected to the jumping probability unit time p using the equation $\tau = p/2$; *i.e.*, the peak frequency (f_{max}) is relative to the jumping probability. With the increase in f_{max} and in the annealing temperature, the jumping or hopping probability increases.

Figure 5 shows, the temperature dependent dc resistivity, represented as (log ρ_{dc}) vs (1000/T), for all the synthesized samples. It is clearly seen that ρ_{dc} in all

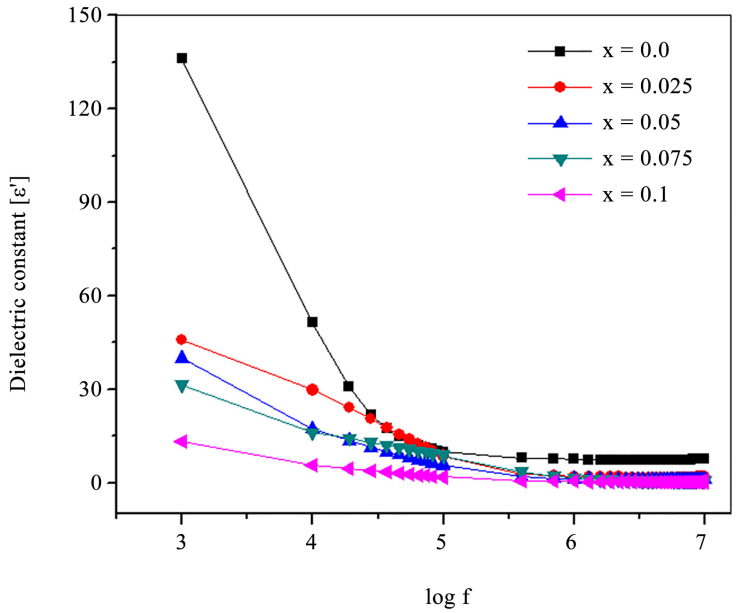

Figure 3. Variations of the dielectric constant for $Ni_{0.5}Cu_{0.25}Zn_{0.25}Gd_xFe_{2-x}O_4$ (x = 0.0, 0.025, 0.05, 0.075, 0.1) ferrites.

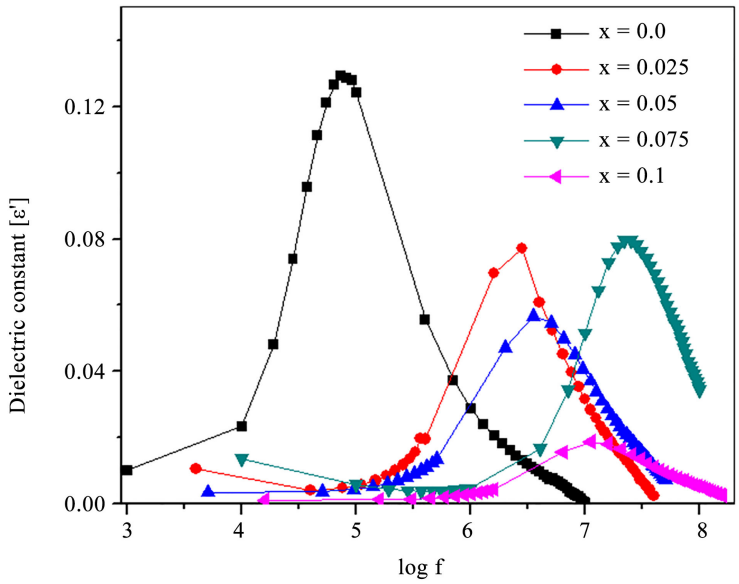

Figure 4. Variations of the dielectric loss tangent for $Ni_{0.5}Cu_{0.25}Zn_{0.25}Gd_xFe_{2-x}O_4$ (x = 0.0, 0.025, 0.05, 0.075, 0.1) ferrites.

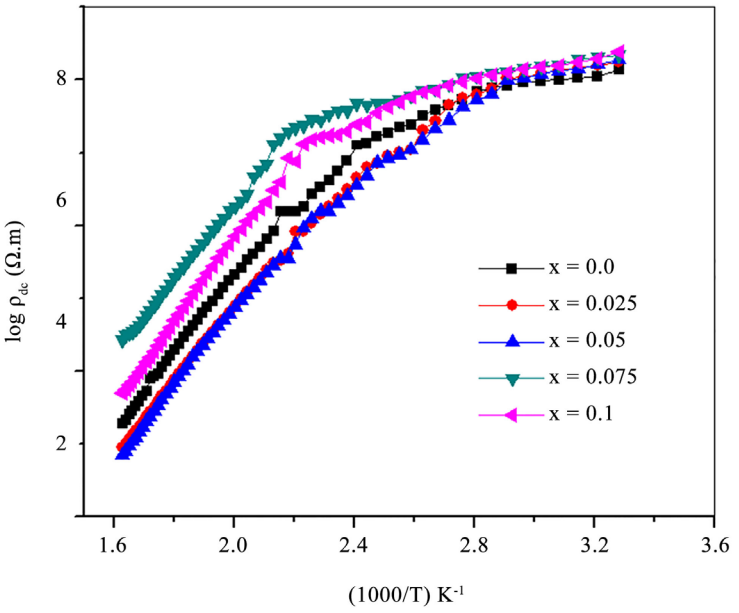

Figure 5. The variation of log (ρ) with 1000/T for $Ni_{0.5}Cu_{0.25}Zn_{0.25}Gd_xFe_{2-x}O_4$ (x = 0.0, 0.025, 0.05, 0.075, 0.1) ferrites.

the samples decreased with the increase in temperature. This kind of results are somewhat common feature among ferrites signifying the normal semiconducting nature explained by Arrheneus relation (1) [29].

$$\rho = \rho_0 \exp\left(\frac{E}{K_B T}\right) \tag{1}$$

where ρ_0 is the resistivity at infinitely high temperature, K is the Boltzmann constant, T is the absolute temperature and ΔE is activation energy.

Further, from **Figure 5** that, each of these curves can be differentiated into the two regions having different activation energies. Contrary, the transition temperatures, Tρ, in between the first region and the second region was observed to be near to the values measured from magnetic measurements [30]. The transition taking place at Tρ is the activation energy at which the magnetic transition is taking place from ferrimagnetic state to the paramagnetic (region one to region second) state. The activation energies in ferrimagnetic and paramagnetic regions were designated as EF and EP. The activation energies values were determined from the least square method as the function of Gd^{3+} ions [31]. The impact of these magnetic transitions of ferrites on the electrical applications was reported in literature by several researchers [32] [33]. It is evident that the EP values are higher than the EF in all the samples accordance with [34]. As a matter of fact, all most all the ferrites exhibit a change at the activation energy around T_C so that EP > EF [32]. The increase in the activation energy near the paramagnetic region compared to the ferrimagnetic region can be accredited to the development of spin polarons [35] [36]. It is observed that the condition for the formation of spin polaron is, the activation energy in EP > 0.2 eV, as in similar to our samples [35] [37]. Therefore our entire sample favors spin polarons formation [38] [39] [40].

4. Conclusion

The $Ni_{0.5}Cu_{0.25}Zn_{0.25}Gd_xFe_{2-x}O_4$ (x = 0.0, 0.025, 0.05, 0.075, 0.1) ferrite samples were successfully prepared using the oxalic method. XRD results showed single phase spinel ferrite structure. The Curie temperature (T_C) was observed to decrease with increasing Gd concentration. The dielectric properties and ρ_{dc} were observed to decrease with the increase in the frequency for all the Gd doped ferrite samples.

Conflicts of Interest

The authors declare no conflicts of interest regarding the publication of this paper.

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