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Electrical Transport Properties of Bi_2O_3-Doped $CoFe_2O_4$ and $CoHo_{0.02}Fe_{1.98}O_4$ Ferrites

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ABSTRACT

Two series of $CoHo_xFe_{2-x}O_4$ (x = 0.0, 0.02) ferrites with Bismuth oxide doping from (0.1 - 0.3)*% were prepared by Co-precipitation technique. X-ray diffraction analysis revealed fcc structure. The lattice constants were found to decrease as the doping of Bi_2O_3 increases in both series. An increase in Bismuth oxide concentration from* (0.1 - 0.3*%*) *in $CoFe_2O_4$, and $CoHo_{0.02}Fe_{1.98}O_4$ ferrites leads to an increase in room temperature resistivity. Temperature dependent resistivity decreases as the temperature increases following the Arrhenius equation. The activation energy increases with the increase of Bi_2O_3-concentration for both $CoFe_2O_4$ and $CoHo_{0.02}Fe_{1.98}O_4$ series. The frequency dependant dielectric constant follows the Maxwell-Wagner type interfacial polarization. The dielectric loss indicates the normal behavior of these ferrites. SEM analysis shows an increase in grain size with increasing Bismuth concentration.*

Keywords: *Cobalt Ferrites*, *Co-Precipitation*, *Bi Doping*, *Electrical Properties*

1. Introduction

The miniaturization of electrical and electronic gadgets demands new materials with nano-size particles for high frequency applications and high density recording. Bi_2O_3 is a potential dopant for improving the magnetic and electrical properties of ferrites. The properties of ferrite materials are known to be strongly influenced by their composition and microstructure which are sensitive to the processing methods used to synthesize them. Over the last decade, the magnetostrictive materials for smart sensors have attracted a great interest due to their wide range of applications in the automotive industry. The cobalt ferrites are well known for its highest magnetostrictive coefficient amongst the oxide-based magnetostrictive materials. Cobalt ferrite nanoparticles have recently become the subject of research interest from the point of view of the synthesis, the magnetic characterization, and the applications [1-4]. Among the various ferrite materials for magnetic recording applications, cobalt ferrite ($CoFe_2O_4$) has been widely studied [5]. Cobalt ferrite ($CoFe_2O_4$) possesses excellent chemical stability, good mechanical hardness and a large positive first-order crystalline anisotropy constant, which made this ferrite a promising candidate for magneto-optical recording media [6]. High electrical resistivity and low eddy current losses make these ferrites an excellent core material for power transformers in electrical and electronic industry, recording heads, antenna rods, loading coils, microwave devices and telecommunication applications [7,8].The magnetic properties of ferrites can be modified by introducing suitable divalent and trivalent oxides as dopants [9]. It is reported [10] the dc electrical conductivity of the Bi_2O_3-doped ferrites was increased. Bi_2O_3 is an alternative sintering aid for lowering sintering temperature of magnesium ferrites. The addition of Bi_2O_3 has significant effect on the resistivity and dielectric properties of ferrites. Moreover the advantageous effect of Bi_2O_3 is attributed to the formation of liquid phase layer due to the low melting point of Bi_2O_3 [11] which enhances the resistivity of the ferrites. Due to the presence of Bi_2O_3 the properties of grain boundaries of ferrite were changed and three dimensional network grain boundary structure formed at grain boundaries [12-15]. Since Bi_2O_3 doping enhances the magnetic and electrical properties of the ferrites, the purpose of this study is to investigate the influence of the Bi_2O_3 doping from 0.1-0.3% by weight on the microstructure, electrical and magnetic properties of $CoFe_2O_4$ and $CoHo_{0.02}Fe_{1.98}O_4$ ferrites. The $CoHo_{0.02}Fe_{1.98}O_4$ was chosen for comparison between $CoFe_2O_4$ and the Rare earth substituted $CoHo_{0.02}Fe_{1.98}O_4$ ferrites after doping Bi_2O_3.

2. Experimental

Bi_2O_3 doped $CoFe_2O_4$, (Bi_2O_3:0.1 - 0.3%) and $CoHo_{0.02}$-$Fe_{1.98}O_4$ (Bi_2O_3:0.1 - 0.3%) ferrites were prepared by using co-precipitation method. The starting materials 99.9% pure, $Co(C_2H_3O_2)_2$,$FeCl_3$ and Ho_2O_3 were used. The stoichometric amounts of selected salts were dissolved in de-ionized water in a 100ml beaker except Ho_2O_3 which is insoluble in de-ionized water. Ho_2O_3 was dissolved in HCl heated at 50 - 60˚C and then added in the solution. The solution so obtained was stirred using magnetic stirrer for 10 hrs. During stirring calculated amount of Na_2CO_3/NaOH were used in the solution as reagent or as precipitating agents to precipitate the metals and hydroxides. The precipitates were thoroughly washed with distilled water until free from chloride ions, which were checked by $AgNO_3$ test. The final product was then filtered with the help of suction flask having an outlet with pump operated on water. The filtered precipitates were dried in oven for 24h at 100ºC. The dried precipitates were then ground in mortar and pestle and Bi_2O_3 was then doped from (0.1 - 0.3) wt% in both set of samples during grinding. The Pellets of ground powders were formed using Paul-Otto Weber hydraulic press under the pressure of (~35 KN·mm^2). The pellets were then sintered in an electric furnace at 1150 - 1200˚C for 10h followed by furnace cooling. For electrical measurements both surfaces of the pellets were polished on the micron paper. The phase formation of the samples was investigated by Shimadzu X-ray diffractometer using CuK_α radiations (λ = 1.5406Å). SEM analysis of the one series of samples was investigated by JEOL-Japan Model JSM 5910 and it was used to observe the microstructure of the sintered specimens. Electrical resistivity was measured by two probe method using source meter model 2400 (Keithley).The dielectric properties were measured using Digi Bridge (GenRad 1689).

3. Results and Discussions

3.1. X-Ray Diffraction

X-ray diffraction patterns of Bi_2O_3 doped $CoFe_2O_4$, (Bi_2O_3:0.1 - 0.3%) and $CoHo_{0.02}Fe_{1.98}O_4$ (Bi_2O_3:0.1 - 0.3%) ferrites are shown in **Figures 1(a**,**b)** respectively. The X-ray diffraction analysis shows the formation of single phase fcc spinel crystal structure for all the samples. The d-values were compared using JCPDS (1998) cards (card No. 01 - 1121) [16]. Apparently no traces of secondary phase were observed [17-19]. The 220, 311, 400, 333, 440 reflections were observed in X-ray diffraction patterns.

3.2. Lattice Constant

The lattice constants of $CoFe_2O_4$ (Bi_2O_3: 0.1 - 0.3%) and $CoHo_{0.02}Fe_{1.98}O_4$ (Bi_2O_3: 0.1 - 0.3%) samples are shown in

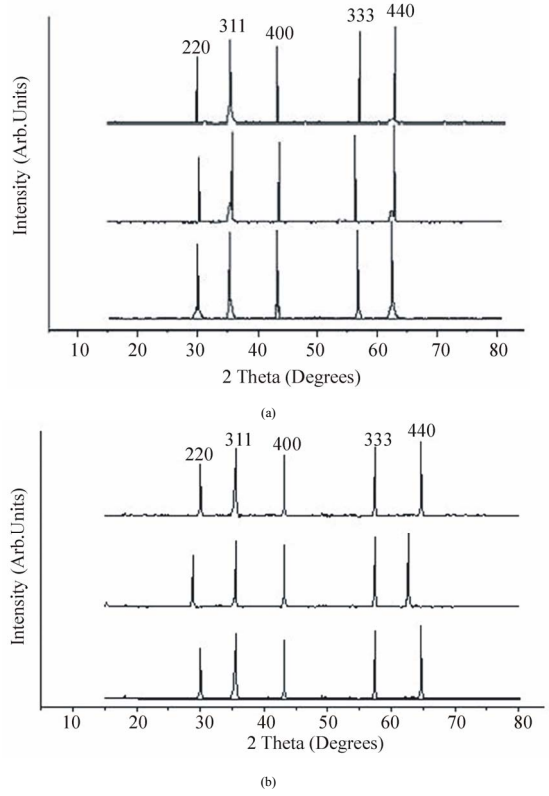


Figure 1. (a) XRD pattern for $CoFe_2O_4$ (Bi_2O_3: 0.1 - 0.3%). (b) XRD pattern for $CoHo_{0.02}Fe_{1.98}O_4$, (Bi_2O_3: 0.1 - 0.3%)

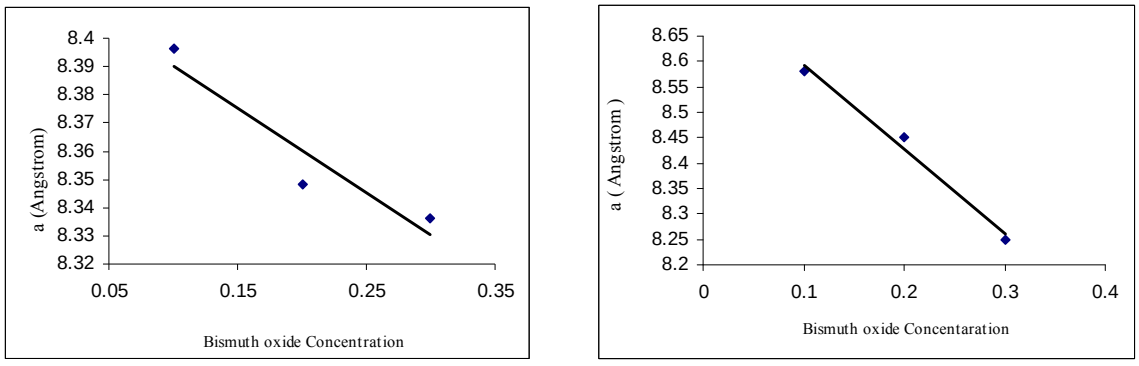
Figures 2(a,**b)**. It is observed that the lattice constants decrease as the doping of Bi_2O_3 increases. The decrease in 'a' is due to the difference in ionic radii of Bi^{3+} (0.74Å) as compared to Fe^{3+} (0.78Å) [7]. The decrease in lattice constant in both series can be explained on the basis of the fact that the Bi_2O_3 enter into the lattice completely during sintering due to very small amount of Bi_2O_3 doping.

3.3. Room Temperature Resistivity

Figures 3(a,**b)** shows the room temperature resistivity versus Bi_2O_3 concentration for both $CoFe_2O_4$ and $CoHo_{0.02}Fe_{1.98}O_4$ ferrite series. It can be observed that as Bi_2O_3-concentration increases, the resistivity increases from 18×10^4 to a maximum value of 42×10^4 Ω-cm for $CoFe_2O_4$, (Bi_2O_3: 0.1 - 0.3%) samples and from 7×10^3 to 32×10^4 Ω-cm for $CoHo_{0.02}Fe_{1.98}O_4$ (Bi_2O_3: 0.1 - 0.3%) ferrites. The increase in Bi_2O_3 concentration leads to an increase in resistivity that might be due to the fact that Bi^{5+} act as scattering centres for the carriers hopping between two octahedral sites [7,10], which hinders the hopping mechanism between the Fe^{2+} and Fe^{3+} ions.

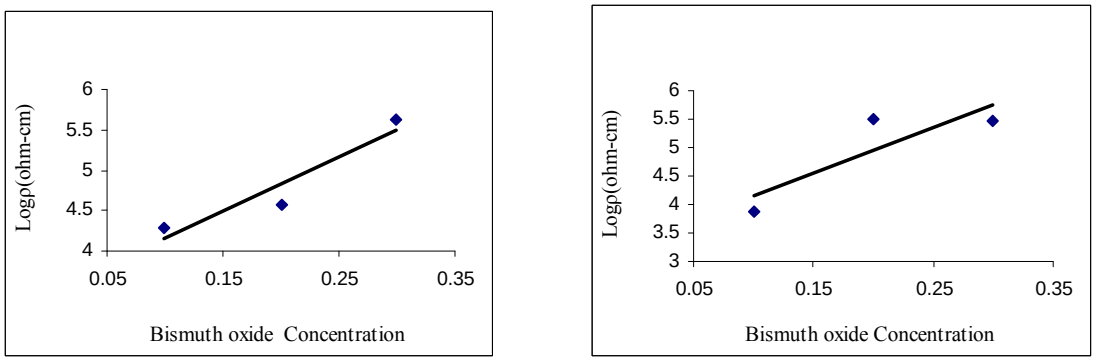
3.4. Temperature Dependant Resistivity

Temperature dependent electrical resistivity of $CoFe_2O_4$



(a) (b)

Figure 2. (a) Lattice constant for $CoFe_2O_4$, (Bi_2O_3: 0.1 - 0.3%); (b) Lattice constant for $CoHo_{0.02}Fe_{1.98}O_4$, (Bi_2O_3: 0.1 - 0.3%).



(a) (b)

Figure 3. (a) Room temperature resistivity (ρ) for $CoFe_2O_4$, (Bi_2O_3: 0.1% - 0.3%); (b) Room temperature resistivity (ρ) for $CoHo_{0.02}Fe_{1.98}O_4$, (Bi_2O_3: 0.1% - 0.3%).

(Bi_2O_3:0.1 - 0.3%) and $CoHo_{0.02}Fe_{1.98}O_4$ (Bi_2O_3:0.1-0.3%) ferrite series was measured in temperature range (40 - 200˚C). The Arrhenius plots of these samples are shown in **Figures 4(a)** and **(b)** respectively. Each sample follows Arrhenius equation,

$$\rho = \rho \exp \Delta E / K_B T \quad (1)$$

where ΔE is the activation energy, T is the absolute temperature, k_B is the Boltzmann's constant. The plots show that the resistivity decreases as the temperature increases indicating the semi conducting nature of the samples [10]. This decrease in resistivity may be due to the excess of electrons released from both sites which reduces the Co^{2+} to their lowest valency and also produce Fe^{2+} ions [20]. The behavior of both type of electric charge carriers can be explained on the basis of Rezlescue model [21]. According to this model, the exchanging of electrons between Fe^{2+} and Fe^{3+} ions and that of holes between Co^{3+} to Co^{2+} ions may be the likely conduction mechanism.

$$Co^{3+} \leftrightarrow Co^{2+} \text{ (Hole conduction)} \quad (2)$$

$$Fe^{2+} \leftrightarrow Fe^{3+} \text{ (Electronic conduction)} \quad (3)$$

3.5. Activation Energy

The activation energy obtained from Arrhenius plots are shown in **Figures 5(a**,**b)** respectively. The activation energy increases with the increase of Bi_2O_3-concentration for both $CoFe_2O_4$ and $CoHo_{0.02}Fe_{1.98}O_4$ samples. It can be observed that the samples having high resistivity value also have high activation energy and vice versa [22] .The result indicates the presence of conduction dependant to the structure [23]. As the activation energy is high and so the resistivity is high due to which conductivity will be lower as Bi_2O_3 is substituted which can be thought of due to phonon-assisted small polaron hopping [21-22].

3.6. Drift Mobility

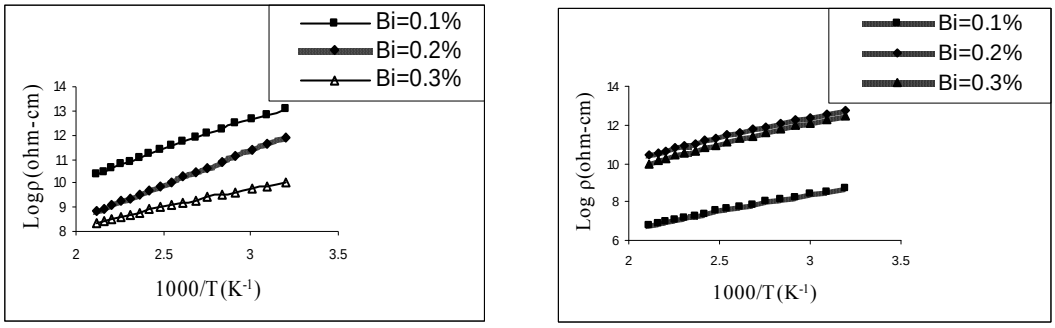
Figures 6(a) and **(b)** respectively shows the variation of drift mobility with temperature for both series. The drift mobility was calculated by using the resistivity data and is calculated with the help of the given formula [23-25].

$$\mu_d = 1/ne\rho \quad (4)$$

where ρ is electrical resistivity, e is charge on an electron and 'n' is the concentration of charge carriers and it is calculated by following relation;

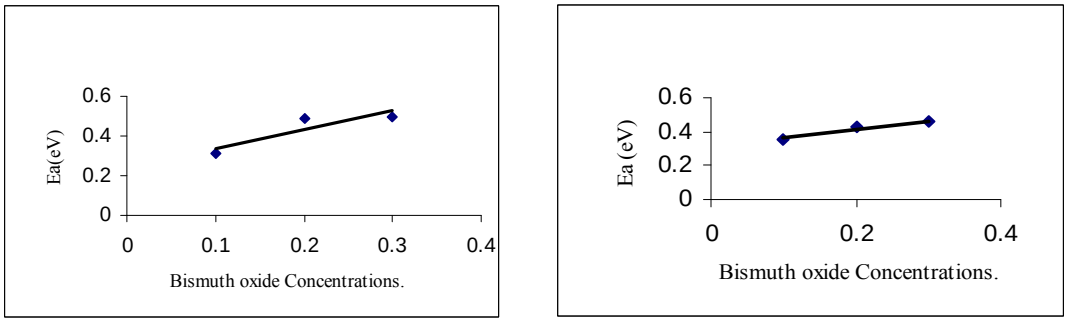
$$n = N_A C_{Fe} \rho_b / M \quad (5)$$

where N_A is Avogadro's number, C_{Fe} is the number of iron atoms in sample, ρ_b is the bulk density and M is the



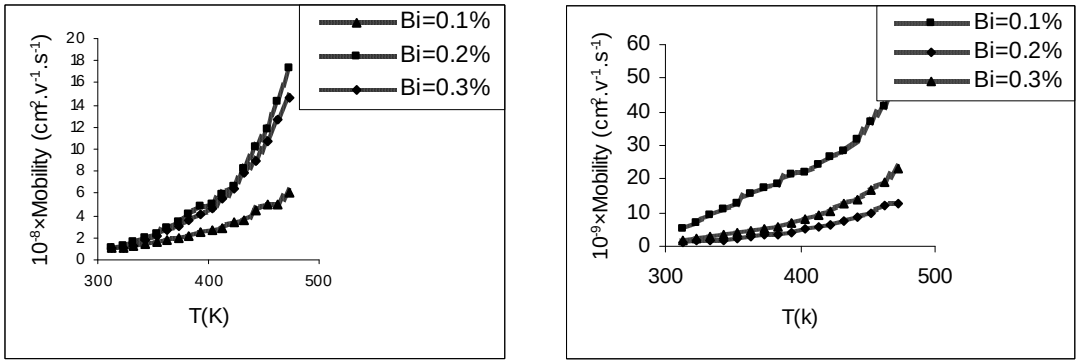
(a) (b)

Figure 4. (a) Temperature dependant resistivity for $CoFe_2O_4$, (Bi_2O_3: 0.1% - 0.3%); (b) Temperature dependant resistivity for $CoHo_{0.02}Fe_{1.98}O_4$, (Bi_2O_3: 0.1% - 0.3%)



(a) (b)

Figure 5. (a) Activation Energy for $CoFe_2O_4$, (Bi_2O_3: 0.1% - 0.3%); (b) Activation Energy $CoHo_{0.02}Fe_{1.98}O_4$, (Bi_2O_3: 0.1% - 0.3%).



(a) (b)

Figure 6. (a) Drift Mobility for $CoFe_2O_4$, (Bi_2O_3: 0.1% - 0.3%); (b) Drift Mobility for $CoHo_{0.02}Fe_{1.98}O_4$, (Bi_2O_3: 0.1% - 0.3%).

molar mass of the samples. It is however observed that drift mobility increases with the increase in temperature. This may be due to the fact that charge carriers start hopping from one site to another as the temperature increases [26]. The temperature dependence of resistivity and mobility shows that the samples are of degenerate type semiconductors.

3.7. Dielectric Properties

3.7.1. Dielectric Constant and Loss Tangent

The dielectric constant Vs frequency of both the series at room temperature 30˚C are shown in **Figures 7(a**,**b)** respectively. The dielectric constant decreases with increasing frequency. At high frequencies the dielectric constant seems to be independent of frequency. This behavior of the samples is in accordance with the Maxwell Wagner model [27-29]. In this model the dielectric structure of ferrite material is assumed to be made up of two layers. First layer being conducting, contains large number of grains and other being grain boundaries which are poor conductor. This bi-layer formation is resulted by high temperature sintering [23]. **Figures 8(a**,**b)** shows

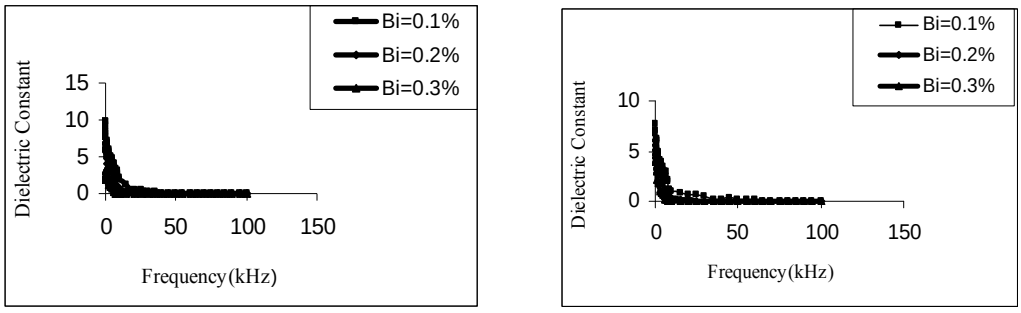


Figure 7. (a) Dielectric Constant vs Frequency for $CoFe_2O_4$, (Bi_2O_3: 0.1 - 0.3%); (b) Dielectric Constant vs Frequency for $CoHo_{0.02}Fe_{1.98}O_4$, (Bi_2O_3: 0.1 - 0.3%).

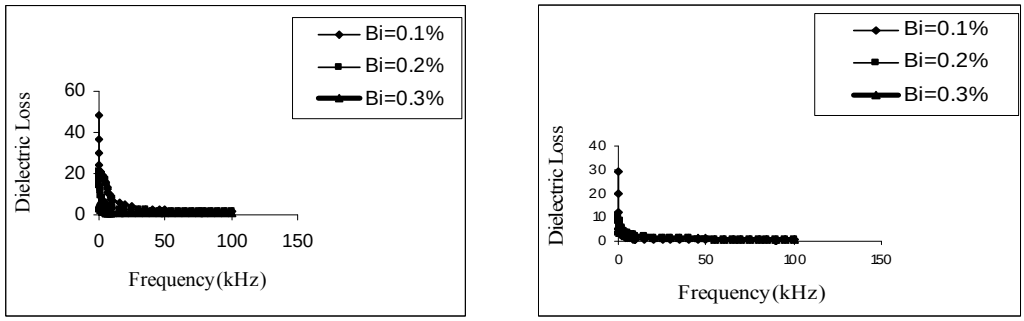


Figure 8. (a) Dielectric vs Frequency loss for $CoFe_2O_4$, (Bi_2O_3: 0.1% - 0.3%); (b) Dielectric Loss vs Frequency for $CoHo_{0.02}Fe_{1.98}O_4$, (Bi_2O_3: 0.1% - 0.3%).

tanδ Vs frequency for both series. The dielectric loss decreases substantially with increasing frequency and reaches a constant value later on [30-32]. When the frequency of applied field is low than the hopping frequency of electrons between ferrous and ferric ions at adjacent octahedral sites, the electron follow the applied field and hence loss is maximum. At higher frequencies the hopping frequency of the electron exchange between ferrous and ferric ions can not follow applied field beyond certain critical frequency and the loss is minimum.

3.8. Scanning Electron Microscopy

Few representative SEM micrographs of $CoFe_2O_4$ (Bi_2O_3: 0.1% - 0.3%) are shown in **Figures 9 (a**,**b**,**c)** respectively. The grain boundaries and grains can be clearly distinguished at 50,000 magnification. The SEM micrographs of the $CoFe_2O_4$ (Bi_2O_3: 0.1% - 0.3%) sintered at 1150˚C for 10 h are shown for various contents of Bi_2O_3. It is however observed that the grain size increases from 141 - 201 nm with the increase in Bi_2O_3 concentration in $CoFe_2O_4$ (Bi_2O_3: 0.1% - 0.3%) ferrites. Uneven grain boun-daries were also observed. These non-uniform grain boundaries seems to be a diffusion induced grain boundary migration [33,34].

4. Conclusions

1) X-ray diffraction analysis reveals that $CoFe_2O_4$ (Bi_2O_3:0.1% - 0.3%) and $CoHo_{0.02}Fe_{1.98}O_4$ (Bi_2O_3: 0.1% - 0.3%) ferrite series clearly indicate formation of spinel fcc single phase crystal structure. The lattice constant 'a' decreases as Bi_2O_3 concentration increases for both series due to the difference in the ionic radii.

2) Room temperature dc resistivity of both series of ferrites increases due to the formation of Bi^{5+} ions.

3) The temperature dependant resistivity and activation energy Vs Bi content follows same trend indicating that the samples with high resistivity have high activation energies and vice versa .

4) The behavior of dielectric constant and loss tangent for both series of ferrite follows the Maxwell Wagner model.

5) The SEM micrographs of the $CoFe_2O_4$ (Bi_2O_3: 0.1% - 0.3%) ferrites shows that the grain size increases from 141 - 201 nm with the increase in Bi_2O_3 concentration.

6) The activation energy shows that the hopping conduction mechanism is established in these samples.

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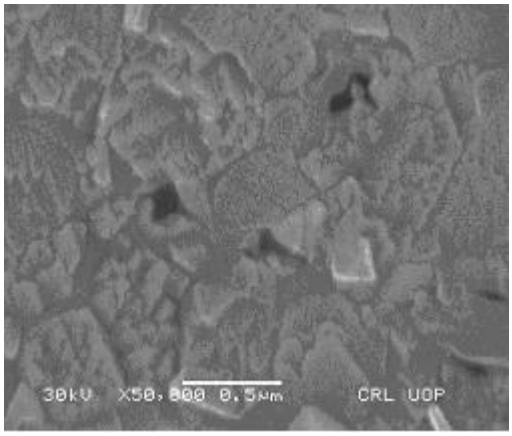
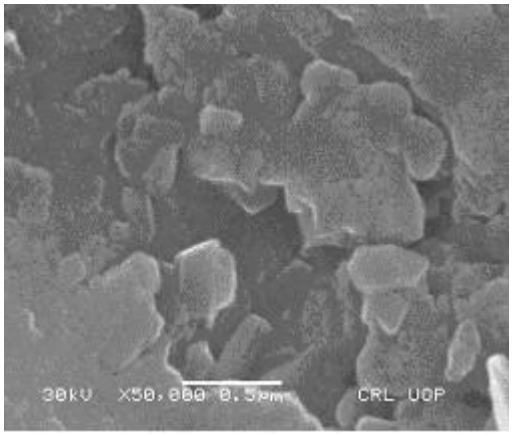
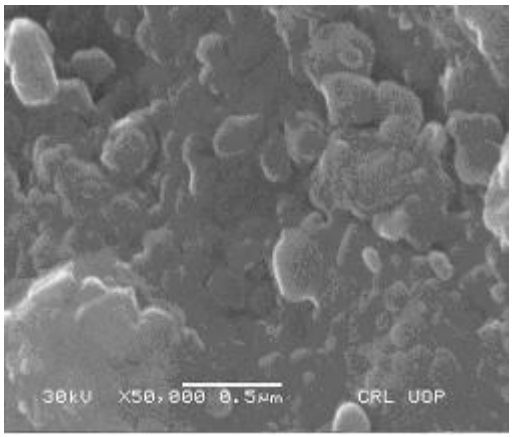


(a)



(b)



(c)

Figure 9. (a) SEM for $CoFe_2O_4$ (Bi_2O_3: 0.1%); (b) SEM for $CoFe_2O_4$ (Bi_2O_3: 0.2%); (c) SEM for $CoFe_2O_4$, (Bi_2O_3: 0.3%).

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