Structural, magnetic and electrical properties of nano NiCr$_x$Fe$_{2-x}$O$_4$ synthesized by flash auto combustion method

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Abstract. The spinel Nano-ferrite system NiCr$_x$Fe$_{2-x}$O$_4$ (where x=0.0, 0.2, 0.4, 0.6, 0.8 and 1.0) have been prepared using flash auto combustion method. The XRD pattern confirms the formation of spinel Ni-Cr ferrite for the as-prepared samples. Electrical properties of as-prepared samples such dielectric constant and AC resistivity were studied in the temperature range (288-773 K). It was observed that the AC resistivity increases by increasing Cr content and decreases by increasing frequency. The DC electrical resistivity was found to increase by increasing Cr content from 25*10$^6$ to 136*10$^6$ Ω.cm, whereas it was found to decrease with increasing temperature. VSM was used to study the magnetic properties of Ni-Cr ferrite at room temperature. It was found that, with increasing Cr content the (Ms) saturation magnetization decreased from 28 to 3 emu/g whereas coercivity increased from 188 to 666 (G). The magnetic moment of the system and Curie temperature decreased by increasing Cr substitution.

1. Introduction

In recent years, nanosize spinel ferrite particles with uniform particle size and narrow size distribution have been attracted much attention because of their broad application in technological fields like medical imaging, magnetic storage devices and electronic devices etc. [1-7]. Among the different spinel ferrites, Nickel ferrite has been widely studied due to their wide range of potential applications as magnetic fluids, data storage, photo catalyst, gas sensors, inductors, magnetic refrigeration and [MRI] magnetic resonance imaging [8-13]. Ni-Cr ferrites are low cost materials and have great interest for high frequency inductive component because of their high resistivity, and low eddy current loss [14-15].

During last years, researches have been searching for soft magnetic materials, which have high saturation magnetization, high permeability and low energy losses for microwave applications [16-20]. It is well known that, the electrical and magnetic properties of most ferrites depend on sintering temperature, sintering time, rate of heating, rate of soaking [21] and on cation distribution at the tetrahedral (A) and octahedral (B) sites in the lattice [16]. The chemical, structural and magnetic properties of nanosize spinel ferrite particles are influenced by their microstructures and composition according to preparation methods such as ceramic [22], hydrothermal [23], co-precipitation [24-26], ball milling [27] sol-gel [28-29], micro emulsion [30-31] and combustion method [32-35].

There is transition in ferrites take place from ferromagnetic to para magnetic state as temperature is increase [36]. It is found that, the activation energy was smaller in ferromagnetic region than para magnetic region, which show the influence of magnetic state on the electrical properties of ferrite. The present work reports the synthesis of nanosize NiCr$_x$Fe$_{2-x}$O$_4$ ferrites by flash combustion method. The results of DC and AC resistivity, activation energy, dielectric constant and magnetic properties of these ferrites were reports here.
2. Experimental producers.
Nano-ferrite samples with the chemical formula NiCr\textsubscript{x}Fe\textsubscript{2-x}O\textsubscript{4} (x=0.0, 0.2, 0.4, 0.6, 0.8 and 1.0) were prepared by using the flash auto combustion method. The analytical grade of Fe(NO\textsubscript{3})\textsubscript{3}.9H\textsubscript{2}O, Cr(NO\textsubscript{3})\textsubscript{3}.9H\textsubscript{2}O, Ni(NO\textsubscript{3})\textsubscript{2}.6H\textsubscript{2}O and Urea [Co(NH\textsubscript{2})\textsubscript{2}] as the fuel were used. The amount of urea was calculated using charge neutrality equations [37]. The chemicals were weighed in the required stoichiometric proportions, good mixed together for a few minutes and then heated on a hot plate magnetic stirrer at 80 °C for half hr. Thermal dehydration resulted in a highly viscous liquid. With more heating, the viscous liquid swelled and auto-ignited to harvest powder. The reaction was very fast and a fine powder was obtained [38]. The structural characterizations of all samples were carried out by X-ray diffractometer (Shimadzu XRD6000, Japan) (XRD) and confirms the formation of spinel structure. XRD data were taken at room temperature using Cu-K\textalpha radiation source (\(\lambda = 1.5405 \ \text{Å}\)).

The AC resistivity and dielectric constant were measured as a function of temperature using RLC Bridge type MC2811C RLC Bridge. The DC resistivity was measured using electrometer type 610C Kethely solid-state electrometer. The magnetic properties were measured at room temperature using vibrating sample magnetometer (VSM) operating system v 1.6 control software Oxford OX8JTL England.

3. Results and Discussion.

3.1. X-ray diffraction analysis
1. Figure 1-a shows the X-ray diffraction patterns for the as-prepared samples of NiCr\textsubscript{x}Fe\textsubscript{2-x}O\textsubscript{4} doped with Cr content with different ratios where x= (0.0, 0.2, 0.4, 0.6, 0.8 and 1.0). As shown in figure, the spinel phase is predominated phase, which confirm the formation of Ni-Cr Ferrite without annealing. This give the evidence that the flash combustion method is successful rout for the preparation of spinel ferrite. Figure 1-b shows the most intense three peaks (311), (400) and (220) in another scale to confirm that the particles are in the Nano range. From the figure, the crystallite size of the prepared samples was calculated using Scherer’s formula [39]:

\[
\frac{1}{t^2} = \frac{0.9 \lambda}{h \cos \theta}
\]

where \(\lambda\) is the wave length of the X-ray for Cu-K\textalpha radiation source (\(\lambda = 1.5405 \ \text{Å}\)), \(\theta\) is the diffraction angle, \(h\) is the full width at half maximum of the peak in radian. The crystallite size ranged from 21 to 32 nm. The intensity of the main diffraction peak (311) is increased by increasing Cr content which mean that the Cr ions were good soluble in the spinel lattice of Ni-ferrite and acts as catalyst for the enhancement of solidstate reaction. The lattice parameter of the spinel ferrite NiCr\textsubscript{x}Fe\textsubscript{2-x}O\textsubscript{4} were calculated and are given in table (1), which was found to decrease by increasing Cr content. As we know that, the Cr ion radius is smaller than the ferric ion radius, which was the reason for the decrease of lattice parameter.

The bulk porosity of the sample was calculated using the equation:

\[
P = 1 - \frac{D_b}{D_s} \% \tag{2}
\]

The X-ray density, bulk density and the porosity were calculated and are given in table 1.
Figure 1. (a) XRD for NiCr_{x}Fe_{2-x}O_{4}, where (x=0.0, 0.2, 0.4, 0.6, 0.8 and 1.0),
(b) XRD of the main three peaks (311), (400) and (220).

Table 1. Values of lattice parameter, particle size, x-ray density, bulk density and porosity for NiCr_{x}Fe_{2-x}O_{4}, (where x=0.0, 0.2, 0.4, 0.6, 0.8 and 1.0).

| Cr content (x) | Lattice parameter a (Å) | Crystallite size t (nm) | X-ray density D_{x} (g/cm\(^3\)) | Bulk density D (g/cm\(^3\)) | Porosity % |
|----------------|-------------------------|-------------------------|-------------------------------|-----------------------------|------------|
| X=0.0          | 8.3459                  | 21.622                  | 5.347                         | 2.327                       | 56.47      |
| X=0.2          | 8.3860                  | 28.563                  | 5.2637                        | 2.310                       | 56.09      |
| X=0.4          | 8.3200                  | 29.365                  | 5.3725                        | 2.425                       | 54.86      |
| X=0.6          | 8.2699                  | 32.296                  | 5.4526                        | 2.477                       | 54.55      |
| X=0.8          | 8.2872                  | 28.971                  | 5.4005                        | 2.434                       | 54.92      |
| X=1            | 8.2870                  | 28.936                  | 5.3829                        | 2.333                       | 56.64      |

3.2. DC resistivity.
The temperature dependence of DC resistivity of NiCr_{x}Fe_{2-x}O_{4} where (x=0.0, 0.2, 0.4, 0.6, 0.8 and 1.0) is shown in Fig. (2) as a function of reciprocal temperature 1000/T (K\(^{-1}\)) according to the relation:
\[ \rho = \rho_0 \exp\left(\frac{\Delta E}{k_B T}\right) \]  

(3)

where \( \rho_0 \) is pre-exponential constant, \( T \) is the absolute temperature (K), \( k_B \) is the Boltzmann constant and \( \Delta E \) is the difference between the activation energies in ferrimagnetic and paramagnetic regions [12, 14]. Figure 2 shows three regions of resistivity belongs to impurities, ferrimagnetic state and paramagnetic state respectively. The break point between ferrimagnetic and paramagnetic state is the Curie temperature are given in Table 2. The resulting decrease in Curie temperature with the increase of Cr content may be due to the reduction in Fe-Fe exchange interaction. The slope of each region depends on its own conductivity mechanism. The conduction below Curie temperature is due to hopping of electrons between Fe\(^{2+}\) and Fe\(^{3+}\) ions, whereas above Curie temperature the conduction is due to hopping of polarons [12]. The DC electrical resistivity around 500 K was found to increase by increasing Cr content from \( 25 \times 10^6 \) to \( 136 \times 10^6 \) \( \Omega \cdot \text{cm} \) except for the sample (x=1.0).

**Figure 2.** The variation of DC resistivity as a function of reciprocal temperature for Ni-Cr ferrite

Figure 3 shows the resistivity as a function of Cr content. In our samples Fe\(^{3+}\) ions are replaced by Cr\(^{3+}\) ions which have strong preference to B site, this leads to the migration of Fe\(^{3+}\) ions to tetrahedral site. It is known that the Cr ion don’t contribute in the conduction process and block the hopping electrons between Fe\(^{2+}\). Fe\(^{3+}\) which results in the increasing of the resistivity. The A-A hopping process doesn’t exist since only Fe\(^{3+}\) ions are present at A site, so the B-B hopping is more dominate process. This explain the increase in resistivity with increasing Cr content.

**Figure 3.** The variation of DC resistivity as a function of Cr content.

The activation energy in the ferrimagnetic and paramagnetic regions were calculated and are listed in Table 2. The activation energy in paramagnetic region is higher than that of ferrimagnetic region \( E_F < E_P \)
which indicate the effect of magnetic order on the conductivity. The value of activation energy increase by increasing Cr content which mean that the activation energy goes hand in hand with resistivity of the ferrite [40-41].

| Composition | TC (K) | Dc Resistivity Ωcm | Activation energy [eV] E_p | E_f |
|-------------|--------|---------------------|-----------------------------|-----|
| 0.0         | 553    | 25.73               | 1.17                        | 0.840 |
| 0.2         | 508    | 136                 | 0.541                       | 0.760 |
| 0.4         | 543    | 31.55               | 0.918                       | 0.624 |
| 0.6         | 538    | 65.4                | 0.740                       | 0.587 |
| 0.8         | 503    | 74.3                | 1.386                       | 0.786 |
| 1.0         | 532    | 10.34               | 0.660                       | 0.622 |

3.3. AC resistivity.
The variation of AC resistivity Vs (1000/T) for all studied samples are shown in figure 4 a and b. It was noticed that the plot has three regions each with its own slope. The conduction at low temperature < 370 K is due to impurities whereas at high temperature > 370 K is due to hopping conduction and polaron model. The temperature dependence of AC electrical conductivity is characterized by low temperature dependent concentration of charge carriers and high temperature dependence of mobility.

![Figure 4](image-url)

**Figure 4.** The variation of AC resistivity as a function of reciprocal temperature at different frequencies 1 kHz and 10 kHz for Ni-Cr ferrite.
Table 3. The Curie temperature from AC resistivity for NiCr$_x$Fe$_{2-x}$O$_4$.

| X  | $T_c$ (K) |
|----|-----------|
| 0.0| 623       |
| 0.2| 573       |
| 0.4| 578       |
| 0.6| 561       |
| 0.8| 548       |
| 1.0| 618       |

It is observed from the figure that the AC resistivity increase by increasing Cr content and decreased by increasing frequency. As the frequency of the applied field increase, hopping of electrons also increase thereby decreasing the resistivity. From table 3, it is observed that the transition temperature from ferrimagnetic region to paramagnetic region decreased by increasing Cr content from 623 to 548 K due to the decrease of B-B exchange interaction.

The real part of AC electrical resistivity consists of two terms [42]:

$$\rho = \rho_1(T) + \rho_2(\omega, T)$$  \hspace{1cm} (4)

The first term is temperature dependent which is related to the thermal activation of mobility of electric charge and follows the Arrhenius relation given by:

$$\rho_1 = \rho_0 \exp\left(\frac{E_a}{k_B T}\right)$$  \hspace{1cm} (5)

The temperature and frequency dependent AC resistivity term which is related to the dielectric relaxation caused by localized charge carriers is given by:

$$\rho_2(\omega, T) = B(T) \omega^n(T)$$  \hspace{1cm} (6)

where B is the parameter having unit and n is dimensionless parameter which can be calculated from the relation:

$$n = \frac{\ln \rho}{\ln \omega}$$  \hspace{1cm} (7)

The dependence of n on temperature is shown in figure 5a and b. It is known that the variation of n with temperature can give information about the type of conduction mechanism. For example, if n increase with temperature, the conduction obeys small polaron tunneling mechanism. The value of n also changed by changing Cr content which indicate the presence of hopping conduction mechanism by different hopping rates. The values of n ranged from 2.243 to 0.753 through the temperature range 300-800 K at 1 kHz which are agreement with the reported value in the previous work [43], and ranged from 1.659 to 0.5959 at 10 kHz also at the same range of temperature.

Figure 5. The variation of n vs. T (K) at different frequencies for Ni-Cr ferrite.
3.4. Dielectric properties of Ni Cr Ferrite.
Generally, there are four types of polarization which affect the value of dielectric constant. They are namely interfacial, dipolar, atomic and electronic [44]. At low frequency the polarization belonging to interfacial type which arises from structural anisotropy of the material are present. At higher frequency 10 kHz the orientation polarization take place. Above this frequency the dielectric constant originated from the atomic and electronic polarization. The dielectric constant decrease by increasing frequency which is explained on the basis of Maxwell-Wagner theory [45] of orientation polarization. In addition, at low frequency the interfacial polarization causes relaxation peak appears for some samples (x=0.4).

The effect of the temperature of dielectric constant of the composition NiCr,Fe2-xO4 at 1 and 10 kHZ in the temperature range 300-800 K are shown in figure 6 (a, b). It is observed that the dielectric constant for all samples gradually increase with increasing temperature up to 570 K for 10 kHz and 500 K for 1 kHz. Above these temperatures the dielectric constant sharply increases and some samples have a peak at nearly 750 K. At high temperature the contribution of dipolar and interfacial dominates and both of these polarizations are temperature dependent. This explains the rapid variation in dielectric constant at high temperature. Another reason for increasing dielectric constant is the increase in drift mobility of charge carriers and thermal activation of hopping electron at high temperature [1].

![Figure 6. The dielectric constant as a function of temperature at different frequencies for Ni-Cr ferrite.](image)

The dielectric constant decrease by increasing Cr content which is in agreement with the result of DC conductivity. That decrease by increasing Cr content. We can say that the mechanism process of polarization is similar to that of conduction process [46-47].

The peak appear at high temperature may be related to the existence of Debye relaxation process which exist in dipolar material due to the orientation polarization [48]. The decrease of dielectric constant by increasing Cr content can be explained as follow: the substitution of Cr ions at B site enforces Fe3+ ions to migrate at tetrahedral site which decrease the local polarization of hopping electrons between Fe3+-Fe2+ resulting in the reduction of dielectric constant.

3.5. Magnetic properties using VSM.

The magnetic hysteresis loop at room temperature of Ni-Cr ferrite at different Cr content are shown in figure 7 a, b and c. From the figure, saturation magnetization (Ms; maximum value of magnetization), remanent magnetization (Mr; magnetization at zero field), coercivity (Hc; magnetic field required to reduce the magnetization of that material to zero after the magnetization of the sample has been driven to saturation) and experimental magnetic moment (μexp) were determined and given in table (4). The area within the hysteresis loop is directly related to hysteresis loss. As the Cr content increase, the area within the hysteresis loop increases indicating the increasing of hysteresis loss. These results can be
confirmed from the Dc conductivity measurements which were found to decrease by increasing Cr content, which lead to the increase of the electric loss by increasing Cr content.

![Figure 7](image-url)

**Figure 7.** M-H curves for NiCr\(_{x}\)Fe\(_{2-x}\)O\(_4\) (x=0.0, 0.4 and 1.0) nanoparticles synthesized by flash combustion method.

**Table 4.** Values of saturation, remanent magnetization, coercive field, remanence ratio, experimental, theoretical magnetic moment and X-ray density for NiCr\(_{x}\)Fe\(_{2-x}\)O\(_4\) (Where x= 0.0, 0.4 and 1.0).

| Composition     | \(M_s\) (emu/g) | \(M_r\) (emu/g) | \(H_c\) (G) | \(M_r/M_s\) | \(\mu_{exp}\) | \(\mu_{th}\) | X-ray density (g/cm\(^3\)) |
|-----------------|-----------------|-----------------|-------------|-------------|--------------|--------------|-----------------------------|
| NiFe\(_2\)O\(_4\) | 28.508          | 3.6113          | 188.10      | 0.12667     | 1.19637      | 1.19637      | 5.461                        |
| NiCr\(_{0.4}\)Fe\(_{1.6}\)O\(_4\) | 21.140          | 3.5599          | 213.61      | 0.16839     | 0.88127      | 1.66         | 5.366                        |
| NiCrFeO\(_4\)   | 3.3239          | 0.99194         | 666.80      | 0.29842     | 0.13719      | 0.7          | 5.426                        |

The hysteresis loop shows the decrease of soft properties of the given ferrite which mean that the ferrite transfers to hard type for the sample NiCrFe\(_2\)O\(_4\). Accordingly, the coercivity is found to increase from 188(G) to 666(G) which is given from the equation:

\[
H_c = \frac{2K}{\mu \mu_s}
\]  

(8)

where K is the anisotropy and \(\mu\) is the initial permeability [49].
According to this relation, \( H_c \) is inversely proportional to \( \mu_s \), and this is really what we found experimentally that \( \mu_s \) decrease from 28 to 3 (emu/g) by increasing Cr content. The experimental magnetic moment was calculated using the formula:

\[
\mu_B = \frac{\mu_s \mu_E}{5585}
\]  

(9)

where \( \mu_s \) is the molecular weight of the sample. As given in Table 4 the experiment magnetic moment decrease by increasing Cr content, the preference of Cr ion to occupy B-site causes the ferric ion Fe\(^{3+}\) to migrate from A-site to B-site which results in the decrease in total magnetic moment. Theoretical and experimental magnetic moment have the same behavior with the increase of Cr content. Whereas the theoretical magnetic moment has greater values than experimental one, which may be due to the deviation from the proposal cation distribution estimated from X-ray diffraction pattern.

The grain size from SEM photograph were found to increase by increasing Cr content. Consequently, the higher the grain size the better the magnetic properties become. The high density of soft magnetic material is very useful to its magnetic properties (high saturation magnetization and low coercivity). As given in Table (4) the sample NiFe\(_2\)O\(_4\) has high magnetic properties which confirm our discussion about the influence of saturation properties on the magnetic properties of the materials.

4. Conclusion.
The auto combustion method is a suitable method for the preparation of Ni-Cr Ferrite in Nano-range. The Cr content enhance the crystallization of Ni Ferrite. The DC and AC resistivity was found to increase by increasing Cr content for most samples. The dielectric constant was increased gradually up to 570 and then increase sharply at higher temperature due to the contribution of dipolar and interfacial polarization while are temperature dependent. The hysteresis loop shows the decrease of soft properties and transfer to hard type by increasing Cr content.

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