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Structural and Electromagnetic Properties of Nanostructured ($Ni_{1-x}Co_xFe_2O_4$) ferrites prepared by Sol-Gel method

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ABSTRACT

Nanoparticles of Nickel-Cobalt ferrites with the chemical formula $Ni_{1-x}Co_xFe_2O_4$ for (x=0, step 0.2 up to 1) were prepared by sol-gel auto combustion technique at temperature (1100ºC). The prepared powder has confirmed the structure of cubic spinel structure. The samples were composed of nearly spherical nanoparticles with an average particle size 53.12nm. The surface structure of samples was investigated by scanning electron microscope (SEM). The electromagnetic properties for prepared samples were investigated using Vector Network Analyzer (VNA) in X-band microwave region.

KEY WORDS: Sol-gel auto combustion, Nickel-Cobalt Ferrites, Spinel Ferrite.

1. INTRODUCTION

Ferrites are classified as ferromagnetic materials so that they are electrically nonconductive material. Their chemical formula is AFe_2O_4 where A refers to any of various metal cations (Carter, 2013). They are two types of ferrites classified according to their magnetic coercivity and demagnetized resistance. The first has high coercivity and it is called hard ferrite, it is difficult to demagnetize, and it is used to make magnets. While the second type is the soft ferrites, it has low coercivity, and it is used to make ferrite cores for inductors and transformers, and in microwave components (Okamoto, 2009).

Spinel ferrites have a closed packed cubic structure (Ozguri, 2009), which are widely studied by the researchers to investigate their structural, electrical and magnetic properties (Augustin, 1996; Tahir, 2009; Aurelija, 2011; Binu, 2011; Al-Nesrawy, 2016; Kooti, 2012, Mohamed, 2003, Arabi, 2013). These ferrites are appropriate for magnetic sensors, microwave applications, and catalytic materials due to their dielectric constant, magnetic permeability, high Curie temperature and low tangent loss, also to chemical stability at low frequencies and mechanical strength (Ahmed, 2003).

Nickel ferrite is a soft magnetic material (Maaz, 2009), which has several applications in electronic devices, such as inductors and transformator cores (Van Uitert, 1956). While cobalt ferrite is recognized as hard ferrite compound with a temperate magnetization and a high coercivity. These features along with its large physical and chemical stabilities make Cobalt ferrite attractive for many practical applications such as digital recording disks, audio and video tapes, etc. (Pallaim, 1996).

2. EXPERIMENTAL METHOD AND MATERIALS

To prepare a one mole of $Ni_{1-x}Co_xFe_2O_4$, for (x=0, 0.2, 0.4, 0.6, 0.8, 1), they have been taken the calculated weights of the metal nitrates and mixed them with ferric nitrate, then dissolved in (100 ml) of distilled water in a glass beaker. The stirring process was carried out for the solution using magnetic hot plat-stirrers model (LMS-1003) to obtain a homogenized solution with complete dissolution for the raw materials. After that, ammonia solution was added to the homogenized solution to raise the pH of the solution to be (~7).

The temperature of hot plat- stirrer increased gradually to reach (50ºC), then it had been kept at this temperature for (15 mints). The temperature increased to (60ºC) for (2 hr), then to (70ºC) for (10 min), then to (80ºC) for (3 hr), the sol was initially transformed into gel. The temperature increased gradually to (90ºC) for (15 min), then to (100ºC), the gel dried at this temperature. The temperature goes gradually to (110ºC) for (10 min), then it becomes gradually to (120 ºC) and up to, then the dried gel was burnt in a self-propagating combustion manner until all gels were completely burnt out to form a fluffy loose structure.

The fluffy material was ground to get ferrite powder using the electric grinder for 10 minutes. The as-burnt ash was calcined at (600ºC) for rate 5ºC / minute, then trying to fix the temperature at (600°C) for one hour, after that the oven leaved to cool gradually, to get better crystallization and homogeneous of the spinel crystallite. Powder is mixing with glycerin material 6 wt% with purity of 88% as a binder. The powder was pressed at (40 kN) by the electrical piston to obtain samples as parallelogram of (2.4×1.2×1.2) cm in dimensions. Attended six samples for each Value of x. The samples were sintered for 6 hr at (1100ºC) sintering temperature with the rate of heating (3ºC/min), with staying at this temperature for (3 hr), then the temperature decreased gradually, it was left to be spontaneously cooling inside the furnace. Refine the samples to make its dimensions (7.5×21.5×9.5) mm.

Theoretical Background:

Structural properties: The structural properties that include the inter plane distance (d) and lattice constant (a) which can be calculated using Bragg's law equations 1 and 2 which is standard relation for the cubic system (Kittel, 2005).

$$n\lambda = 2d\ sin\theta \quad (1)$$

$$a = d\sqrt{h^2 + k^5 + l^2} \quad (2)$$

Where (hkl) are the miller indices, λ is the wavelength of X-ray radiation and θ is the Bragg angle. The crystalline size (C.S) is calculated using Scherer's formula:

$$\text{C.S} = \frac{0.9\ \lambda}{\beta\ cos\theta} \quad (3)$$

Where β is the FWHM (full width half maxima) of the peak at angle θ**.**

The percentage porosity (p) is calculated depending on values of X-ray density (ρ_{x-ray}) and bulk density (ρ_{bulk}) using the following equation (Cullity, 1956):

$$p = \left(1 - \frac{\rho_{bulk}}{\rho_{x-ray}}\right)\% \quad (4)$$

Electromagnetic properties: According to Nicolson-Ross-Weir method for a sample with a thickness d with scattering parameters S_{11} and S_{21} varied with current frequency (ω), The relation that govern the reflection coefficient (Γ) and transmission coefficient (T) are begin with the functions V_1 and V_2, and these are:

$$V_1 = S_{21} + S_{11} \quad (5)$$

$$V_2 = S_{21} - S_{11} \quad (6)$$

These functions submit a new function which is:

$$X = \frac{1 - V_1 V_2}{V_1 - V_2} \quad (7)$$

Then

$$\Gamma = X \pm \sqrt{X^2 - 1} \quad (8)$$

$$T = \frac{V_1 - \Gamma}{1 - V_1\Gamma} \quad (9)$$

And the appropriate sign is chosen so that $|\Gamma| < 1$.

The relative permittivity and permeability are given by (Nicolson, 1970):

$$\mu_r = \sqrt{c_1 c_2} \quad (10)$$

$$\epsilon_r = \sqrt{\frac{c_2}{c_1}} \quad (11)$$

Where,

$$c_1 = \frac{\mu_r}{\epsilon_r} = \left(\frac{1 + \Gamma}{1 - \Gamma}\right)^2 \quad (12)$$

and

$$c_2 = \mu_r \epsilon_r = -\left[\frac{c}{\omega d}\ ln\left(\frac{1}{T}\right)\right]^2 \quad (13)$$

Where c is the light speed, ω is the angular frequency, and d is the sample thickness.

Keep in mind that because the s-parameters are complex number then all other properties calculated from s- parameters are complex. Such as complex permeability and permittivity are thus obtained from measurement of the transmission and reflection scattering coefficients of a slab of the material.

3. RESULTS AND DISCUSION

Structural properties:

XRD characterization: Fig.1, shows the X-ray diffraction (XRD) patterns of the samples of $Ni_{1-x}Co_xFe_2O_4$ where (x=0, 0.2, 0.4, 0.6, 0.8, 1). The XRD patterns shows well developed diffraction line assigned to pure cubic spinel phase. The all measured XRD peaks match well with the standard patterns of inverse spinel ferrite. The crystalline size (C.S) for the prepared samples was found in the range of (~50-~55) nm. The percentage porosity (p) was found in range of (~43%-~55%)

Table.1, contains the values of structural parameters. The intensity & width of the Bragg's peak conforming, good crystallinity and nanoparticle size.

Table.1. Structural parameters of $Ni_{1-x}Co_xFe_2O_4$

x	a (nm)	C.S (nm)	D (nm) (311)	Porosity %	x	a (nm)	C.S (nm)	D (nm) (311)	Porosity %
0	0.833	52.42	0.244	55.19	0.6	0.836	54.87	0.211	51.21
0.2	0.834	54.56	0.211	53.73	0.8	0.837	49.91	0.211	42.98
0.4	0.835	53.41	0.245	48.97	1	0.838	53.53	0.245	54.49

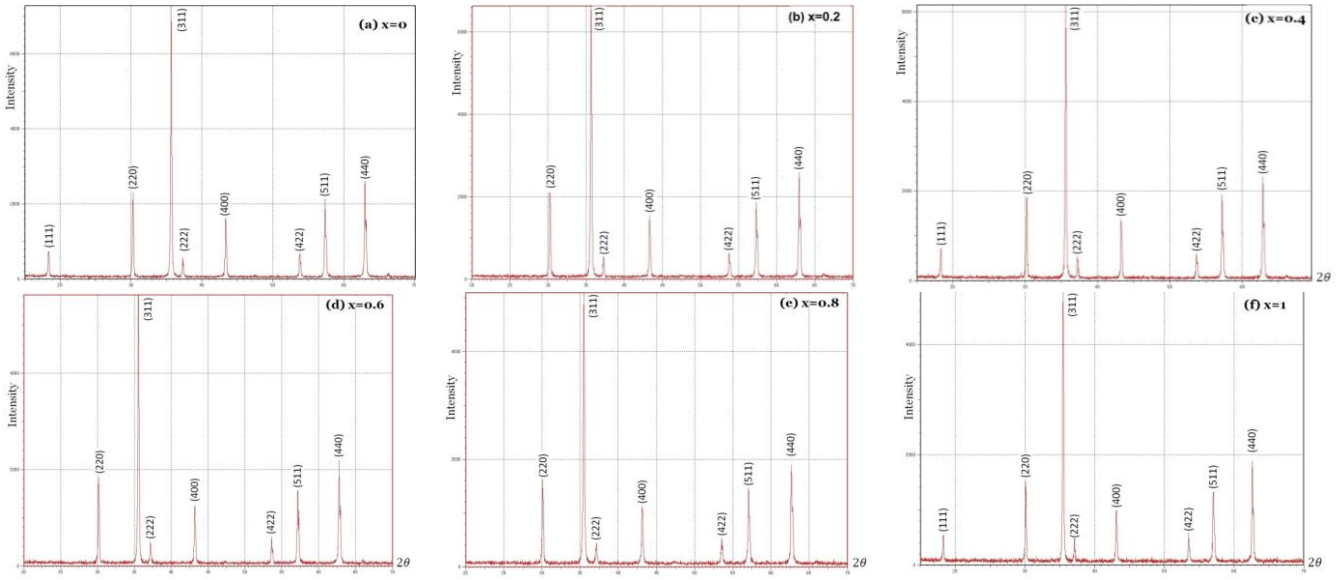

Fig.1. X-ray patterns of $Ni_{1-x}Co_xFe_2O_4$

Scanning Electron Microscope: The morphological patterns of $Ni_{1-x}Co_xFe_2O_4$ samples are shown in Fig.2, which taken by scanning electron microscope (SEM). SEM images confirmed the formation of Nanosize crystallites. The average grain size was found to be in range of 0.45-0.6 μm using linear intercept method.

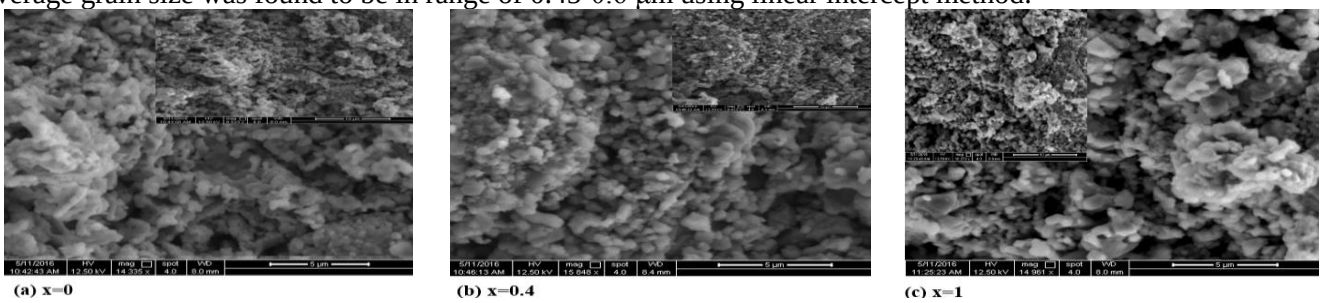

Fig.2. SEM images for $Ni_{1-x}Co_xFe_2O_4$ at magnification (5μm)

Electromagnetic properties: It was utilized, a sample of flat faces with 7.5 mm in thickness. The scattering parameters were measured Anritsu vector network analyzer at X-band microwave region. The real and imaginary permittivity and permeability were taken as a function of cobalt content as shown in Figs.3-6, respectively.

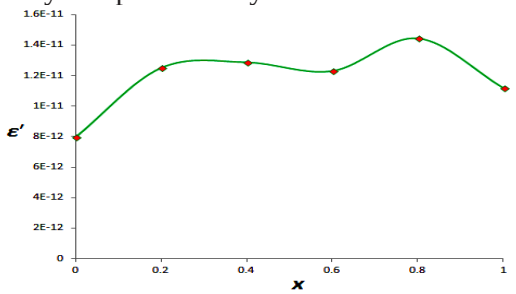

Fig.3. Real permittivity of $Ni_{1-x}Co_xFe_2O_4$ as a function of Co content

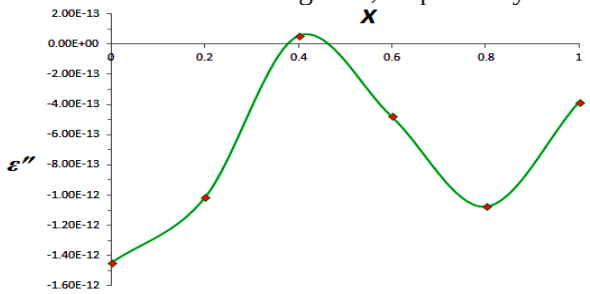

Fig.4. Imaginary permittivity of $Ni_{1-x}Co_xFe_2O_4$ as a function of Co content

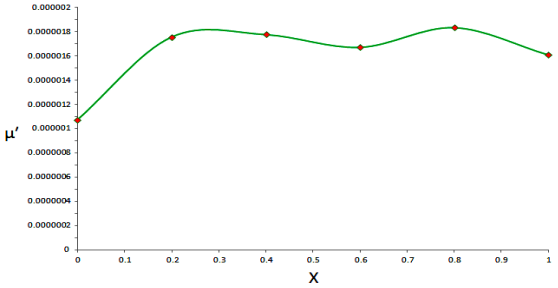

Fig.5. Real permeability of $Ni_{1-x}Co_xFe_2O_4$ as a function of Co content

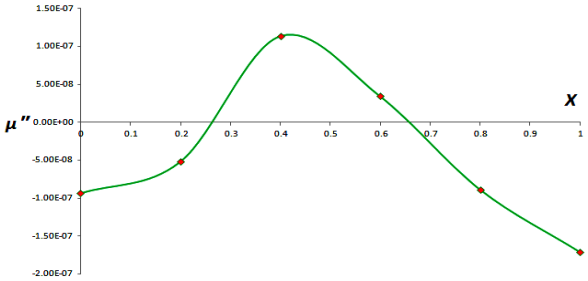

Fig.6. Imaginary permeability of $Ni_{1-x}Co_xFe_2O_4$ as a function of Co content

The reflection and transmission coefficients for the ferrite samples were taken as a function of cobalt content as shown in Figs.7 and 8 respectively, it is clear from Fig.7, that reflection coefficient increases slowly with increasing of cobalt content, while transmission coefficient decreases, and that behavior is due to the decreasing in electron density with increasing of cobalt content.

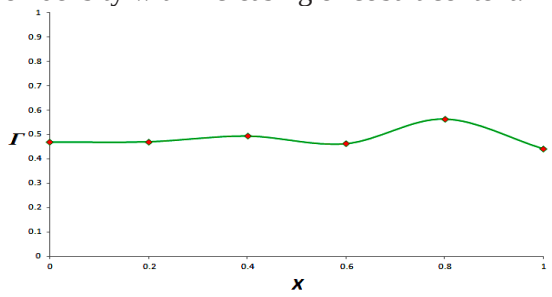

Fig.7. Reflection coefficient of $Ni_{1-x}Co_xFe_2O_4$ as a function of Co content

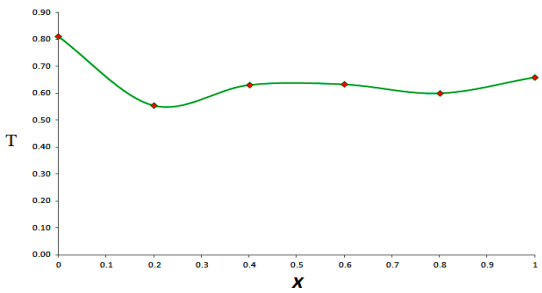

Fig.8. Transmission coefficient of $Ni_{1-x}Co_xFe_2O_4$ as a function of Co content

The other quantities had been investigated are relative dielectric constant, relative permeability and loss tangent which shown in Figs.8-10, respectively each one of them is plotted as a function of cobalt content.

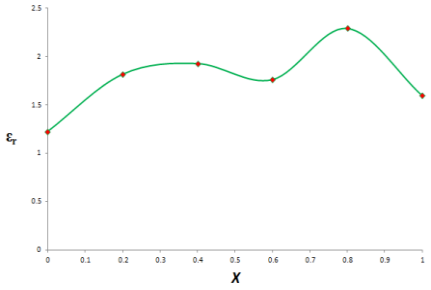

Fig.8. Relative dielectric constant of $Ni_{1-x}Co_xFe_2O_4$ as a function of Co content

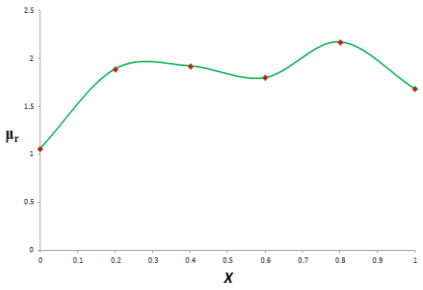

Fig.9. Relative permeability of $Ni_{1-x}Co_xFe_2O_4$ as a function of Co content

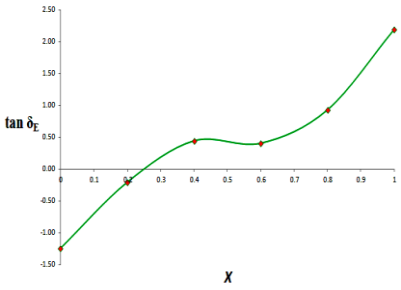

Fig.10: Loss tangent of $Ni_{1-x}Co_xFe_2O_4$ as a function of Co content

4. CONCLUSIONS

The $Ni_{1-x}Co_xFe_2O_4$ (x=0, 0.2, 0.4, 0.6, 0.8, 1), nanosize were prepared using sol-gel technique. The change in the Co^{2+} content gives the significant effects in electrical and magnetic properties of the material. The crystalline size is obtained 50 to 55 nm. X-ray diffraction results showed the presence of all characteristic reflections (111), (220), (311), (222), (400), (422), (511), (440) which confirmed the structure of cubic spinel ferrite. The lattice parameter increases with increasing Co^{2+} content in Ni.

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