



A study on the dielectric properties of La^{+3} doped nickel ferrite

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ABSTRACT

In the present work, the Ni-La ferrite chain ($\text{NiLa}_x\text{Fe}_{2-x}\text{O}_4$) with ($x=0.0- 0.2$), were synthesized using sol-gel auto-combustion as a modern chemical methods. Nitrates, citric acid and ammonia were used in order to get ferrite nano powders at temperature of $(200)^\circ\text{C}$, which characterized a higher dispersion and homogeneity with particle size in the range of (20-39) nm. The dielectric properties are measured using (LCR) meter in the frequency range of (50Hz –5MHz). Dielectric constant (ϵ_r'), the loss tangent ($\tan\delta$) and the loss factor (ϵ_r'') are decreases with increasing frequency, while the Conductivity ($\sigma_{a.c}$) increases with increasing the rare earth ion (La) concentration in Ni ferrite.

Keywords: Ni-La ferrite, spinel ferrite materials, ac conductivity of ferrites, and dielectric of ferrites.

دراسة الخصائص العزلية لفرايت النيكل المطعمة بعنصر La^{+3}

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الملخص

في العمل الحالي تم تحضير سلسلة فرايت النيكل لثنائيوم ($Ni La_x Fe_{x-2}O_4$) عند قيم تراوحت (x من الصفر الى 0.2) باستخدام الطريقة الكيميائية الحديثة (sol-gel auto combustion). وقد استخدمت النترات وحامض الستريك والامونيا كمواد اولية داخلية في التفاعل للحصول على مسحوق فرايتي ذات دقائق نانوية عند درجة حرارة (200) م. تتميز بكونها دقائق متجانسة ذات احجام في المدى (20-39 نانومتر). تم قياس ثابت العزل الكهربائي بجزئيه الحقيقي والخيالي وفقد الظل باستخدام (LCR meter) عند المدى الترددي (50 هرتز - 5 ميكا هرتز). لوحظ بان ثابت العزل الكهربائي وفقد الظل وفقد العازل (ثابت العزل الخيالي) تتناقص مع زيادة التردد. كما اوضحت النتائج بان الموصلية الكهربائية المتناوبة تزداد مع ازدياد محتوى عنصر الاتربة النادرة (لانثانيوم) في فرايت النيكل.

الكلمات الدالة: نيكل لثنائيوم فرايت، مواد أسبيل فرايت، التوصيلية المتناوبة للفرايت ، وعزلية الفرايت.



1. Introduction

The complex oxide, which contains trivalent iron ion as the main ingredient is generally called Ferrite [1]. Magnetic ceramics, or ferrites, are a very well established group of magnetic materials. Research activity on ferrites, especially intense in the last 50 years has led to the establishment of many theories and models additional to or complementing those obtained from research on metallic materials [2]. The name "Ferrite" deduces that iron oxide Fe_2O_3 is generally the common oxide to all ferrite. The magnetic properties arise from interactions between metallic ions occupying particular positions relative to the oxygen ions in the crystal structure of the oxide. Ferrites or Ferromagnetic are oxides ceramic have brown, black and gray color with spinel structure general formula (MFe_2O_4) , where (M) is divalent metal ion like (Ni, Mn, Mg, Zn, Cu, Co, Fe, or a mixture of such ions) possess the properties of ceramic materials, such as being hard, brittle and chemically inert. The electromagnetic properties of ferrites can be changing by controlling the manner of preparation and type of materials used, amount of material added and sintering temperatures,[3]. The resistivity of ferrites at room temperature can vary from $(10^{-2}$ to $10^{11}) \Omega\text{-cm}$, depending on their chemical composition [4]. They are considered superior to other magnetic materials because they have low eddy current losses and high electrical resistivity. Ferrites have good magnetic properties at the same time are the insulating materials so it is preferred in industries which operate within the high frequency up to (10) MHz , they do not readily conduct electricity. This also gives them an advantage over iron, nickel and other transition metals that have magnetic properties in many applications because these metals conduct electricity [5,6]. Due to rapid progress in the fabrication and processing of nanostructures, ferrite magnetic materials can be made in the range of nanometer (1-100) nm. The chemical and physical properties of nano -materials can significantly differ from those of bulk materials of same chemical composition [7]. Suitable control of properties and response of nanostructures can lead to new devices and technologies. Several research groups are involved in the investigations of spinel oxide nano particles because of their potential applications in magnetic devices, microwave technology and high-density magnetic recording media, etc [8].

2. Material and Method

2.1. Material

Ferrite powders were prepared by sol-gel auto combustion method. Analytical grade Iron (III) nitrate $[\text{Fe}(\text{NO}_3)_3 \cdot 9\text{H}_2\text{O}]$ with purity ($>99\%$) Nickel Nitrate $[\text{Ni}(\text{NO}_3)_2 \cdot 6\text{H}_2\text{O}]$ with purity ($>99.9\%$), Lanthanum nitrate hexahydrate $[\text{La}(\text{NO}_3)_3 \cdot 6\text{H}_2\text{O}]$ with purity ($>99\%$), ammonia solution and citric acid $[\text{C}_6\text{H}_8\text{O}_7 \cdot \text{H}_2\text{O}]$ with purity ($>99\%$), were used as raw materials in auto combustion method.

2.2. Ni ferrite nano particle Preparation

After the weight of nitrates, appropriate amount of distilled water was added to them, according to the percentage standard stoichiometric weight: two moles of iron nitrate, one mole of nitrate (Lanthanum and Nickel) and three moles of citric acid (the mole ratio of metal nitrates to citric acid is equal to one) to provide increased fuel to the mixture of ferrite series ($\text{Ni La}_x\text{Fe}_{2-x}\text{O}_4$), where x take the values (0.0 to 0.2), All these are collected in a glass beaker to become a total solution and mixed well at room temperature by magnetic stirrer with high velocities and after a short period until solution becomes smooth and a slimy red-colored as shown in **Fig. (1a)**. Ammonia solution was slowly added in the form of drops into the mixed solution to control its pH until reach the value of (6.9 to 7) with continuous stirring and so the solution become a dark brown color as shown in the **Fig.(1b)**.

The temperature of solution was raised subsequently to $(60)^\circ\text{C}$ for a period of one hour and then increased to $(85)^\circ\text{C}$. After that the size of the solution in the beaker glass be less with high viscosity and after 30 minutes, the solution viscosity is very high, hence the beginning of gel formation on the surface of the solution, particularly in the middle and then all the solution convert to gel, and even this moment, the solution is still on the magnetic stirrer and temperature $(85)^\circ\text{C}$ as shown in the **Fig. (1c)**.

After the completion of the solution turned to gel, the temperature drops to the room temperature and this gel becomes dry and dark brown. Where the weight of the gel putting in glass beaker by a sensitive balance and then put it inside the oven at a temperature $(120)^\circ\text{C}$ for three hours to become dry gel and decreasing in weight as shown in **Fig. (1d)**. Then evaporation of some of the material at raise the temperature of the dried gel to $(200)^\circ\text{C}$, after 15 minutes the dry gel began to change shape appear convexity in the center of the glass beaker as` shown in

Fig. (1e).After a short period we note a flame at the top of convexity where the flame spread in all directions to burn the top layer of dried gel and turn into a grid of columns converging and intersecting at random and ascend to the top and then burn another layer of dried gel and ascend to the top also, and so until the gel flammable in complete and be the end of the flame in the bottom of the glass beaker and thus becomes all the dried gel after combustion to a fine powder with a dark gray color, which marks the beginning of formation of high purity ferrite as shown in Fig. (1f).

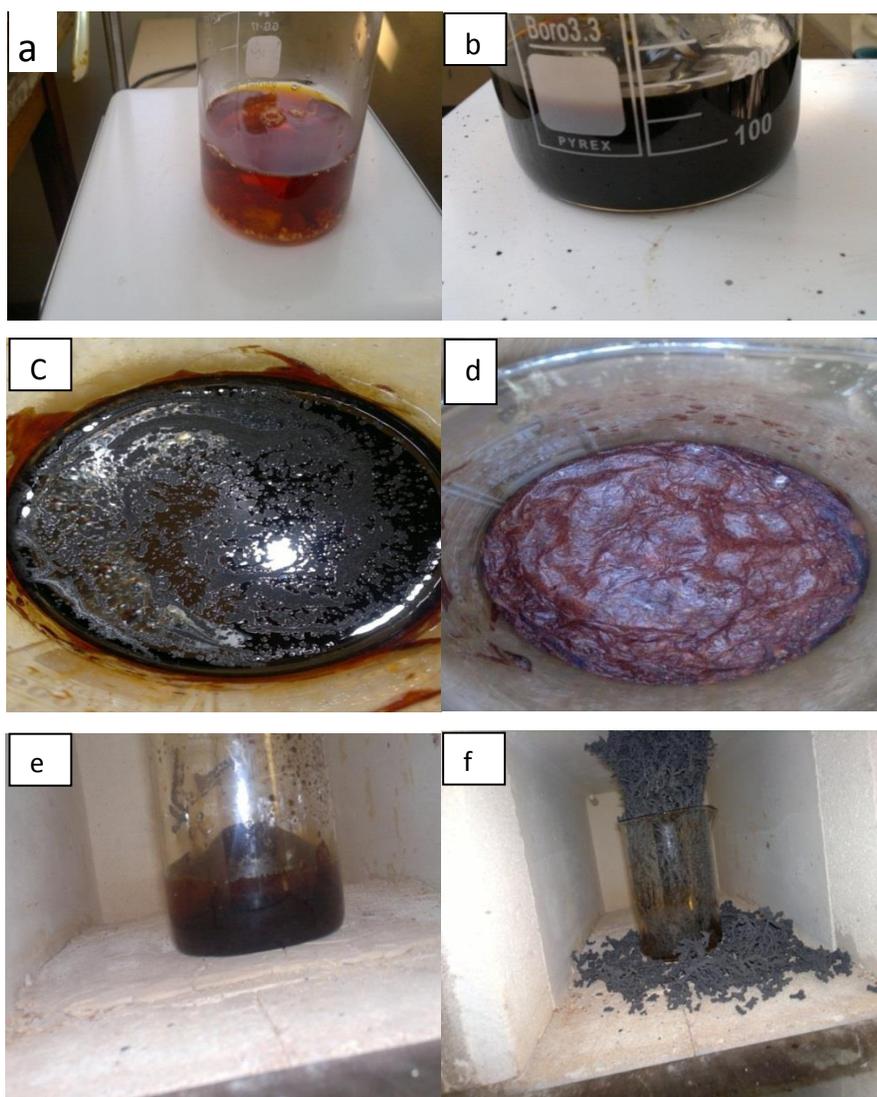


Fig. (1): Photographs of (a) Nitrate-citrate solution, (b) The solution after the adding of ammonia, (c) Dry gel, (d) Dry bulk temperature of (120)°C, (e) Dry bulk temperature of (200)°C and (f)Auto combustion and become nanopowder ferrite

Briefly, we can describe the state that our saw in the following; the dried gel was placed on a hot plate at (200)°C. Upon ignition, dried gel 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. The as-burnt ash was calcined at different temperatures (400, 600 and 800)°C for three hours to get better crystallization and homogeneous cation distribution in the spinel and finally ground to get NiLa_xFe_{2-x}O₄ ferrite nanopowders. To prepare the samples of the ferrite for dielectric measurement, pressed the powders into pellets with (12)mm diameter and (2)mm in thickness by using mechanical hydraulic press with pressure of (2.5) ton/ cm² .Finally these are sintered at temperature of (1000)°C for three hours.

3. Experimental characterization

3.1. Electrical properties

The investigations into electrical properties of sintered samples are essential for their use in various applications. Dielectric behaviors (dielectric constant, dielectric loss factor, and dielectric loss tangent) were studied as a function of frequency, and AC conductivity was studied as a function of frequency.

3.1.1. Dielectric constant

Capacitance are one of the many components used in electronic circuits. The basic constriction of capacitor is dielectric material sandwiched between tow electrode capacitance in the measurer of the quantity of electrical charge (Q) that can be held (stored)between two electrodes. The charge on the capacitance is[9];

$$Q = C * V \quad \dots\dots\dots (1)$$

Where (V) is the applied voltage and (C)is the capacitance .The dielectric constant is calculated from the capacitance value of the material, with different frequency measured by the LCR meter model (889B bench), by the formula given below[9]:

$$\epsilon'_r = C / C_0 \quad \dots\dots\dots(2)$$

(C) is the capacitance of the material and The (C₀) is the capacitance of air

$$C_0 = \epsilon_0 A/ d \quad \dots\dots\dots (3)$$

Where, (ε₀) is the permittivity of the air and has a value of (8.854*10⁻¹²)F/m. Equation becomes:

$$\epsilon_r' = Cd / A\epsilon_0 \dots\dots\dots(4)$$

(C) is the capacitance of the pellet in farad, (d) is the thickness of the pellet in meters, (A) is the cross-sectional area of the flat surface of the pole and (ϵ_0) is the constant of permittivity for free space [11] This equation is used for the calculation of the dielectric constant from the measured capacitance of the sample from (LCR)meter.

$$\tan \delta = \epsilon_r'' / \epsilon_r' \dots\dots\dots(5)$$

and

$$\epsilon_r'' = \tan \delta \times \epsilon_r' \dots\dots\dots(6)$$

where, ($\tan \delta$) is tangent loss dispersion factor and (ϵ_r'') is imaginary dielectric constant [10].

3.1.2 A.c. Conductivity

The a.c. conductivity of these samples was calculated from the values of dielectric constant and dielectric loss factor using the relation [11]

$$\sigma_{ac} = \omega \epsilon_0 \epsilon_r' \tan \delta \dots\dots\dots(7)$$

Where (σ_{ac}) is the ac conductivity, (ω) is the angular frequency, (ϵ_0) is the permittivity of free space, (ϵ_r') is the dielectric constant and ($\tan \delta$) is the dielectric loss factor of the sample.

4. Results and discussion

Real, imaginary dielectric constant and tangent loss in ferrites are contributed by several structural factors. Figures (2) , (3) and (4) show that the real, imaginary dielectric constants and loss tangent of all Ni-La ferrite samples decrease with increasing frequency and the maximum values are in the range of (10-25) for real and imaginary parts, while the maximum values for loss tangent are in the range of (0.4-1.0) for all samples. These value decreased rapidly at the lower frequencies as compared with high-frequencies region, as already reported [12]. It is observed that at very high frequency, both dielectric constant and loss factor value do not depend on frequency, which is a normal behavior for semiconductors and follows the Debye-type dielectric dispersion process [13]. This is due to the fact that hopping frequency does not follow up the field variation at high frequency. It can also be explained by the Maxwell-Wagner interfacial type polarization, which is also fully in agreement with Koop’s phenomenological theory [14].

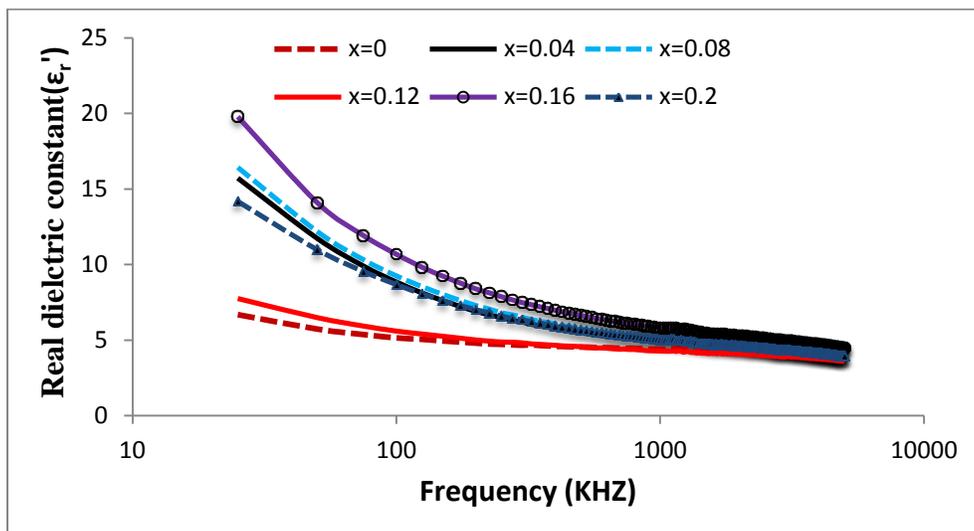


Fig. (2): Real dielectric constant (ϵ_r') as a function of frequency with different La- ion concentration

The loss factor decreases with the increase in frequency from 100 Hz to 5 MHz as shown in Fig. (3). The values of ($\tan \delta$) depend on a number of factors such as stoichiometry, Fe^{3+} content and structural homogeneity, which in turn depend on the composition and sintering temperature of the samples.

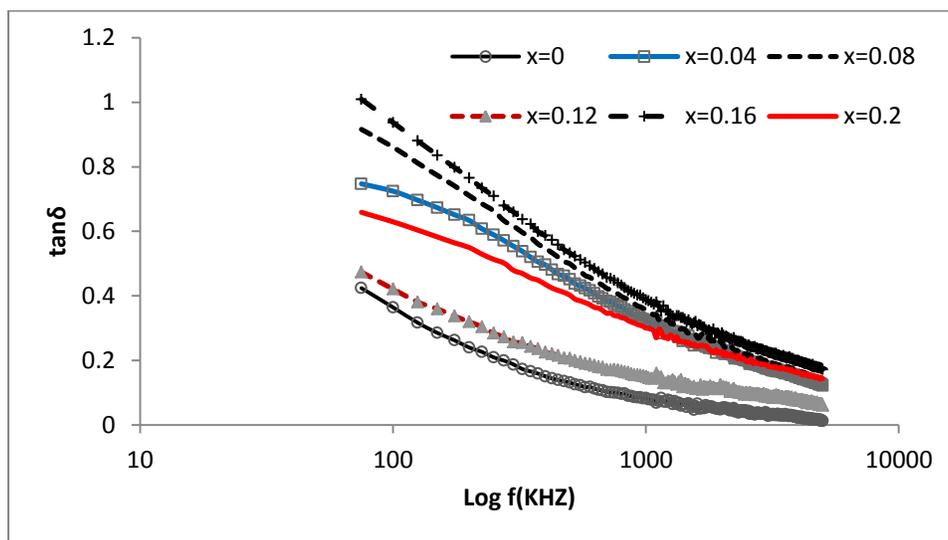


Fig. (3): Loss tangent ($\tan \delta$) of Ni-La ferrites system with different concentration..

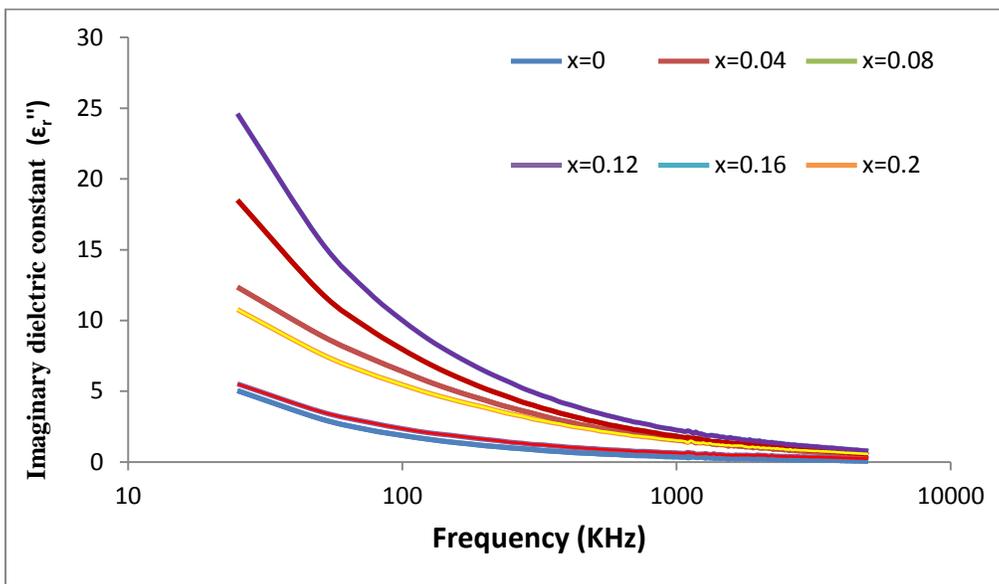


Fig. (4): Imaginary dielectric constant (ϵ_r'') of Ni-La ferrites system with different concentration

Fig.(4) shows continuous decrease in dielectric loss(imaginary part) with applied frequency, this decrease with increasing frequency for all sample is attributed to the decrease in the polarization of the sample because the dipoles cannot follow up the field variation. The loss factor is similar to those of the real part of dielectric constant

The a.c conductivity of the sintered ferrite samples was calculated from the values of dielectric constant and dielectric loss factor using the relation (7). It is observed that σ_{ac} increases with the increase in (La^{+3}) concentration, and with the decrease in (Fe^{+3}) ion concentration in Ni $La_xFe_{2-x}O_4$ system for all samples as shown in Fig. (5)

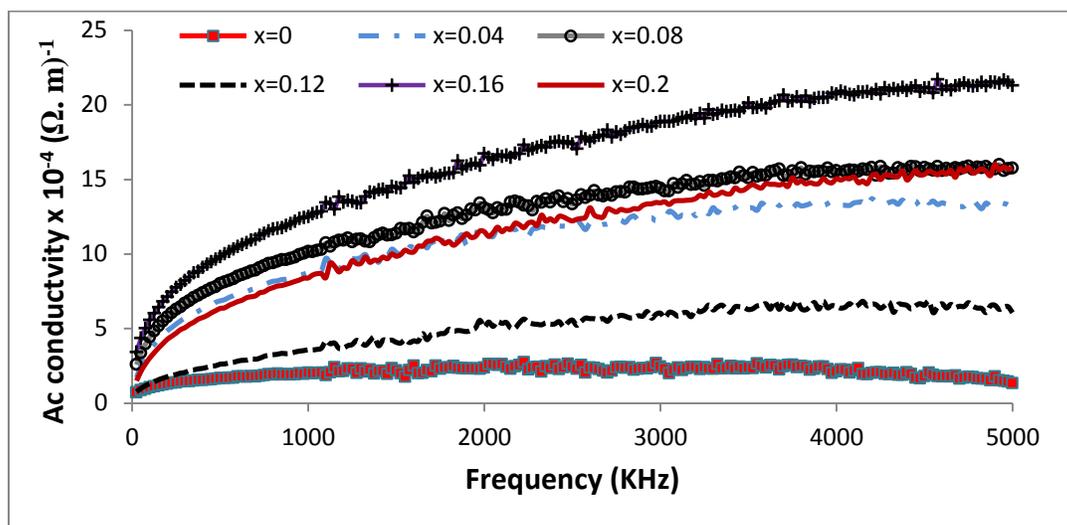


Fig. (5): A.C. conductivity (σ_{ac}) of Ni-La ferrites with different concentration

5. Conclusions

- 1- The real dielectric constant, imaginary dielectric constant and loss tangent decrease with increasing frequency, while the dielectric constant increases with increasing La-ion concentration in Ni-La ferrite samples.
- 2- The imaginary dielectric constant and tangent loss decrease with increase in La- ions concentration.
- 3- An a.c. conductivity (σ_{ac}) increases with increasing frequency and concentration of Lanthanum ions added to the Ni ferrite .

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