

Synthesis and Studying Dielectric Properties of Mixed Nanoferrites

Dr. Fadhil A. Chyad

Materials Engineering Department, University of Technology/Baghdad.
fchyad_2009@yahoo.de

Dr. Mohammed S. Hamza

Materials Engineering Department, University of Technology/Baghdad.

Zainab I. Dhary

Materials Engineering Department, University of Technology/Baghdad.

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ABSTRACT

Based on a low temperature route (sol-gel autocombustion) a nano-ferrite CoFe_2O_4 and $\text{Li}_{0.5}\text{Fe}_{2.5}\text{O}_4$ and different percentages of cobalt-ferrite are synthesized. The crystalline structure by XRD and dielectric properties with TEM micrographs are presented. The XRD (crystallite size and lattice parameter) are determined which affected by cobalt content. TEM micrographs show a nano-particles in the range of (8-20) nm which correlate with crystallite size. Dielectric properties (dielectric constant, loss tangent and dielectric loss factor) are decreased with frequency and cobalt content.

Keywords: ferrite, crystallite size, dielectric properties, TEM, sol-gel.

تحضير ودراسة بعض الخواص الميكانيكية المترابطة

الخلاصة:

استخدمت طريقة السول-جيل (الاحتراق الذاتي) لتحضير فيرايت الكوبلت والليثيوم النانوي وكذلك خلط نسب مختلفة من الكوبلت مع الليثيوم. تم دراسة التركيب البلوري باستخدام حيود الاشعة السينية والتركيب المجهرى باستخدام المجهر الالكتروني النافذ بالاضافة الى قياس الخواص العزلية. تم حساب الحجم البلوري و ثابت الشبكية حيث تآثر كليهما باضافة فيرايت-الكوبلت وان صور المجهر الالكتروني النافذ بينت ان معدل الحجم الحبيبي كان من (8-20) نانومتر والتي تكون متقاربة مع الحجم البلوري. اما الخواص العزلية (ثابت العزل، ظل فقدان الزاوية، معامل فقدان العزلي) فقد تناقصت مع التردد مع محتوى الكوبلت.

INTRODUCTION

Historically ferrite attracted the attention of the physicists and technologies since they were magnetic semiconductors which exhibit interesting properties that could be used in many electronic devices.

The increasing demand for low loss ferrites results in detailed investigation of the various aspects of electrical conductivity and on the influence of various substitutions. Nano-sized spinel ferrite particles have received enormous attention during the past several years because of their chemical and physics properties [1-2].

Lithium-ferrite, is an unusual and, in the same respect, a remarkable material, where, a several research programs have been understating to study its fundamental properties and to develop high-power micro-wave materials from it.

Reddy[3] studied the mixed ferrite $(Li_{0.5}Fe_{0.5})_{1-x}Ni_xFe_2O_4$. Electrical conductivity and thermoelectric power as a function of composition and temperature were measured. Charge carriers mobility, dielectric constant and loss tangent were studied at different frequencies.

Sun et al [4], studied the ferrite system $(Ni_{0.5}Zn_{0.5}Fe_{2-x}R_xO_4)$ where R= La or Gd and $x= 0-0.04$. The structural, magnetic and dielectric properties were investigated. It is found that the increasing of R_2O_3 the relating density decreased and lattice parameters increased. The dielectric properties decreased with frequency while increasing with rare earth content.

Wende and langbein[5], were studied the thermal decomposition of freeze-dried Li-Mn(II)Fe(III) formatted precursor by different thermal analysis (DTA) and DTG. It was found that the thermal decomposition is a suitable method of preparing single phase solid solution ferrite $(Li_xMn_{1-x}Fe_{2-2x}O_4)$ with 1×1 at relieving low temperature.

Guilherme et al[6], were studied the structural investigation of MFe_2O_3 (M= Fe, Co). Ferrites of this type were prepared by the traditional co-precipitation method. XRD and TEM analysis was showed that the magnetic nano-particles (non-modified and modified) present diameters in the range (10-15) nm.

Naidak et al [7], were studied the structure and morphology of spinel MFe_2O_4 (M= Co, Ni) nano-particles, chemically synthesized from hetrometallic complexes. They found that the nanoparticles range from (9-25) nm. As a consequence, the magnetic properties of this ferrite are closer to the aimed room temperature super paramagnetic behavior, than are those of other ferrites obtained by a mixture of salts.

Arana et al [8], were studied the Li-substituted Mn-Zn ferrite structural and magnetic properties after different thermal treatment. Spinel ferrites of composition $Zn_{0.6}Mn_{0.4}Fe_2O_4$ and $Li_{0.2}Mn_{0.4}Fe_{2.5}O_4$ were prepared by the self-combustion sol-gel method crystalline lattice increases by lithium incorporation.

Mohapatra et al [9], were studied the surface controlled synthesis of MFe_2O_4 (Mn, Co, Ni and Zn) nano-particles are magnetic and showed cubic-spinel at characterized by XRD and Raman spectroscopy.

Theoretical part:

The dielectric measurements were carried out at room temperature over the frequency range (50Hz-5MHz). The values of capacitance (C), dielectric constant, loss tangent and loss factor were noted directly at different frequencies.

The dielectric constant (ϵ) was calculated using the following relation:

$$\epsilon = \frac{Cd}{\epsilon_0 A} \quad \dots (1)$$

Where

d is the distance between the plates of the capacitors, A is the surface area of the plates, ϵ_0 is the permittivity of the vacuum (8.85×10^{-12} f/M).

Dielectric loss tangent ($\tan \delta$) was calculated using the following relation:

$$\tan \delta = \frac{\epsilon''}{\epsilon'} \quad \dots (2)$$

Where

ϵ'' is the dielectric loss factor.

The crystallite sizes were calculated by Scherer's formula using the full width at half maximum (FWHM) value of indexed peak in the X-ray pattern[10]:

$$D = \beta \frac{0.9\lambda}{\cos \theta_\beta} \quad \dots (3)$$

Where

λ is the wave length incident X-ray (1- 5405 Å), θ_β is the diffracted angle and β is the FWHM value at θ_β in radius.

Experimental part

Most of studies are preparing the nano-ferrite as oxides, while this work used another method which mixed cobalt-ferrite of different percentages with lithium-ferrite.

The raw materials used to prepare the ferrite were, lithium nitrate, cobalt nitrate, iron nitrate, citric acid as fuel, ammonia, distilled water and ethanol. Cobalt and lithium nitrate and iron nitrate solution prepared as 0.5M. The cobalt solution was mixed with iron solution according to definite chemical stoichiometric ratio as (Fe: Co=2:1) by using magnetic stirrer while for lithium was (Fe: Li=5:1).

Different volume fractions of cobalt ferrite were added to the lithium-ferrite (2,5, 10, 15, 20). Glucose was added as a surfactant material with continuous mixing, then ammonium hydroxide dropped to the mixed solution until the gel bed was formed at PH 5. Addition of citric acid that leads to hear notifying the combustion of citric acid which helps in reducing the particle size of producing gel. The last step was filtrating the gel with washing by distilled water to get out the gel. The received gel was calcined at 800 °C for 2 hours.

XRD, TEM were used to evaluate the crystallite size and particle size. Dielectric parameters were measured.

Result and Discussion

Figures (1-5) show the XRD patterns of the synthesized powders of lithium ferrite and cobalt-ferrite alone and that with different percentages of cobalt-ferrite after calcined at 800 °C for 2 hours. The diffraction peaks of all the samples assigned to cubic fluorite are consistent with the JCPDS (Joint committee on powder diffraction standard).

The peaks are appreciably sharpened which indicate higher crystallinity. Adding the percentages of cobalt-ferrite have shifted the peak for higher diffraction angles which can also give information of the diameter of crystal.

The crystallite sizes were calculated by Scherer's formula. Table (1) shows the effect of the cobalt-ferrite content on the crystallite size of the mixed ferrite. 10% has the lowest value while in the other percentages the crystallite size increased.

TEM micrographs as shown in figure (6) illustrates the size and shape of the particles extracted from the powder reduced at 800 °C for 2 hours. This figure shows the range of particle sizes was (8-20) nm and the particles showed regular and nearly round shape appearance and in good contact to each other. Some appreciable formation of heavy agglomeration powders were present and this was attributed to the large surface area of these nanoparticles. This range of sizes is in good agreement with XRD crystallite size.

Figures (7-12) represent the dielectric properties of the mixed ferrite. Figure (7) shows the relation of dielectric constant with frequency at different percentages of cobalt-ferrite. Generally dielectric constant decreases with increasing frequency and that may be due to the basis of the mechanism of polarization process in ferrite, which is similar to that in the conduction process. The whole polarization in ferrite is mainly contributed by the space charge polarization, which is governed by the number of space charge carriers and the conductivity of the material [11].

Figure (8) shows the effect of cobalt additives on the dielectric constant of the mixed ferrite at 1MHz. The increasing of cobalt content decreased the dielectric constant, having the lowest value at 10%. The loss tangent has the same behavior of dielectric constant which decreases with frequency and the cobalt content as it is shown in figures (9 and 10).

Conclusions:

- 1-Mixed ferrites are successfully synthesized by sol-gel auto combustion technique.
- 2-The microstructure of ferrite prepared by the technique, consists of crystalline which is clearly seen spinel cubic form.
- 3-Structural studies proved the nanocrystalline nature of the samples.
- 4-Dielectric properties of the mixed ferrite show inverse trend with frequency at different percentages of cobalt addition which decreases rapidly with frequency.
- 5-Dielectric properties decreases as the cobalt content increases.

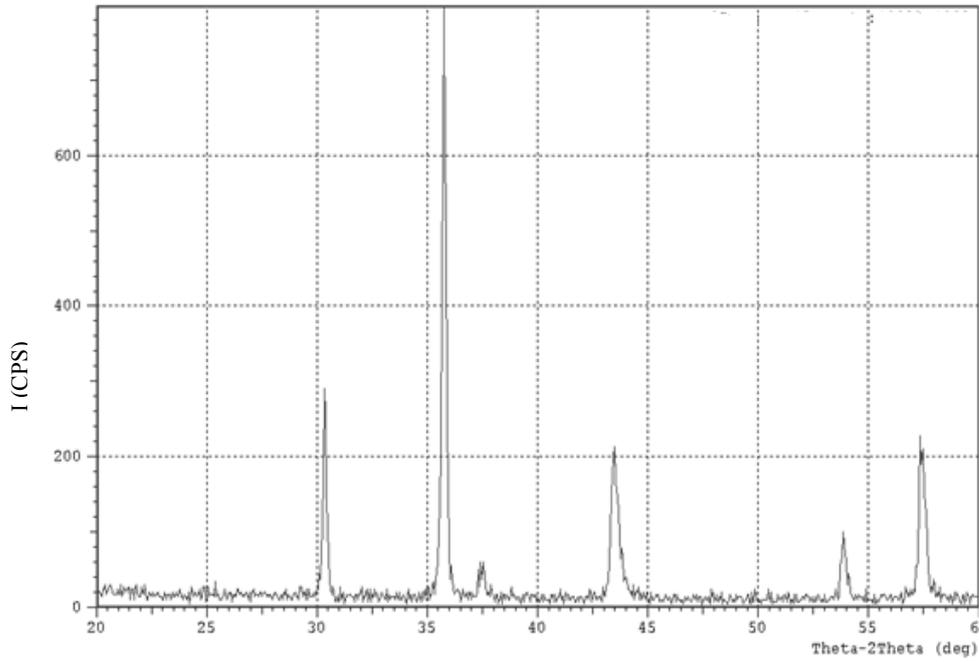


Figure (1): XRD pattern of the prepared lithium-ferrite powder at 800 °C.

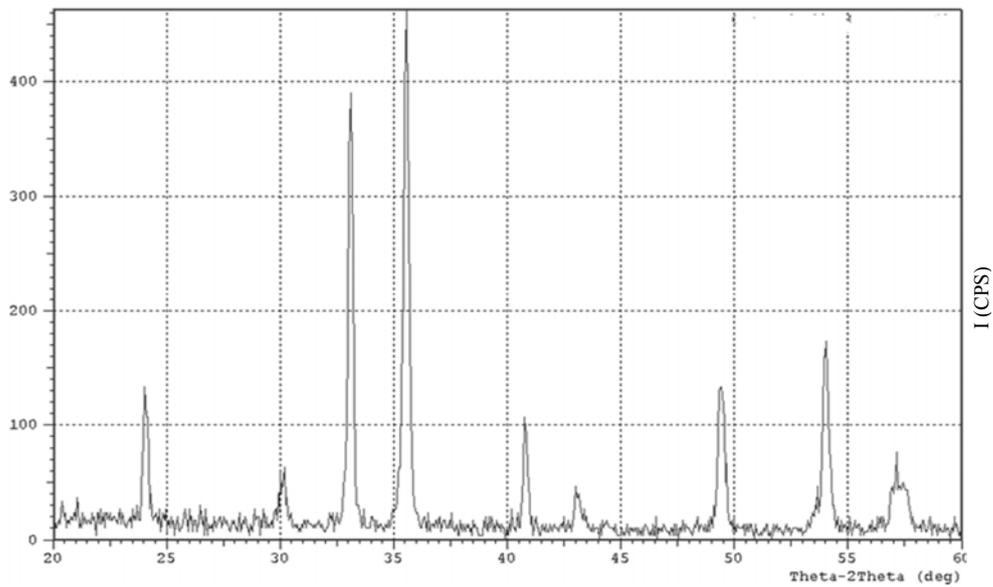


Figure (2): XRD pattern of the prepared cobalt-ferrite powder at 800 °C.

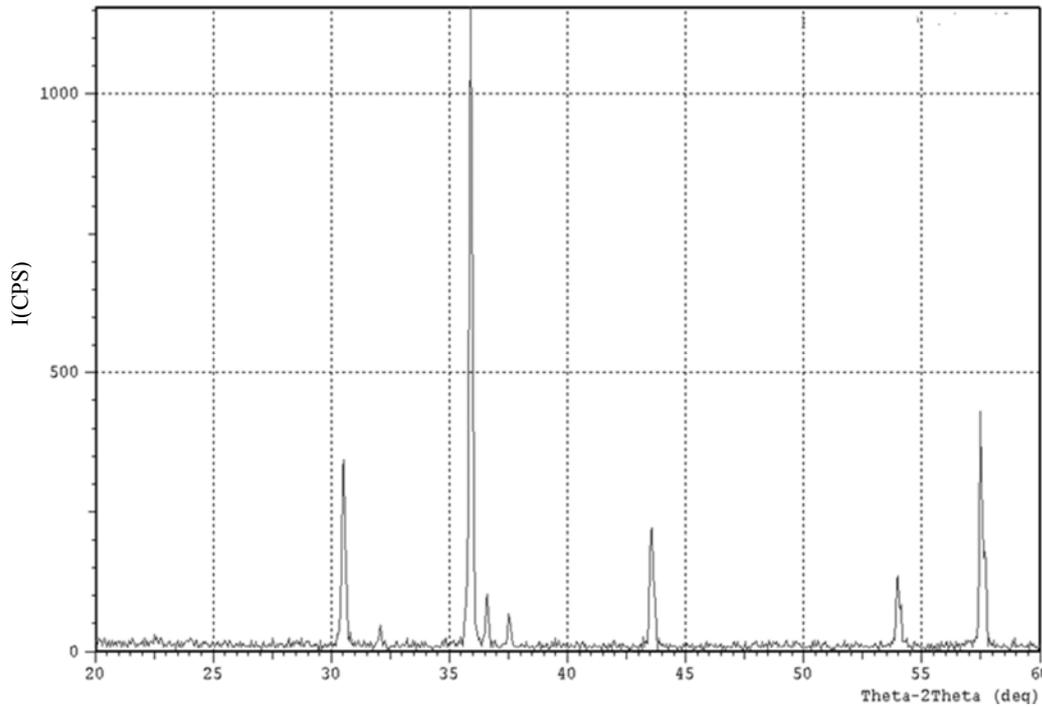


Figure (3): XRD pattern of the prepared lithium ferrite doped with 2% cobalt-ferrite at 800 °C.

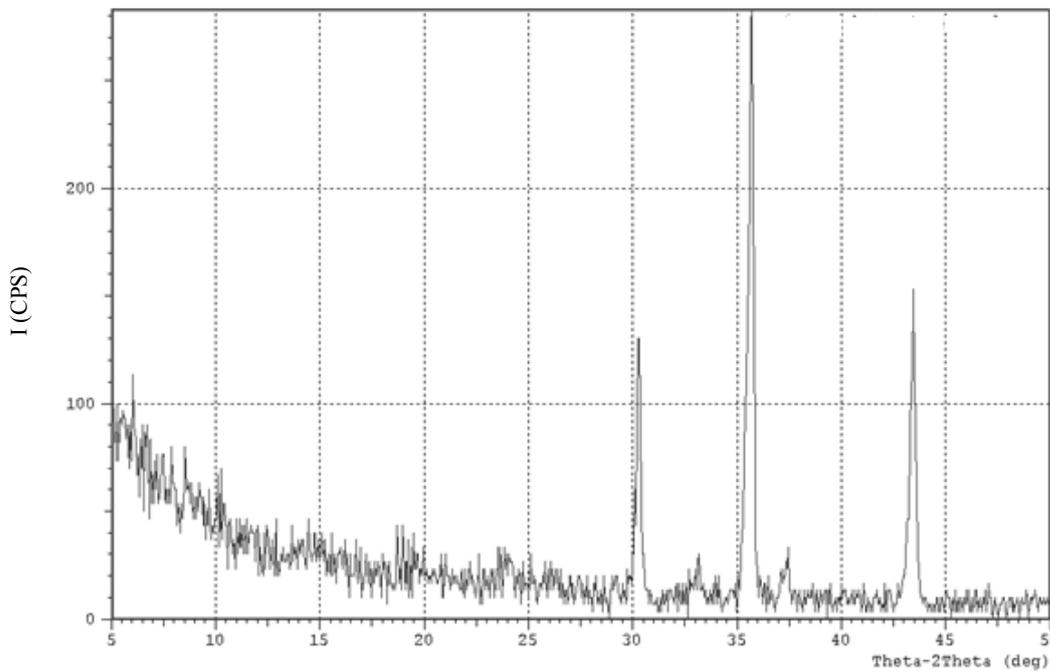


Figure (4): XRD pattern of lithium-ferrite doped with 10% cobalt-ferrite at 800 °C.

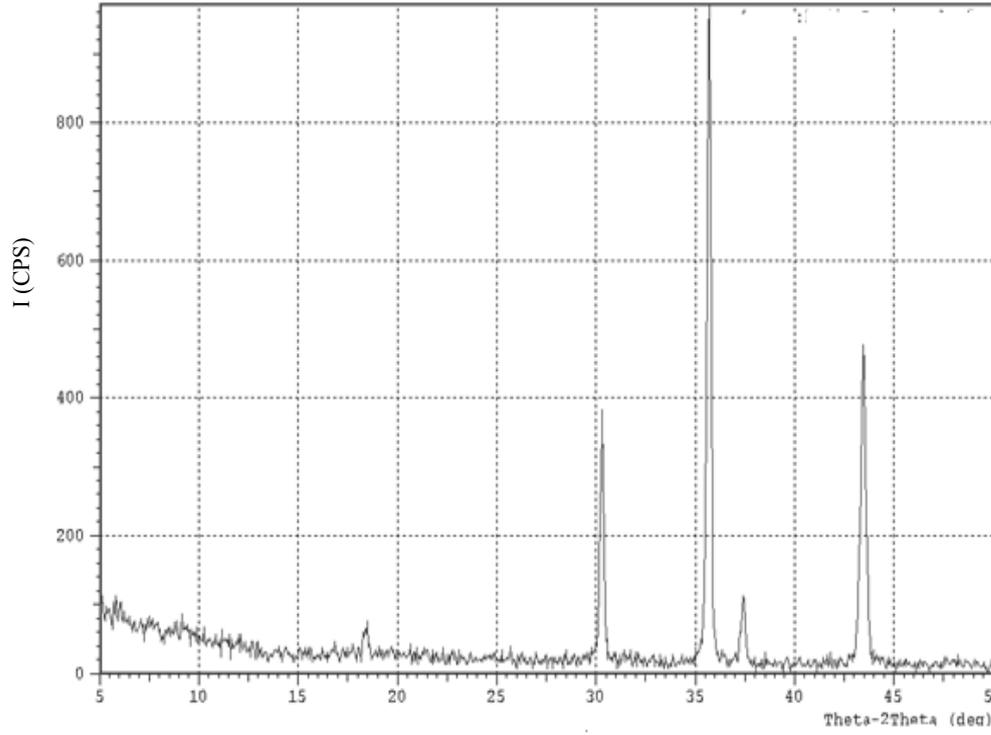
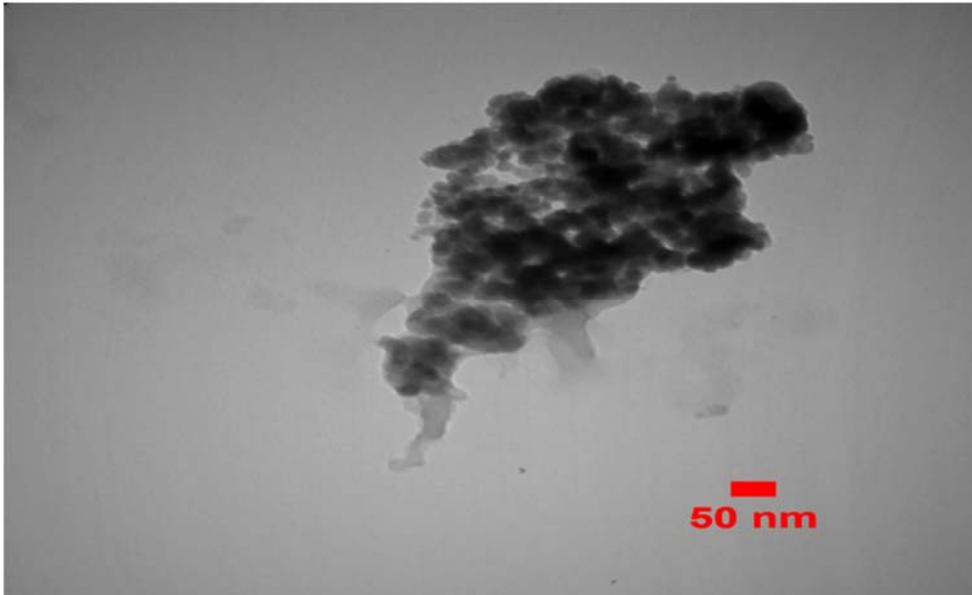
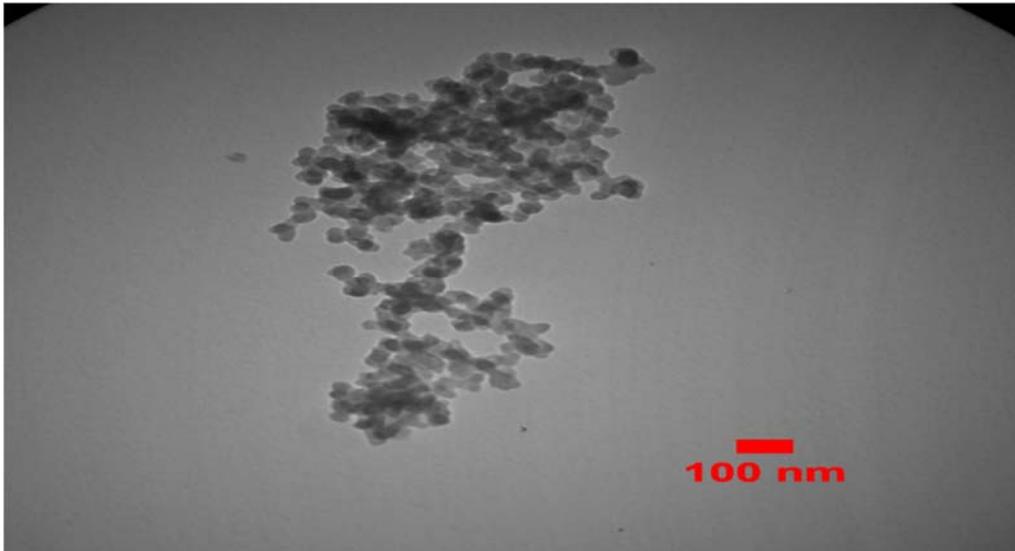


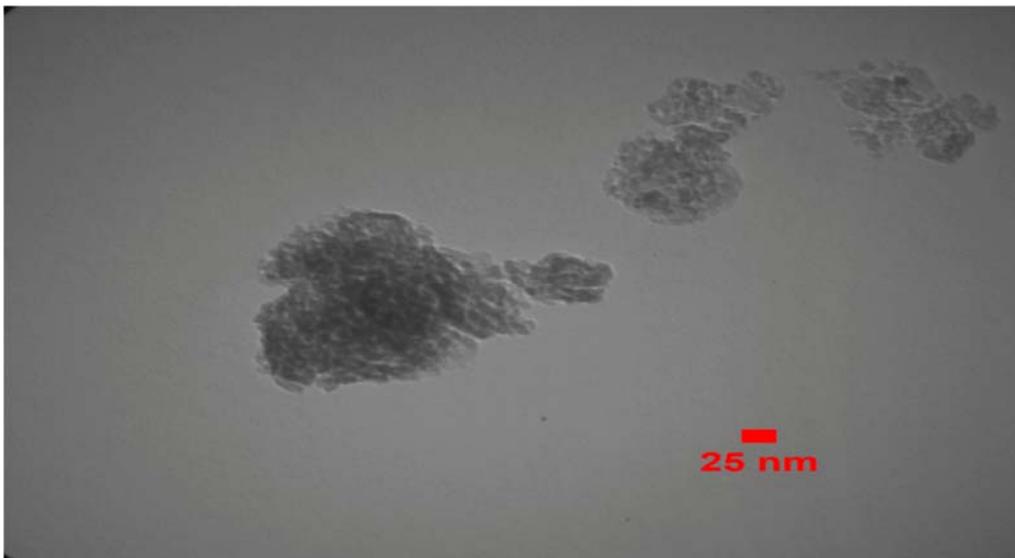
Figure (5): XRD pattern of lithium-ferrite doped with 20% cobalt-ferrite at 800°C.



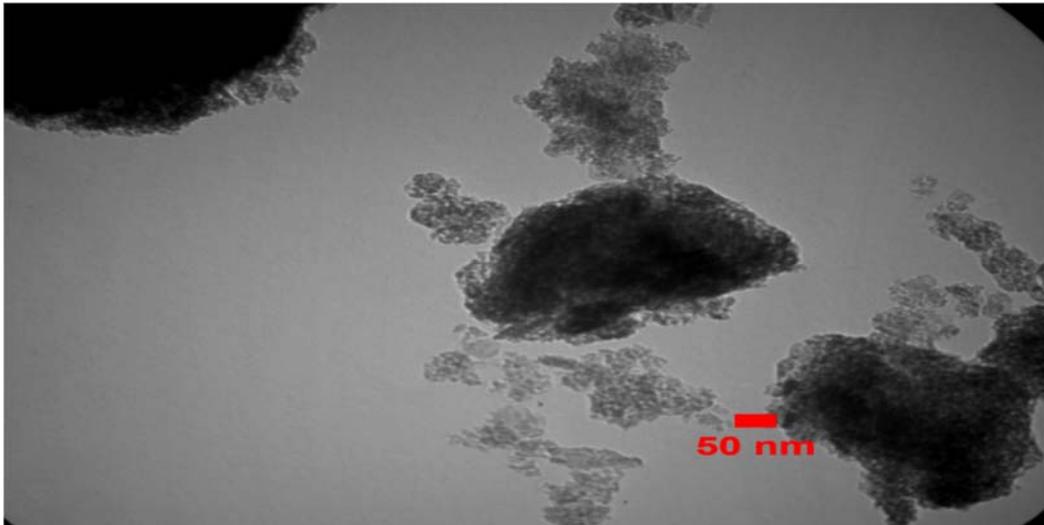
a



b



c



d

Figure (6): TEM images for prepared nano ferrite powder (10 %) after calcination at 800°C for 2 hr.

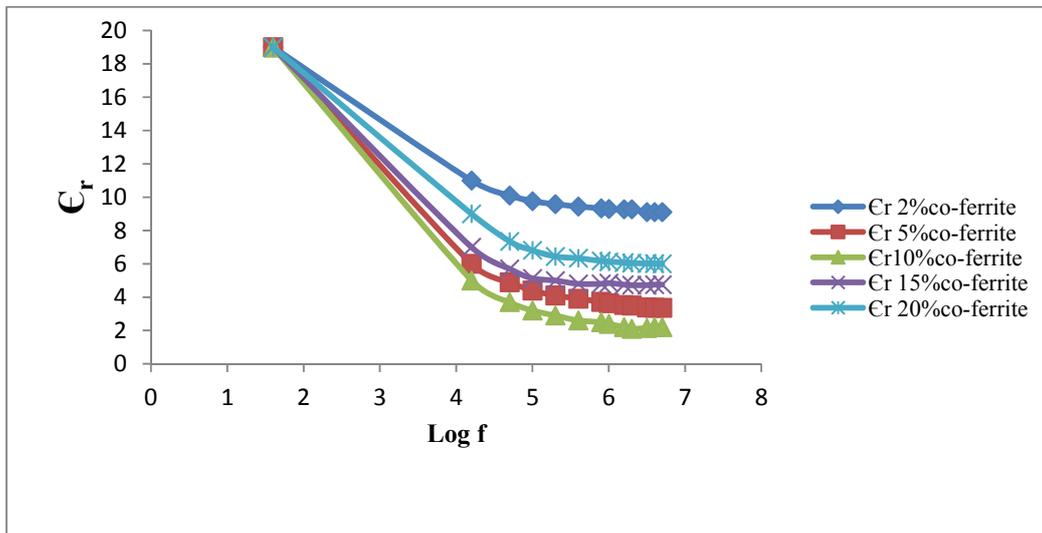
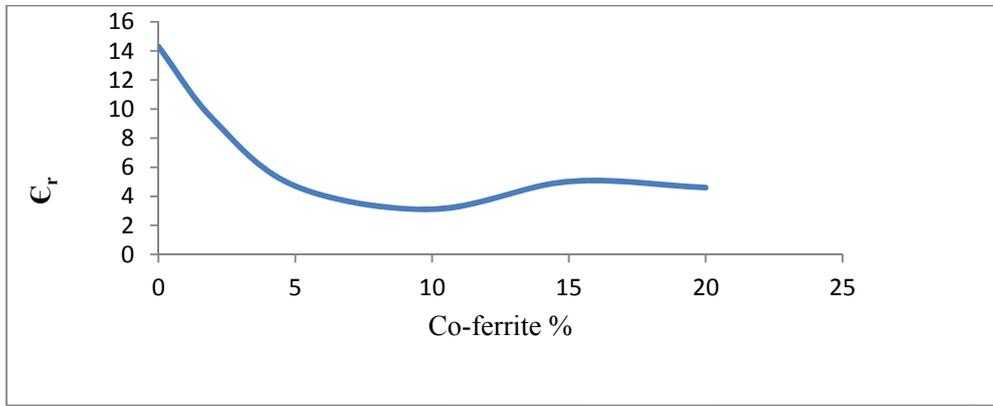


Figure (7): The relation between dielectric constant and frequency of lithium-ferrite with different percentage of cobalt-ferrite.



Figure(8): The dielectric constant of lithium-ferrite with different percentage of cobalt-ferrite at 1MHz.

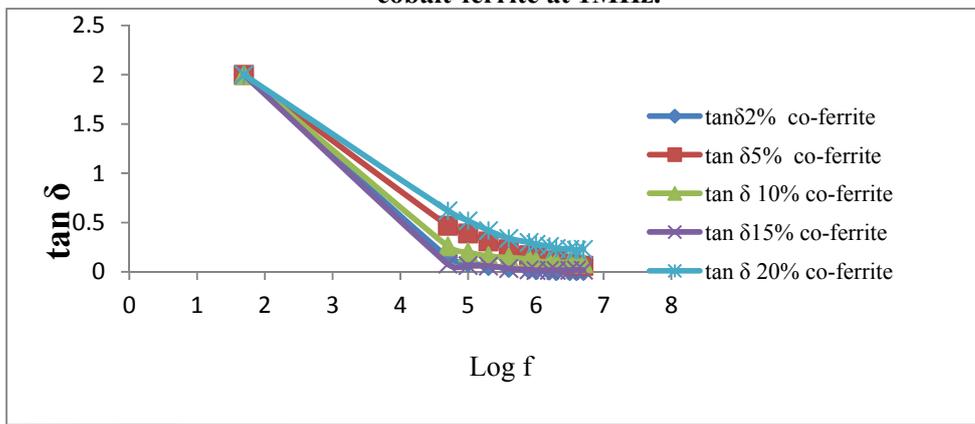


Figure (9): The relation between loss tangent ($\tan \delta$) and frequency of lithium-ferrite with different percentage of cobalt-ferrite.

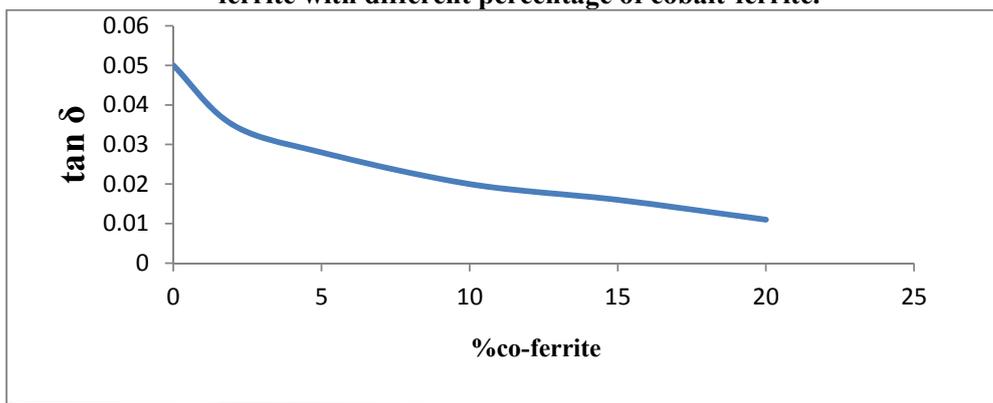


Figure (10): The loss tangent ($\tan \delta$) of lithium-ferrite with different percentage of cobalt-ferrite at 1MHz.

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