Eng. &Tech.Journal, Vol. 33,Part (A), No.1, 2015

Synthesis and studyingdielectric 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.

Received on: 6/4/2014 & Accepted on: 4/12/2014

ABSTRACT

Based on a low temperature route (sol-gel autocombustion) a nano-ferrite $CoFe_2O_4$and $Li_{0.5}Fe_{2.5}O_4$ and different percentages of cobalt-ferrite have been synthesized. The crystalline structure by XRD and dielectric properties with TEM micrographs have been presented. The XRD (crystallite size and lattice parameter) have been determined which affected with the content of 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) have been decreased with frequency and cobalt content.

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

تحضير ودراسة بعض الخواص الميكانيكية المتراكبة

الخلاصة:

استخدمت طريقة السول-جيل (الاحتراق الذاتي) لتحضير فيرايت الكوبلت والليثيوم النانوي وكذلك خلطنسب مختلفة من الكوبلت مع الليثيوم.

تم دراسة التركيب البلوري باسنخدام حيود الاشعة السينية والتركيب المجهري باستخدام المجهر الالكتروني النافذ بالاضافة الى قياس الخواص العزلية .تم حساب الحجم البلوري و ثابت الشبكية حيث تاثر 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 الفقدان العزلي) فقد تناقصت مع التردد وكذلك نسب اضافة فيرايت-الكوبلت.

https://doi.org/10.30684/etj.33.1A.2
2412-0758/University of Technology-Iraq, Baghdad, Iraq

INTRODUCTION

Historically ferrite attracted the attention of the physicists and technologies since they are 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 electricalconductivity and on the influence of various substitutions. Nano-sized spinel ferrite particles have received enormous attention during the past several years because of their 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.

Reedy[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 have been measured. Charge carriers mobility, dielectric constant and loss tangent have been studied at different frequency.

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], have been 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 x 1 at relieving low temperature.

Guilherme et al[6], have been studied the structural investigation of MFe2O3 (M= Fe, Co). Ferrites of this type have been prepared by the traditional co-precipitation method. XRD and TEM analysis have shown that the magnetic nano-particles (non-modified and modified) present diameters in the range (10-15) nm.

Naidak et al [7], have been 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 otherferrites obtained by a mixture of salts.

Arana et al [8], have been studied the Li-substituted Mn-Zn ferrite structural and magnetic properties after different thermal treatment. Spinel ferrites of composition $Zn0.6Mn0.4Fe_2O_4$ and $Li0.2Mn0.4Fe_{2.5}O_4$ were prepared by the self-combustion sol-gel method crystalline lattice increases by lithium incorporation.

Mohapatra et al [9], have been studied the surface controlled synthesis of MFe_2O_4 (Mn, Co, Ni and Zn) nano-particlesare magnetic and show 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 (Є) was calculated using the following relation:

$$Є= \frac{Cd}{Є_o A} \qquad \text{.... (1)}$$

Where

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

Dielectric loss tangent (tanδ) was calculated using the following relation:

$$\tan \delta=\frac{Є''}{Є} \qquad \text{... (2)}$$

Where

Є″ is the dielectric loss factor.

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

$$D = \beta \frac{0.9\lambda}{cos\theta_\beta} \qquad \text{...(3)}$$

Where

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

Experimental part

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

The row 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 stoichometric 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 has been added as a surfactant material with continuous mixing, then ammonium hydroxide droppedto the mixed solution until the gel bed was formed at PH 5.Addition of citric acid that lead 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 ^{o}Cfor 2 hours.

XRD, TEM have been used to evaluate the crystallite size and particle size. Dielectric parameters have been 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 percentage of cobalt-ferrite after calcined at 800 ºC 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 peak are appreciably sharpened which indicate higher crystallinity. Adding the percentages of cobalt-ferrite have shifted the peak for higher angles diffraction can also give information of the diameter of crystal.

The crystallite sizes have been calculated from 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 ºC 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 percentage of cobalt-ferrite. Generally dielectric constant decreased with increasing frequency and that may 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 decreased with frequency and the cobalt content as shown in figures (9and10).

Conclusions:

1-Mixed ferrites were successfully synthesized by sol-gel auto combustion technique**.**
2-The microstructure of ferrite prepared by the technique, consist of crystalline which 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 decreased rapidly with frequency.
5-Dielectric properties decreased as the cobalt content increased.

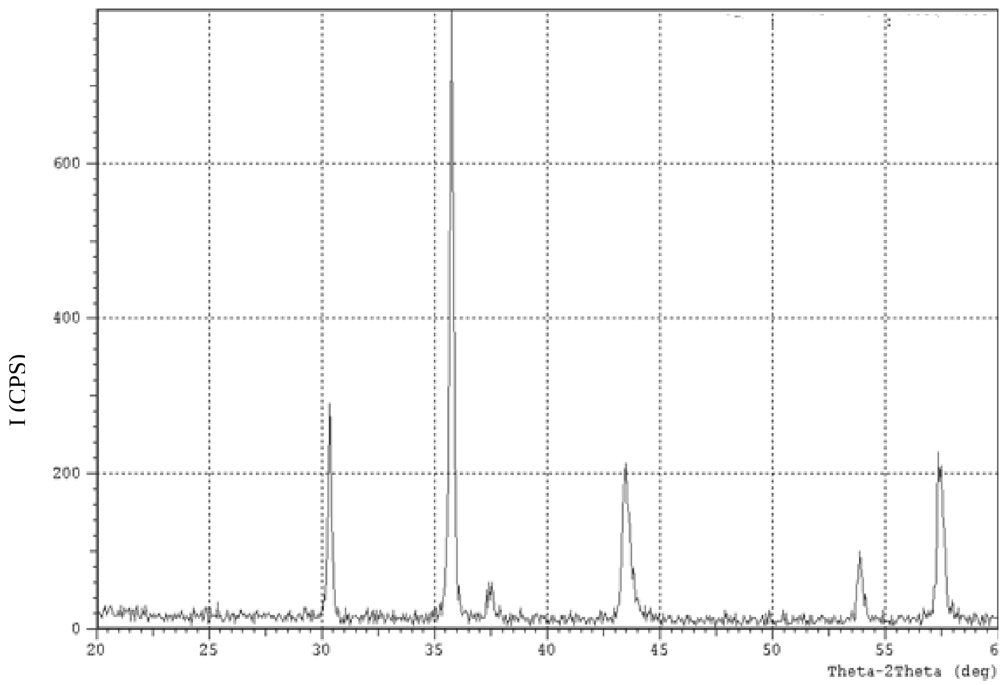


Figure (1): XRD pattern of the prepared lithium-ferrite powder at 800 ^oC.

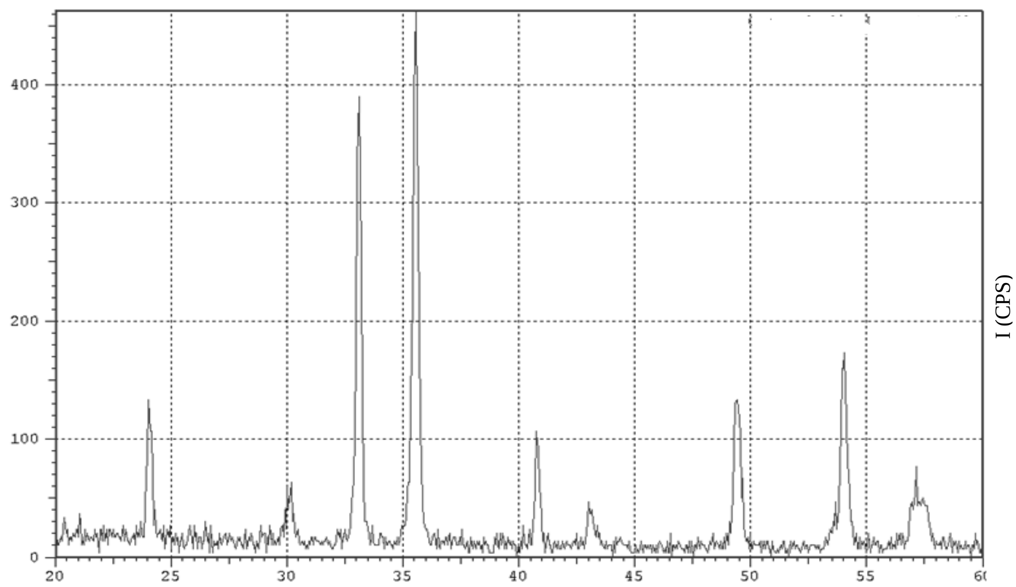


Figure (2): XRD pattern of the prepared cobalt-ferrite powder at 800 ^oC.

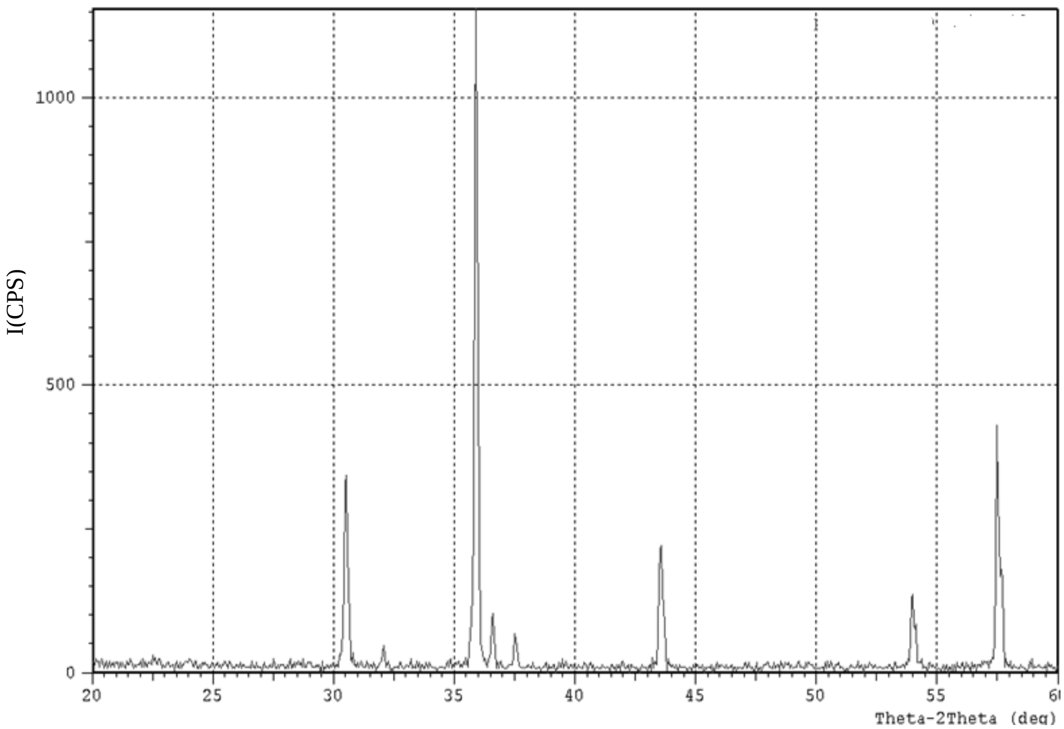


Figure (3): XRD pattern of the prepared lithium ferrite doped with 2% cobalt-ferrite at 800 °C.

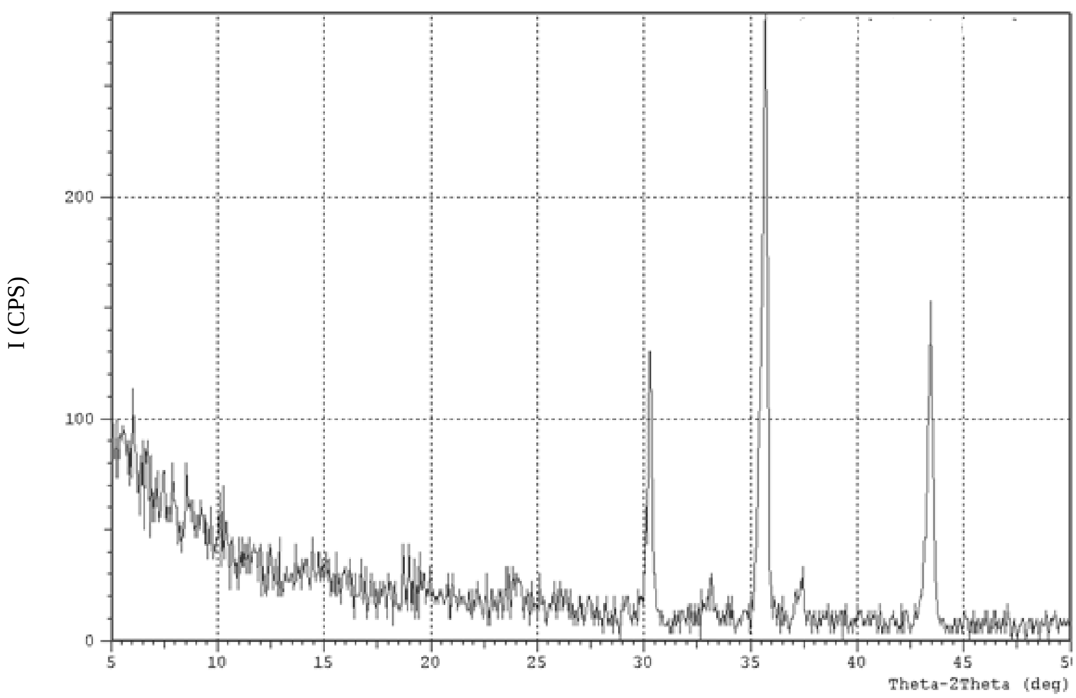


Figure (4): XRD pattern of lithium-ferrite doped with 10%cobalt-ferrite at 800 °C.

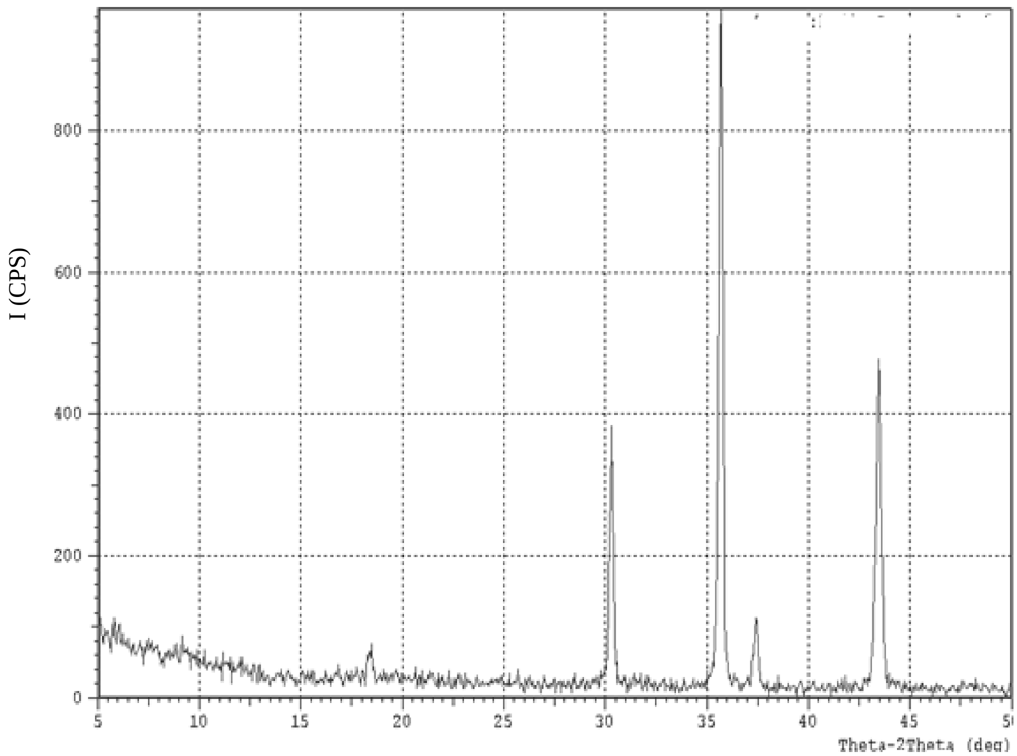
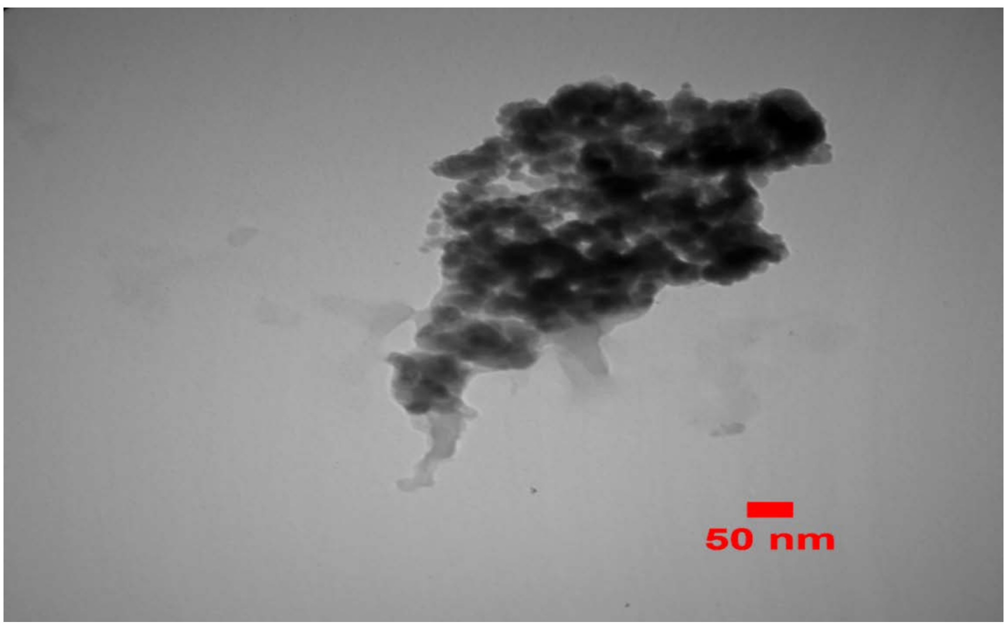
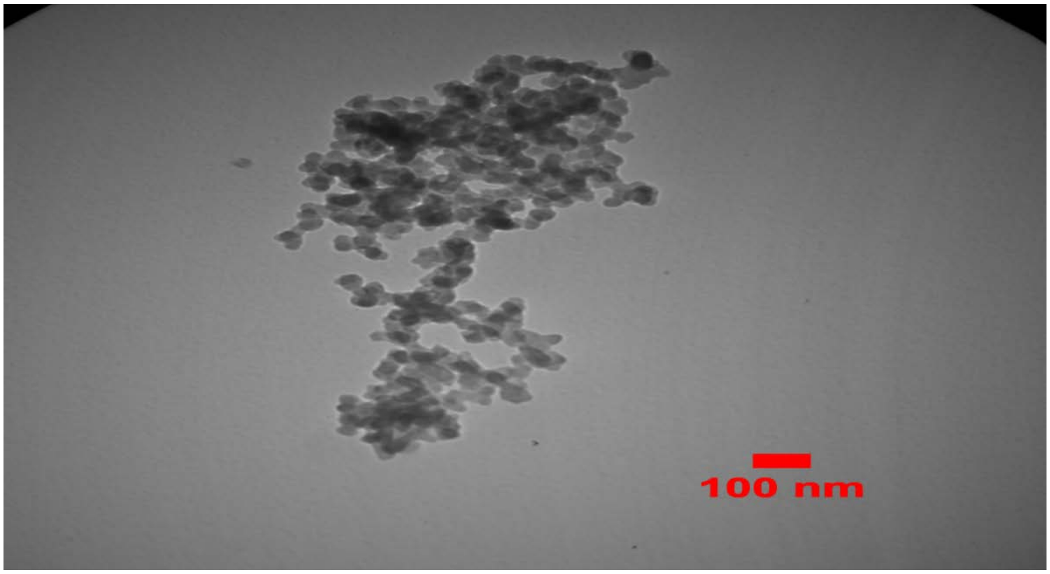


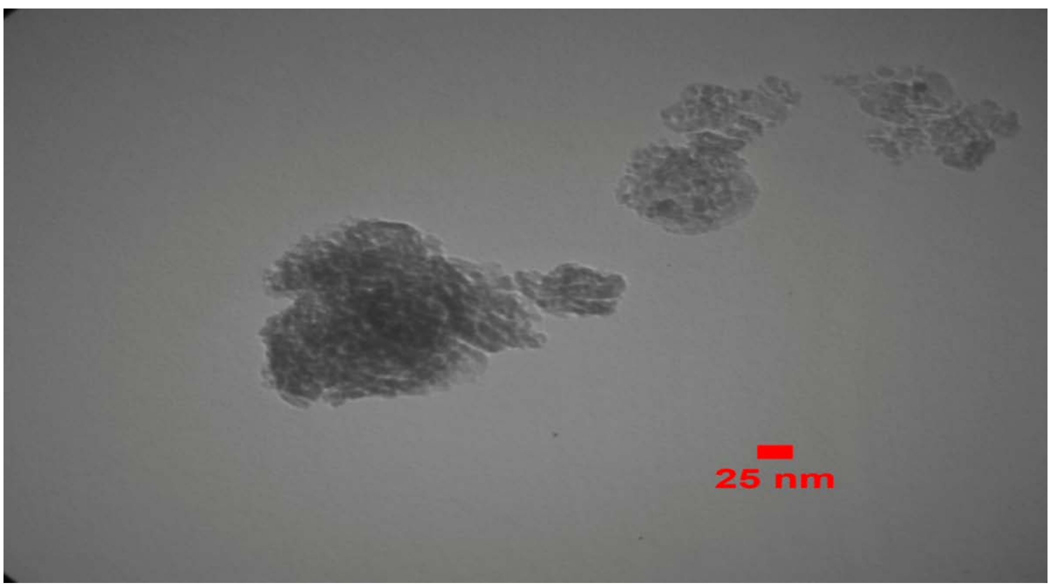
Figure (5): XRD pattern of lithium-ferrite doped with 20%cobalt-ferrite at 800^{o}C.



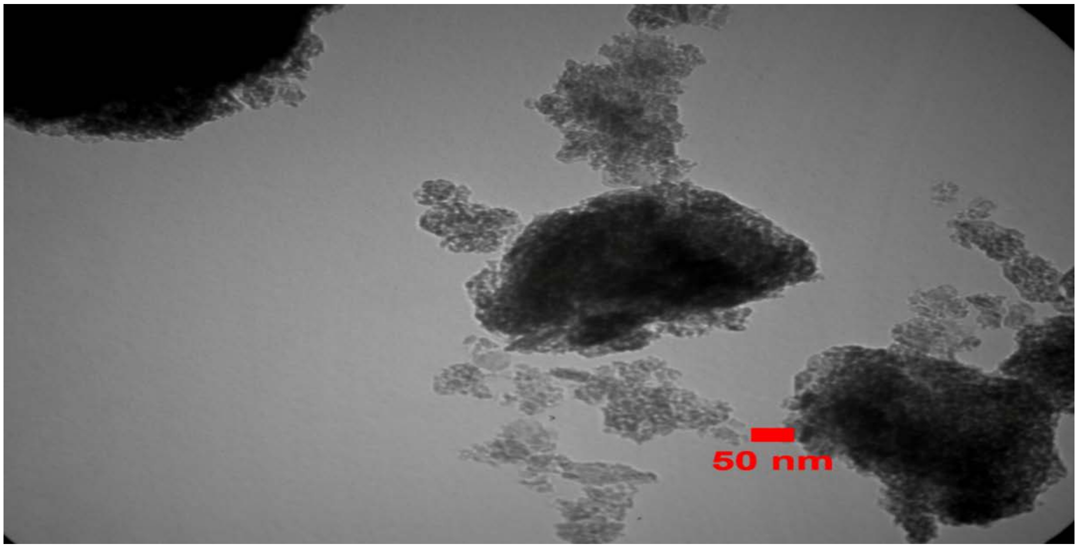
a



b



c



d

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

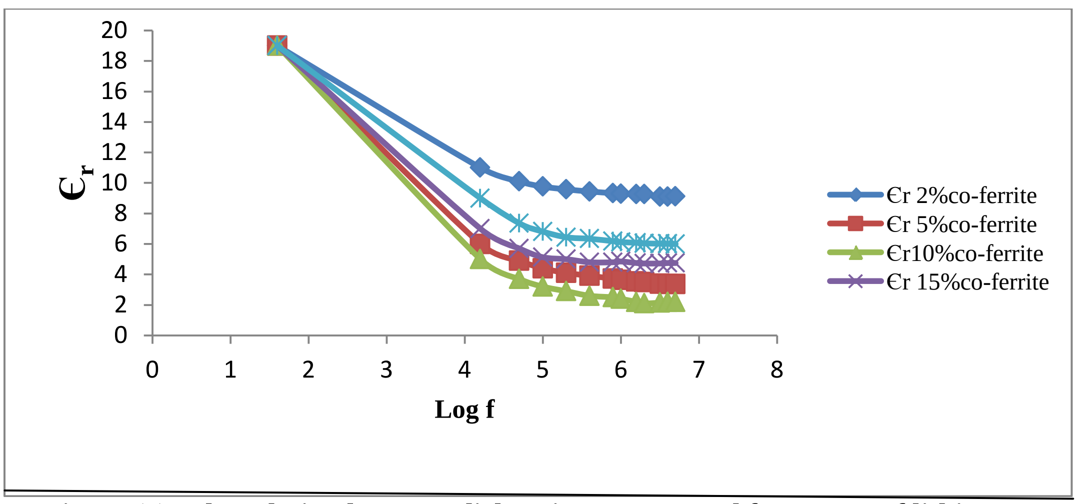
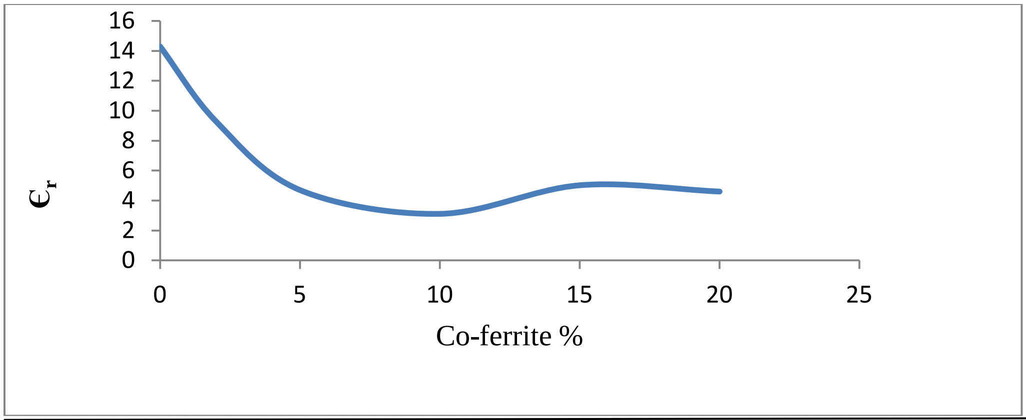


Figure (7): The relation between dielectric constant and frequency of lithium-ferrite with different percentage of cobalt-ferrite.



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

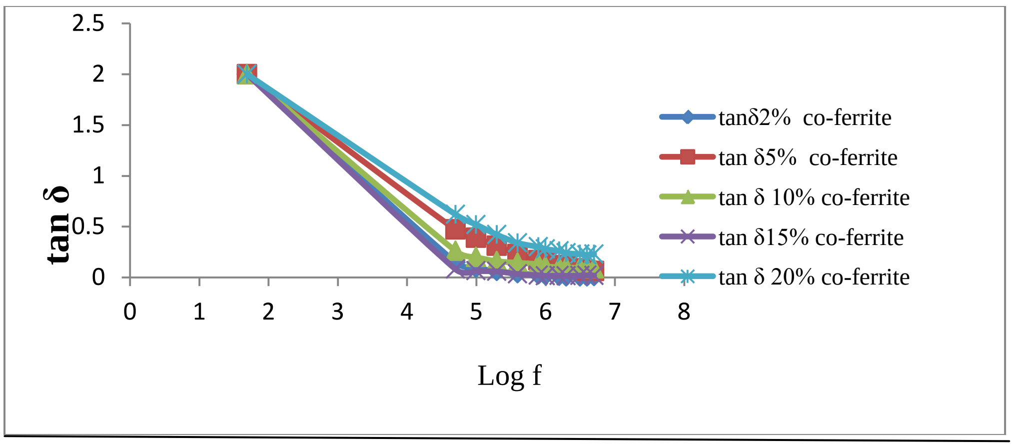


Figure (9): The relation between loss tangent (tanδ) and frequency of lithium-ferrite with different percentage of cobalt-ferrite.

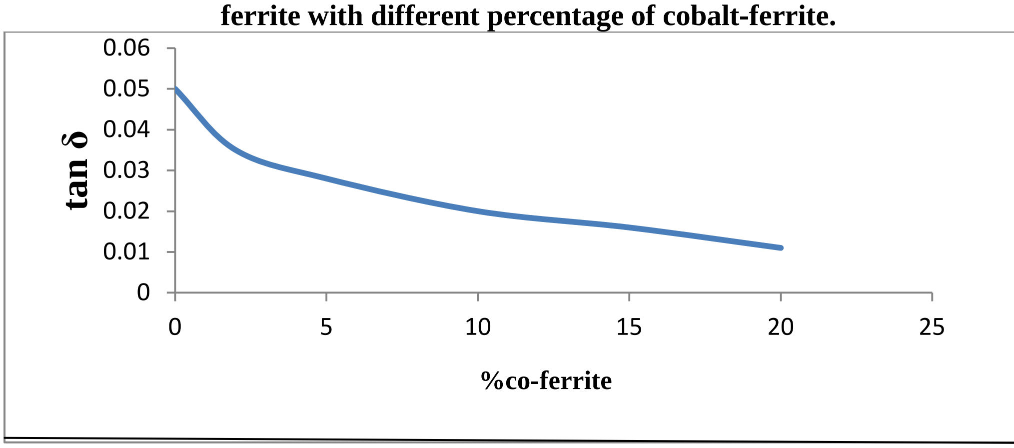


Figure (10): The loss tangent (tan δ) of lithium-ferrite with different percentage of cobalt-ferrite at 1MHz.

REFERENCE

[1].V.Kassabova-Zhetcheva, L. Pavlovol and V.Popova, “Preparation of magnetic Mg-Zn ferrite ceramic and evaluation of some important magnetic characterization”, J. of the university of chemical technology and metallurgy , Vol.44.3, pp(243-248), 2009.

[2].J. Guangbin, T. Shaolong, X. Baolong, G. Benxi, D. Youwei,” Synthesis of $CoFe_2O_4$ nanowire arrays by sol–gel template method” , Vol. 379,PP(484–489) , 2003.

[3].P.V.Reddy, “Charge transport in Li-Ni ferrospinels”, J. Apple. Phys., Vol.63, No.8, pp (3783-3785), 1988.

[4].S.Sun, L. Jianbao and G. Sun, “Effect of Li_2O_3 and Gd_2O_3 on some properties of Ni-Zn ferrite”, J. of magnetism at magnetic materials”, Vol.250, pp. (20-24), 2002.

[5].C.Wenda and H. Lengbein, “Synthesis and characterization of compounds $Li_xMn_{1-x}Fe_{2-2X}O_4$with spinel structure in the quasiternary system $LiO_{0.5}MnO_xFeO_x$”, Cryst.Res. Technd., Vol.41, No.1, pp (16-18), 2006.

[6].G.V.M.Jacintho, A.G.Brolo, P.Corio, P.A.Z.Surez and J.C. Rubim, “Structural investigation of MFe_2O_4 magnetic fluids”, J.Phys. Chem. C. , Vol. 113, pp(7684-7691), 2009.

[7].K. P. Naidek, F. Bianconi, T. C. Rocha, D. Zanchet, J. A. Bonacin, M. A. Novak , M. G. Vaz, H. Winnischofer,”Structure and morphology of spinel MFe_2O_4 (M=Fe, Co, Ni) nanoparticles chemically synthesized from heterothallic complexes”, J. Colloid Interface Science, Vol. 358,pp (39-46), 2011.

[8].M.Arana, P. G. Bercoff and S. E. Jacobo ,”Li-substituted Mn-Zn ferrite :structural and magnetic properties after different thermal treatments”, Pro. materials science, Vol.1, pp(620-627), 2012.

[9].J. Mohapatra, A. Mitra, D. Bahadur and M.Aslam, “Surface controlled synthesis of MFe_2O_4 (M= Co, Ni and Zn) nanoparticles and their magnetic characterization”, Cryst. Eng. Comm, Vol.15, pp(524-532), 2013.

[10].D. Ravmder and K.V. Jaykumar, Bull. Mater. Sco.Vol.5, No.505, 2001.

[11].B.P.Rao, K.H.Rao, K.Trinadh and O.F.Caltun, J.Optoelec. Adv. Mater. , Vol.3, pp951, 2004.