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Ferroelectric Ferromagnetic composite materials

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يَقُولُ تَعَالَى عَزَّ وَجَلَّ:

بِسْمِ اللَّهِ الرَّحْمَنِ الرَّحِيمِ

وَيَسْأَلُونَكَ عَنِ الرُّوحِ قُلِ
الرُّوحُ مِنْ أَمْرِ رَبِّي وَمَا
أُوتِيتُمْ مِنَ الْعِلْمِ إِلَّا
قَلِيلًا ﴿٨٥﴾

صدق الله العظيم

الآية

سورة الإسراء
{85}

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Ali A. H.

DEDICATION

To the three pillars of my life: my God, my father and my mother. Without you, my life would fall apart. I might not know where the life's road will take me, but

Walking with You:

My God, through this journey has given me Strength.

My Father, I would like to lift your forehead high in response your grace to me.

My Mother, if love was a human it shall be you, thanks for your faith in me.

My brothers and friends, the source of my happiness and tranquility

Thanks for inspiring. We made it...



Ali A. H.

ABSTRACT

In the present study, $\text{BaTiO}_3+\text{CuFe}_2\text{O}_4$ composite materials were synthesized using sol-gel auto combustion chemical method, in which pre-synthesized Barium Titanate was combined with the copper ferrite, using Titanium oxide, Barium nitrate, Ferric nitrate, Citric acid, Copper nitrate Tri hydrate and ammonium solution. BaTiO_3 was burned at (800°C) for 3 hours then it out the oven and mixing in strung again, put in crucible and burned at 1200°C and cooled at room temperature. CuFe_2O_4 was burned in $(200-220^\circ\text{C})$ and cooled at room temperature, then Calcine at (400°C) for three hours. This process was repeated to obtain ferrimagnetic and ferroelectric composites $[\text{xBaTiO}_3+(1-\text{x})\text{CuFe}_2\text{O}_4]$ with different weight fractions ($\text{x}= 0, 0.1, 0.2, 0.3, 0.4, 0.5, 0.6, 0.7, 0.8, 0.9, 1$). Then composite powders with different weight, burn each mix at 800°C for three hours. Then composite powder with different weight percent were milled and pressed into pellets 1.2cm, and then darkened at 950°C for three hours.

The Structural, electrical and dielectric properties of $\text{BaTiO}_3+\text{CuFe}_2\text{O}_4$ composite materials nanopowders have been studied using FTIR, XRD, AFM, SEM and LCR-meter analysis.

FTIR-spectra of $\text{BaTiO}_3+\text{CuFe}_2\text{O}_4$ composite materials powders calcined at different temperatures (950°C) where shown Fourier transform infrared model barium titanate nanoparticles prepared having packets within the scope of the (522 cm^{-1}) to (547 cm^{-1}) belonging to the association of Aoxgen- metal (titanium), it featured packages within the scope of the (459 cm^{-1}) to (584 cm^{-1}) belonging to the association of oxygen - a metal (copper), as well as packages have emerged within the range $(550-$

600 and 400-450 cm^{-1}) belonging to the association of oxygen - a metal (titanium - copper)..

The chemical phase analysis that has been used X-ray diffraction measurement device XRD confirms be copper ferrite (CuFe_2O_4 spinel powder), Barium Titanate (Barovskait) and materials overlapped between them. Where the study of X-ray diffraction showed that the crystal size increases with increasing temperature calcination powder (copper ferrite). While finding that the crystal size increases with the concentration of Barium Titanate decrease concentration (copper ferrite) and also found that the lattice constant decreases (at least) with the increase concentration Barium Titanate. The theoretical ferrite powder density decreases with increase Barium Titanate content.

From SEM micrographs, cobalt ferrite showed particle size is less than 100nm, also noted an increase in the particle shapes with increasing calcination temperatures. AFM figures proved that the particle size is very small, homogeneous and in the range of nano size.

Electric and dielectric properties were studied in all pelletized and sintered samples at temperature 950 $^{\circ}\text{C}$, and the frequency range of (50 Hz – 5MHz).

AC Resistivity of all samples (sintered at 950 $^{\circ}\text{C}$) have been measured as a function of frequency and found it decreases with increase in frequency. Dielectric constant (ϵ_r'), the loss tangent ($\tan\delta$) and the loss factor (ϵ_r'') are calculated from capacitance data, and showed that the dielectric parameters decreases with increasing frequency. This behavior is typical of ferrites as explained by Koop's model, conductivity increased with increase Barium Titanate content in $\text{BaTiO}_3+\text{CuFe}_2\text{O}_4$ composite materials, dielectric loss were change between increase and decrease with increase Barium Titanate, while conductivity increases with frequency increase.

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LIST OF SYMBOLS

Symbol	Definition	Unit
f	Voltage frequency	Hz
P	Polarization	C/m²
q	Electric charge	C
N	Number of dipoles per unit volume	1/m³
d	distance	Mm
E	Electric field	V/cm
ϵ_0	Permittivity of vacuum (8.85x10⁻¹²)	F/m
ϵ_r	Relative permittivity (dielectric constant)	---
C₀	Vacuum capacitance	F
C	Capacitance of dielectric material	F
ϵ	Permittivity of the dielectric material	F/m
X_c	Impedance of capacitor	Ω
ω	Angular frequency (2πf)	Rad.s⁻¹
ϕ	Phase angle	Degree
δ	Loss angle	Degree
I_a	Active current (resistive current)	A
I_r	Reactive current (capacitive current)	A
ϵ'_r	Real dielectric constant	---
ϵ''_r	Imaginary dielectric constant	---
tanδ	Tangent loss angle	---
Q	Quality factor	
α	Thermal Coefficient to Dielectric losses Factor	K⁻¹
T	absolute temperature	°K
UP	Unsaturated polyester	---

β	Is the broadening of diffraction line measured at half its max.	rad
D	Crystallite Size	Nm
a	Lattice constant	Å
ℓ	Thickness	M
A	Area	m ²
(hkl)	Miller indices	-

List of Abbreviations

SEM	Scanning electron microscope
XRD	X-ray diffraction
HRXRD	High resolution X-ray diffraction
TEM	Transmission electron microscope
HRTEM	High resolution Transmission electron microscope
TMAH	Tetramethylammonium hydroxide
FESEM	Field Emission Scanning electron microscope
CTAB	Hexadecyl trimethylammonium bromide
TGA	Thermo-gravimetric analysis
EDS	Energy dispersive spectrometry
MPS	Methacryloxy propyltrimethoxy silane
FTIR	Fourier Transform Infrared
FWHM	Full Width at Half Maximum
T_N	Neel Temperature
T_C	Curie Temperature
ICDD	International Centre for Diffraction Data
JCPDS	Joint Committee on Powder Diffraction Standards
LCR	Inductance(L), Capacitance(C), and Resistance(R)

1.1 Introduction

The science of nanotechnology is the study of the phenomena and processed materials in terms of the balance of atomic, molecular and molecules, these characteristics are significantly different from those on a large scale in the advantages [1]. The reason that makes sense to regulate nanotechnology as a separate category is that materials, nanotechnology work differently from bulk materials [2], as the basis of nanotechnology depended on the rearrangement of atoms to make new molecules with new specifications and planned. Nano-scale material appears properties that differ from the properties in micron-sized bulky due to the large surface area [3]. When the specific surface area increases, rates of interaction of particles increase. This means that the inverse proportionality between particle size and specific surface area. Nano-size materials particle be few compensation atoms compared to bulk particles and this leads easily to link the materials. Nano-sized materials possess little surface stability and average highest binding energy per atom because of low consistency [4, 5]. The goal of knowing the nanotechnology and how to be entered in the field of manufacturing including devices and systems with accurate sizes and compositions for generating important functions and characteristics of new and better results shows us this technical feature [6]. There are many applications for nanotechnology and we can set then at figure (1-1). Recently, many applications depended on magnetism and magnetic materials, [7] and one of the materials has electrical and magnetic properties are ferrites. Ferrites are polycrystalline magnetic oxides that can be described by the general chemical formula " $XO \cdot Fe_2O_3$ " in which X is a divalent ion such as Co^{2+} or Mn^{2+} [8]. In new fields, many applications found for ferrites in nanocrystalline form like magnetically guided drug

delivery, magnetic resonance imaging (MRI), catalyst, humidity and gas sensors. [9-11]

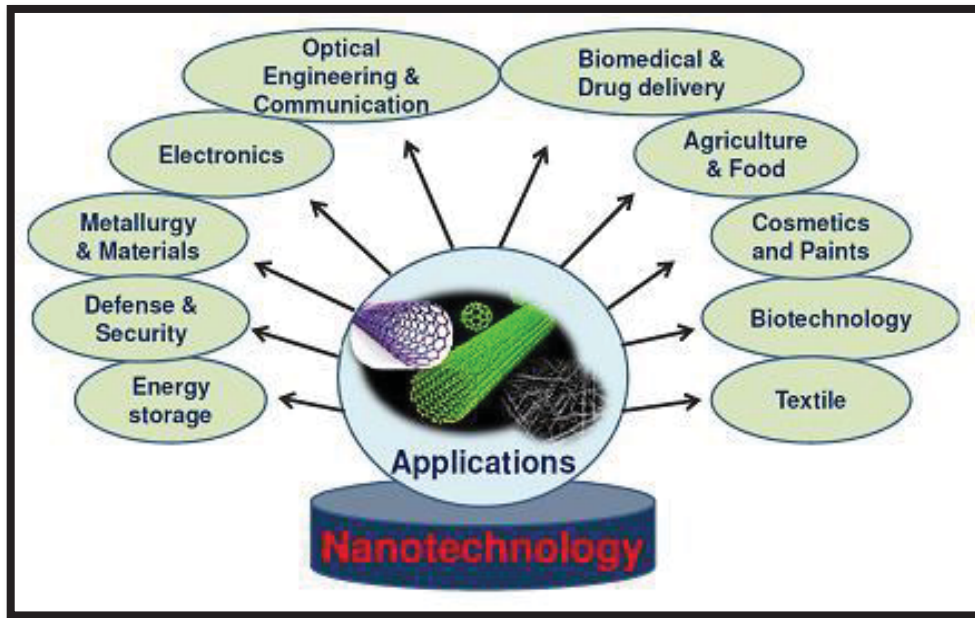


Fig (1-1): Applications of nanotechnology

One of the important categories of types of ferrite is copper ferrite which is a good magnetization, coercive force is high contrast and high magnetic field (HA) as well as premium chemical constancy and also corrosion resistance [12]. copper ferrite powder with a tight particle size distribution is to be a benefit of the media as the registration of high density and lack of size because it is usually desirable to dramatically increase the absorption of information storage and limit the issuance of the medium fuss [13, 14]. The copper ferrite is an inverse spinel with the general formula CuFe_2O_4 , where the Cu^{+2} commute some of the iron cations in the structure [15, 16]. Its structure is like the magnetite, so far with partly various of chemical and physical properties due to the existence of copper. The metal was found in an uncovered ore dump, on the monarchy of Consolidated Rambler Mines confined near Baie Verte, New found land [17, 18]. It can

be used in a large number of applications like gas sensing, catalytic, Li-ion batteries, high density magnetoelectric register devices, colour imaging, bioprocessing, magnetic refrigeration and ferrofluids [19-24]. Moreover, CuFe_2O_4 conduct it is a major significance due to high thermal stability, high electric conductivity and high catalytic efficiency for O_2 progression from alumina–cryolite system used for aluminum fabrication [25]. Barium titanate is a ferroelectric oxide is subject a transition from a ferroelectric tetragonal phase to a paraelectric cubic phase when heating above 120°C due to its low loss and high dielectric constant characteristics. It has been used in applications, like capacitors, multilayer capacitors doped barium titanate has been found a wide application in PTC theorist and piezoelectric devices semiconductors, semiconductors, and has become one used of the most important ferroelectric ceramics [26,27]. There is a new class of physical properties of composite materials called "product Properties". After the notion of productive property as submitted from before Van Suchtelen, a proper combination of magnetostrictive material, piezoelectric material can allow altitude to the magneto-electric effect. The composites exhibiting a magneto-electric impact are described as magneto-electric composite. Its impact in such composites leads to the strain induced at the ferrite juncture being mechanical connected to a stress induce in the ferroelectric juncture. Conjugation results in an electrified voltage [28,29]. Ferrites are mixed metal oxides with iron(III) oxide as main component. Ferrites crystal in three crystal types: spinel (cubic crystal structure where $\text{Me}^{1/4}\text{Fe}$, Mn, Mg, Ni, Zn, Cd, Co, Cu, Al or a mixture of these), garnet (cubic crystal structure where $\text{Me}^{1/4}\text{Y}$, Sm, EU, Gd, Th, Dy, Ho, Er, Tm.or Lu) and magnetoplumbite type (hexagonal crystal structure where $\text{Me}^{1/4}\text{Be}$ or Sr) [30]. Preparations and properties of a bulk composite such as $\text{BaTiO}_3\text{--Ni}_{0.5}\text{Zn}_{0.5}\text{Fe}_2\text{O}_4$, $\text{PLZT--Ni}_{0.5}\text{Zn}_{0.5}\text{Fe}_2\text{O}_4$, $\text{BaTiO}_3\text{--CoFe}_2\text{O}_4$, $x\text{NiFe}_2\text{O}_4\text{--}(1-$

x)BaTiO₃ have been recently reported [30,31]. The main problem in preparing of these composite materials is possible reaction at the interfaces between the ferroelectric and magnetic phases during sintering. Therefore, the optimization of sintering process should be performed in order to obtain di-phase composite material of desired composition. The preparation method and properties of composite materials containing barium titanate (BT) as a ferroelectric phase and Copper ferrite (CF) as a magnetic phase is presented in this work.

As a matter of fact, there have been a few attempts made to overcome this obstacle, such as spark plasma sintering technique,[32]. FM/FE core-shell precursors and powder in-sol wet chemistry method. [32,33]. They are either expensive or relatively complicate. More importantly, for Pb-free FE/FM composites (e.g., BaTiO₃ based composites) those often require high calcination temperature and thus possess high inter-phase reactive activity, their effects are so far not as good as those observed in Pb(ZrTi)O₃ based composites[32-35]

1.2 Literature review

A composite made from two or more constituent materials with a difference of chemical properties or physical when collecting articles and so material it produces different properties of individual components as the individual substances shall be separate and distinct within the final composition and be a favorite for many reasons, and advantage materials are stronger and lighter and also less expensive compared to traditional point of view[36]. Many of the earlier researchers may have collected articles for various compounds as the following :

In 2000, MAHAJAN et al [37], studied dielectric behavior, conductivity and magneto-electric effect in copper ferrite–barium titanate composites. Composites of CuFe_2O_4 and BaTiO_3 were prepared using a conventional ceramic double sintering process. The presence of both phases was confirmed by X-ray diffraction. The variations of resistivity and (thermo emf) with temperature in these samples were studied. All the composites showed (n-type) behavior. The variation of dielectric constant (ϵ'') in the frequency range was between 100 Hz to 1 MHz and with the temperature at constant frequency were studied. The conduction phenomenon was explained on the basis of a small Polaron mobility model. Also confirmation of this phenomenon was made with the help of A.C conductivity measurements. The static value of the magneto-electric conversion factor, i.e. d.c. $(\text{ME})_H$ was studied as a function of intensity of the magnetic field. The maximum value of the ME coefficient has been observed in 75% phase Ferroelectric compound.

In 2002, MAHAJAN et al [38], studied dielectric behavior and magnetoelectric effect in cobalt ferrite CoFe_2O_4 – BaTiO_3 composites and their electrical properties. Cobalt Ferrite(CF) combining with Barium Titanate(BT) composites were prepared using(conventional ceramic double sintering methods) with various compositions by using measurements (XRD,L.C.R meter). It was confirmed that presence of two phases in composite materials using(X-ray diffraction). The D.C resistivity, A.C conductivity, dielectric constant (ϵ'') and loss tangent $\tan(\delta)$ and thermo emf as a function of temperature in the temperature range 300 K to 600 K were measured. Where the frequency was between ranged 100 Hz to 1 MHz and also with the temperature at a constant frequency of 1 KHZ has been studied. It was discussed the nature of conductivity on the

basis of a small Polaron mobility model. The study has a fixed value of the magneto-electric conversion factor as a function of magnetic field.

In 2003, Arya et al. [39], studied dielectric behavior properties of nanometer sized(Barium Strontium Titanates(BST)) prepared by(the polymerization citrate precursor method). Since these oxides were found with a cubic structure, which is retained until after heating at 800 °C. Particle size is almost nanoscale reasonably stable. They found that the dielectric constant of sintered of these oxides decreases from 510 for BaTiO₃(BT) to 190 for SrTiO₃(ST) at 100 KHz.

In 2004, Xiwei Qi et al [40], studied dielectric behavior, magnetic and electric properties of ceramic ferroelectric–ferromagnetic composite(xPMZNT·(1 –x) NiCuZn), in which x varies as (0, 0.1, 0.2, 0.4, 0.6, 0.9, and 1.0) using a standard ceramic technique.

In 2005, J. G. Wan et al [41], studied structure,magnetic and electric properties of Magneto-electric CoFe₂O₄–Pb(Zr,Ti)O₃ composite thin films .The Magnetolectric (ME) CoFe₂O₄–Pb(Ti, Zr)O₃ composite thin films have been prepared by a spin-coating technique and sol-gel methods and spin-coating technique . The use of X-ray diffraction and scanning electron microscopy was noted that there is a gathering of atoms and the separation of the phases of the Pb(Zr,Ti)O₃ and CoFe₂O₄ phases in the thin films. Ferroelectric test unit, Vibrating sample magnetometer , and magnetolectric measuring device were used to describe the ferroelectric and magnetic properties, likewise the ME effect of the films. It turns out that each of the films exhibit magnetic properties of ferroelectric good, as well as the impact of ME. It is noted the initial presence of magnetolectric high voltage coefficient of the film.

In (2007) , Cui et al [42], prepared Nb-doped BaTiO₃ nanocrystalline powders and ceramics by a simple sol–gel process, using H₃[Nb(O₂)₄] as a precursor. The powders and ceramics were characterized by XRD, SEM and TEM, while dielectric properties of the ceramics were also determined. The results indicated that the powders synthesized by sol–gel process were in nanometer scale, which were mainly composed of cubic BaTiO₃ with small amount of BaCO₃.

In 2007, A. KUMAR RAY [43], studied the synthesis and characterization of BaTiO₃ powder prepared by combustion synthesis process. This study described a simple low temperature combustion synthesis method of barium titanate powders. XRD diffraction pattern of BaTiO₃ calcined in air at (600,700,800,900°C), the peaks become sharpened and the phase pure BaTiO₃ is found at 900°C through citrate precursor method. Further, the oxidation of citrate precursor by HNO₃ was accompanied by the evolution of CO₂ , NO₂ and water vapour and the gas evolution helped the product to result in a fine-grained structure. Citric acid and HNO₃ presented in the solution play the key role for the synthesis of shaped barium titanate at a low temperature. The average particle size of BaTiO₃ at different temperatures was calculated using Scherrer's formula and it was found to be around 24nm. Scanning electron micrograph of this powder considerable amount of a nanorod formed.

In (2008), Ramajo et al. [44], synthesized and studied BaTiO₃ powder from the Pechini method. The synthesis of BaTiO₃ starts at 150°C by the thermal dehydration of organic precursors. The usual inevitable formation of barium carbonate during the thermal decomposition of the precursor could be retarded at lower calcination temperatures and optimized heating rates. The organic precursors were treated at temperatures between 200 and 400°C. Samples were then calcined at 700 and 800 °C for 4 and 2 h,

respectively. The resulted ceramic powders were characterized by gravimetric and differential thermal analyses, X-ray powder diffraction and infrared spectroscopy. It was found that depending on the heating rate and final temperature of the thermal treatment, high amounts of BaCO₃ and TiO₂ could be present due to the higher concentration of organics in the final calcination step.

In 2008 Habib et al. [45], synthesized and studied barium titanate (BaTiO₃) nanopowder using two TiO₂ powder precursors with different particle sizes and barium hydroxide via hydrothermal route. They studied the effect of temperature, time and particle size on barium titanate using transmission electron microscopy (TEM), scanning electron microscopy (SEM) and X-ray diffraction (XRD) techniques. TEM observation of low reaction temperature samples (60°C) supports in situ transformation or short range dissolution precipitation reaction mechanism. The fine grained TiO₂ (~25 nm) precursor reacted faster than coarse grained TiO₂ (~110–125 nm) precursor. The phase of the obtained BaTiO₃ in all samples was found to be cubic.

In 2008, K. Kamishima et al [46], studied the simple process synthesis of BaTiO₃–(Ni,Zn,Cu)Fe₂O₄ Ceramic Composite. Ceramic composites (2Ni 0.41 Zn 0.41 Cu 0.18 Fe₂O₄ –BaTiO₃) were successfully prepared by a direct solid-state reaction of raw materials (BaCO₃, CuO, α-Fe₂O₃, NiO, TiO₂, and ZnO). The X-ray diffraction (XRD) and electron probe micro analysis (EPMA) measurements were performed on these samples and it is confirmed that the composites consist of spinel ferrite and BaTiO₃ phases. The composites are so homogeneous that the ferrite and BaTiO₃ grains do not react with each other and have radius in the range of 1–5 μm. Hexagonal BaTiO₃ (h-BaTiO₃) can be made in this composite form with a sintering temperature of 1200 °C, although h-BaTiO₃ can be usually synthesized above 1460 °C. The freezing-point depression of

BaTiO₃ takes place due to the mixing with the spinel ferrite, which may result in the formation of h-BaTiO₃ at a low temperature of 1200 °C.

In 2010, Ramana et al [47], studied and prepared the ferromagnetic-dielectric Ni_{0.5}Zn_{0.5}Fe_{1.9}O_{4-δ}/PbZr_{0.52}Ti_{0.48}O₃ particulate composites electric, magnetic, mechanical, and electromagnetic properties. Novel ferromagnetic-dielectric particulate composites of Ni_{0.5}Zn_{0.5}Fe_{1.95}O_{4-δ} (NZF) and PbZr_{0.52}Ti_{0.48}O₃ (PZT) were by using the conventional ceramic method. The presence of two phases in composites was confirmed by XRD technique. The variations of dielectric constant with frequency in the range of 100 kHz–1 MHz at room temperature and also with temperature at three different frequencies (50 kHz, 100 kHz, and 500 kHz) were studied. Detailed studies on the dielectric properties were done to confirm the magnetoelectric interaction between the constituent phases may that be resulted in various anomalies in the dielectric behaviour of the composites. It is proposed that interfaces play an important role in the dielectric properties, causing space charge effects and Maxwell-Wagner relaxation, particularly at low frequencies and high temperatures.

In 2011, Khamkongkao et al [48], studied frequency-dependent magnetoelectricity of CoFe₂O₄-BaTiO₃ particulate composites. CoFe₂O₄-BaTiO₃ particulate composites were prepared by wet ball milling method, and their magnetoelectric (ME) effect was studied as a function of their constituents and modulation frequency. The results show that the ME coefficient increases as a function of modulation frequency from 400 to 1000 Hz and the ME characteristics of ME curves are also modified because the electrical conductivity of the CoFe₂O₄ phase is sensitive to the increase in frequency between 400 and 1000 Hz. The third phase Ba₂Fe₂O₅ formed during the sintering tends to reduce the ME effect.

In 2012, Bhuiyan et al [49], the Synthesis and Characterization of Barium Titanate (BaTiO_3) Nano particle. Barium titanate (BaTiO_3) nanoparticles were synthesized via an electrochemical route from Ti metal plate at room temperature. Structural, compositional and optical properties were characterized by XRD, SEM, EDX, FTIR, UV-Vis and photoluminescence (PL) spectroscopy. The X-ray diffraction (XRD) confirmed the preferential growth of BaTiO_3 nano particles that width is ~ 15 nm in the (110) orientation. The SEM image shows the synthesized BaTiO_3 were nanowires in shape. The EDX measurements confirm that the composition of the samples was Ba, Ti and O elements. UV – Vis Spectroscopy shows absorption peak at ~ 330 nm. PL measurements reveal an intense and broad band at around the green colour emission region.

In 2012, Joshi et al [50], studied the synthesis and dielectric behavior of nano-scale Barium Titanate. In this study, an effort has been made to synthesize nano-scale barium titanate powder by sol-gel and hydrothermal methods. Characterization of the synthesized Barium Titanate is conducted by using X-ray diffraction for crystallite size, Transmission Electron Microscopy for particle size and scanning electron microscopy for surface morphology. It is observed that the powders prepared by Sol-gel and hydrothermal routes have almost similar average crystallite size of 34 ± 2 nm. Electrical properties such as dielectric constant, dielectric dissipation factor and electrical resistivity have also been measured. Hydrothermal process has enabled the synthesis of material with higher dielectric constant of 4000 compared to the Sol-gel route value of 1600. The dielectric dissipation factor is measured and found to be less 0.3 than a wide range of frequencies for powders generated using both processes.

In 2013, Bochenek et al [51], studied the Ferroelectric–Ferromagnetic composites based on PZT type powder and ferrite Powder. A ferroelectric-

ferromagnetic composites based on PZT powder have been obtained in presented work. The main aim of combination of ferroelectric and magnetic powders was to obtain material showing both electric and magnetic properties. Ferroelectric ceramic powder (in amount of 90%) was based on the doped PZT type solid solution while magnetic component of the composite was nickel-zinc ferrite $Ni_{1-x} Zn_x Fe_2 O_4$ (in amount of 10%). The synthesis of components of ferroelectric-ferromagnetic composite was performed using the solid phase sintering. Final densification of synthesized powder has been done using free sintering. For obtained of ferroelectric-ferromagnetic composites the XRD, the microstructure, EDS, dielectric, magnetic, internal friction and electrical hysteresis loop investigations were performed. Obtained results showed the correlations between the magnetic subsystem and the electrical subsystem of the ferroelectric-ferromagnetic composites. Such properties of obtained composites give the possibility to use them in memory applications of new type.

In 2014, Haffer et al [52], studied the synthesis notion of a nanostructured of $BaTiO_3/ CoFe_2O_4$ composite across multiferroics. The mixture of a periodically ordered, nanostructured composite consisting of $BaTiO_3$ (BT) and $CoFe_2O_4$ (CF) was presented. In a first step, mesoporous $CoFe_2O_4$ (CF) was prepared by the structure, replication method (Nanocasting) using [Mesoporous KIT-6 silica as a structural fungus]. Subsequently, BT was created inside the pores of CF by the citrate route, give rise to in a well - ordered composite material of jointly phases. The two components are known for their distinct Ferroic properties, namely Ferrimagnetism (CF) and Ferroelectricity (BF), on the ranking. Therefore, this proof of synthesis concept offers new Point of views in the invention of composite materials with Multiferroic properties.

In 2015, Alexander et al[53], studied magnetic and crystal properties of multiferroic composites $[(x)\text{MFe}_2\text{O}_4 + (1-x)\text{BaTiO}_3]$, where $(\text{M} = \text{Ni}, \text{Co})$. Multiferroic composites of $[(\text{BaTiO}_3)_{1-x} + (\text{CoFe}_2\text{O}_4)_x]$ with $x = 0.2$, and $0.4[(1-x)\text{BaTiO}_3 + (x)\text{NiFe}_2\text{O}_4]$ with $x = 0.2, 0.3$ and 0.4 and $[(\text{CoFe}_2\text{O}_4)_x + (\text{BaTiO}_3)_{1-x}]$ with $x = 0.2$, and 0.4 $[(x)\text{NiFe}_2\text{O}_4 + (1-x)\text{BaTiO}_3]$ with $x = 0.2, 0.3$ and 0.4 have been synthesized by mixing CoFe_2O_4 (NiFe_2O_4) spinel and BaTiO_3 piezoelectric. Distribution of Co (Ni) ions on 8a and 16d positions of the spinel lattice (space group $Fd\bar{3}m$) is determined by (neutron powder diffraction). The Magnetic structure of wave vector of the spinel structure is $(k = 0)$. The dielectric permittivity of the composites was measured in the frequency range (102 – 105 Hz). Where the dielectric permittivity decreased with low frequencies from(~ 940 for $x = 0.2$ to ~ 360 for 0.4).

1.3 Applications of ME composites

The device, the electric composite is classified into three types based on the magneto:

- 1) The device using ferroelectric and magnetic properties separately.
- 2) The device that employing the magnetic and ferroelectric properties, but without any ME interaction.
- 3) The device whose action is based on ME effect The Faraday phase invertors operating in the microwave region, Also the reversing optical modulators and the optical processors came under the devices of the second type and the first type, which is not considered significant in the present text. The third type of devices is based on the ME effect. It is the tool for the conversion of energy from magnetic to the electric form. The gyrator was proposed by Tellegen [54] using the ME materials, as well as Austin suggested some species of an applications of ME material, Respectively, Optical diodes, Amplification, ME data storage and

switching, Spin wave generation, Modulation of amplitudes, polarizations and phases of optical waves and Frequency conversion [54].

1.4 Objective of the reseach

1- Preparing BaTiO₃, CuFe₂O₄ nanoparticle and composite [(BaTiO₃)_x+ (CuFe₂O₄)_{1-x}] with different composition where, x takes a value (0.1 to 0.9) by using sol-gel auto-combustion method.

2- Studying the structural properties using XRD, AFM, SEM and FTIR for some powder that calcined at the different temperatures.

3- Studying the dielectric properties as a function of frequency in the range 50 Hz-5MHz for all samples.