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Magnetodielectric Properties of Cobalt Ferrite-Silica Composites Prepared by a Sol-Gel Technique

A Thesis
Submitted to the Council of the College of Science- University of Diyala in Partial Fulfillment of the Requirements for the Degree of Master of Science in Physics

By

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2021 AD

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بِسْمِ اللَّهِ الرَّحْمَٰنِ الرَّحِيمِ

﴿وَأَنْزَلْنَا الْحَدِيدَ فِيهِ بَأْسٌ شَدِيدٌ وَمَنَافِعُ لِلنَّاسِ

وَلِيَعْلَمَ اللَّهُ مَن يَنصُرُهُ وَرُسُلَهُ بِالْغَيْبِ ۚ إِنَّ اللَّهَ

قَوِيٌّ عَزِيزٌ﴾

صدق الله العظيم

سورة الحديد

الاية ٢٥

Dedication

My M.Sc. is dedicated to...

My merciful parents.

My supporters brother.

Mohammed B. Jumaa

2021

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List of Symbols

Symbol	*Definition*
θ	Bragg's angle
λ	Wavelength
M	Magnetization
H	Magnetic field
M_S	Saturation magnetization
H_c	Coercivity
M_r	Remanence magnetization
n_B	Magnetic moment
K	Magnetic anisotropy
T	Temperature
'a'	Lattice parameters
ε'	Dielectric constant
ε''	Dielectric loss factor
$tan\ \delta$	Dielectric loss angle
σ_{ac}	ac conductivity
T_c	Curie temperature
T_N	Neel temperature,
μ_r	Relative permeability
μ_o	Vacuum permeability
μ	Permeability of specific medium
χ	Susceptibility
χ_m	Magnetic susceptibility
C	Curie constant.
P	Polarization
C	Capacitance

C_o	Capacitance of air
f	Frequency
d	Spacing between the atomic planes
D	Crystallite size
hkl	Miller indices
L	Hopping length
ρ_x	X-ray density
M	Molecular weight
℃	Degrees celsius
P_e	Electric polarization
P_i	Ionic polarization
P_o	Orientation polarization
P_S	Space-charge polarization
D_c	Critical diameter
α	Polarizability
E_a	Activation energy

List of Abbreviations

Abbreviations	*Definition*
NPs	Nanoparticles
NCs	Nanocomposites
XRD	X-ray diffraction
TEM	Transmission electron microscopy
FT-IR	Fourier transform infrared spectroscopy
SEM	Scanning electron microscopy
FE-SEM	Field emission-scanning electron microscopes
VSM	Vibrating sample magnetometer
MRI	Magnetic resonance imaging
FWHM	Full width at half-maximum
ICSD	Inorganic crystal structure database
PXRD	Powder X-ray diffraction
GMR	Giant magnetoresistance

Abstract

The aim of the thesis is to synthesize and study the structural, magnetic and electrical properties of cobalt ferrites nanoparticles with the stoichiometric formula $Co_xFe_{3-x}O_4$ (x= 0.8, 0.9, 1, 1.1, and 1.2) respectively, were prepared using the sol-gel auto-combustion process. After combustion, the as-burnt powders were calcined at 500, 600, and 700 ℃ for 3 hrs to increase homogeneity and remove organic waste, where the as-burnt specimens and the specimens that calcined at 500, 600, and 700 ℃ added with a four-drop PVA as a binder to press it into circular pellets of diameter 13 mm with thickness about 2 mm. The prepared pellets were sintered at 350, 600, 700, and 800 ℃ for 3 hrs to intensify of the specimens and, slowly allowed to be cooled naturally to examine the dielectric properties. Another purpose of this work is to blend and study the structural, magnetic and electrical properties of cobalt ferrite-silica nanocomposites at 600 ℃ using the formula $CoFe_2O_4/SiO_2$ in different ratios (35%, 40%, 45%, and 50%) respectively, which were prepared using the conventional ceramic method. The specimens, that calcined at 600 ℃, added with a four-drop PVA as a binder to press it into circular pellets of diameter 13 mm with thickness about 2 mm. The prepared pellets were sintered at 700 ℃ for 3 hrs to intensify of the specimens and slowly allowed to be cooled naturally to examine the dielectric properties.

The XRD diffraction analysis for $Co_xFe_{3-x}O_4$ nanoferrites showed all the major peaks corresponding to the single spinel structure. The size of the formed crystallite of ferrite specimens increases with increasing temperature calcination and cobalt content. On the other hand, the XRD patterns of $CoFe_2O_4/SiO_2$ nanocomposites revealed all of the major peaks corresponding to the spinel structure's single phase. Based on its amorphous nature, there are no signatures in XRD for SiO_2. The formation of a cubic spinel structure is revealed by fourier transform infrared (FT-IR)

spectra, which uncovered two main absorption bands in the 600-400 cm^{-1} range. In $Co_xFe_{3-x}O_4$ nanoferrites a similar pattern of shifting peaks for the bands v_1 towards the higher frequencies side whereas a similar pattern of shifting of the peaks for the bands v_2 is observed towards the lower frequencies side have observed with the increasing of the calcined temperature and cobalt content. The FT-IR spectra, on the other hand, showed major absorption bands at 468.702, 800.459 and 1103.28 cm^{-1}, indicating the formation of amorphous SiO_2. In $CoFe_2O_4/SiO_2$ nanocomposites a similar trend in shifting of the (Fe-O) (v_1) and (Co-O) (v_2) bands towards the lower frequencies have noticed with the increase mixing SiO_2. According to images taken with a Field Emission-Scanning Electron Microscope of cobalt ferrite nanoparticles $Co_xFe_{3-x}O_4$, the particle size increases as the calcination temperature and cobalt content rises (FE-SEM) and shows agglomerated with homogenous spherical and polyhedral particles. The presence of Co, Fe, and O in all specimens is confirmed using the Energy Dispersive Spectrum (EDS). On the other hand, according to images taken with a Field Emission-Scanning Electron Microscope of cobalt ferrite-silica nanocomposites $CoFe_2O_4/SiO_2$, the grain size increases with the increase in the mixing ratio of silica and shows that all specimens contain a compact order of homogeneous nanoparticles with a spherical form and polyhedral particles. The presence of Co, Fe, O, and Si in all specimens is proved using the Energy Dispersive Spectrum (EDS). At room temperature, the magnetic characteristics are measured with a VSM in an applied field of ±15 kOe ranges. The saturation magnetization (M_s), remanence magnetization (M_r), and magnetic moment (n_B) of $Co_xFe_{3-x}O_4$ nanoferrites are found to go up with increasing calcination temperature. This behavior is linked to spin canting and disturbance in the surface spin. When Co^{2+} content increases, also the saturation magnetion and magnetic moment at calcined temperatures (600

and 700 ℃) increases except for x=1.2. Whereas the remnant magnetization decreases with increasing content Co^{2+} of as-burnt and calcined specimens. In another term, the saturation magnetization (M_s), remanence magnetization (M_r) and magnetic moment (n_B) of $CoFe_2O_4/SiO_2$ nanocomposites decrease with the increase of the SiO_2 mixing ratio. The dielectric properties are measured using a (LCR) meter in the frequency range of (50Hz-2MHz) at room temperature. The dielectric constant (ε'), dielectric loss angle ($tan\delta$) and dielectric loss factor (ε'') for $Co_xFe_{3-x}O_4$ nanoferrites and $CoFe_2O_4/SiO_2$ nanocomposites are found to decrease with increasing frequency. This behavior is typical of ferrites as explained by Koop's theory. The dielectric constant was found to decrease with increasing temperature and increases with increasing of the SiO_2 mixing ratio. It was also found that the dielectric loss angle and dielectric loss factor increases with increasing temperature and decreases with an increasing of the SiO_2 mixing ratio. The ac conductivity (σ_{ac}), gradually increased as the frequency increased, it was also found that the ac conductivity (σ_{ac}) increases with increasing temperature and decreases with an increasing of the SiO_2 mixing ratio.

Chapter One

Concept of Nanoparticles and Literature

Review

1.1 Introduction

Researchers are constantly working on new materials that could be used in a variety of industries. Wood, fabric, glass, alloys, metals, ceramics, petroleum fuels, radioactive materials, coal, polymers, stone, semiconductors, and other materials have ushered in significant advances in humanity's history. The investigation of innovative materials with superior properties, on the other hand, stretches back to the Stone Age. The study of material synthesis and properties has only recently emerged as a distinct field of science with technical and practical ramifications.

In the domains of physics and other sciences, nanotechnology is one of the most important and intriguing technologies. It has made a significant contribution to the events of great scientific revolutions that are hoped to change the course of technologies and applied sciences, as it provides a high ability to settings and control in the composition of matter at the level of atomic dimensions, as well as a high potential in nanofabrication, resulting in amazing physical qualities and properties. Because of this, nanotechnology has been used to create systems and devices with unique features by manipulating the form and size of the nanosphere [1]. Due to the enormous ratio of the surface of the grains to their size, magnetic materials in general, and nanoferrite in particular, have a significant impact on physical, electrical, and magnetic properties. Due to its magnetic properties and a wide range of uses, nanoferrite has sparked interest in the sphere of science and technology in recent years [2]. High frequency transformer cores, antenna bars, and choke coils are all made of ferrites [3, 4]. Nanoelectronic devices, integrated circuits, and magnetic resonance imaging (MRI) are all examples of this [5-8]. The typical formula for ferrites is (MFe_2O_4), where represents M, one of the divalent metallic elements (Zn^{+2},Cu^{+2},Fe^{+2},Mg^{+2}). The ferrites are divided into three groups based on their chemical composition: Garnet, Hexagonal and Spinal ferrite [9]. Because of its strong electrical and magnetic properties and wide range of uses, we will concentrate our research on this last type. Spinal ferrites are materials with good magnetic and electrical properties that are highly influenced by the distribution pattern of positive ions (cations) between the tetrahedral and octahedral sites [10]. One of the most important methods of preparing nanoferrite is the sol-gel method auto-combustion since it is simple to prepare, takes little time, and does not require high temperatures [11]. Ferrite is made from a powder that is compressed and sintered to take the desired shape. It is one of the simplest and cheapest materials to make, and its properties are determined by a number of factors, including the

shape and size of the grains, the method of preparation, the sintering temperature, the type of materials that make up ferrites, and their quantity [12].

1.2 Literature Review

The study of nanospinel ferrite has attracted researcher's interest in recent years owing to its unique features and vast range of uses. Some of the properties of nanospinel ferrites that have been studied, including structural, electrical, and magnetic properties, are reviewed below:

N. M. Deraz and A. Aarifi, (2011) [13]; They used sol-gel auto-combustion to make nanocrystalline Zn-substituted cobalt ferrite powders, Co_{1-x} $Zn_xFe_2O_4$ (x = 0, 0.25, 0.5, 0.75, and 1). The specimens had a cubic spinel structure, and the X-ray diffraction investigation revealed that the crystallite size decreased from 70 to 51nm as the zinc content was increased to (x=1).The lattice constant increased from (0.8370 to 0.8400 nm) with increasing the concentration of zinc to (x=1), whilst the X-ray density increased from (5.293 to 5.381 g/cm^3) as the concentration of zinc increased to (x=1). The saturation magnetization of Co-Zn nanoferrites was examined using a vibrating sample magnetometer (VSM) at room temperature, and the results showed that the saturation magnetization increased as zinc substitution increased. Increased Zn concentrations resulted in a drop in coercivity (H_c) from 807.7 to 46.0 Oe.

K. M. Batoo and M. S. Ansari, (2012) [14]; They used an auto-combustion approach to make nanoparticles of polycrystalline $Ni_{0.7-x}Zn_xCu_{0.3}Fe_2O_4$ (x= 0, 0.05, and 0.2) ferrites in the form of a powder, with average crystallite sizes ranging from 28 to 32 nm. The cubic spinel structure of ferrites was revealed by X-ray diffraction study of powder specimens sintered at 600 °C for 4 hrs. Two absorption bands in the range of (600-400 cm^{-1}) were observed using fourier transform infrared (FT-IR), which are related to the stretching vibration of tetrahedral and octahedral sites. Then they took electrical measurements and discovered that when doping with zinc 10%, each of (ε', ε'', $tan\delta$, σ_{ac}) reaches its maximum value.

M. Zhang et al., (2013) [15]; They used the sol-gel method to prepare (9) specimens of the chemical $Ni_{0.5}Zn_{0.5}Fe_2O_4$. After completing an X-ray diffraction investigation, it was discovered that all specimens of the produced ferrite compound formed the spinel phase, with average crystallite sizes ranging from 9 to 96 nm, if the average crystallite size increasing with the increase in the annealing temperature. When the annealing temperature is raised, the lattice constant decreases. The results of the magnetic measurements performed on the specimens of the synthesized chemical revealed that all of the specimens were paramagnetic. It was also discovered that when particle size rises, saturation magnetization increases, which can be explained by cation redistribution on tetrahedral A and octahedral B sites, as well as domain wall motion.

M. Mozaffari et al., (2014) [16]; They used the sol-gel method to make nanoparticles of nickel substituted cobalt ferrite $Ni_xCo_{1-x}Fe_2O_4$ (x= 0.1, 0.3, 0.5, 0.7, and 0.9). The crystallite size of the specimens was estimated to be around 30 nm using X-ray diffraction. The mean particle sizes in the SEM pictures were in the range of 70-160 nm, indicating that each particle contains many crystallites. As the nickel concentration was increased, the lattice parameter of the specimens decreased from 8.350 to 8.300 Å. Magnetic measurements were performed on the specimens, and the results demonstrate that as nickel content increases, saturation magnetization drops from 70.8-37.3 emu/g. With increasing nickel concentration, the materials coercivity reduces from (1188 to 321 Oe), as evidenced by variations in magneto crystalline anisotropy.

A. V. Raut et al., (2014) [17]; Utilizing citric acid as a fuel, $Co_{1-x}Zn_xFe_2O_4$ ($0.0 \leq x \geq 1.0$) was synthesized using the sol-gel auto-combustion approach. X-ray diffraction studies revealed the production of a single phase cubic spinel structure, with the lattice constant and X-ray density increasing as the Zn^{2+} concentration increased within the expected range. The development of nanocrystalline grains was revealed by SEM examination. The specimens ferrite composition was revealed in the FT-IR spectra, which showed two prominent bands between 400 cm^{-1} and 600 cm^{-1}. With increasing Zn^{2+} concentration, different magnetic characteristics such as saturation magnetization, remanence magnetization, coercivity, and squareness ratio decrease. In the nanoparticles investigated, a decreasing squareness ratio indicated the presence of a single unreactive field particle with cubic contrast.

M. Lakshmi et al., (2015) [18]; They used the sol-gel method to make nanoparticles of Cr-substituted Zn ferrites with the chemical formula Cr_x Zn Fe_{2-x} O_4 (x= 0.0-0.5). And its average grain size is between 43 and 63 nanometers, They then looked at the structural and magnetic properties after calcining at 900 ℃ for 3 hrs, because they discovered that as Cr (x) cont increases, both crystallite size and lattice parameter decrease. And the amount of chromium has an active effect on the magnetization current, as they discovered that the magnetization current is highest at (x= 0), drops dramatically at (x= 0.1), and then progressively rises at subsequent values of x. Infrared microscopy confirms the formation of spinel structure. The creation of a single-phase spinel structure with cubic symmetry was confirmed by X-ray diffraction study of Cr-Zn ferrite. Nanoparticles with a restricted size distribution were seen in SEM micrographs.

R. S. Yadav et al., (2015) [19]; They used a starch-assisted sol-gel auto-combustion approach to make $Co_{1-x}Zn_xFe_2O_4$ (x= 0.0 and 0.5) spinel ferrite nanoparticles at 800 ℃. The development of the ferrite phase was confirmed using fourier transform infrared (FT-IR) spectroscopy. The XRD examination revealed that $Co_{1-x}Zn_xFe_2O_4$ (x= 0.0 and 0.5) spinel ferrite nanoparticles develop in a single phase. The generation of nanosized spherical particles with spherical morphology was revealed by FE-SEM analysis. The decrease in nanocrystalline size and cation distribution in spinel ferrite explain the observed shift in saturation magnetization and coercivity.

T. Dippong et al., (2016) [20]; Using the sol-gel process, they created a $Co_xFe_{3-x}O_4$ (x=0.5-2.5) system embedded in a silica matrix. FT-IR observations revealed the production of silica matrix and oxidic phases, which were also detected by XRD studies. The XRD data support the nanocrystallites modest size: 10 nm for 700 ℃ calcined specimens and 20-25 nm for 1000℃ calcined specimens. Hysteresis and magnetization derivative data were used to explore the genesis of magnetic phases. The hysteresis loops show a low coercive field, but the magnetization derivatives show broad peaks, which could indicate the presence of a poorly crystallized secondary magnetic phase and connected magnetic phases.

T. Dippong et al., (2017) [21]; They used the sol-gel process to make nanocomposites $xCoFe_2O_4/(100\text{-}x)SiO_2$, (x=10, 30, 50, 70, and 90%) and then annealed them at 1100 °C. The production of single-phase $CoFe_2O_4$ was revealed by the X-ray diffraction pattern and FT-IR spectra for all specimens. The creation of the amorphous silica matrix was also suggested by the FT-IR spectra (SiO_2). The agglomerated shape of $CoFe_2O_4$ distributed in the silica matrix was visible in SEM pictures. Magnetic $CoFe_2O_4$ nanoparticles distributed in a silica matrix are spherical in shape and range in size from 6-35 nm. The saturation magnetization of the examined nanocomposites increased as the $CoFe_2O_4$ concentration (x) in the silica matrix increased, according to the vibrating sample magnetometer (VSM). The saturation magnetization (M_s)and coercivity (H_c) of $CoFe_2O_4$ nanocrystals embedded in silica matrix were also found to have a linear relationship with the mean crystallite size, according to studies.

T. Dippong et al., (2017) [22]; They used the sol-gel process to make cobalt ferrite nanocrystallites embedded in a silica matrix, $CoFe_2O_4$:SiO_2. XRD examination revealed that annealing at 400-1100 °C resulted in the development of single-phase $CoFe_2O_4$ embedded in the silica matrix. The production of the precursor in the pores of the silica matrix was confirmed using fourier transform infrared spectroscopy (FT-IR). The particles were formed on the substrate and interconnected with each other, forming enormous crystals, according to scanning electron microscopy studies. $CoFe_2O_4$ nanoparticles embedded in the silica matrix had a high potential to form agglomerates. Magnetic experiments revealed a direct link between annealing temperature and saturation magnetization in a constant coercive field for the investigated magnetic hysteresis loops. The annealing temperature has a significant impact on the particle size of ferrite powders.

R. Zhang et al., (2018) [23]; A sol-gel auto-combustion process was used to successfully synthesize single phase cobalt ferrite powders with a cubic spinel structure, and the pellets were sintered at various temperatures for 2 hrs. The average grain size grew from 0.26 μm to 0.83 μm when the sintering temperature climbed from 900 to 1300 °C, according to SEM examination. Due to electron hopping between ferrous (Fe^{2+}) and ferric (Fe^{3+}) ions, the dielectric constant and loss tangent decreases with increasing frequency and becomes constant at high frequencies. The crystallinity and grain size of materials have a big impact on their magnetic characteristics. As a result of the change in crystallinity and grain size, the maximum magnetization was greatly influenced by the sintering

temperature. The coercivity drops sharply as the grain size exceeds the domain size.

V. P. Senthil et al., (2018) [24]; Using the auto-combustion sol-gel approach, they were able to successfully synthesis cobalt ferrite ($CoFe_2O_4$) nanocrystals. At varied calcined temperatures of 600, 700, and 800 ℃, X-ray diffraction patterns revealed single phase production of $CoFe_2O_4$ spinel ferrite nanoparticles. Fourier transform infrared spectroscopy analyses further confirmed the production of $CoFe_2O_4$. The saturation magnetization and coercivity increase with increasing calcination temperature, according to tests made with a vibrating sample magnetometer (VSM). The magnetic characteristics of the $CoFe_2O_4$ nanoparticles have shown significant fluctuations, which appear to be caused by the calcination temperature of the spinel ferrite nanoparticles.

A. Hossain et al., (2018) [25]; They used the citrate-gel auto combustion approach to make semi-soft ferrimagnetic $CoFe_2O_4$ nanoparticles, which they investigated. The creation of cubic spinel $CoFe_2O_4$ nanoparticles was confirmed by X-ray diffraction, with the average crystallite size of as-obtained specimens being approximately (30 nm). Within the spinel lattice, FT-IR spectra of ferrites revealed the presence of tetrahedral and octahedral group complexes. The synthesized particles are formed as octahedron and tetrahedron nanoscale in size with some micrometer phases, according to SEM pictures. Using a vibrating sample magnetometry (VSM), the magnetic characteristics of $CoFe_2O_4$ nanoparticles (NPs) at room temperature were determined. The results show that all specimens had soft ferrimagnetic behavior. In addition, the specific saturation magnetization M_S= 60 emu.g^{-1} of single domain particles measured and low coercivity 620 Oe. The ordered single-domain magnetic nanoparticles are responsible for the higher saturation magnetization, while the decrease in interparticle interactions and magneto-elastic anisotropy is responsible for the reduced magnitude of coercivity. The dielectric constant and dissipation factor of a series of ferrites $Co_{1-x}Fe_{2-x}O_4$ are low with increasing frequency, as seen in the results of an electrical conduction research.

T. Dippong et al., (2019) [26]; They used a modified sol-gel approach to make $Ni_xCo_{1-x}Fe_2O_4/SiO_2$ nanocomposites (x = 0, 0.25, 0.50, 0.75, and 1.00). The X-ray diffraction refers to the formation of single phase cubic spinel structure for all the compositions, the X-ray diffraction (XRD) patterns revealed a decrease of lattice constant (8.438-8.318 Å), unit cell

volume (600.8-575.5 $Å^3$), average crystallite size (31.7-18.2 nm) and hopping length in A (3.654-3.602 Å) and B (2.983-2.941 Å) sites and an increase of X-ray density (5.188-5.411 g/cm^3) and relative crystallinity (81.6-100 a. u.) with the increase of nickel content for the specimens annealed at 1100 ℃. Due to the influence of cation stoichiometry and specific site occupancy, magnetic studies revealed that saturation magnetization, remanent magnetization, coercivity, magnetic moments per unit cell, and anisotropy decrease with increasing nickel content but increase with increasing annealing temperature.

S. Dabagh and G. Dini, (2019) [27]; They used the sol-gel auto combustion method to make monophase $Ag_xCo_{1-x}Fe_2O_4$ ($0 \leq x \leq 0.08$) powders having cubic spinel structure, spherical-shaped particles, and an average size of around (20-25 nm). The sol-gel technique was then used to coat a specimen with a monophase structure with an amorphous silica layer on the surfaces of the generated $Ag_{0.08}Co_{0.92}Fe_2O_4$ NPs with optimal magnetic characteristics. The properties of silica-coated Ag-Co-ferrite NPs were studied using fourier transform infrared (FT-IR) spectroscopy, which confirmed the existence of the silica coating on the Ag-Co-ferrite NPs' surfaces. The saturation magnetization (M_S) value of the silica coated specimen was somewhat lower than the uncoated value. When cobalt ions are replaced with diamagnetic silver ions in the spinel structure of Co-ferrite nanoparticles, the magnetization of the nanoparticles decreases.

T. Dippong et al., (2019) [28]; They evaluated the structural and magnetic properties of $Ni_xZn_{1-x}Fe_2O_4/SiO_2$ nanocomposites synthesized using the sol-gel method. The production of Ni-, Zn-, and Fe-succinate precursors and their breakdown into ferrites were detected by FT-IR spectroscopy. The development of ferrites embedded in SiO_2 matrix was further confirmed by FT-IR spectra. Scherrer's formula yields average crystallite sizes of nanocomposites in the ranges of (48.3-15.9 nm) (1200 °C), (12.8-5.1 nm) (800 °C), and (5.8-3.8nm) (500 °C). By annealing at $1200°C$, X-ray diffraction revealed the production of highly crystalline single-phase ferrite in specimens with high Ni content and crystalline ferrite in specimens with low Ni and high Zn content. With Ni x=1, the saturation magnetization was found to be the maximum value of the specimen, however the coercivity decreased as the Ni content in the specimens increased due to a reduction in the amount of lattice defects and internal strain. The magnetic anisotropy constant in Ni-Zn ferrite nanopowders increases exponentially as the Ni content rises. They also

discussed about the hysteresis loop results, which revealed that the NCs were superparamagnetic and ferromagnetic.

V. R. Bhagwat et al., (2019) [29]; They used the sol-gel auto combustion approach to successfully synthesize spinel cobalt ferrite ($CoFe_2O_4$) nanoparticles. All specimens contain a cubic spinel structure, with computed average crystallite sizes in the range of (15-22 nm).With aggregation of particles, the SEM images appear to be homogeneous. Regardless of the fuels used, the sponge-like spherical morphology was seen. The average grain size was discovered to be between 65 and 86 nanometers. The superparamagnetic behavior of the specimens was validated by the increased saturation magnetization (M_s) and coercivity (H_c) obtained by the magnetic hysteresis (M-H) loop. The size of the nanoparticles increased the saturation magnetization, coercivity, and remanent magnetization.

T. Dippong et al., (2020) [30]; They investigated the structural and magnetic properties of $\alpha Cu_{0.6}Co_{0.4}Fe_2O_4/(100\text{-}\alpha)SiO_2$ nanocomposites (α =0,25, 50, 75, and 100%) produced by sol-gel technique and thermally treated at 200, 500, 800, and 1200 °C. Low crystallinity of NCs with low ferrite concentration is indicated by low intensity, widened diffraction peaks, most likely due to reduced crystallite size. The Sherrer formula was used to calculate the average crystallite sizes (D_C) for sintered specimens, which ranged from 34 to 110 nm. The development of SiO_2 matrix and Cu-Co ferrite was revealed by fourier transform infrared spectroscopy (FT-IR) analysis. The amorphous SiO_2 matrix has the smallest nanoparticles (30 nm), ensuring that the nanostructure of Cu-Co ferrite powders is refined. With increasing SiO_2 matrix percentage, the size of round shaped Cu-Co ferrite nanoparticles decreases. With increasing Cu-Co ferrite content embedded in SiO_2 matrix, they found that saturation magnetization and remanent magnetization improved while coercivity and magnetic anisotropy decreased. They also discovered that when Cu^{2+} is doped in the Co ferrite structure, less Co^{2+}-Fe^{3+} ions pairs develop, resulting in a lower magnetization value, because Cu^{2+} prefers tetrahedral positions and has a lower magnetic moment than Co^{2+}.

C. C. Naik and A. V. Salker, (2020) [31]; They used the sol-gel auto-combustion method to make $CoFe_{2-x}Sb_xO_4$ (x = 0.00, 0.03, 0.06, and 0.09). The development of a single-phase cubic structure free of impurities was confirmed by X-ray diffraction. The distinctive absorption bands corresponding to M-O stretching vibrations were visible in infrared

spectra. The inductively coupled plasma-atomic emission spectroscopy technique was used to confirm the precise chemical composition. Studies using X-ray photoelectron spectroscopy confirmed the chemical state of the current metal ion species. Scanning electron microscopy (SEM) and transmission electron microscopy (TEM) studies were used to determine the morphology of the specimens as well as their particle sizes. At both 300 K and 50 K, the saturation magnetization decreased after Sb replacement, with a significantly greater value at 50 K. Due to thermal energy or heat mediated disturbance in the alignment at 300 K, the magnetic parameters measured at 50 K revealed greater values than those obtained at 300 K. The semiconducting property of the materials was revealed by DC electrical resistivity measurements, which revealed an increasing trend with Sb^{3+} ions content. The dielectric constant increased as the concentration of Sb^{3+} ions and temperature increased.

G. R. Patta et al., (2020) [32]; They used a polyethylene glycol-assisted sol-gel technique to make high coercive single-domain cobalt ferrite $CoFe_2O_4$ nanoparticles, which they then annealed at varied temperatures (400, 600, 700, and 800 ℃). XRD and FT-IR methods were used to characterize the produced specimens. The size and shape of the particles are significantly dependent on the annealing temperature, according to X-ray diffraction analysis. The produced cobalt ferrite nanoparticles are extensively distributed and have a consistent shape. The octahedral vibrational band of the Fe-O bond is identified at about 587 cm^{-1} in the FT-IR spectra. The tetrahedral vibrational band of the Co-O bond is observed at roughly 420 cm^{-1} in the spectra of all the specimens. For cobalt ferrite particles of size 12.8 nm, the highest values of saturation magnetization M_s and remnant magnetization M_r were recorded, with 85.5 emu/g and 49 emu/g, respectively. For the cobalt ferrite containing nanoparticles of average size ~10 nm, the coercive field (H_c) showed non-monotonic behavior with unique maximum (2214 Oe) and modest magnetization, 62.7 emu/g, indicating that the particles are restricted to single-domain. For the materials annealed at 800 ℃, the magneto-crystalline anisotropy constant, K, determined using the Stoner-Wohlfarth relationship, the maximum value was 10.74×10^6 erg/cm^3.

T. Dippong et al., (2020) [33]; Using a modified sol-gel process and high temperature annealing (1000 ℃), they created copper substituted cobalt ferrite $Cu_xCo_{1-x}Fe_2O_4$ (x= 0.00, 0.25, 0.50, 0.75, and 1.00) nanoferrites embedded in SiO_2 matrix. The prepared specimens have a single-phase cubic spinel structure, according to X-ray diffraction

examination. The Scherrer equation was used to determine crystallite sizes ranging from 20 nm ($CoFe_2O_4$) to 60 nm ($CuFe_2O_4$). Fourier transformed infrared spectroscopy (FT-IR) was used to analyze the reaction progress. The FT-IR spectroscopy confirmed the consumption of nitrogen oxides up to 200 ℃. Fe_3O_4 emerges in all specimens at 200 ℃, according to PXRD analysis, while Copper oxide appears in those with significant copper concentration. Scanning electron microscopy (SEM) was used to examine the particle size and form. The SEM images revealed an agglomeration of homogeneous, regular, and yeast-like form particles on the specimen surfaces.

1.3 Aim of the Study

1- Synthesis of $Co_xFe_{3-x}O_4$ nanoferrites by a sol-gel auto-combustion method and $CoFe_2O_4/SiO_2$ by the conventional ceramic method.

2- Studying the effect of increasing the content (x) and calcination, temperature of $Co_xFe_{3-x}O_4$, and studying the influence of amorphous silica (SiO_2) concentration on the structural, magnetic, and dielectric properties (XRD, FT-IR and FE-SEM EDS).

3- Studying the magnetic parameters such as (saturation magnetization (M_s), remanence magnetization (M_r), and coercivity (H_c)) of synthesized ferrite nanoparticles in applied field ±15 kOe through vibrating sample magnetometer (VSM) at room temperature.

4- Studying the electrical properties (dielectric constant (ε'), dielectric loss angle ($tan\ \delta$), dielectric loss factor (ε''), and ac conductivity (σ_{ac})) of synthesized nanoferrite with the frequency from (50 Hz) to (2MHz) by using LCR meter at room temperature.