

Republic of Iraq Ministry of Higher Education and Scientific Research University of Diyala College of Science Department of Physics



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

Mohammed Burhan Jumaa B. Ed. in Physics (2017-2018)

Supervised By

Prof. Dr. Tahseen H. Mubarak Assist. Prof. Dr. Ali M. Mohammad

2021 AD

1443 AH



﴿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

Acknowledgement

First and foremost, I would like to thank Almighty Allah for giving me the strength, knowledge, ability and opportunity to undertake, persevere and complete this research. Without his blessings, this achievement would not have been possible.

There are many people I need to thank for their support and encouragement.

I would like to express my heartfelt thanks to my supervisors,

Prof. Dr. Tahseen H. Mubarak and *Assist. Prof. Dr. Ali M. Mohammad*, for their guidance, inspiration, and encouragement. I am very grateful for both their expertise and commitments throughout the course of my research.

I express my thanks to head of Physics Department *Asst. Prof. Dr. Ammar A. Habeeb* and all members, especially postgraduate faculty members and staffs of the college for their cooperation.

It certainly would have been very difficult completing this research without the support from *my parents and my brother*. Their encouragements and supports inspired me to hold out to the end.

I express my deep gratitude to my best friend *Saad Sh. Habeeb* for his encouragement and cooperative.

I am also grateful to the Physics Department, College of Science at Sulaimani University for their kind help during the laboratory work and LCR meter tests also special thanks for the Education Physics Department in the University of Garmian for their kind help during the laboratory work.

I certainly cannot mention the names of everyone who contributed towards the success of this research, but as they have always known, I am very grateful.

> Mohammed B. Jumaa 2021

Supervisors Certification

We certify that this thesis entitled "*Magnetodielectric Properties of Cobalt Ferrite-Silica composites Prepared by a Sol-Gel Technique*" for the student (**Mohammed Burhan Jumaa**), was prepared under our supervisions at the Department of Physics, College of Science, Diyala University in part. One of the prerequisites for the awarding of the *M.Sc in Physics*.

Signature:	Signature:		
Name: Dr. Tahseen H. Mubarak	Name: Dr. Ali M. Mohammad		
Title: Professor	Title: Assist. Professor		
Address: College of Science	Address: College of Education		
University of Diyala	University of Garmian		
Date: / / 2021	Date: / / 2021		

Head of the Physics Department

In view of available recommendation, I forward this thesis for debate by the examining committee.

Signature: Name: *Dr. Ammar A. Habeeb* Title: *Assist. Professor* Head of the Physics Department Address: College of Science, University of Diyala Date: / / 2021

Linguistic Amendment

I certify that the thesis entitled "*Magnetodielectric Properties of Cobalt Ferrite-Silica Composites Prepared by a Sol-Gel Technique*" presented by student (**Mohammed Burhan Jumaa**) has been corrected linguistically, therefore, it is suitable for debate by examining committee.

Signature:

Name: *Dr. Karim H. Hassan* Title: *Professor* Address: *University of Diyala /College of Sciences*

Date: / / 2021

Scientific Amendment

I certify that the thesis entitled "*Magnetodielectric Properties of Cobalt Ferrite-Silica Composites Prepared by a Sol-Gel Technique*" presented by student (**Mohammed Burhan Jumaa**) has been evaluated scientifically, therefore, it is suitable for debate by examining committee.

Signature:

Name: **Dr.** *Nadir F. Habubi* Title: *Professor* Address: *Al-Mustansiriyah University* Date: / / 2021

Scientific Amendment

I certify that the thesis entitled "*Magnetodielectric Properties of Cobalt Ferrite-Silica Composites Prepared by a Sol-Gel Technique*" presented by student (**Mohammed Burhan Jumaa**) has been evaluated scientifically, therefore, it is suitable for debate by examining committee.

Signature:

Name: *Dr. Shihab A. Zaidan* Title: *Professor* Address: *University of Technology* Date: / / 2021

Examination Committee Certificate

We certify that we have read this thesis entitled "*Magnetodielectric Properties of Cobalt Ferrite-Silica Composites Prepared by a Sol-Gel Technique*" and, as an examining committee, we examined the student (Mohammed Burhan Jumaa) on its content, and in what is related to it, and that in our opinion it meets the standard of a thesis for the degree of master in Physics Sciences.

Signature:

Name: **Dr. Enas Muhi Hadi** Title: Professor Address: University of Technology Date: / / 2021 (Chairman)

Signature:

Name: Dr. Widad Hano Abbas Title: Assistant. Professor Address: Al-Mustansiriyah University Date: / / 2021 (Member)

Signature:

Name: Zena Mohammed Ali Abbas Title: Assistant. Professor Address: University of Diyala Date: / / 2021 (Member)

Signature:

Name: *Dr. Tahseen H. Mubarak* Title: *Professor* Address: *University of Diyala* Date: / / 2021 (Member / Supervisors) Signature:

Name: Dr. Ali Mustafa Mohammed Title: Assistant. Professor Address: University of Garmian Date: / / 2021 (Member / Supervisors)

Approved by the Council of the College of Science.

(The Dean)

Signature: Name: *Dr. Tahseen H. Mubarak* Title: *Professor* Date: / / 2021

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List of Symbols

Symbol	Definition
θ	Bragg's angle
λ	Wavelength
М	Magnetization
Н	Magnetic field
M_S	Saturation magnetization
H _c	Coercivity
M _r	Remanence magnetization
n_B	Magnetic moment
K	Magnetic anisotropy
Т	Temperature
'a'	Lattice parameters
ε′	Dielectric constant
arepsilon''	Dielectric loss factor
tan δ	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
Χm	Magnetic susceptibility
С	Curie constant.
Р	Polarization
С	Capacitance

Co	Capacitance of air
f	Frequency
d	Spacing between the atomic planes
D	Crystallite size
hkl	Miller indices
L	Hopping length
$ ho_x$	X-ray density
М	Molecular weight
°C	Degrees celsius
P _e	Electric polarization
P _i	Ionic polarization
Po	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 $Co_x Fe_{3-x}O_4$ (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 °C 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 °C 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 °C 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 °C using the formula CoFe₂O₄/SiO₂ in different ratios (35%, 40%, 45%, and 50%) respectively, which were prepared using the conventional ceramic method. The specimens, that calcined at 600 °C, 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 °C 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 SiO₂. 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}O₄ 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 SiO₂. In CoFe₂O₄/SiO₂ 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 SiO₂. According to images taken with a Field Emission-Scanning Electron Microscope of cobalt ferrite nanoparticles Co_xFe_{3-x}O₄, 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 CoFe₂O₄/SiO₂, 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 $Co_x Fe_{3-x}O_4$ nanoferrites are found to go up with increasing calcination temperature. This behavior is linked to spin canting and disturbance in the surface spin. When Co²⁺ content increases, also the saturation magnetion and magnetic moment at calcined temperatures (600

700 °C) increases except for x=1.2. Whereas the remnant and magnetization decreases with increasing content Co²⁺ of as-burnt and calcined specimens. In another term, the saturation magnetization (M_s) , and magnetic moment (n_B) of remanence magnetization (M_r) CoFe₂O₄/SiO₂ nanocomposites decrease with the increase of the SiO₂ 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 δ) and dielectric loss factor (ϵ'') for Co_xFe_{3-x}O₄ nanoferrites and CoFe₂O₄/SiO₂ 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 SiO₂ 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 SiO₂ 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 SiO₂ 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 (MFe₂O₄), 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 autocombustion 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/cm³) 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 autocombustion approach to make nanoparticles of polycrystalline Ni_{0.7}. _xZn_xCu_{0.3}Fe₂O₄ (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 δ , 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}Fe₂O₄ (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, $Co_{1-x}Zn_xFe_2O_4$ ($0.0 \le x \ge 1.0$) was synthesized using the sol-gel autocombustion 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} O₄ (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 °C 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 autocombustion approach to make $Co_{1-x}Zn_xFe_2O_4$ (x= 0.0 and 0.5) spinel ferrite nanoparticles at 800 °C. 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 °C calcined specimens and 20-25 nm for 1000°C 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 xCoFe₂O₄/(100-x)SiO₂, (x=10, 30, 50, 70, and 90%) and then annealed them at 1100 °C. The production of single-phase CoFe₂O₄ 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 (SiO₂). The agglomerated shape of CoFe₂O₄ distributed in the silica matrix was visible in SEM pictures. Magnetic CoFe₂O₄ 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 CoFe₂O₄ concentration (x) in the silica matrix increased, according to the vibrating sample magnetometer (VSM). The saturation magnetization (M_s) and coercivity (H_c) of CoFe₂O₄ 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, $CoFe_2O_4$:SiO₂. 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 μ m to 0.83 μ m when the sintering temperature climbed from 900 to 1300 °C, according to SEM examination. Due to electron hopping between ferrous (Fe²⁺) and ferric (Fe³⁺) 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 (CoFe₂O₄) nanocrystals. At varied calcined temperatures of 600, 700, and 800 °C, X-ray diffraction patterns revealed single phase production of CoFe₂O₄ spinel ferrite nanoparticles. Fourier transform infrared spectroscopy analyses further confirmed the production of CoFe₂O₄. 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 CoFe₂O₄ 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 approach to make semi-soft ferrimagnetic CoFe₂O₄ combustion nanoparticles, which they investigated. The creation of cubic spinel CoFe₂O₄ 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 CoFe₂O₄ 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 Å³), 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/cm³) and relative crystallinity (81.6-100 a. u.) with the increase of nickel content for the specimens annealed at 1100 °C. 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 $Ag_xCo_{1-x}Fe_2O_4$ ($0 \le x \le 0.08$) 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}Fe₂O₄/SiO₂ 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 SiO₂ 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 (CoFe₂O₄) 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 $\alpha Cu_{0.6}Co_{0.4}Fe_2O_4/(100-\alpha)SiO_2$ 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 SiO₂ 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 SiO₂ matrix percentage, the size of round shaped Cu-Co ferrite nanoparticles decreases. With increasing Cu-Co ferrite content embedded in SiO₂ 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+} -Fe³⁺ ions pairs develop, resulting in a lower magnetization value, because Cu²⁺ 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 autocombustion 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 Sb³⁺ ions content. The dielectric constant increased as the concentration of Sb³⁺ ions and temperature increased.

G. R. Patta et al., (2020) [32]; They used a polyethylene glycolassisted sol-gel technique to make high coercive single-domain cobalt ferrite CoFe₂O₄ nanoparticles, which they then annealed at varied temperatures (400, 600, 700, and 800 °C). 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 nonmonotonic 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 °C, the magnetocrystalline anisotropy constant, *K*, determined using the Stoner-Wohlfarth relationship, the maximum value was 10.74×10^6 erg/cm³.

T. Dippong et al., (2020) [33]; Using a modified sol-gel process and high temperature annealing (1000 °C), 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 SiO₂ 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 (CoFe₂O₄) to 60 nm (CuFe₂O₄). 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 °C. Fe₃O₄ emerges in all specimens at 200 °C, 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 (SiO₂) 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.

Chapter Two

Theoretical Part

2.1 Introduction

The need for new technology is growing at a quicker rate than ever before as society evolves. As a result, information technology, which has grown at an exponential rate over the last half-century, is one of the cornerstones of this advancement. In recent years, the demand for smaller transistors and other important components has shifted emphasis to various approaches to boost computer performance without drastically reducing structural dimensions. One solution to this problem has been to use multiferroics instead of single-ferroic materials in the process. Multi-ferroic materials contain two or more of the major ferroic orders [34]. The magnetic properties of nanoparticles are influenced by a variety of circumstances. Chemical composition, particle size and design, type and severity of crystal lattice defects, and shape are among the most important of these variables. The magnetic and electric properties of a material based on nanoparticles can be tailored by altering some dimensions and compositional elements. In any case, controlling these variables during the production of nanoparticles that are virtually equal in size and chemical composition is unachievable; as a result, the attributes of nanoparticles of the same type can vary significantly.

Ferrites are magnetic iron oxides that contain other elements such as cobalt, nickel, zinc, barium, manganese, and strontium. The ferrite core is made using a variety of ceramic methods. As a result, they're brittle and hard. Ferrites are divided into two categories: hard and soft. Hard ferrites can be made in two main ways: isotropic and anisotropic. Isotropic ferrite materials are created without orientation and can be oriented in any applied magnetic direction. The direction of the applied magnetic field, on the other hand, affects anisotropic ferrite materials [35]. Cobalt ferrite is a highhardness magnetic material. Ferrite has a high saturation magnetization, high coercivity, strong anisotropy, good mechanical hardness, and chemical stability. Cobalt ferrite is ferrimagnetic due to its high curie temperature of 793 K [36]. It is normal for it to be half metallic [37]. Cobalt ferrite is a good conductor for magnetic recording media and can be utilized in giant magnetoresistance (GMR) device [38]. Magnetostrictive cobalt ferrite is employed in sensor applications. Cobalt ferrite is used in (MRI), drug delivery systems, and the separation of bioactive molecules including enzymes and proteins [39].

2.2 Magnetic Properties of Materials

Magnetic materials may be found in many kinds of places, including soils, animals, plants, and even the human mind. The chemical and crystallographic structure of bulk/gigantic magnetic materials determines their inherent magnetic properties, such as magnetization reversal in nanodots, coercive force, and Curie temperature. Whereas in nanoparticles, these properties are influenced by their finite size and geometry. The advent of nanoparticles has enabled researchers to investigate magnetic properties at all scales, from bulk to atomic [40].

Differences in internal and external flux, as well as variations in magnetization M or magnetic induction B, can be utilized to identify magnetic materials when a magnetic field is applied [41, 42]. The two factors that relate M and B to H are magnetic susceptibility (χ_m) and permeability (μ); The permeability (μ) in the International system is measured in (Henry/m). Susceptibility is the increase in magnetic moment caused by an applied field, whereas permeability is the proportional increase in flux caused by the presence of the magnetic material. At least one of these types of magnetism exists in most materials, and the behavior is determined by the reaction of atomic magnetic dipoles and electron to an external applied magnetic field.

A magnetic field, like gravitational and electrical fields, is a force field that surrounds a source of potential and has a contoured sphere of effect or field. The parent of the magnetic potential is known as magnetic dipoles. The two electronic movements that accompany the atom and cause magnetism are the orbital movement of the electron and the spin movement of the electron. For macroscopic purposes, these little currents created by these movements may be termed magnetic dipoles; however, due to the random orientation of the atoms, they usually cancel each other out. A net alignment of these magnetic dipoles occurs when a magnetic field is applied, and the medium becomes magnetically polarized [43, 44].

2.3 Types of Magnetic Materials

The most common magnetic material classes, as shown in table 2.1, are diamagnetism, paramagnetism, ferromagnetism, antiferromagnetism, and ferrimagnetism [45]. Magnetic behavior can be utilized to classify all materials. All of the periodic table elements magnetic properties are accounted for at room temperature by the most common two forms of magnetism materials: diamagnetic and paramagnetic. Usually, these

elements are referred to as not magnetic, whereas ferromagnetic are in fact those which are referred to as magnetic materials. The other form of magnetism is the only antiferromagnetism seen in pure elements at room temperature. Finally, magnetic materials can be classified as ferrimagnetism, however this is only seen in compounds like Barium ferrite, not in pure elements (BaFe₁₂O₁₉).

Type of Magnetism	Susceptibility	Example
Diamagnetism	Small & negative	Au, Cu
Paramagnetism	Small & positive	Pt, Mn
Ferromagnetism	Large & positive, function of applied field, microstructure dependent	Fe, Co
Antiferromagnetism	Small & positive	Cr, MnO
Ferrimagnetism	Large & positive, function of applied field, microstructure dependent	BaFe ₁₂ O ₁₉ , Fe ₃ O ₄

Table 2.1 Types of Magnetism [45].

2.3.1 Diamagnetic Materials

Diamagnetism, or a weak, negative magnetic susceptibility, can be found in all materials to some extent. In a diamagnetic substance, a magnetic field induces a magnetic moment that opposes the applied magnetic field that generates it [46]. A diamagnetic substance's atoms have no net magnetic moment when no applied field exists. The electrons spin under the influence of an applied field (H), and this motion, which is a type of electric current, produces a magnetization (M) in the opposite direction of the applied field. The diamagnetism effect can be found in all materials, however it is typically overshadowed by the bigger paramagnetic or ferromagnetic word. The value of susceptibility is unaffected by temperature. Michael Faraday discovered and named diamagnetism in September 1845 [45]. Figure 2.1 depicts the atomic magnetic dipole configuration for a diamagnetic material in the presence and absence of an external field. In the figure, the arrows represent atomic dipole moments.



Figure 2.1 Atomic dipole configuration with and without an external magnetic field for a diamagnetic materials [47].

2.3.2 Paramagnetic Materials

Some of the atoms or ions in this class of materials exhibit a net magnetic moment due to unpaired electrons in partially filled orbitals. When a paramagnetic material is exposed to an external magnetic field, it becomes magnetic. In the presence of a field (Figure 2.2), the atomic magnetic moments are partially aligned in the field's direction, resulting in net positive magnetization and susceptibility χ_m is > 0 (order of 10⁻⁵ to 10⁻²) for paramagnetic materials [48]. Thermal motion becomes more important as the temperature rises, and paramagnetic materials susceptibility decreases. Higher temperatures necessitate bigger fields to produce the same magnetization because thermal agitation affects the alignment of magnetic moments. The temperature dependence of paramagnetic susceptibility is described by the relationship (2.1).

$$\chi = \frac{C}{T} \tag{2.1}$$

This behavior is known as the Curie law, where C is a material constant called the Curie constant [49].

Actually, the Curie law is a subset of the more general Curie-Weiss law (equation 2.2), which contains a temperature constant (θ) and is derived from the Weiss theory, which was suggested for ferromagnetic materials and involves the interaction between magnetic moments.

$$\chi = \frac{C}{T - \theta} \tag{2.2}$$

In this equation, (θ) might be positive, negative, or zero. The Curie-Weiss law is identical to the Curie law when $\theta = 0$. When θ is non-zero, nearby magnetic moments interact, and the material becomes paramagnetic only above a certain transition temperature. If θ is positive, the material is ferromagnetic below the transition temperature, and the value of θ corresponds to the transition temperature (Curie temperature, T_c). If θ is negative, the material is antiferromagnetic below the transition temperature (Néel temperature, T_N), but the value of θ has nothing to do with T_N . It is vital to keep in mind that this equation only works when the material is paramagnetic. Because the electrons that contribute to the magnetic moment aren't in a fixed location, it doesn't apply to many metals. The law does, however, apply to specific metals, such as rare earths. Above the T_c , ferromagnetic materials become paramagnetic. Liquid Oxygen, Aluminum, Barium, Uranium, Aluminum, Platinum, Sodium, Strontium, and Calcium are all paramagnetic materials [45].



Figure 2.2 Atomic dipole configuration with and without an external magnetic field for a paramagnetic materials [47].

2.3.3 Ferromagnetic Materials

Ferromagnetism is a phenomenon of spontaneous magnetization. The alignment of a considerable part of molecular magnetic moments in the crystal in a proper direction characterizes it. Ferromagnetism is only visible below a certain temperature, known as the Curie temperature. Moments are directed randomly above Curie temperature, resulting in zero net magnetization [50]. A ferromagnet has a spontaneous magnetization even when no applied field is present. The magnetic moments are focused in one direction alone [46]. Transition metals such as Gd, Fe, Ni, and Co are ferromagnetic, but other elements and alloys containing transition or rare-

earth elements are also ferromagnetic due to their 3d and 4f shells. These materials have a large and positive magnetic susceptibility to an external magnetic field. They exhibit a powerful attraction to magnetic fields and are able to retain their magnetic properties after the external field is removed. As the heat agitation rises, the substance eventually becomes paramagnetic. Magnetic susceptibilities of ferromagnetic materials (Gd,Fe,Ni,Co) approach 10⁶. Ferromagnetic materials act like paramagnetic materials at the aforementioned temperature, and their susceptibility is determined by the Curie-Weiss equation [51].

Defined as

$$\chi = \frac{C}{(T - \theta_f)} \tag{2.3}$$

where C: material constant, T : temperature, θ_f : Curie temperature.

Only when atoms are arranged in a lattice and their atomic magnetic moments interact to align parallel to one another does ferromagnetism arise (Figure 2.3). even in the absence of an external field [49].



Figure 2.3 Mutual alignment of atomic dipoles in a ferromagnetic material, which exists even when no external field exists [47].

2.3.4 Antiferromagnetic Materials

Chromium is the sole element in the periodic table that is antiferromagnetic at room temperature. Antiferromagnetic materials are similar to ferromagnetic materials, but the atomic magnetic moments align anti-parallel due to the exchange interaction between neighboring atoms. As a result, the magnetic field cancels out, and the material behaves similarly to a paramagnetic material. These materials like ferromagnetic materials become paramagnetic above a transition temperature, known as the Néel temperature, T_N . (Cr: $T_N=37$ °C) [45]. A crystal lattice is subdivided into two or more atomic sublattices which order in such a way that their net magnetization is zero (Figure 2.4). The antiferromagnetic axis along which the two sublattice magnetizations lie is determined by magnetocrystalline anisotropy, and the magnetic response below T_N depends on the direction of H relative to this axis. An anti-ferromagnetic has no form anisotropy due there can be no demagnetizing field when M =0 [52].



Figure 2.4 Antiparallel alignment of spin magnetic moments for antiferromagnetic materials [47].

2.3.5 Ferrimagnetic Materials

Ferrimagnetism is only found in compounds with more complicated crystal structures than pure elements. Within these materials in that the exchange coupling between adjacent magnetic ions leads to antiparallel alignment of the localized moments. The material divides into magnetic domains similarly to ferromagnetic materials, and the magnetic behavior is nearly identical, with the exception that ferrimagnetic materials have lower saturation magnetizations (Figure 2.5). For example in this kind of magnetism occurs in materials such as ferrites which are basically the oxides of various metal elements. These ionic materials may be represented by the chemical formula MFe_2O_4 , in which M represents metallic elements. The prototype ferrite is Fe_3O_4 or $FeO-Fe_2O_3$, the mineral magnetite. Ferrimagnetic materials become paramagnetic above a certain Curie temperature [45].



Figure 2.5 The spin magnetic moment configuration of Fe^{2+} and Fe^{3+} ions in Fe_3O_4 (Ferrimagnetism) [47].

2.4 Composite Materials

A composite material is made by combining two or more materials with vastly different properties that do not dissolve or mix. The different components of the composite work together to give it its specific characteristics [53]. Today, the composites industry is continuously evolving, with much of the growth focused on renewable energy [54-57]. Fibreglass was the first modern composite material, and it has a number of advantages, including corrosion resistance, strength, light weight, durability, improved productivity, design flexibility, and lower material costs. It is the most extensively used in composite applications [58]. A composite material consists of three phases: a continuous matrix, discontinuous or dispersed reinforcements, such as fibers and particles, and a fine interphase area, commonly referred to as the interface [59, 60]. The continuous phase is the matrix, which can be formed of polymer, metal, or ceramic. Polymers have a low strength and hardness, metals have intermediate strength and hardness but high ductility, and ceramics have high strength and hardness but are brittle. The matrix (continuous phase) is in charge of a variety of crucial functions, including maintaining the right orientation and spacing of the fibers as well as protecting them from abrasion and the environment. Fibres are the most commonly used in composite applications, and they have the greatest impact on the properties of composite materials. Particulate composites are often weaker and less rigid than continuous fiber composites, although they are usually far less expensive. Continuous fibers have lengthy aspect ratios, while discontinuous fibers have teeny aspect ratios. Discontinuous fibers, on the other hand, have a random direction, but continuous fiber composites often have a preferred orientation. Continuous reinforcements include unidirectional, woven cloth, and helical winding, whereas discontinuous reinforcements include chopped fibers and random mat. The mechanical characteristics of the final composite material can be greatly influenced by the interface between the matrix and reinforcing phases. The interface can be studied by a number of surface analysis methods and the interaction modelled [61, 62].

2.5 Silicon

Swedish chemist Jones Jacob Berzelius was the first to isolate silicon in 1824 [63]. Silicon is a principal ingredient of glass, a low-cost material with good mechanical, optical, thermal, and electrical properties. For hightemperature work, it's a beneficial refractory material [64]. The most common component of sand is silicon oxide, which is a critical component in microelectronics and computer chips. Silicon, the seventh most abundant element in the universe and the second most abundant element on the planet, after oxygen in nature, silicon is no longer (Figure 2.6). Silicon dioxide, often known as silica, is formed when it is combined with a pair of oxygen molecules. Non-crystallized silica makes up quartz, a major component of sand. Silicon is neither a metal nor a non-metal; it's a metalloid, a substance that falls somewhere in between the two. Metalloid is a bit of a gray area, with no clear definition of what qualifies, despite the fact that metalloids share features with both metals and non-metals. They have a metallic appearance, but only conduct electricity to a limited extent. Silicon is classified as a semiconductor, which implies it can conduct electricity. Unlike an instance metal, however, silicon enhances its capacity to conduct electricity as temperature rises (metals get worse at conductivity at higher temperatures) [63]. Silicon dioxide (SiO₂) exhibits substantial magnetic properties at room temperature. The dazzling white brightness of porous silicon is also well-known [63, 65]. Silicon is found in a variety of low-tech items, including bricks and ceramics. But it's in the high-tech stuff where the element really shines. Silicon is used to make transistors, which amplify or convert electrical currents and constitute the backbone of electronics ranging from radios to smartphones. Silica is the only stable oxide of silicon, and has the chemical formula SiO₂. Silica is not a silicon atom with two dual bonds to two oxygen atoms. Silica is made up of one silicon atom linked by four single bonds to four oxygen molecules [63].



Figure 2.6 The structure of silicon dioxide.

2.6 Ferrite Materials

Ferrites are magnetic materials made up of oxides that contain iron oxide as a component [35]. Ferrites, which have a dark brown or black hue and are exceedingly hard and brittle, are electrically non-conductive ferrimagnetic ceramic materials [66]. Ferrites have long piqued the interest of scientists and technologists because they are magnetic semiconductors. Both semiconductors and magnetic materials with high resistivity and low eddy current losses with magnetic losses and low dielectric have electrical properties that could be used in electronic devices [67-69]. Spinel, garnet, and hexagonal ferrite are the three most common forms of ferrites, based on their crystal structure and constructions [70].

2.6.1 Spinel Ferrite

In 1915, Bragg [71] and Nishikawa independently discovered the spinel ferrite structure [72]. The typical formula for spinels is Mo. Fe₂O₃, where M is a divalent metal ion like Mn^{2+} , Ni^{2+} , Fe^{2+} , Co^{2+} , Zn^{2+} , Cu^{2+} . The spinel lattice is made up of a close-packed oxygen arrangement in which 32 oxygen ions form the unit cell. These anions are packed in a face-centered cubic (FCC) arrangement, which leaves two types of spaces between anions: tetrahedral coordinated sites (A), which are surrounded by four nearest oxygen atoms, and octahedral coordinated sites (B), which are surrounded by six nearest neighbor oxygen atoms. The unit cell has 32 oxygen ions, 64 tetrahedral sites, and 32 octahedral sites. Metal ions occupy just 8 tetrahedral places and 16 octahedral sites, resulting in an electrically unbiased structure. Magnetically, spinel ferrites exhibit a ferrimagnetic ordering. The magnetic moments of the cations at the A and B-sites are aligned parallel to one another. Because there are twice as many B-sites as A-sites, the crystal has a net moment of spins yielding, resulting

in ferrimagnetic ordering. The ion distribution between sites A and B, as well as the metal cation used, demonstrate an adjustable magnetic system [73]. Spinel ferrites are classified into three categories based on cation distribution: normal, inverse, and random spinel ferrites.

2.6.1.1 Normal Spinel

A material with the typical spinel structure has M^{2+} on tetrahedral sites and Fe³⁺ on octahedral sites, (M²⁺) [Fe³⁺], structural formula of such ferrites is M^{2+} [Fe₂ ³⁺] O_4^{2-} . In zinc ferrites Zn²⁺ [Fe²⁺ Fe³⁺] O_4^{2-} , this sort of distribution occurs [74].

2.6.1.2 Inverse Spinel

In an inverse spinel structure, all M^{2+} ions are found on B sites, whereas Fe^{3+} ions are uniformly distributed over A and B sites per unit cell. Ferrimagnetic properties are present in these ferrites. $Fe^{3+}[M^{2+}Fe^{3+}]O_4^{2-}$ is the formula for an inverse spinel structure. $CoFe_2O_4$ and $NiFe_2O_4$ ferrites has inversed spinel structure [75].

2.6.1.3 Random Spinel

In this case, M^{2+} ions and Fe^{3+} ions are distributed randomly over A and B sites. The general relation may be used $M_{1-\delta}^{2+}Fe_{\delta}^{2+}[M_{\delta}^{2+}Fe_{2-\delta}^{3+}]O_4^{2-}$ to represented the whole range of possible M^{2+} and Fe^{3+} ions distribution on A and B sites, where δ is a metric for the degree of inversion between positive ion ratios M^{2+} and Fe^{3+} , an example of this type is manganese ferrite with the formula $[M_{0.8}^{2+}Fe_{0.2}^{2+}]_{tet}[M_{0.2}^{2+}Fe_{1.8}^{3+}]_{oct}O_4^{2-}$ [76].

2.6.2 Garnet Ferrite

Garnets are a type of ferrimagnetic material that can be used in a wide range of high-frequency applications. Yttrium iron garnet ($Y_3Fe_5O_{12}$) is a well-known garnet ferrite that has attracted a lot of interest due to its technological importance in a number of applications such as phase shifters, high-quality filters, circulators, phase isolators, and a variety of electronics magnetic optical devices (Figure 2.7) [77]. Garnet ferrites ($A_3B_5O_{12}$) have intriguing and unique electromagnetic, mechanical, thermal, and magneto-optical properties [78]. In this structure, there are three types of sites: (a), (b), and (c). It has been found that among three lattice sites $24Fe^{3+}$ ions occupy tetrahedral sites, $16Fe^{3+}$ ions occupy octahedral sites and $24R^{3+}$ ions goes on the dodecahedral sites, whereas oxygen ions are distributed to the interstitial sites [79].



Figure 2.7 Unit cell of yttrium iron garnet (Y₃Fe₅O₁₂)[80].

2.6.3 Hexagonal Ferrite

Ferrites have been used in a range of technological applications due to their unique structural and magnetic properties. Such ferrites are used in many home appliances, stealth technologies, electrical devices, radar absorbent materials, electronic equipment, high frequency applications, and electric motors [81]. Because of their high magnetic, high Curie temperature, large coercivity, large resistivity, and other properties, hexaferrites are among the most important ferrites. Hexagonal ferrites having the formula $MFe_{12}O_{19}$ (M = Sr, Ba), where M is usually replaced with another divalent element (Figure 2.8). Hexaferrites are classified into six categories based on their crystal structure and chemical compositions: M, W, X, Y, Z, and U. M-type hexaferrite belongs to the family of ferrimagnetic oxides, which have various properties such as strong corrosion resistance and low manufacturing costs, as well as superior electrical resistance, chemical stability, large dielectric and magnetic loss, and so on. Hexagonal ferrites, in comparison to garnet ferrite, have a lot of ions and are created by substituting oxygen ions. Br and Sr make up the majority of these larger ions [82, 83].



Figure 2.8 Structure of barium hexaferrite [84].

2.7 Magnetization of Ferrite

Soft and hard ferrite can be separated into two groups based on the persistence of their magnetization. This classification is based on whether they can be magnetized or not [85].

2.7.1 Soft Ferrite

Soft magnetic materials are easy to magnetize and demagnetize. Soft magnetic materials have a low coercive field and a high magnetization, which are both relevant in a range of applications. Because they have a tight hysteresis loop (Figure 2.9-b), the magnetization nearly follows the applied field disparition without hysteresis loss. Ni, Fe, Mn, Co, and other metals, for example, are used in inductors, transformer cores, microwave devices, recording heads, and medical devices [86]. Soft ferrites have a cubic structure and magnetic properties such as saturation, resonance frequency in megahertz, excellent chemical resistance to oxidation, low coercivity, and high magnetic permeability. Because of these advantages, soft ferrites are superior than all other magnetic materials [87].

2.7.2 Hard Ferrite

The difficulties of magnetizing and demagnetizing hard ferrites is well known. They're employed as permanent magnets. The hysteresis loop in hard ferrite is quite large, and the coercivity is quite high (Figure 2.9-a). Household products such as refrigerator magnets and alnico are examples of rare earth metal alloys. The development of permanent magnets began in the 1950s with the progression of hard ferrites. The magnets can also be used in permanent magnet motors if they are exposed to sufficient demagnetizing fields. Both strontium ferrite SrFe₁₂O₁₉ and barium ferrite BaFe₁₂O₁₉ contain hexagonal ferrite. These hexagonal ferrites have the magneto-plumbite crystal structure PbFe₁₂O₁₉, while Strontium ferrites have significantly better magnetic properties [49].



Figure 2.9 Comparison of hysteresis loops for: (a) hard ferrites; (b) soft ferrites[88].

2.8 Magnetic Domains

In 1907, Pierre Weiss was the first to characterize the magnetic structure of a bulk-ferromagnet by the existence of sub-domains comprising of aligned moments inside each domain but unaligned moments between various domains (Figure 2.10) [89]. In this sight, extremely turbulent areas known as domain walls clearly divide these magnetic domains. This multidomain structure is favoured as long as the magnetostatic energy is greater than the domain wall energy. Because the magnetostatic and the domain wall energy proportional to the volume and the surface of the material, respectively, the S/V ratio determines the relative extent of these two energy contributions. Particularly when the particle size is decreased to a critical value known as "critical diameter" (D_c) , which is generally within a few tens of nanometers and depends on the material properties. Because of the high wall domain energy, a multi-domain magnetic structure is no longer energetically convenient due to the contribution from multiple anisotropy energy terms. To make it smaller, a single-domain magnetic structure is produced, in which each particle is made up of a single evenly magnetized zone and is defined by a magnetic moment [90].

According to the equation, the critical diameter, D_c , is reliant on material properties such as anisotropy constants (A, K_{eff}) , μ_o the vacuum permeability, and saturation magnetization (M) [91].



Figure 2.10 Schematic illustration of a ferromagnetic particle's interior structure, with Bloch barriers between Weiss domains [92].

2.9 Hysteresis Loop and Magnetic Parameters

The hysteresis loop can teach us a lot about magnetic materials. A hysteresis loop illustrates the relationship between the induced magnetic flux density (B) and the magnetizing force (H). It is often referred to as the B-H loop (Figure 2.11) [93].



Figure 2.11 Hysteresis loop in magnetic materials [94].

When measuring the magnetic flux of a ferromagnetic material, the loop is generated by adjusting the magnetizing force. A ferromagnetic substance that has never been magnetized or is totally demagnetized will follow the dashed line when H is increased. The stronger the magnetic field in the component (B+), as represented by the line, the higher the amount of current applied (H+). Because nearly all of the magnetic domains are aligned at point "a," increasing the magnetizing force will only result in a minor increase in magnetic flux. This area is known as the domain growth region. The magnetization is on the verge of reaching saturation. When the magnetic field returns to zero, the curve will movement from point "a" to point "b." There is still some magnetic flux in the material at this point, even though the magnetizing force is zero. The remanence or level of residual magnetism in the material is shown by the point of retentivity on the graph (The magnetic field no longer aligns the spins). As the magnetizing force inverts, the curve moves to point "c," where the flux has been reduced to zero. On the curve, this is known as the point of coercivity. (Because the invert magnetizing force has flipped enough domains in the material, the materials net flux is zero). The materials coercive force, or coercivity, is the force required to remove the remaining magnetism from the material. When the magnetizing force is increased in the negative direction, the material will once more become magnetically saturated (M_s) , but in the opposite direction (point "d"). When H is set to zero, the curve reaches point "e." It will have the same level of residual magnetism (M_r) as it does in the opposite direction. Restoring (B) to zero by increasing (H) in a positive direction. The curve did not return to the graph's origin because residual magnetism required some force to eliminate. The curve will take a different path back to the saturation point from point "f," completing the loop [94]. The most commonly used parameters to define the characteristics of a specimen are saturate magnetization (M_s) , remanent magnetization (M_r) , and coercive field (H_c) . The remnance or squareness ratio, which is the ratio of remnant magnetization and saturation magnetization, is a characteristic parameter of magnetic materials that indicates how the direction of magnetization reorients to the nearest easy axis magnetization direction after the magnetic field is turned off [95], as shown by the following relationship:

Remnance ratio =
$$\left(\frac{M_r}{M_s}\right)$$
 (2.5)

The magnetic moment (n_B) per unit at the magnetic moment can be calculated with the assistance of the saturation magnetization value from the following relationship [96].

$$n_B = \frac{M_{wt} x M_s}{5585} \tag{2.6}$$

where, M_{wt} is the molecular weight.

Further, the anisotropy constant is evaluated using following relation [96].

$$H_c = \frac{0.96 \ x \ K}{M_s} \tag{2.7}$$

where, K is magnetic anisotropy, H_c is the coercivity.

2.10 Vibrating Sample Magnetometer

For decades, the vibrating sample magnetometer (VSM) has been the most widely used method of measuring magnetic moment. Unlike an alternating gradient magnetometer, the VSM is unconcerned about specimen mass and size up to a certain range. The specimen to be studied is placed in a constant magnetic field in this scheme, and the specimen is magnetized as a result of the constant magnetic field, as shown in this diagram. The external magnetic field is disturbed by the oscillation of the magnetized specimen. To measure these disturbances, a collection of coils or some magnetic field sensors can be placed around the specimen. For example in the case of coils, magnetic flux piercing the coils will change resulting in the formation of an electro motive force (emf) in coils. The electro motive force (emf) created in coils is dependent on (a) the external magnetic field, (b) the frequency and amplitude of vibration, and (c) the magnetization of the specimen for a given coil shape. We can deduce the value for magnetisation from electro motive force (emf) with proper manipulation. Figure 2.12 illustrates a VSM schematic [97, 98].



Figure 2.12 VSM schematic [99].

2.11 Chemical Composition of Spinel Ferrite

Spinel ferrites, which have a general chemical composition $MeFe_2O_4$, offer favorable magnetic, optical, and electrical properties [100]. MgAl₂O₄, also known as Spinel, is the source of the spinel structure. Molecules of the spinel structure have the general formula $MeFe_2O_4$, where Me is a divalent transition metal or a mixture of transition metal ions (Co, Mn, Ni, Fe, Cu, and Zn), which are likely either magnetic or non-magnetic and have an ionic radius of 0.6 to 1Å. It is possible to combine of these ions and it can be named as a solid solution of two ferrites or mixed spinel ferrites, meaning that a wide range of spinel oxides is possible [101, 102]. Among these oxides, magnetite Fe₃O₄ is an important compound, the spinel ferrites generated by the trivalent iron ions (Fe³⁺) in MeFe₂O₄ can be replaced fully or partially by another trivalent ion such as Cr^{3+} or Al^{3+} , resulting in mixed crystals including chromites and aluminates. If a large number of nonmagnetic ions are absent, these compounds are also ferrimagnetic at room temperature. When ferric ions are substituted with a tetravalent ion like Ti^{4+} , Fe^{3+} is transformed to Fe^{2+} in the same proportion. In ferrimagnetic oxide with spinel structure, a large range of chemical compositions is possible [103].

2.12 Crystal Structure of Spinel Ferrite

The crystal structure was determined by Bragg and independently by Nishikawa [104]. A mineral with the chemical formula $MeFe_2O_4$ is known as spinel. Spinel has a face-centered cubic crystal structure. The structure

is derived from that of mineral spinel. Each unit cell has eight formula form interstitial sites of two types: four oxygen neighbors (tetrahedral or A-sites) and six oxygen neighbors (octahedral or B-sites) per unit cell. In all there are 64 tetrahedral and 32 octahedral sites, with cations taking up 8 of the tetrahedral sites as illustrated in figure 2.13. The valency combinations 6-1 (rare), 4-2, and 2-3 are possible because one Me cation and two Fe cations must have a total positive charge of eight units. In the figure, the valencies of the Me and Fe^{3+} cations are depicted [105-107]. The cubic unit cell is made up of 56 atoms: 32 oxygen anions arranged in a cubic close-packed structure and 24 cations occupying eight of the 64 possible tetrahedral sites (A- sites) and sixteen of the 32 possible octahedral sites (B-sites). As a result, the cell contains eight formula units per cubic unit cell (Z=8). The location of (A-sites) and (B-sites) is always the identical and is independent on the nature of component cations. On the other hand, the equilibrium distribution of cations in the spinel structure is determined by electrostatic energies, electronic configuration, and ionic radii, whereas the nonequilibrium distribution of cations in the spinel structure can influence the intrinsic magnetic properties of these materials [108].



Figure 2.13 Structure of cubic spinel ferrite [109].

2.13 Cobalt Ferrite (CoFe₂O₄)

Because of its potential applications in high-density magnetic recording, magnetic nanoparticles have gotten a lot of attention. Because of its extraordinary chemical stability and mechanical hardness, cobalt ferrite (CoFe₂O₄) has been investigated extensively among the numerous ferrite materials. The ability to control crystal size within superparamagnetic and single domain limits is critical to the practical application of cobalt ferrite (CoFe₂O₄) [39]. Cobalt ferrite (CoFe₂O₄) is a hard magnetic material with low eddy current loss and significant magnetic anisotropy, as well as moderate magnetization and high coercivity. Cobalt ferrite is suitable for a wide range of applications, including videotape and audio, high-density digital recording disks, and so on, due to these characteristics, as well as its good physical and chemical stability [110]. The magnetic properties of $CoFe_2O_4$ are influenced by the distribution of iron (Fe) and cobalt (Co) ions in the A and B sites. Magnetic anisotropy and magnetic moments can be significantly altered by even minor changes in the cationic distribution [111]. $CoFe_2O_4$ nanoparticles have an effective uniaxial anisotropy, according to a system of non-interacting single domain particles. And ferrite powder coercivity is strongly dependent on annealing temperatures and may be related to the sizes difference between cobalt ferrite particles [39].

2.14 Magnetic and Electric Properties of Cobalt Ferrite

In ferrites, the metallic ions occupy two crystallographic sites, i.e. the tetrahedral (A site) and the octahedral (B site). Three forms of magnetic interactions between metallic ions are produced by the super exchange mechanism: A-A interaction, B-B interaction, and A-B interaction. These interactions occur occasionally via anion and occasionally via direct interaction [112]. For A-B interaction, small distances exist in two formations. Only the first of the two B-B interaction formations works, since the distance between the metal ion and the oxygen ion in the second formation is too great. The A-A interaction is the weakest due to the great distance. As a result, A-B interactions are the most important. The ferromagnetic crystal lattice, according to Neel, can be divided into two sub-lattices: tetrahedral (A-site) and octahedral (B-site). The individual moments of the A and B-sub-lattice arrange themselves anti-parallel (opposite direction) to one another in figure 2.14. Within an A or B sublattice, however, individual magnetic moments are arranged parallel to one another [113]. The resultant saturation magnetization M_S is expressed as $M_{S} = M_{B} - M_{A}$ because the saturation magnetic moments of the B-site (M_B) are larger than those of the A-site (M_A). This non-equilibrium distribution amends the cations ratio at octahedral and tetrahedral sites, reducing the tetrahedral-octahedral interaction while improving the octahedraloctahedral interaction [114]. Ferrites are also excellent dielectric materials, with a high electrical resistance that makes them appropriate for highfrequency applications. They are also used as microwave absorbers. Some ferrites absorb microwave by losing interaction of the electric and magnetic field vector of the incident wave and in the process transfer microwave

energy into thermal energy. The order of magnitude of electrical conductivity has a significant impact on the dielectric and magnetic behavior of ferrites. Cobalt ferrite has an inverse spinel structure, with the degree of inversion depending on the thermal treatment [115].



Figure 2.14 The ferrimagnetic order of a partial spinel ferrite unit cell [73].

2.15 Dielectric Materials

Dielectrics are materials with a low conductivity (typically oxides). If it has the ability to store energy when an external electric field is applied. These qualities vary based on mixing, orientation, temperature, frequency, pressure, and the molecular structure of the substance. The bulk density of the air-particle mixture is another component that influences the dielectric properties of grain or particulate materials. Of course, the dielectric properties of a material are heavily influenced by its chemical composition, particularly the permanent dipole moments associated with any molecules that make up the substances of interest. There is no net dipole moment in the majority of solids because there is no net separation of positive and negative charges. Solids have molecules organized in such a way that the unit cell of the crystal has no net dipole moment. When an electric field is applied to such a solid, it induces a field in the solid that is the polar opposite of the applied electric field. This field is formed by two factors: a distortion of the electron cloud of the atoms or molecules, and the motions of a few atoms themselves. The average dipole moment per unit volume induced in a solid is characterized as electrical polarization (P) and is proportional to the applied electric field (E) [116]. The polarizability (α) of the dielectric material is determined by

$$P = \alpha E \tag{2.8}$$

Where P is the dipole moment induced by local electric field, (E).

The Polarizability is split into four types, each with its own process dependent on the sort of dipole moment established.

$$\alpha = \alpha_e + \alpha_i + \alpha_o + \alpha_s \tag{2.9}$$

where, α_e is electronic polarizability, α_i is an ionic polarizability, α_o is orientation polarizability and α_s is space charge polarizability [117].

2.16 Polarization

If electric field is applied to a medium made up of large number of atoms or molecules, the positive charge center is relocated along the direction of the applied field, while the center of the negative charge is displaced in the opposite direction. As a result, the medium gets polarized. Electric polarization is defined as the dipole moment per unit volume. Polarization is a macroscopic amount because it involves averaging of the dipole moments over a volume which contains much dipoles [118].

With an externally applied electric field, polarization is induced via a number of atomic mechanisms. Electron polarization, ionic polarization, orientation polarization, and space charge polarization are the four basic types of dielectric polarization. Depending on the materials and the method of external field application, dielectric materials normally exhibit at least one of these polarization types [119].

2.16.1 Electronic Polarization

Electronic polarizability occurs in all solids, and in some, such as glass, it is the only contribution to the dielectric constant due to the lack of additional contributors [48]. Electrons and positively charged nuclei will be displaced in such a way that electrons move in the positive field direction and nuclei in the opposite direction, as shown in figure 2.15-a, or is defined as the dipole moment per unit volume. This is given by the relation:

$$P_e = N\alpha_e E_{loc} \tag{2.10}$$

where *N* is the number of atoms or molecules, α_e is the electronic polarizability, and E_{loc} is the local electric field that is exposed to the atom or molecules. Thus the applied field is proportional to the induced dipole moment [120, 121].

2.16.2 Ionic Polarization

Only dielectric materials with ionic bonds between atoms exhibit ionic polarization. When a solid is exposed to an external electric field, the anions and cations in the substance separate. Leading to a net dipole moment. Figure 2.15-b depicts the situation. It is the principal source of polarization in ionic crystals, and ionic polarisability is determined by the relationship

$$P_i = N\alpha_i E_{loc} \tag{2.11}$$

where N is the number of formula units per unit volume, and α_i is the ionic polarizability.

Induced dipoles are formed by both electronic and ionic polarization [120, 121].

2.16.3 Orientation Polarization

If the material contains complex ions or molecules with a permanent dipole moment, the dipoles will align in the field direction. As seen in figure 2.15-c, the net effect will be to induce a dipole moment within the solid. Non-polar materials lack this polarizability [122]. The orientation polarization is give by

$$P_o = N\alpha_o E_{loc} \tag{2.12}$$

A polar material may have a zero dipole moment due to thermal randomization of dipoles, but an applied electric field will yield a net dipole moment. As a result, polarization is closely related to the temperature-dependent thermal motion of molecules [120, 121, 123].

2.16.4 Space-Charge Polarization

Space-charge polarization, also known as interfacial polarization, is the last type of polarization. Space charge occurs when charge accumulates at the electrodes at the interface in a multiphase material, as shown in figure 2.15-d. In response to the applied field, the ions spread over the concrete distance. This leads to a redistribution of charges in the dielectric medium. Not just in semiconductor field-effect devices, but also in ceramics with electrically conducting grains and insulating grain borders, space charge polarization is critical, especially at high temperatures. Space polarization



occurs at low frequencies because it takes a long time. Space-charge polarization is also known as migrational polarization [120, 124].

Figure 2.15 Schematic diagram of (a) Electronic polarization, (b) Ionic polarization, (c) Orientation polarization, (d) Space-charge polarization [120].

The total polarization of dielectric substances is the sum of all of the aforementioned contributions, as determined by the relationship.

$$P = P_e + P_i + P_o + P_s \tag{2.13}$$

In contrast to extrinsic contributions, the contributions from the lattice are referred to as intrinsic contributions.

2.17 Dielectric Characterization and Parameters

Materials dielectric properties are important because they show how electromagnetic energy interacts with the material at microwave frequency as well as radio frequency. Therefore, it is fundamental to study their dielectric behavior at different frequencies by using a LCR meter device. The term LCR meter indicates that it is a measuring device for inductance (L), capacitance (C), and resistance (R). LCR meters are commonly used to analyze components such as transformers, inductors, electromechanical devices, and capacitors in a simple and precise manner. For dispersion factor, resistance, and production test environments, the ability of LCR meters to apply specific measurement conditions is critical [125]. The dielectric constant, also known as the relative permittivity, is used to evaluate insulators like plastics, powders, and rubber. It is used to determine the ability of an insulator to store electrical energy.

The dielectric constant is relative permittivity (real part) of the material was calculated from the measured values of capacitance using the formula;

$$\varepsilon' = \frac{C}{C_o} \tag{2.14}$$

where C is the capacitance of the pellet in farad, and C_o is the capacitance of free space, and

$$C_o = \varepsilon_o \frac{A}{d} \tag{2.15}$$

where A is the cross-sectional area of the flat surface of the pellet, d is the thickness of the pellet in meter and ε_0 the constant of permittivity of free space and has a value of (8.854 x 10⁻¹² F. m⁻¹).

The equation 2.15 becomes

$$\varepsilon' = \frac{Cd}{\varepsilon_o A} \tag{2.16}$$

The dielectric constant is calculated using this equation using the observed capacitance of the specimens from the LCR meter. The fact that dielectric is not loss free can generally be denoted by a complex dielectric constant (ϵ), as follows:

$$\varepsilon = \varepsilon' - i\varepsilon'' \tag{2.17}$$

where, ϵ' and ϵ'' are real and imaginary part of a relative permittivity.

At ac voltages, the charge stored on a dielectric has both real (in phase) and imaginary (out of phase) components due to resistive leakage or dielectric absorption. The loss is calculated by dividing the out of phase component by the in phase component. This is the dissipation factor or the dielectric loss, commonly known as the loss tangent or $tan \delta$ in the literature. And the loss tangent may be expressed as [126].

$$tan\delta = \varepsilon'' / \varepsilon' \tag{2.18}$$

The ac conductivity was calculated from the values of dielectric constant and dielectric loss factor using the relationship [127]:

$$\sigma_{ac} = \omega \varepsilon_o \varepsilon'' \tag{2.19}$$

where σ_{ac} is the ac conductivity, ω is the angular frequency.

2.18 Electrical Conductivity

Due to its high electrical resistivity, ferrites are useful in a variety of technical applications. The electrical conductivity of spinel ferrites is important because it reveals important details about the conduction mechanism. Electrical conductivity is believed to be caused through the nearest-neighbor hopping mechanism. These systems exhibit a lot of conductivity due to the presence of iron with a different valence state at crystallographically equivalent lattice points. Cation distribution in sites by magnetic and non-magnetic replacements, chemical composition, grain size, sintering circumstances, method of preparation in general and their structure in particular, all have a significant impact on ferrites' electrical characteristics [128]. The formation of a Coulomb gap, according to Phillips et al. [129], is due to electron interactions caused by the large amount of excess charges in Fe²⁺ ions at the B site. On the other hand, with small Fe²⁺ concentrations, electrical conductivity has been explained as depending on nearest-neighbor hopping at high temperatures and on variable range hopping at low temperatures. Ferrites are well-known as good base materials for magnetic devices that run at high frequencies, such as inductor/transformers used in power electronics and magnetic heads of video tape recorders, due to their much reduced electrical conductivity compared to metals [130]. The conduction mechanism of ferrites has been compared to a magnetite (Fe₃O₄) derivative molecule. A metal-insulator transition (Verway transition) occurs at 120 K, which is believed to be connected with electron charge ordering and electron-electron interactions [131]. As a result of the small impurities, the transition temperature drops suddenly or vanishes [132]. As the concentration of ions substituted in magnetite increases, thermally activated Fe²⁺ parity states lead to electrical conduction via a hopping mechanism [130]. A variety of models have been presented to account for the electrical characteristics. Jonker used the hopping conduction model, which is based on localized electron levels, to obtain the mobility equation. A band polaron model based on the electron transition between localized cells has also been presented [133]. Based on the fact that ferrites are ferrimagnetic materials and their magnetic properties would effect their electrical properties.

The variance of conductivity becomes temperature dependent and increase with the increase in temperature according to the general exponential relationship [134].

$$\sigma = \sigma_o e^{\left[-E_a/KT\right]} \tag{2.20}$$

Where σ_o is the pre-exponential constant and E_a is the activation energy, which is the energy required to release an electron from the ion for a leap to the neighbouring ion, and leads to the rise the electric conductivity, and *K* is Boltzmann constant.

2.19 X-ray Diffraction (XRD)

Max von Laue and Co. discovered in 1912 that crystalline substances act as three-dimensional diffraction gratings for X-ray wavelengths similar to the spacing of planes in a crystal lattice [135]. The X-ray diffraction method is a quick analytical technique that can provide information on unit cell dimensions and is commonly used for crystalline material phase determination [136]. An X-ray diffractometer is made up of three basic parts: an X-ray source, a sample holder, and an X-ray detector. X-ray diffraction is based on a crystalline sample and a constructive interference of monochromatic X-rays. These X-rays are generated by a cathode ray tube, filtered to produce monochromatic radiation, collimated to concentrate, and directed toward the sample (Figure 2.16).



Figure 2.16 Schematic of an X-ray diffractometer [137].

A particular arrangement of atoms for each crystalline substance that diffracts X-rays in a distinct pattern. Bragg's diffraction from parallel planes is seen in figure 2.17.



Figure 2.17 Constructive interference from the parallel planes [138].

When Bragg's law is satisfied, incident rays interact with the specimen, resulting in constructive interference (and a diffracted ray) [139].

$$n\lambda = 2d \sin\theta \tag{2.21}$$

where *n* is the order of the reflection and can any whole number, λ is the wavelength of the X-rays, *d* is the spacing between the atomic planes, and θ is the diffraction angle. This law links the wavelength of electromagnetic radiation to the diffraction angle and lattice spacing in a crystalline specimen. As a result, the scanning of range angles of the reflection by a detector is givens a pattern of peaks with specific intensities and spacing [140].

The crystallite size (*D*) can be estimated from the peak with Scherrer's formula [141]:

$$D = \frac{K\lambda}{\beta \cos\theta} \tag{2.22}$$

where *D* is the crystallite size, *K* its value constant (0.9), λ is the X-ray wavelength, θ is Bragg's angle (2 θ) and β the full width at half-maximum (FWHM) in radians.

The lattice constant 'a' is calculated according to the following equation [142]:

$$a = d_{hkl}\sqrt{(h^2 + k^2 + l^2)}$$
(2.23)

where d_{hkl} is the interplanar distance of each plane and (hkl) are Miller indices.

The X-ray density (ρ_x) is dependent on the molar mass of the synthesized compounds and the lattice constant '*a*' which was calculated using the relationship [143]:

$$\rho_x = \frac{8M}{Na^3} \tag{2.24}$$

where M is the molecular weight of the specimens, N is the Avogadro's number $(6.022 * 10^{23} mol^{-1})$ and a is the lattice constant. Each cell has 8 formula units.

The hopping length (L_A and L_B) between magnetic ions (the distance between the ions) in the tetrahedral A-site and octahedral B-site can be calculated using the relations below [144]:

$$L_A = 0.25 \ a\sqrt{3} \tag{2.25}$$

$$L_B = 0.25 \ a\sqrt{2} \tag{2.26}$$

2.20 Fourier Transform Infrared Spectroscopy (FT-IR)

Fourier transform infrared (FT-IR) is the preferred method of infrared spectroscopy. Is a measurement technique that provides detailed information on the rotation, molecular structure, and vibration of chemical bonds, making it useful in a range of analysis. A sample's fingerprint is an infrared spectrum, with absorption peaks matching to vibrational frequencies between the bonds of the atoms making up the material. No two compounds have the same infrared spectrum since each material is made up of a different combination of atoms. As a result, infrared spectroscopy can positively identify each type of material (qualitative analysis), and infrared is a premium instrument for quantitative analysis utilizing modern software algorithms [145, 146]. When IR radiation is passed through a specimen in infrared spectroscopy, the specimen absorbs radiation at frequencies corresponding to molecule vibrational frequencies but passes (transmits) all other frequencies. The frequencies of radiation absorbed are measured by an infrared spectrometer, and the resulting

planner from the absorbed energy against frequency is known as the infrared spectrum of the material. Because different materials have different vibrations and create different infrared radiation, a substance can be identified. Furthermore, it is likely to detect whether distinct chemical groups are present or not in a chemical structure using absorption frequencies [146, 147]. Figure (2.18) shows a schematic of the (FT-IR) spectrometer [148].



Figure 2.18 A Schematic diagram of FT-IR spectrophotometer [148].

2.21 Field Emission-Scanning Electron Microscopes (FE-SEM)

Zworykin developed the first genuine scanning electron microscope (SEM) in 1942, proving that secondary electrons (SE) produced topographic contrast. Recent improvements in electron microscopy have been heavily subsidized by nanotechnology, with demands for not only for increasing resolution but also for more information from the specimen. The field emission scanning electron microscope (FE-SEM) is a type of electron microscope that images the specimen surface by raster scanning over it with a high-energy beam of electrons and high resolution down to about few nanometers [149, 150]. Electrons are liberated from a field emission source and accelerated in a high electrical field gradient. Within the high vacuum column, these so-called primary electrons are focussed and obliqued by electrostatic or magnetic lenses to form a narrow scan beam that shells the target. As a result, secondary electrons are emitted from echo spot on the target. The speed and angle of these secondary electrons were connected to the surface structure of the target. A detector captures the secondary electrons, resulting in an electrical signal. Backscattered electrons, secondary electrons, diffracted backscattered
electrons, photons, visible light, and heat are examples of these signals. This signal is amplified and turned into a video scan image that may be viewed on a monitor or digital images that are easier to store and handle [151]. The job of the electron gun is to deliver a large, constant current in a tiny beam. Two types of emission sources are the thermionic emitter and the field emitter. A field emission gun produces an electron beam with an extremely high current density by applying an intense electric field to a tungsten single crystal with a needle-shaped tip. Unlike the conventional thermal emission gun this requires ultra high vacuum (10^{-10} Torr). The FEG is known as the high brightness gun because the electrons emission source is relatively small and the number of electron guns [152, 153]. The schematic diagram of field emission scanning electron microscopy is shown in figure (2.19) [154].



Figure 2.19 Schematic diagram of a field emission scanning electron microscope (FE-SEM) [154].

Chapter Three

Experimental Part

3.1 Introduction

This chapter discusses the practical steps involved in preparing the ferrite models $Co_xFe_{3-x}O_4$ (x= 0.8, 0.9, 1, 1.1, and 1.2) and $CoFe_2O_4/SiO_2$ (35%, 40%, 45%, and 50%) as well as the materials used in specimens preparation the preparation method, calcination and compression, sintering, structural, electrical and magnetic tests and the devices annexed.

3.2 Materials Used in Preparing Specimens

The materials used in the preparation of the ferrite compound by sol-gel auto combustion method are listed in table 3.1 with their full description.

Sr. No.	Compounds	Chemical formula	Mol. mass (g mol ⁻¹)	Purity %	Supplier
1	Iron (III) nitrate	Fe(NO ₃) ₃ .9H ₂ O	404.00	97.00	India
2	Cobalt (II) nitrate	Co(NO ₃) ₂ .6H ₂ O	291.03	97	India
3	Silicone dioxide	SiO_2	50.08	99.5	USA
4	Citric acid	C ₆ H ₈ O ₇ .H ₂ O	210.14	99.00	USA
5	Ammonia solution	NH ₃	17.03	25	India

Table3.1 Materials used in preparing specimens with percentage purity.

3.3 Instruments Used

Glassware, sensitive electronic balance, magnetic stirrer with hot plate are among the primary instruments used in the production of $Co_xFe_{3-x}O_4$ nanoparticles and $CoFe_2O_4/SiO_2$ nanocomposites (LMS-1003, Daihan Lab Tech, Korea). Stirring can be done simultaneously with heating to maintain homogeneity and the required temperature, digital pH meter (Starter 2000,Ohaus, USA) to maintain the pH of the reaction mixture, electric furnace (muffle furnace LAC, LMH 07/12, Kladská) to calcined and sintered the specimens at varied required temperatures, hydraulic press (Specac atlas 15ton, Britain), and mortar with pestle.

3.4 Preparation Method

The steps for preparation are as follows:

- 1- Metals nitrate and citric acid in mole ratio of (citric acid: nitrates=1:1) are dissolved separately in distilled water according to the molar ratios of each specimen.
- 2- The nitrate solution is mixed with the acid solution in a suitable heatresistant flask (Pyrex), and the components are mixed using a magnetic mixer with the acid function set to a value of (pH=7) by adding drops of ammonia.
- 3- Stir the materials together for half an hour at room temperature to ensure homogeneity.
- 4- The temperature is gradually raised and then fixed at 90 °C, with constant stirring until the gel form is achieved.
- 5- The viscous gel of $\text{Co}_x\text{Fe}_{3-x}\text{O}_4$ is placed in the oven and heated to 250 °C, then crushed using an agate mortar to activate an auto combustion reaction and create as-burnt ferrite powder. The flow diagram for the manufacture of ferrite nanoparticles is shown in figure 3.1.
- 6- Using the conventional ceramic method, mixing silica with cobalt ferrite in various amounts at 600 °C.

3.5 Calcinations and Pellet Formation

The resultant powder is placed in a porcelain bowl and then calcined at 500,600,700 °C for 3 hrs to remove reaction leftovers such as water molecules or carbon dioxide coming from combustion, resulting in the necessary ferrite powder. The as-burnt specimens and the specimens that were calcined at 500, 600, and 700 °C added with a four-drop PVA as a binder to press it into circular pellets with a diameter of 13 mm and a thickness of 2 mm. This is accomplished by exerting a pressure of 2 Tons for 1 min using a hydraulic press utilizing the dry pressing method, then sintered at 350, 600, 700, and 800 °C for 3 hrs to intensify of the specimens and, slowly allowed to be cooled naturally to examine the dielectric properties.



Figure 3.1 Flowchart of sol-gel auto combustion synthesis of $Co_xFe_{3-x}O_4$ nanoferrites and $CoFe_2O_4/SiO_2$ nanocomposites.

3.6 Structural Characterization

3.6.1 XRD Measurements

Because each crystalline substance has its own individual powder pattern, powder X-ray diffraction is an important technique that has been widely employed in qualitative phase analysis; this method is frequently referred to as the powder fingerprint method. In the middle of DayPetronic co., Tehran, Iran, the crystal structure test were carried out of the synthesized $Co_x Fe_{3-x}O_4$ ferrite nanoparticle specimens (x = 0.8, 0.9, 1, 1.1, and 1.2) for as-burnt and calcined powders at various temperatures 500,600, and 700 °C and CoFe₂O₄/SiO₂ nanocomposites (35%, 40%, 45%, and 50%) at 600 °C were characterized at room temperature. Measurements are made with an X-ray diffraction powder (XRD type PANalytical (X'pert Pro, Netherlands) fitted with a high-intensity source of Cu k α radiation $(\lambda=0.154 \text{ nm}, 40 \text{ mA}, 40 \text{ kV} \text{ in the } 2\theta \text{ range } (20^{\circ}-70^{\circ})$. The X-ray Diffractometer instrument utilized in this study is shown in figure (3.2). By comparing the results with the international centre for diffraction data (ICDD) cards, it is possible to verify the accuracy of the resulting ferrite, and this technique can also give other important structural information such as diffraction angles (2θ) , the perpendicular distance between $(hk\ell)$ miller planes (d_{hkl}) as well as the full width at half maximum (FWHM).



Figure 3.2 Photograph of X-ray diffraction instrument.

3.6.2 Fourier Transform Infrared Spectroscopy (FT-IR)

Fourier transform infrared spectroscopy is used to investigate the produced ferrite structures structurally. The obtained (FTIR) spectrum is utilized to determine absorption band positions. Fourier transforms infrared spectroscopy spectra for as-burnt and calcinated powders at various temperatures 500, 600, and 700 °C and calcinated powders at temperature 600 °C in the wave number range of 300 to 4000 cm⁻¹ were obtained for both synthesized Co_xFe_{3-x}O₄ ferrite nanoparticle and CoFe₂O₄/SiO₂ nanocomposites specimens. KBr pellets were used to confirm the spinel structure of the materials using an FTIR spectrophotometer (Shimadzu, IRAffinity-1, Japan). The Fourier transform infrared instrument is depicted in figure 3.3. This test was carried out in research Lab., physics department, college of science at Diyala University.



Figure 3.3 Photograph of fourier transform infrared spectroscopy.

3.6.3 Field Emission Scanning Electron Microscope Measurement (FE-SEM)

This technique, which operates on the principle of electronic scanning of electrons that bounce off the model's surface, provides us with a threedimensional image of the model's surface. This means that the device does not take an image of the model's surface just once. Rather, it takes images of each point in a cascade and then collects them to give us a comprehensive black-and-white image of the model's surface. Using a FE-SEM model (Mira3-XMU, TESCAN, Japan), an image of as-burnt and calcined specimens of $Co_xFe_{3-x}O_4$ nanoferrites and $CoFe_2O_4/SiO_2$ nanocomposites was taken at the center of DayPetronic Co., Tehran, Iran. Figure 3.4 depicts the situation.



Figure 3.4 Photograph of field emission scanning electron microscopy instrument.

3.6.3.1 Energy - Dispersive X - ray Spectroscopy (EDS)

This method is used to determine the kind of elements utilized in the model's construction. It also relies on the X-ray principle, which is based on the mutual effect of charged particles, such as a bundle of electrons, with the model's material, and the resulting X-rays are characteristic of the model materials constituent elements, allowing the chemical composition of the model to be determined. Each chemical element has a peak in its X-ray spectrum, and to produce the model's unique X-ray, the atoms must first be stimulated, which is done by bombarding the model with a bundle of electrons, which liberates an electron from the atom's inner orbits. Another electron launched the transition from a higher orbit to occupy this vacuum in order to fill the void created by the liberated electron, putting out X-rays with an energy equal to the energy differential between the planes of the orbits, and this team is distinct for each element.

3.7 Magnetic Characterizing

The examination of the magnetization curve (Hysteresis loops) of the produced materials using a vibrating sample magnetometer measuring (VSM) device is one of the magnetic properties.

3.7.1 Vibrating Sample Magnetometer (VSM)

The VSM is a method for calculating the magnetic moment, the most fundamental quantity of magnetism, in magnetic specimens. When a specimen material is placed in a homogeneous magnetic field, it produces a dipole moment proportional to the product of specimen susceptibility and applied field. The magnetic properties of both as-burnt and calcined $Co_xFe_{3-x}O_4$ nanoferrites specimens, as well as the cobalt ferrite-silica nanocomposites specimens calcined at 600 °C are calculated using a vibrating sample magnetometer (VSM) (LBKFB model Meghnatis Daghigh Kavir Company). The tests were carried out in the applied field at room temperature in the range of ±15 kOe in the center of DayPetronic Co., Tehran, Iran. Figure 3.5 depicts the vibrating sample magnetometer apparatus utilized in this study.



Figure 3.5 Photograph of vibrating sample magnetometer (VSM) instrument.

3.8 The Electrical Properties Measurement

The set for the electrical tests is created and prepared, and the two sides of the specimen are smoothed with a smoothing paper (sandpaper), which is accomplished by rubbing and polishing the two sides of the specimen to ensure the equal thickness at each of the two opposite points on the surfaces of the model. It is also attached to the electrodes of the instrument (LCR meter) that is used to measure electrical qualities. And is of the type (KEYSIGHT E4980A) that is connected to a computer device to display results directly on the screen. The electrical properties of $Co_xFe_{3-x}O_4$ nanoferrites sintered at 350, 600, 700 and 800 °C, as well as cobalt ferritesilica nanocomposites sintered at 700 °C, were measured in the research lab., physics department, college of science at Sulaimani University, using an LCR meter (KEYSIGHT E4980A) on a frequency range of (50Hz-2MHz). This device measures both the capacitance of the pellet (C_n), dielectric loss angle (tan δ), and capacitance of air with the same thickness as the pellet. Both (ϵ') and (ϵ''), as well as (σ_{ac}) may be estimated as a function of the frequency range (50Hz-2MHz) using the information provided by the device, as shown in figure 3.6.



Figure 3.6 Photograph of LCR meter instrument.

Chapter Four

Result and Discussion

4.1 Introduction

This chapter presents the results and discussions of the preparation of $Co_xFe_{3-x}O_4$ nanoparticles and $CoFe_2O_4/SiO_2$ nanocomposite, as well as the structural and magnetic characteristics (XRD, FE-SEM, FT-IR, and VSM) and electrical characteristics (dielectric constant, dielectric loss angle, dielectric loss factor, and ac conductivity), all of which are supported by diagrammatic planning and specimen representation.

4.2 Co_xFe_{3-x}O₄ Nanoparticles Characterization

4.2.1 Structural Properties

The auto-combustion sol-gel method was used to make the ferrite with the chemical formula $Co_xFe_{3-x}O_4$ for the values (x= 0.8, 0.9, 1, 1.1, and 1.2). The generated materials were characterized for structural and morphological features using X-ray diffraction (XRD), Fourier transform infrared (FT-IR) spectroscopy, and field emission scanning electron microscopy (FE-SEM).

4.2.1.1 X-ray Diffraction Studies

The XRD patterns of $Co_xFe_{3-x}O_4$ (x = 0.8 – 1.2, with a step of 0.1) for as-burnt and the calcined specimens at different temperature 500, 600 and 700 °C are shown in figures (4.1-4.5). The XRD pattern shows that all the reflection peaks corresponding to (111), (220), (311), (222), (400), (422), (511) and (440) planes of a cubic unit cell. The peak position in XRD patterns are precisely suited the standard pattern with reference code ICSD 00-001-1121. Specimens with x = 0.8 and 0.9 showed few traces of a second phase of hematite (iron oxide) at 600 and 700 °C. Our findings reveal that when the calcination temperature rises, the diffraction peaks get sharper and narrower, and their intensity rises. This points to an increase in crystallinity as a result of the increased crystalline volume ratio caused by the nuclei's particle size enlargement [155], similar observations were reported by B. Purnama et al. and T. Prabhakaran et al. [156, 157].

The XRD study of synthesized specimens of cobalt ferrite shows that crystalline and single spinel phases are formed under the experimental circumstances.



Figure 4.1 XRD patterns of $Co_xFe_{3-x}O_4$ nanoparticles (x=0.8) for as-burnt and calcined at 500,600 and 700 °C.



Figure 4.2 XRD patterns of Co_xFe_{3-x}O₄ nanoparticles (x=0.9) for as-burnt and calcined at 500, 600 and 700 °C.



Figure 4.3 XRD patterns of $Co_xFe_{3-x}O_4$ nanoparticles (x=1) for as-burnt and calcined at 500, 600 and 700 °C.



Figure 4.4 XRD patterns of $Co_xFe_{3-x}O_4$ nanoparticles (x=1.1) for as-burnt and calcined at 500, 600 and 700 °C.



Figure 4.5 XRD patterns of Co_xFe_{3-x}O₄ nanoparticles (x=1.2) for as-burnt and calcined at 500, 600 and 700 °C.

4.2.1.1.1 Crystallite Size (D)

The crystallite size (D) of each specimen was calculated from Scherrers formula (equation 2.22) using the peak's FWHM (311).

Table 4.1 explicitly illustrates how the crystallite size of the all specimens sharply increases with calcination temperature when cobalt content (x) remains constant, and increases basically with increasing cobalt content when calcination temperature stays the same, with the exception of (x=1.2). This increase can be attributed to the difference in cationic radii. Previous studies reported an increase in crystallite size due to increased calcination temperature and cobalt content [32, 156, 158, 159].

4.2.1.1.2 Lattice Parameter 'a'

Equation (2.23) was used to measure the lattice constant 'a' of the prepared specimens with the same peaks for different calcination temperatures. Table 4.1 shows how the lattice parameters of the specimens changed as the amount of cobalt increased, which does not appear to be a simple linear function and the lattice parameter is affected by a variety of factors such as atoms size and interactive forces between atoms [160].

As shown in table 4.1, the lattice parameters for as burnt increase with increasing cobalt content in the prepared cobalt ferrite specimens. However, the lattice parameter showed a drop at x=1 and x>1, because the ionic radius of Co^{2+} is greater than that of Fe^{3+} at either A-site or B-site [159].

Table 4.1 shows the variation of lattice parameters as a function of calcination temperatures. The lattice parameters reveal a non-monotonic trend with increasing temperature. A.M. Mohammad et al. observed similar non-monotonic behavior in lattice parameter for nanoparticles of Mg-substituted cobalt ferrite [161].

4.2.1.1.3 X-Ray Density (ρ_x)

The X-ray density (ρ_x) values were determined from the XRD data using equation (2.24). The value of X-ray density for as burnt increases monotonically with rising cobalt content except for x= 0.9 as shown in table 4.1, which was observed in an earlier report [159]. The decrease in the lattice constant, which outweighs the decrease in mass, is the cause of this increase. Table 4.1 shows the variation in X-ray density (ρ_x) as a function of calcination temperatures. We observe that with increasing temperature, the X-ray density (ρ_x) shows a non-linear trend. Similar nonlinear behavior in the X-ray density has been observed for the nanoparticles of Mg-substituted cobalt ferrite by A.M. Mohammad et al. [161].

4.2.1.1.4 Hopping Length (L)

The hopping length L (distance between magnetic ions) affects the physical properties of ferrite specimens; which was determined using equations (2.25 and 2.26) for A-site L_A (tetrahedral) and B-site L_B (octahedral) respectively.

Table 4.1 shows the measured hopping length L_A and L_B values for the different formulations. It is noted that the hopping length values decrease with an increase in the ratio of cobalt for as-burnt specimens except for x=0.9. The rate at which Fe³⁺ substitutes into the tetrahedral and octahedral sites changes as Fe³⁺ concentration increases, which could affect the value

of hopping length, which is related to the variation in the ionic radius of Co^{2+} and Fe^{3+} ions.

It can be also observed that L_A is bigger than L_B ; implying that the electron hopping between ions at tetrahedral (A-site) and octahedral (B-site) is less likely than that between octahedral (B-site) and octahedral (B-sites) [95].

Table 4.1 also shows the measured hopping length L_A and L_B values for the different calcination temperatures. We observe the hopping length L_A and L_B shows a non-linear trend with increasing temperature [161].

x	Composition	<i>Temp</i> .°C	2Theta	FWHM	D(nm)	a(Å)	$\rho_x(g/cm^3)$	L _A (Å)	$L_B(\mathring{A})$
0.8	(Co _{0.8} Fe _{2.2} O ₄)	as- burnt	35.590	0.34	24.530	8.360	5.330	3.6202	2.9559
		500	35.552	0.30	27.798	8.368	5.315	3.6235	2.9585
		600	35.618	0.23	36.265	8.353	5.344	3.6171	2.9533
		700	35.641	0.17	49.067	8.348	5.354	3.6148	2.9515
0.9	(Co _{0.9} Fe _{2.1} O ₄)	as-burnt	33.550	0.31	26.901	8.368	5.315	3.6236	2.9587
		500	35.710	0.24	30.886	8.369	5.314	3.6237	2.9587
		600	35.536	0.22	37.904	8.372	5.308	3.6251	2.9599
		700	35.551	0.19	43.891	8.369	5.315	3.6237	2.9587
1	(Co1Fe2O4)	as-burnt	35.590	0.31	26.904	8.359	5.333	3.6196	2.9554
		500	35.600	0.27	30.891	8.358	5.335	3.6191	2.9550
		600	35.686	0.22	37.920	8.338	5.374	3.6104	2.9478
		700	35.581	0.21	39.714	8.362	5.328	3.6207	2.9563
1.1	(Co _{1.1} Fe _{1.9} O ₄)	as-burnt	35.620	0.26	32.080	8.353	5.344	3.6171	2.9534
		500	35.710	0.24	34.763	8.332	5.384	3.6080	2.9459
		600	35.640	0.20	41.707	8.349	5.353	3.6150	2.9517
		700	35.617	0.16	52.130	8.353	5.343	3.6172	2.9534
1.2	(Co _{1.2} Fe _{1.8} O ₄)	as-burnt	35.660	0.31	26.909	8.343	5.363	3.6127	2.9497
		500	35.561	0.30	27.798	8.366	5.319	3.6226	2.9578
		600	35.670	0.23	36.270	8.341	5.368	3.6116	2.9489
		700	35.560	0.21	39.712	8.366	5.319	3.6227	2.9579

Table 4.1 Values of crystallite size (*D*), Lattice parameter '*a*', X-ray density (ρ_x), hopping length (L_A) and (L_B) for Co_xFe_{3-x}O₄ nanoparticles with (x = 0.8 - 1.2).

4.2.1.2 Fourier Transform Infrared Spectroscopy (FT-IR)

Based on vibrational modes, the FT-IR spectrum analysis is a valuable technique for determining where ions are situated in the crystal structure [162]. Ferrites are permanently bound crystals, which means that the atoms are bonded to all of nearest neighbors by equal powers (Ionic, Covalent or Van der Waals). According to the geometrical structure of the oxygen closest neighbours, metal ions are located in two various sub-lattices specified in ferrites: tetrahedral (A-site) and octahedral (B-site). Frequency band ν_1 represents the intrinsic lattice vibration of tetrahedral group complexes (M-O), while frequency band ν_2 represents the octahedral group complexes (M-O). Differences in bond lengths between oxygen and metal ions (Fe) at tetrahedral (A-site) and octahedral (B-site) may explain the difference in frequency between typical vibrations ν_1 and ν_2 [163].

Figures (4.6-4.10) demonstrate the FT-IR spectra of $Co_xFe_{3-x}O_4$ nanoferrites for as-burnt and calcined at 500, 600, and 700 °C.

The FT-IR spectrum of as-burnt $\text{Co}_{x}\text{Fe}_{3-x}\text{O}_{4}$ nanoferrites shows two main absorption bands in the range of 600-400 cm⁻¹, the first of which is between 569.001- 570.930 cm⁻¹ (high frequency ν_{1}) and the second band is between 368.404 - 372.261 cm⁻¹ (low frequency ν_{2}). These bands (ν_{1} and ν_{2}) were assigned due to the vibrations of the metal and oxygen ions complexes in the tetrahedral and octahedral positions. The band position differences (ν_{1} and ν_{2}) arise due to difference in the metal-oxygen distance for tetrahedral and octahedral complexes, indicating the presence of a cubic spinel structure [164].

Table 4.2 shows the positions of bands ν_1 and ν_2 as a function of different calcination temperatures, and except for (x=1.2) at 700 °C, we notice a similar pattern of shifting peaks for bands ν_1 towards the higher frequencies side. Whereas a similar pattern of shifting of the peaks for the bands ν_2 is observed towards the lower frequencies side except for (x=1)

at 500 °C, (x=1.1) at 700 °C, and (x=1.2) at 600 °C, clearly indicating the mixed spinel state of $Co_xFe_{3-x}O_4$. This could be attributed to the change in the bond lengths between the metal ions and the oxygen ions present at the tetrahedral and octahedral sites [165].

The standard mode of vibration observed at the tetrahedral site is higher than that observed at the octahedral site, which Waldron attributes to the tetrahedral bond's shorter length than the octahedral bond's [166].

Table 4.2 summarizes the FT-IR spectrum findings for $Co_xFe_{3-x}O_4$ nanoferrites as-burnt and calcined at 500, 600, and 700 °C.



Figure 4.6 FTIR spectra of $Co_x Fe_{3-x}O_4$ (x = 0.8) for as-burnt and calcined at 500, 600 and 700 °C.



Figure 4.7 FTIR spectra of $Co_x Fe_{3-x}O_4$ (x = 0.9) for as-burnt and calcined at 500, 600 and 700 °C.



Figure 4.8 FTIR spectra of $Co_xFe_{3-x}O_4$ (x = 1) for as-burnt and calcined at 500, 600 and 700 °C.



Figure 4.9 FTIR spectra of $Co_x Fe_{3-x}O_4$ (x = 1.1) for as-burnt and calcined at 500, 600 and 700 °C.



Figure 4.10 FTIR spectra of $Co_x Fe_{3-x}O_4$ (x = 1.2) for as-burnt and calcined at 500, 600 and 700 °C.

Table 4.2 FT-IR spectral bands for $Co_x Fe_{3-x}O_4$ nanoparticles (x=0.8, 0.9, 1, 1.1 and 1.2) for as-burnt and calcined at 500, 600 and 700 °C.

Commentation of the second sec	Toma °C	FTIR frequency bands (cm ⁻¹)			
Composition	<i>Temp</i> . C	<i>v</i> ₁	<i>v</i> ₂		
	as-burnt	569.001	368.404		
	500	569.001	366.475		
$Co_{0.8}Fe_{2.2}O_4)$	600	570.930	362.617		
	700	570.930	362.617		
	as-burnt	569.001	366.475		
	500	572.858	364.546		
$(CO_{0.9}Fe_{2.1}O_4)$	600	572.858	362.617		
	700	572.858	362.617		
	as-burnt	569.001	370.332		
	500	574.787	387.692		
$(\text{CO}_1\text{Fe}_2\text{O}_4)$	600	576.716	366.475		
	700	576.716	362.617		
	as-burnt	570.930	368.404		
$(\mathbf{C}_{\mathbf{c}}, \mathbf{E}_{\mathbf{c}}, \mathbf{O})$	500	574.787	366.475		
$(CO_{1.1}Fe_{1.9}O_4)$	600	574.787	362.617		
	700	576.716	387.692		
	as-burnt	569.001	372.261		
	500	574.787	366.475		
$(CO_{1.2}Fe_{1.8}O_4)$	600	578.644	385.763		
	700	572.858	360.689		

Meanwhile, the FT-IR spectra also displays other absorption bands in the region of 1400 to 3600 cm⁻¹ for the prepared compounds are attributed to NO_3^- ions, the CO₂ stretching frequency, and bending vibrational modes of absorbed or free water molecules O-H respectively, as illustrated in figures 4.6-4.10. Due to the high temperature generated during the combustion process, all carboxyl, hydroxyl, and nitrate present groups appear with decreased intensity, as indicated in previous studies [167, 168].

4.2.1.3 Field Emission Scanning Electron Microscopy (FE-SEM)

The findings of FE-SEM investigations for the compound Co_xFe_{3-x}O₄ showed that the particles have a spherical structure with a narrow distribution of nanoparticle sizes, indicating that the particles were formed and porosity of the nanoparticles. We also note the homogeneous agglomeration of the fine spherical particles, as particles of smaller size have permanent magnetization, therefore each particle has permanent magnetization, which appears as a result of particle agglomeration and grouping. Figures (4.11- 4.15) shows the results of the tests FE-SEM for values (x = 0.8, 0.9, 1, 1.1, and 1.2) at a temperatures (as-burnt, 500, 600, and 700 °C) respectively, and indicates an increase in particle size with increasing temperature across the forms. Table 4.3 displays the approximate diameters of $Co_x Fe_{3-x}O_4$ nanoparticles (x = 0.8, 0.9, 1, 1.1, and 1.2) for as-burnt and calcined specimens at 500, 600, and 700 °C with comparatively well-crystallized grains and a mean particle size smaller than 26.707, 29.964, 39.062, 53.652, 28.748, 31.203, 41.275, 46.666, 29.178, 32.134, 42.664, 44.655, 34.420, 38.561, 44.922, 55.247, 32.803, 36.350, 38.895, and 43.126 nm respectively determined by Image J Software. All the data correspond relatively well with the XRD results.

The particle size, which was estimated by FE-SEM micrographs, is found to be larger than that estimated using XRD data. The XRD method could be responsible for the molecular structural disruption and lattice strain generated by different ionic radii and/or nanoparticle collection. As a result, it has a more stringent requirement, which results in smaller sizes [169].

The particle size increased larger as the calcination temperature was raised, as shown by the FE-SEM image. Figures 4.11-4.15 show a FE-SEM image of cobalt ferrite nanoparticles after being as-burnt and calcined at 500, 600, and 700 °C. Spherical ferrite particles were moderately agglomerated and separated at 500 °C. At higher calcination temperatures, bigger spherical and elongated particles were also detected [170].

Table 4.3 shows that, except for x=1.2, the particle size of as-burnt and calcined specimens increases as the concentration of (x) increases at 500 and 600 °C. Because of its permanent magnetic moment, each particle is magnetized all of the time and prefers to clump together. Co-substituted nanoparticles have a greater magnetic moment, which causes more clustering [171, 172]. With the exception of x=1.1, the particle size and agglomeration of the calcined specimens at 700 °C decreases as the (x) content increases. This could be due to the magnetic cations being replaced with a non-magnetic dopant, or it could be due to the synthesis conditions [173].



Figure 4.11. FE-SEM micrographs of $Co_{0.8}Fe_{2.2}O_4$ nanoferrites for (a) as-burnt (b) calcined specimen at 500 °C (c) at 600 °C and (d) at 700 °C.



Figure 4.12. FE-SEM micrographs of $Co_{0.9}Fe_{2.1}O_4$ nanoferrites for (a) as-burnt (b) calcined specimen at 500 °C (c) at 600 °C and (d) at 700 °C.



Figure 4.13. FE-SEM micrographs of Co₁Fe₂O₄ nanoferrites for (a) as-burnt (b) calcined specimen at 500 °C (c) at 600 °C and (d) at 700 °C.



Figure 4.14. FE-SEM micrographs of Co_{1.1}Fe_{1.9}O₄ nanoferrites for (a) as-burnt (b) calcined specimen at 500 °C (c) at 600 °C and (d) at 700 °C.



Figure 4.15. FE-SEM micrographs of Co_{1.2}Fe_{1.8}O₄ nanoferrites for (a) as-burnt (b) calcined specimen at 500 °C (c) at 600 °C and (d) at 700 °C.

In addition, cobalt alters the shape of cobalt ferrite, as shown in figure 4.13 (b) and figure 4.14 (c). The morphology of the cobalt ferrite specimen is regular variance and distribution, as shown in the micrograph. Such behavior could be a feature of cobalt ferrite made by the auto combustion sol-gel method. Cobalt concentration has a noticeable impact on grain size and smooth morphology. When the concentration of cobalt rises, grains appear to consolidate into bigger grains with smooth surfaces [174].

x	Composition	Temp.°C	D(nm) XRD	D(nm) FE-SEM
	(Co _{0.8} Fe _{2.2} O ₄)	as- burnt	24.530	26.707
0.0		500	27.798	29.964
0.0		600	36.265	39.062
		700	49.067	53.652
	(C_{2}, F_{2}, O_{1})	as-burnt	26.901	28.748
0.0		500	30.886	31.203
0.9	(C00.9Fe2.104)	600	37.904	41.275
		700	43.891	46.666
		as-burnt	26.904	29.178
1	$(\mathbf{C}_{\mathbf{Q}},\mathbf{F}_{\mathbf{Q}},\mathbf{Q}_{\mathbf{v}})$	500	30.891	32.134
1	$(C0_11 C_2O_4)$	600	37.920	42.664
		700	39.714	44.655
		as-burnt	32.080	34.420
11	$(\mathbf{C}_{\mathbf{C}}_{\mathbf{C}_{\mathbf{C}}_{\mathbf{C}_{\mathbf{C}}_{\mathbf{C}}_{\mathbf{C}_{\mathbf{C}}_{\mathbf{C}_{\mathbf{C}}_{\mathbf{C}_{\mathbf{C}}}}}}}}}}$	500	34.763	38.561
1.1	(COL1 Pel.904)	600	41.707	44.922
		700	52.130	55.247
	(Co _{1.2} Fe _{1.8} O ₄)	as-burnt	26.909	32.803
12		500	27.798	36.350
1,4		600	36.270	38.895
		700	39.712	43.126

Table 4.3 Average crystallite size and particle size of $Co_xFe_{3-x}O_4$ nanoferrites (x = 0.8, 0.9, 1, 1.1, and 1.2) for (a) as-burnt (b) calcined specimen at 500 °C (c) calcined at 600 °C and (d) calcined specimen at 700 °C determined from XRD and FE-SEM.

4.2.1.3.1 Energy Dispersive Spectroscopy (EDS)

Energy dispersive spectroscopy (EDS) is a technique for determining the chemical or elemental makeup of a material. It is based on the interaction of an X-ray excitation source with a specimen. Its ability to characterize elements is largely due to the basic premise that each element has a unique atomic structure that allows for a different set of peaks on its electromagnetic emission spectrum. The EDS pattern of nanocrystalline $Co_xFe_{3-x}O_4$ (x= 0.8, 0.9, 1, 1.1, and 1.2) ferrites is shown in figures 4.16 to 4.20. In the absence of any impurities, the relative atomic abundance of Co, Fe, and O species present in the upper surface layers of specimens.

According to EDS analysis, the iron element decreases from 55.9% to 44.4 at 500 °C and from 52.0% to 44.7 at 600 °C as the content of (x) increase, whereas the cobalt element increases from 21.9% to 31.0% at 600 °C as the concentration of (x) increases. The proportions of elements are shown in figures 4.16 - 4.20. As shown in figures 4.16, 4.17, and 4.20, the cobalt element increases as the calcined temperature rises, with the exception of 700 °C at (x= 0.8, 0.9, and 1.2).



Figure 4.16 EDS spectra of $Co_{0.8}Fe_{2.2}O_4$ nanoferrites for (a) as-burnt (b) calcined specimen at 500 °C (c) at 600 °C and (d) at 700 °C.



Figure 4.17 EDS spectra of Co_{0.9}Fe_{2.1}O₄ nanoferrites for (a) as-burnt (b) calcined specimen at 500 °C (c) at 600 °C and (d) at 700 °C.



Figure 4.18 EDS spectra of Co₁Fe₂O₄ nanoferrites for (a) as-burnt (b) calcined specimen at 500 °C (c) at 600 °C and (d) at 700 °C.



Figure 4.19 EDS spectra of Co_{1.1}Fe_{1.9}O₄ nanoferrites for (a) as-burnt (b) calcined specimen at 500 °C (c) at 600 °C and (d) at 700 °C.



Figure 4.20 EDS spectra of Co_{1.2}Fe_{1.8}O₄ nanoferrites for (a) as-burnt (b) calcined specimen at 500 °C (c) at 600 °C and (d) at 700 °C.

4.2.2 Magnetic Properties

Under a magnetic field of ±15 KOe, the magnetic characteristics of produced $\text{Co}_x\text{Fe}_{3-x}\text{O}_4$ nanoparticles (x= 0.8, 0.9, 1, 1.1, and 1.2) were measured at room temperature for as-burnt and calcined specimens at varied temperatures 500, 600, and 700 °C. The remanent magnetization (M_r) , saturation magnetization (M_s) , and coercive field (H_c) values are calculated using the hysteresis loops mentioned in section (2.9). Moreover, the equations (2.5, 2.6, and 2.7) were used to quantify the remanence ratio (M_r/M_s) , magnetic moment (n_B) , and magnetic anisotropy (K), respectively.

4.2.2.1 Saturation Magnetization

The saturation magnetization (M_s) of a magnetic material refers to its greatest value of magnetization [175].

Figures 4.21-4.25 displays a model hysteresis loops registered for $Co_xFe_{3-x}O_4$ nanoferrites (x= 0.8, 0.9, 1, 1.1 and 1.2) for as-burnt and various calcination temperatures and the values of saturation magnetization and magnetic moments are recorded in tables 4.4 - 4.8.

The value of saturation magnetization (M_s) and magnetic moment (n_B) in the present measurement are increased with the increasing of the calcination temperature, being 59.37-71.93 emu.g⁻¹ (2.49-3.02 μ_B) for x=0.8, and 60.79-78.22 emu.g⁻¹ (2.55-3.28 μ_B) for x=1.2. This increase in (M_s) as the calcination temperature was raised may be due to spin canting and surface spin disturbance in these nanoferrites [32, 165]. The magnetic moment (n_B) increased in nanoferrites as the calcining temperature increased because Fe³⁺ ions have a greater magnetic moment than Co²⁺ ions, resulting in the predominant Fe³⁺ ions being redistributed at B-sites [156].

When the Co²⁺ concentration is increased, the saturation magnetization increases for as-burnt specimens, except for x >1, and the saturation magnetization increases at calcined temperature (500 °C), except for x=1 and 1.2. Except for x=1.2, the saturation magnetization increases at calcined temperatures (600 and 700 °C). The decrease in (M_s) value can be attributed to the fact that the magnetic moment of Co²⁺ is lower than that of Fe³⁺, resulting in a reduction in B-site magnetization [159].

The magnetic moment increases as the Co^{2+} concentration is raised for as-burnt specimens except for x >1, and increases for calcined specimens at (500, 600, and 700 °C) except for x=1.2. The lower (n_B) value could be attributable to higher Co^{2+} occupancy at B-sites [176].

4.2.2.2 Remanent Magnetization

Remanent magnetization (M_r) which is the magnetization that remains in ferrimagnetic materials after an external magnetic field has been removed.

The remnant magnetization has increased as the calcination temperature has increased (tables 4.4 - 4.8), being 31.04 - 36.02 emu.g⁻¹ for x=0.8, and 28.84 - 40.13 emu.g⁻¹ for x=1.2. This increase in (M_r) is due to Fe³⁺ having a greater magnetic moment than Co²⁺ [159].

Tables 4.4 - 4.8 show the values of the squareness ratio ($S = M_r/M_s$) that were achieved. The magnetocrystalline anisotropy and superexchange interactions of the investigated nanoferrites are reflected in *S* values [177]. The exchange-coupling is present when the squareness ratio is bigger than 0.5. It's also been suggested that the squareness ratio is smaller than 0.5 in the presence of non-interacting single domain particles [17].

When the Co²⁺ content is raised, the remnant magnetization increases for as-burnt specimens except for x =1 and x >1, as well as the remnant magnetization at calcined temperature (500 °C) except for x =1 and 1.2, and the remnant magnetization at calcined temperatures (600 and 700 °C) except for x >1. The decrease in (M_r) value could be due to Co²⁺ having a smaller magnetic moment than Fe³⁺ [159].

4.2.2.3 Coercivity

The coercivity (H_c) is the size of the field that must be applied in the negative direction to return the specimen magnetization to zero [178]. The increase in the coercive force (H_c) with calcination (at 500 °C) is shown as in figures 4.21- 4.25. It is evident from tables 4.4 - 4.8, the (H_c) value increased from 1854.6 O_e to 2474.5 O_e for x= 0.8 and 1151.6 O_e to 1707.5 O_e for x=1.2. Nonetheless, with the exception of x= 0.9 at 700 °C, the

coercivity values (H_c) decrease as the calcining temperature rises (at 600 and 700 °C). An increase in magnetic crystallographic anisotropy may be responsible for the increase in coercivity value for x= 0.8, 0.9, 1, 1.1, 1.2 at 500 °C [159]. The decrease in coercivity as calcining temperature rises (at 600 and 700 °C) may be due to a reduction in anisotropy field in order to reduce domain wall energy [165].

When the Co^{2+} content is increased, the coercivity increases for as-burnt specimens except for x= 1 and 1.2, while the coercivity increases for calcined temperatures (500, 600, and 700 °C) except for x=1 and x >1. This could be due to a decrease in the anisotropy field, which lowers the domain wall energy [159].

The values of the anisotropy constant (K) alter with an increase in temperature, either decreasing or increasing, as indicated in the tables (4.4-4.8). Because of the drop in average crystalline size, cationic redistribution in the magnetic domain regime changed, affecting magnetocrystalline anisotropy [170].

When the Co^{2+} content is increased, for as-burnt specimens and calcined specimens at 500, 600 and 700 °C the anisotropy constant decreases except for x=0.9. For the same reason mentioned above [159].

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Figure 4.21. Hysteresis curves of $Co_{0.8}Fe_{2.2}O$ nanoferrites for as-burnt and different calcination temperatures.



Figure 4.22. Hysteresis curves of Co_{0.9}Fe_{2.1}O₄ nanoferrites for as-burnt and different calcination temperatures.



Figure 4.23. Hysteresis curves of Co₁Fe₂O₄ nanoferrites for as-burnt and different calcination temperatures.



Figure 4.24. Hysteresis curves of Co_{1.1}Fe_{1.9}O₄ nanoferrites for as-burnt and different calcination temperatures.



Figure 4.25. Hysteresis curves of Co_{1.2}Fe_{1.8}O₄ nanoferrites for as-burnt and different calcination temperatures.

Table 4.4. Variation in saturation magnetization (M_s) , remanence magnetization (M_r) coercivity (H_c) magnetic moment (n_B) , squareness ratio (M_r/M_s) , and magnetic anisotropy (K) of Co_{0.8}Fe_{2.2}O₄ nanoferrites for different calcination temperature.

Temp °C	M _s	M _r	H _c	n_B	M /M	$K \times 10^3$
<i>1 cmp</i> . C	$(emu. g^{-1})$	$(emu. g^{-1})$	(O_e)	(µ _B)	m_r/m_s	$(emu . O_e g^{-1})$
As-burnt	59.37	31.04	1854.6	2.49	0.52	114.70
500	63.71	34.41	2474.5	2.68	0.54	164.22
600	70.31	35.41	1239.7	2.95	0.50	90.80
700	71.93	36.02	1228.4	3.02	0.50	92.03

						1
Temn °C	M _s	M _r	H _c	n_B	M /M	$K \times 10^3$
Temp. C	$(emu. g^{-1})$	$(emu. g^{-1})$	(O_e)	(µ _B)	$n_{\gamma} n_{S}$	$(emu . O_e g^{-1})$
As-burnt	66.30	35.49	2045.3	2.79	0.54	141.32
500	69.83	38.50	2872.3	2.93	0.55	208.93
600	70.60	38.66	2699.5	2.96	0.55	198.53
700	74.41	40.49	2785.4	3.12	0.54	215.90

Table 4.5. Variation in saturation magnetization (M_s) , remanence magnetization (M_r) coercivity (H_c) magnetic moment (n_B) , squareness ratio (M_r/M_s) , and magnetic anisotropy (K) of Co_{0.9}Fe_{2.1}O₄ nanoferrites for different calcination temperature.

Table 4.6. Variation in saturation magnetization (M_s) , remanence magnetization (M_r) coercivity (H_c) magnetic moment (n_B) , squareness ratio (M_r/M_s) , and magnetic anisotropy (K) of Co₁Fe₂O₄ nanoferrites for different calcination temperature.

Temn °C	M _s	M _r	H _c	n_B	M /M	$K \times 10^3$
Temp. C	$(emu. g^{-1})$	$(emu. g^{-1})$	(O_e)	(µ B)	m _r m _s	$(emu . O_e g^{-1})$
As-burnt	69.23	33.77	1139.4	2.91	0.49	82.17
500	66.92	35.13	2619.5	2.94	0.50	190.79
600	75.35	40.78	2522.4	3.16	0.54	197.98
700	75.79	41.44	2450.5	3.18	0.55	193.46

Table 4.7. Variation in saturation magnetization (M_s) , remanence magnetization (M_r) coercivity (H_c) magnetic moment (n_B) , squareness ratio (M_r/M_s) , and magnetic anisotropy (K) of Co_{1.1}Fe_{1.9}O₄ nanoferrites for different calcination temperature.

Temn °C	M _s	<i>M_r</i>	H _c	n_B	M_r/M_s	$K \times 10^3$
Temp. C	$(emu. g^{-1})$	$(emu. g^{-1})$	(O_e)	(µ _B)		$(emu . O_e g^{-1})$
As-burnt	67.20	33.45	1164.4	2.82	0.50	81.50
500	69.97	36.56	2170.3	2.94	0.52	158.18
600	75.79	39.95	1764.5	3.18	0.53	139.31
700	79.02	41.21	1619.6	3.32	0.52	133.31

Tomn °C	M _s	M_r	H _c	n_B	M_r/M_s	$K \times 10^3$
<i>1 emp</i> . C	$(emu. g^{-1})$	$(emu. g^{-1})$	(O_e)	(µ _B)		$(emu . O_e g^{-1})$
As-burnt	60.79	28.84	1151.6	2.55	0.47	72.93
500	68.65	33.49	1707.5	2.88	0.49	122.10
600	74.11	36.81	1199.3	3.11	0.50	92.59
700	78.22	40.13	1149.5	3.28	0.51	93.66

Table 4.8. Variation in saturation magnetization (M_s) , remanence magnetization (M_r) coercivity (H_c) magnetic moment (n_B) , squareness ratio (M_r/M_s) , and magnetic anisotropy (K) of Co_{1.2}Fe_{1.8}O₄ nanoferrites for different calcination temperature.

4.2.3 Electrical Properties

The study of the dielectric properties of ferrites as a function of electric field frequency was included in the electrical properties, and the study of the dielectric properties of ferrite nanoparticles provided us with information about the electrical conduction mechanism in terms of the dielectric response in the applied electric field. Various factors influence these properties, including the method of manufacture, chemical composition, porosity, charge ionic, grain size, and the distribution of cations between surface compositions, the real dielectric constant in ferrite materials behaves according to the dielectric polarization process. This is comparable to the electrical conductivity behavior of the materials themselves, where the behavior of the dielectric depends on the amount of abundance of the presence of positive ions Fe^{2+} and Fe^{3+} specified by the polarization, resulting in the emergence of the dielectric constant. Within the frequency range (50Hz -2 MHz), the real part of the dielectric constant (ε') , imaginary part of the dielectric constant (ε'') or dielectric loss factor, and ac conductivity (σ_{ac}) were computed using the relations discussed in detail in section (2.17).

4.2.3.1 Dielectric Constant

The dielectric constant of cobalt ferrite Co_xFe_{3-x}O₄ was calculated for the prepared specimens within a range of (50Hz-2MHz) using the equation (2.16) at different temperatures (350, 600, 700 and 800 °C) at values of (x =0.8, 0.9, 1, 1.1 and 1.2) respectively. The dielectric constant decreases with increasing frequency and the decreasing trend becomes slow in higher frequency region, showing frequency dispersion at low frequency range. This behavior is normal for the polar dielectric materials as shown in figures 4.26 - 4.30. The high values of dielectric constant at low frequency region may be due to presence of all type of polarizations (electronic, ionic and orientation) including space charge which dominates at low frequency. The decrease in dielectric constant with frequency shows the dispersion is Maxwell-Wagner type and is consistent with Koop's theory of dielectrics [22]. We also notice that as the temperature rises, the dielectric constant decreases, except for at $(800 \,^{\circ}\text{C})$ for x=1.1 and 1.2, which is due to an increase in grain growth, accompanied by a decrease in grain boundaries, and results in a decrease in the real and imaginary dielectric constant values [179].



Figure 4.26 Variation of dielectric constant (ε') with frequency for Co_{0.8}Fe_{2.2}O₄ spinel nanoparticles.



Figure 4.27 Variation of dielectric constant (ϵ') with frequency for Co_{0.9}Fe_{2.1}O₄ spinel nanoparticles.



Figure 4.28 Variation of dielectric constant (ε') with frequency for Co₁Fe₂O₄ spinel nanoparticles.



Figure 4.29 Variation of dielectric constant (ϵ') with frequency for Co_{1.1}Fe_{1.9}O₄ spinel nanoparticles.



Figure 4.30 Variation of dielectric constant (ϵ') with frequency for Co_{1.2}Fe_{1.8}O₄ spinel nanoparticles.

4.2.3.2 Dielectric Loss Factor (ϵ'')

The dielectric loss factor is an important factor in applications of ferrites materials. This is to determine the operating frequency in the magnetic materials, as almost identical behavior was observed in all the prepared specimens, where it was found that the dielectric loss factor is highest at low frequencies and decreases as the frequency of the applied electric field increases, as shown in figures 4.31-4.35. This decrease can be explained by the relationship (2.18). This inverse proportional between the dielectric loss factor and the frequency is due to the large energy that the dipoles acquire from the electric field applied to them, which in turn increases the possibility of collisions between these diodes as they rotate at the low frequencies, resulting in a weakening of the dielectric characteristic. When increasing the frequency of the applied field, the ability of the dipoles will decrease to follow the change of the field, reducing the energy absorbed by the dipoles from the applied field, and resulting in a decrease in dielectric loss factor [95].



Figure 4.31 Variation of dielectric loss factor (ϵ'') with frequency for Co_{0.8}Fe_{2.2}O₄ spinel nanoparticles.



Figure 4.32 Variation of dielectric loss factor (ϵ'') with frequency for Co_{0.9}Fe_{2.1}O₄ spinel nanoparticles.



Figure 4.33 Variation of dielectric loss factor (ε'') with frequency for Co₁Fe₂O₄ spinel nanoparticles.



Figure 4.34 Variation of dielectric loss factor (ϵ'') with frequency for Co_{1.1}Fe_{1.9}O₄ spinel nanoparticles.



Figure 4.35 Variation of dielectric loss factor (ε'') with frequency for Co_{1.2}Fe_{1.8}O₄ spinel nanoparticles.

Figures 4.31-4.35 show the variation of dielectric loss factor as a function of frequency for different temperatures for all the specimens in the frequency range 200KHz to 2MHz summarized in tables 4.9-4.13. It turns out that the dielectric loss factor increases with an increase in temperature, except for (at 600 and 700 °C) values of (x= 1.1 and 1.2) at all the above mentioned frequencies, and this increase is due to the increase in ac conductivity [180].

4.2.3.3 Dielectric Loss Angle $(tan \delta)$

The change of the loss angle was studied as a function of the frequency within (50Hz-2MHz) for cobalt ferrite $Co_xFe_{3-x}O_4$, with the results showing that the shadow of the dielectric loss angle decreases with increasing frequency for all specimens, which is the normal behavior of any ferrous materials, at high frequencies that correspond to the low resistance due to the presence of grains, a small amount of energy is needed for the electrons to hopping between the ions Fe^{2+} , Fe^{3+} at the octahedral site (B), while in

the low-frequency region, the boundaries of the grains are active, causing a high resistance. As a result, the electrons need a lot of energy to hopping between the ions Fe from the tetrahedral site (A) [181].

Figures 4.39 and 4.40 show the variance in dielectric loss angle as a function of frequency for various temperatures for values (x= 1.1 and 1.2). The dielectric loss angle at 1MHz, 1.5MHz, and 2MHz decreases as the temperature increases, with the exception of 800 °C. The relaxation effects may be responsible for the decrease in dielectric loss tangent with increases temperature [182]. Whereas the dielectric loss angle increase with increasing the temperature for values (x= 0.8, 0.9, and 1) at 200KHz, 500KHz, 1MHz, 1.5MHz and 2MHz. The increase in the loss angle with temperature can be attributed to the resulting electrical conductivity being larger than that of relaxation effects [183].



Figure 4.36 Variation of dielectric loss angle $(tan \delta)$ with frequency for Co_{0.8}Fe_{2.2}O₄ spinel nanoparticles.



Figure 4.37 Variation of dielectric loss angle $(tan \delta)$ with frequency for Co_{0.9}Fe_{2.1}O₄ spinel nanoparticles.



Figure 4.38 Variation of dielectric loss angle $(tan \delta)$ with frequency for Co₁Fe₂O₄ spinel nanoparticles.



Figure 4.39 Variation of dielectric loss angle $(tan \delta)$ with frequency for Co_{1.1}Fe_{1.9}O₄ spinel nanoparticles.



Figure 4.40 Variation of dielectric loss angle $(tan \delta)$ with frequency for Co_{1.2}Fe_{1.8}O₄ spinel nanoparticles.

4.2.3.4 AC Conductivity (σ_{ac})

The ac electrical conductivity was calculated by using the relationship (2.19) for temperatures (350, 600, 700, and 800 °C) in the frequency range (50Hz-2MHz). The figures 4.41-4.45 show the increase in the values of the ac electrical conductivity by increasing the frequency of the electric field applied to all the prepared specimens, and then releasing them from their positioning centers to contribute to the electrical conduction [183].

It is worth noting that the electrical conduction process depends mainly on the transition of charges between Fe^{2+} , Fe^{3+} , therefore at low frequencies the charges cannot cross the grain boundaries because they do not have sufficient energy, and with the increase in frequency, the charges are able to move between Fe^{2+} , Fe^{3+} , because it acquires sufficient energy to cross the grain boundaries, then the number of transported charges increases, which leads to an increase in ac electrical conductivity.



Figure 4.41 Variation of ac conductivity (σ_{ac}) with frequency for Co_{0.8}Fe_{2.2}O₄ spinel nanoparticles.



Figure 4.42 Variation of ac conductivity (σ_{ac}) with frequency for Co_{0.9}Fe_{2.1}O₄ spinel nanoparticles.



Figure 4.43 Variation of ac conductivity (σ_{ac}) with frequency for Co₁Fe₂O₄ spinel nanoparticles.



Figure 4.44 Variation of ac conductivity (σ_{ac}) with frequency for Co_{1.1}Fe_{1.9}O₄ spinel nanoparticles.



Figure 4.45 Variation of ac conductivity (σ_{ac}) with frequency for Co_{1.2}Fe_{1.8}O₄ spinel nanoparticles.

Figures 4.40-4.45 show the variance in ac conductivity as a function of frequency for various temperatures for all the specimens, and table 4.9-4.13 summarizes their values at different frequencies (200 kHz, 500 kHz, 1MHz, 1.5 MHz, and 2 MHz). Except for (at 600 and 700 °C) values of (x = 1.1 and 1.2), the ac conductivity increases with rising temperature at all the above-mentioned frequencies. The increase in ac conductivity with temperature can be ascribed to the greater variety of thermally activated charge carriers, which leads to more electron transfer from one ion to another [184].

The decrease in the ac conductivity was observed at the temperatures of 600 and 700 °C, as shown in tables 4.12 and 4.13. Due to charge carriers accumulating around grain boundaries a hopping mechanism prevents charge transfer in these accumulated charge carriers [185].

Table 4.9.	Values of dielectric constant (ε'), dielectric loss angle (tan δ), dielectric
loss factor	(ε'') and ac conductivity (σ_{ac}) at 200 KHz, 500 KHz, 1 MHz, 1.5 MHz and
2 MHz for	Co _{0.8} Fe _{2.2} O ₄ sintered at 350 °C, 600 °C, 700 °C, and 800 °C.

Dielectric parameters	350 °C	<i>600</i> °С	<i>700</i> °С	<i>800</i> °C
ε' (200KHz)	10.3	9.39	9.38	8.92
tan δ (200KHz)	0.360	0.458	0.500	0.536
ε'' (200KHz)	3.75	4.35	4.74	4.83
$\sigma_{ac} (200 KHz) \times 10^{-5}$	4.12	4.78	5.22	5.33
ε' (500KHz)	9.12	8.34	8.23	8.03
tan δ (500KHz)	0.224	0.268	0.354	0.516
ε'' (500KHz)	2.09	2.29	2.96	4.19
σ _{ac} (500KHz)×10 ⁻⁵	5.69	6.22	8.11	11.5
ε' (1MHz)	8.51	7.78	7.64	7.50
tan δ (1MHz)	0.153	0.177	0.246	0.466
ε'' (1MHz)	1.35	1.43	1.92	3.55
$\sigma_{ac} (1MHz) \times 10^{-5}$	7.26	7.66	10.4	19.5
ε' (1.5MHz)	8.08	7.30	7.24	7.12
tan δ (1.5MHz)	0.122	0.139	0.194	0.420
ε'' (1.5MHz)	0.98	1.01	1.45	3.03
$\sigma_{ac} (1.5MHz) \times 10^{-5}$	8.22	8.44	11.7	24.9
ε' (2MHz)	7.63	6.97	6.87	6.74
tan δ (2MHz)	0.101	0.113	0.161	0.382
ε'' (2MHz)	0.77	0.79	1.15	2.62
$\sigma_{ac} (2MHz) \times 10^{-5}$	8.42	8.75	12.2	28.5

Table 4.10. Values of dielectric constant (ε'), dielectric loss angle (tan δ), dielectric
loss factor (ϵ'') and ac conductivity (σ_{ac}) at 200 KHz, 500 KHz, 1 MHz, 1.5 MHz and
2 MHz for Co _{0.9} Fe _{2.1} O ₄ sintered at 350 °C, 600 °C, 700 °C, and 800 °C.

Dielectric parameters	<i>350</i> °C	<i>600</i> °С	<i>700</i> °С	<i>800</i> °C
ε' (200KHz)	12.6	12.4	12.1	11.6
tan δ (200KHz)	0.492	0.501	0.565	1.03
ε'' (200KHz)	6.23	6.23	6.75	11.9
σ _{ac} (200KHz)×10 ⁻⁵	6.89	6.89	7.50	13.3
ε' (500KHz)	10.5	9.99	9.32	8.48
tan δ (500KHz)	0.328	0.353	0.438	0.677
ε'' (500KHz)	3.47	3.58	4.11	5.80
σ _{ac} (500KHz)×10 ⁻⁵	9.52	9.83	11.3	16.0
ε' (1MHz)	9.47	8.94	8.03	7.44
tan δ (1MHz)	0.224	0.245	0.333	0.435
ε'' (1MHz)	2.17	2.25	2.72	3.29
$\sigma_{ac} (1MHz) \times 10^{-5}$	11.8	12.2	14.9	18.0
ε' (1.5MHz)	8.92	8.36	7.36	6.93
tan δ (1.5MHz)	0.176	0.194	0.279	0.330
ε'' (1.5MHz)	1.62	1.67	2.10	2.33
$\sigma_{ac} (1.5 MHz) \times 10^{-5}$	13.1	13.6	17.1	19.1
ε' (2MHz)	8.43	7.87	6.82	6.53
tan δ (2MHz)	0.146	0.161	0.242	0.269
ε'' (2MHz)	1.28	1.31	1.71	1.81
$\sigma_{ac} (2MHz) \times 10^{-5}$	13.7	14.0	18.4	19.5

Table 4.11. Values of dielectric constant (ε'), dielectric loss angle (tan δ), dielectric
loss factor (ϵ'') and ac conductivity (σ_{ac}) at 200 KHz, 500 KHz, 1 MHz, 1.5 MHz and
2 MHz for Co ₁ Fe ₂ O ₄ sintered at 350 ℃, 600°C, 700 ℃, and 800 ℃.

Dielectric parameters	350 °C	600 °C	<i>700</i> °С	800 °C
ε' (200KHz)	15.6	13.3	13.2	13.2
tan δ (200KHz)	0.302	0.505	0.661	0.696
ε'' (200KHz)	4.73	6.75	8.80	9.22
$\sigma_{ac} (200 KHz) \times 10^{-5}$	5.21	7.46	9.75	10.2
ε' (500KHz)	12.2	10.9	10.3	9.09
tan δ (500KHz)	0.197	0.330	0.551	0.679
ε'' (500KHz)	2.44	3.64	5.74	6.22
σ _{ac} (500KHz)×10 ⁻⁵	6.64	9.99	15.8	17.2
ε' (1MHz)	10.5	9.86	8.94	6.99
tan δ (1MHz)	0.142	0.215	0.404	0.604
ε'' (1MHz)	1.54	2.16	3.66	4.26
$\sigma_{ac} (1MHz) \times 10^{-5}$	8.28	11.8	20.1	23.5
ε' (1.5MHz)	9.60	9.30	8.19	6.07
tan δ (1.5MHz)	0.117	0.162	0.319	0.536
ε'' (1.5MHz)	1.17	1.55	2.65	3.30
σ_{ac} (1.5MHz) ×10 ⁻⁵	9.38	12.5	21.7	27.1
ε' (2MHz)	8.92	8.79	7.60	5.48
tan δ (2MHz)	0.099	0.130	0.266	0.485
ε'' (2MHz)	0.89	1.19	2.07	2.71
$\sigma_{ac} (2MHz) \times 10^{-5}$	9.83	12.7	22.4	29.6

Table 4.12. Values of dielectric constant (ε'), dielectric loss angle (tan δ), dielectric
loss factor (ϵ'') and ac conductivity (σ_{ac}) at 200 KHz, 500 KHz, 1 MHz, 1.5 MHz and
2 MHz for Co _{1.1} Fe _{1.9} O ₄ sintered at 350 °C, 600 °C, 700 °C, and 800 °C.

Dielectric parameters	350 °C	600 °C	<i>700</i> °С	800 °C
ε' (200KHz)	12.8	11.4	10.8	23.4
tan δ (200KHz)	0.387	0.419	0.428	0.535
ε'' (200KHz)	4.95	4.79	4.63	12.3
σ _{ac} (200KHz)×10 ⁻⁵	5.40	5.28	5.10	14.2
ε' (500KHz)	10.9	9.72	9.26	17.4
tan δ (500KHz)	0.275	0.279	0.271	0.515
ε'' (500KHz)	3.04	2.76	2.56	8.95
σ _{ac} (500KHz)×10 ⁻⁵	8.30	7.53	6.99	25.3
ε' (1MHz)	9.91	8.91	8.55	13.9
tan δ (1MHz)	0.207	0.196	0.185	0.465
ε'' (1MHz)	2.09	1.78	1.63	6.50
$\sigma_{ac} (1MHz) \times 10^{-5}$	11.4	9.72	8.80	36.4
ε' (1.5MHz)	9.31	8.41	8.10	12.2
tan δ (1.5MHz)	0.174	0.158	0.147	0.419
ε'' (1.5MHz)	1.66	1.38	1.24	5.09
$\sigma_{ac} (1.5 MHz) \times 10^{-5}$	13.5	11.1	9.90	43.1
ε' (2MHz)	8.78	7.95	7.68	11.0
tan δ (2MHz)	0.151	0.133	0.122	0.380
ε'' (2MHz)	1.38	1.11	0.94	4.20
$\sigma_{ac} (2MHz) \times 10^{-5}$	14.8	11.7	10.4	47.2

Table 4.13. Values of dielectric constant (ε'), dielectric loss angle ($tan \delta$), dielectric
loss factor (ϵ'') and ac conductivity (σ_{ac}) at 200 KHz, 500 KHz, 1 MHz, 1.5 MHz and
2 MHz for Co _{1.2} Fe _{1.8} O ₄ sintered at 350 ℃, 600 ℃, 700 ℃, and 800 ℃.

Dielectric parameters	350 °C	600 °C	<i>700</i> °С	800 °C
ε' (200KHz)	24.0	16.3	14.5	23.6
tan δ (200KHz)	0.546	0.548	0.699	0.584
ε'' (200KHz)	13.6	9.83	10.0	13.8
σ _{ac} (200KHz)×10 ⁻⁵	14.6	11.1	11.0	15.1
ε' (500KHz)	18.0	12.2	11.1	16.6
tan δ (500KHz)	0.519	0.472	0.491	0.597
ε'' (500KHz)	9.35	5.78	5.50	9.96
σ _{ac} (500KHz)×10 ⁻⁵	26.0	16.2	15.2	26.7
ε' (1MHz)	14.4	10.1	9.82	12.8
tan δ (1MHz)	0.471	0.371	0.342	0.541
ε'' (1MHz)	6.81	3.79	3.39	6.96
$\sigma_{ac} (1MHz) \times 10^{-5}$	38.0	21.1	18.6	37.4
ε' (1.5MHz)	12.7	9.29	9.13	11.2
tan δ (1.5MHz)	0.429	0.309	0.273	0.477
$\varepsilon^{\prime\prime}(1.5MHz)$	5.48	2.88	2.51	5.38
$\sigma_{ac} (1.5 MHz) \times 10^{-5}$	45.7	23.9	20.8	43.1
ε' (2MHz)	11.5	8.60	8.57	10.3
tan δ (2MHz)	0.394	0.265	0.232	0.436
ε'' (2MHz)	4.57	2.21	2.05	4.51
$\sigma_{ac} (2MHz) \times 10^{-5}$	50.8	25.5	22.0	46.3

4.5 CoFe₂O₄/SiO₂ Nanoparticles Characterization

4.5.1 Structural Properties

The conventional ceramic method was used to make nanocomposites of cobalt ferrite mixed SiO₂ with different concentrations of $CoFe_2O_4/SiO_2$ (0%, 35%, 40%, 45%, and 50%). The prepared specimens were characterized by powder X-ray diffraction (XRD), Fourier transform infrared (FT-IR) spectroscopy and Field emission scanning electron microscopy (FE-SEM) for their structural and morphological characteristics.

4.5.1.1 X-ray Diffraction Studies

The XRD patterns of nanocomposites $CoFe_2O_4/SiO_2$ (0%, 35%, 40%, 45%, and 50%) for the calcined specimens at 600 °C are shown in figure 4.46. In all of the specimens, the index peaks are cubic spinel type, and there are no signs of impurities.

Figure 4.46. The diffraction peaks associated with Bragg's reflections from (111), (220), (311), (222), (400), (422), (511), and (440) planes a good match to the standard structure of $CoFe_2O_4$ pure (ICSD 98-011-1281) while there are no signatures for the SiO₂ in XRD due to its amorphous nature [21, 186, 187]. As a result of mixing, all of the major peaks in the $CoFe_2O_4/SiO_2$ nanocomposites that correspond to the spinel structures remained unaltered.



Figure 4.46 XRD pattern of uncomposited CoFe₂O₄ nanoparticles, and nano composite CoFe₂O₄/SiO₂ in ratio (35%, 40%, 45, and 50%).

4.5.1.1.1 Crystallite Size (D)

The crystallite size (D) for each specimen was calculated by using Scherrer's formula (equation 2.22) using FWHM for peak (311).

The value of crystallite size increases with increasing SiO_2 mixing in SiO_2 mixed cobalt ferrite nanoparticles, implying that SiO_2 mixing enhances grain growth in spinel phase formation. The nature of the crystallite size variation is inconsistent with recent findings in the literature for cobalt ferrite/silica nanocomposites [186]. While the crystallite size is decreases with increasing SiO_2 ions at 35%, the grain growth is limited due to the large number of nucleation sites supplied by silica [186].

4.5.1.1.2 Lattice Parameter 'a'

For varied mixing concentrations, the lattice constant 'a' of prepared specimens with the same peaks was measured using equation (2.23). The values of the lattice constant 'a' determined for SiO_2 mixed are shown in table 4.14. With the exception of 40% and 50% SiO_2 mixed, the lattice

parameters for calcined specimens at $600 \,^{\circ}\text{C}$ rise as the SiO₂ mixed increases. The increased lattice constant could be attributed to the significant size fraction of the interface structure [188].

4.5.1.1.3 X-Ray Density (ρ_x)

The X-ray density (ρ_x) values were determined from the XRD data using equation (2.24). The value of X-ray density (ρ_x) for silica mixed cobalt ferrite decreases as the value of silica increases, with the exception of 40% and 50%, as indicated in table 4.14. It is directly linked to a decrease in molecular weight.

However, this was attributed to the actuality that silica atomic weight is lower than that of cobalt [189].

4.5.1.1.4 Hopping Length (L)

Equations (2.25 and 2.26) were used to measure the hopping length in A-site L_A (tetrahedral) and B-site L_B (octahedral).

As the silica ion is mixed, the hopping length increases, which is related to cation redistribution between the tetrahedral (A) and octahedral (B) sites. Furthermore, at 40% and 50%, the hopping length reduces as the amount of silica in the mixture increases. This decrease is due to a change in the physical properties of the ferrite composite [190, 191].

Mixing ratio	2Theta	FWHM	D(nm)	a(Å)	$\rho_x (g/cm^3)$	$L_A({A})$	L _B (Å)
0%	35.686	0.22	37.920	8.338	5.374	3.6105	2.9479
35%	35.593	0.22	37.910	8.359	5.333	3.6195	2.9553
40%	35.608	0.20	41.703	8.355	5.340	3.6180	2.9541
45%	35.519	0.19	43.887	8.376	5.301	3.6268	2.9613
50%	35.545	0.187	44.595	8.370	5.312	3.6242	2.9592

Table 4.14 Values of crystallite size (*D*), Lattice parameter '*a*', X-ray density (ρ_x), hopping length (L_A) and (L_B) for CoFe₂O₄/SiO₂ nanocomposites.

4.5.1.2 Fourier Transform Infrared Spectroscopy (FT-IR)

Fourier transform infrared (FT-IR) spectroscopy can be used to explore the nature of local chemical bonding in a substance. Because XRD does not provide evidence of amorphous SiO₂ surface coating, we used FT-IR spectroscopy to demonstrate the CoFe₂O₄/SiO₂ interactions. The FT-IR absorption spectrum of KBr palletized CoFe₂O₄/SiO₂ nanocomposites in the range 400-4000 cm⁻¹ is shown in figure 4.47. The intrinsic lattice vibration of the chemical (O-M_{Oct}.-O) bond at octahedral (B-site) and chemical (O-M_{tet}.-O) bond at tetrahedral (A-site) of CoFe₂O₄ nanoparticle causes the absorption bands at v_1 and v_2 , respectively [192]. The presence of these absorption bands at 468.702, 800.459, and 1103.28 cm⁻¹ correspond to the symmetric and asymmetric expansion Si-O-Si vibrations, confirming the formation of amorphous SiO₂ [192, 193].

Figure 4.47 and table 4.15 show that when SiO₂ nanoparticles are mixed, the absorption bands (Fe-O) (v_1) and (Co-O) (v_2) shift towards lower frequencies compared with that in pure CoFe₂O₄. This is likely due to SiO₂ amendment onto CoFe₂O₄ [194].

0%

35%

40%

45%

50%

576.716

574.787

572.858

572.858

570.929



Figure 4.47 FTIR spectra of CoFe₂O₄/SiO₂ nanocomposite specimens with different SiO₂ weight percentage.

,	different SiO2 weig	ght.				
Mining antis			FTIR fre	quency bands	(cm ⁻¹)	
Mixing ratio	V 1	<i>V</i> 2	V 3	V 4	<i>v</i> 5	

366.475

366.475

364.546

362.617

362.617

468.702

468.702

468.702

468.702

800.459

800.459

800.459

800.459

1103.28

1103.28

1103.28

1103.28

Table 4.15 FT-IR spectral bands for CoFe₂O₄/SiO₂ nanocomposite specimens with

4.5.1.3 Field Emission Scanning Electron Microscopy (FE-SEM)

FE-SEM images were used to analyze the surface morphology and particle size of $CoFe_2O_4/SiO_2$ nanocomposites calcined at 600 °C, as shown in the figures 4.48 (a, b, c, d and e). All of the specimens contain a compact order of homogenous nanoparticles with a spherical form and polyhedral particles, as illustrated in figure 4.48. Figure 4.48 (a) shows the image of pure $CoFe_2O_4$ without the addition of SiO_2 . By fashioning a dense structure visible from the shape, spherical crystallites with the majority of crystals aligned in the same direction can be formed. All of the crystals appear to be roughly the same size. Figure 4.48 (b) depicts a homogeneous microstructure with smaller crystallites distributed uniformly. When compared to the images in figure 4.48 (b), figure 4.48 (c) shows the bigger grains. In comparison to the previous image, figure 4.48 (d and e) exhibited a morphology consisting of agglomerated crystallites of diverse forms and sizes [195].

Table 4.16 shows the estimated diameters of $CoFe_2O_4/SiO_2$ nanocomposites for calcined specimens at 600 °C with comparatively wellcrystallized grains and a mean particle size less than 42.664, 42.028, 43.600, 45.472, and 47.523 nm respectively calculated by Image J Software. All data correspond relatively well with the XRD findings.

The particle size calculated using FE-SEM micrographs is bigger than that estimated using XRD data. The molecular structural disruption and lattice strain caused by different ionic radii and/or nanoparticle gathering can be attributed to the XRD technique. As a result, it has a stricter standard, resulting in smaller sizes [169].



Figure 4.48 FE-SEM images of CoFe₂O₄/SiO₂ nanocomposites calcined at 600 °C for; (0%, 35%, 40%, 45% and 50%).

Table 4.16 Average particle sizes of $CoFe_2O_4/SiO_2$ nanocomposites calcined at 600 °C with (0%, 35%, 40%, 45%, and 50%) calculated from XRD and FE-SEM.

Mixing ratio	D(nm) XRD	D(nm) FE-SEM
0%	37.920	42.664
35%	37.910	42.028
40%	41.703	43.600
45%	43.887	45.472
50%	44.595	47.523

4.5.1.3.1 Energy Dispersive Spectroscopy (EDS)

The elemental compositions of nanoparticles were investigated using an energy dispersive spectroscopy (EDS) approach triggered by an electron beam (10 keV), as illustrated in figure 4.49. The peaks at 0.5 keV are oxygen peaks (O). While silica has the highest peak at 1.8 kev (Si). The peaks at 0.8, 6.9 and 7.7 keV belong to cobalt (Co). The peaks at 0.7 and 6.5 kev belong to iron (Fe). EDS analysis confirms the presence of Co, Fe, O, and Si as major elements in the produced specimens. As the Si mixing concentration rises, the peak of Si appears to rise. The weight ratios of Co, Fe, O, and Si, as determined by EDS, are shown in the insets of figure 4.49.



Figure 4.49 EDS spectra of $CoFe_2O_4/SiO_2$ nanocomposites for; (0%, 35%, 40%, 45% and 50%)

4.5.2 Magnetic Properties

Magnetic measurements were carried out at room temperature utilizing a vibrating sample magnetometer (VSM) with a maximum applied field of ± 15 kOe. The M-H loops of CoFe₂O₄/SiO₂ nanocomposites are shown in figure 4.50 (at 35%, 40%, 45% and 50%). The saturation magnetization (M_s), remanent magnetization (M_r), and coercive field (H_c) values are calculated using the hysteresis loops mentioned in section (2.9). While the equations (2.5, 2.6, and 2.7) were used to measured the remanence ratio (M_r/M_s), magnetic moment (n_B), and magnetic anisotropy (K).

4.5.2.1 Saturation Magnetization

In the current measurement, the value of saturation magnetization (M_s) and magnetic moment (n_B) decreases when SiO₂ mixing increases (Figure 4.50), being 75.35 emu.g⁻¹ (3.16 μ_B) for pure and 41.296 emu.g⁻¹ (1.73 μ_B) for 50%. The presence of vacancies, interatomic spacing, and a low coordination number can explain the decrease in (M_s) with increasing SiO₂ mixing [186]. The values of magnetic moments (n_B) for SiO₂ mixed cobalt ferrites are listed in table 4.17. Pure cobalt ferrite (3.16 μ_B) had the greatest value of (n_B), which decreased with increased SiO₂ mixing and became (1.73 μ_B) at 50%. This result could be attributed to higher SiO₂ occupancy at B sites [161].

4.5.2.2 Remanent Magnetization

Remanent magnetization (M_r) is the magnetization that remains in ferrimagnetic materials after an external magnetic field has been removed. The remnant magnetization behavior of SiO₂ mixed cobalt ferrite (Figure 4.50) exhibits a decreasing trend with increasing SiO₂ mixing from (40.78 emu.g⁻¹) at pure cobalt ferrite to (22.728 emu.g⁻¹) at 50%, and follows the same pattern as saturation magnetization (Table 4.17). This decrease may be attributed to the non-crystalline SiO_2 formation on the surface of cobalt ferrite nanoparticles [196].

The squareness ratio (M_r/M_s) of a material determines its magnetic hardness, which is based on the system's anisotropy. The level of squareness determines whether or not grain exchange takes place. Because (M_r/M_s) values are greater than 0.5, our findings indicate that the nanoparticles have exchange coupled interaction [165].

4.5.2.3 Coercivity

The coercive force values (H_c) decrease from 2522.4 O_e (at pure) to 2399.5 O_e (at 40%) with an increase of mixing content SiO₂. Nonetheless, the coercive force values (H_c) increase from 2399.5 Oe (at 40%) to 2438.8 O_e (at 50%) with an increase of mixing content SiO₂.

It is possible that the decrease in coercivity when SiO_2 mixing increases is related to a decrease in anisotropy field, which reduces domain wall motion [197, 198]. The increase in $CoFe_2O_4/SiO_2$ coercivity values (at 45% and 50%) could be attributable to an increase in magnetic crystalline anisotropy [186].

Table 4.17 lists the measured magnetic anisotropy (*K*). The magnetic anisotropy (*K*) values in cobalt ferrite fall as the SiO₂ mixing increases, from 197.98 to 104.91×10^3 emu.O_e g⁻¹. The high magnetic anisotropy of cobalt ferrite is mainly owing to the presence of Co²⁺ ions on the octahedral (B) sites, and this observation may be understood of in terms of the impact of SiO₂ mixing on the site occupancies of the cations. As a result, *K* is reduced if SiO₂ enters the octahedral (B) site and at least some Co²⁺ ions are dislodged from the octahedral (B) site to the tetrahedral (A) site [199].



Figure 4.50 Hysteresis loops of $CoFe_2O_4/SiO_2$ nanocomposites with (0%, 35%, 40%, 45% and 50%) calcined at 600 °C.

Table 4.17 Variation in saturation magnetization (M_s) , remanence magnetization (M_r) , coercivity (H_c) , magnetic moment (n_B) , squareness ratio (M_r/M_s) , and magnetic anisotropy (K) for CoFe₂O₄/SiO₂ nanocomposites.

Mixing ratio	M _s	M _r	H _c	n_B	м /м	$K \times 10^3$
	$(emu. g^{-1})$	$(emu. g^{-1})$	(O_e)	(µ _B)	m_r/m_s	$(emu . O_e g^{-1})$
0%	75.35	40.78	2522.4	3.16	0.54	197.98
35%	56.775	30.890	2414.5	2.38	0.54	142.80
40%	51.309	27.996	2399.5	2.15	0.55	128.25
45%	47.658	25.937	2438.7	2.00	0.54	121.07
50%	41.296	22.728	2438.8	1.73	0.55	104.91

4.5.3 Electrical Properties

The dielectric parameters such as pellet capacitance, dielectric loss angle ($tan \delta$), and capacitance of air with the same thickness as the pellet of SiO₂ mixed cobalt ferrite CoFe₂O₄/SiO₂ (0%, 35%, 40%, 45%, and 50%) were calculated using an LCR meter at room temperature in the range of applied frequencies (50Hz -2 MHz).

The real part of the dielectric constant (ε'), the imaginary part of the dielectric constant (ε'') or dielectric loss factor, and the ac conductivity (σ_{ac}) are computed using the relationships shown in Section (2.17).

4.5.3.1 Dielectric Constant

The dielectric constant (ε') is a measure of the ability of an electrically insulating object or substance to store electric potential energy under the influence of an electric field [200]. Figure 4.51 demonstrates that increasing the frequency lowers the dielectric constant, and this effect is common in all types of ferrites, according to several researchers [186].



Figure 4.51 Variation of dielectric constant (ε') as a function of frequency of CoFe₂O₄/SiO₂ nanocompostie specimens with (0%, 35%, 40%, 45% and 50%).
To explain this behavior, it is necessary to look at the mechanisms of electrical polarization, which can be divided into four categories: Polarization of space charge, atomic or ionic polarization, orientational polarization, and electronic polarization are all examples of polarization. Due to the large mass of the two diodes, the polarization of the space charge is believed to own the largest part of a contribution to the total polarization. Moreover, when the frequency of the external electric field increases, these diodes are unable to spin with it, causing it to become regular and arranged in its direction. It subsequently ceases to be a part of the total electrical polarization, resulting in a reduction in electrical insulation [201]. At various frequencies (200 kHz, 500 kHz, 1 MHz, 1.5 MHz, and 2 MHz), the values of the dielectric constant (ε') for CoFe₂O₄/SiO₂ are measured as shown in table 4.18.

In addition, the current study found that the dielectric constant (ε') increases when SiO₂ composition increases from 0 to 50%, which is consistent with an earlier reports by Q.M. Ahkam et al. and A.B. Kadam et al. [193, 202]. The incident frequency that causes the dielectric constant to vary can be controlled via space charge polarizations. When an electric field is applied, the dielectric constant rises due to charge trapping, causing space charge to accumulate at grain boundaries, leading the grain boundaries to become active. This could account for why nanostructured materials have greater dielectric constants than bulk materials.

4.5.3.2 Dielectric Losses

The dielectric loss is described in terms of dielectric loss angle $(tan \delta)$ and dielectric loss factor (ε'') and represents the electromagnetic energy dissipate in a dielectric material [103]. Figure 4.52 display the variation of dielectric loss angle as a function of frequency of CoFe₂O₄/SiO₂ nanocomposite specimens (at a mixing ratio 35%, 40%, 45% and 50%) and

figure 4.53 show the variation of dielectric loss factor as a function of frequency of CoFe₂O₄/SiO₂ nanocomposite specimens and their values at various frequencies (200 kHz, 500 kHz, 1 MHz, 1.5 MHz, and 2 MHz) summarized in table 4.18. It is possible to see that the dielectric loss angle $(tan \delta)$ and dielectric loss factor (ε'') appear a decreasing direction with increasing frequency and it then stays constant at high values, which is a natural behavior for any ferrite material. The Koop's phenomenological model illustrates the decrease in the values of both $(\tan \delta)$ and (ε'') with increasing frequency [203]. Only a small amount of energy is required for electron hopping between Fe²⁺ and Fe³⁺ ions at the octahedral B-site at high frequencies, which correspond to low resistivity (due to the grains). Although grain boundaries in the low frequency district are energetic due to high resistivity, electron hopping between Fe^{2+} and Fe^{3+} ions at the Bsite needs a large amount of energy. As a result, one may conclude that the dielectric behavior of the studied ferrite specimens is caused by a two-layer of the type [204].



Figure 4.52 Variation of dielectric loss angle $(tan \delta)$ as a function of frequency of CoFe₂O₄/SiO₂ nanocomposite specimen with (0%, 35%, 40%, 45% and 50%).



Figure 4.53 Variation of dielectric loss factor (ε'') as a function of frequency of CoFe₂O₄/SiO₂ nanocomposite specimen with (0%, 35%, 40%, 45% and 50%).

The dielectric loss (tan δ) and (ε'') have high values for pure cobalt ferrite, but these values decrease with increasing SiO₂ mixing for CoFe₂O₄/SiO₂ nanocomposite (35-50%).

This shows that the energy losses at high frequencies decrease as the SiO_2 content increases. As a result, they are good for high-frequency applications. In the interfacial areas, such as the materials surface, grain boundaries have many defects or faults compared to the intrinsic grain area. Many factors influence the dielectric loss angle and factor, including the technique of production, Fe^{2+} content, density, structural homogeneity, and sintering history [205].

4.5.3.3 AC Conductivity (σ_{ac})

The frequency dependence of the ac electrical conductivity (σ_{ac}) measured at room temperature for CoFe₂O₄/SiO₂ nanocomposites is shown in figure 4.54. The conductivity obviously increases with frequency, and Q. M. Ahkam et al. discovered a similar ac conductivity (σ_{ac}) pattern for cobalt-doped SiO₂ [193].



Figure 4.54 Variation of ac conductivity (σ_{ac}) as a function of frequency of CoFe₂O₄/SiO₂ nanocomposite specimen with (0%, 35%, 40%, 45% and 50%).

The increase in ac electrical conductivity with increase in frequency is related with the magnitude of the electronic exchange of Fe^{2+}/Fe^{3+} ion pairs present at the octahedral sites [95].

The frequency dependent variation of ac electrical conductivity has been elucidated using the Maxwell-Wagner double layer model for dielectrics. Grain boundaries are more effective at lower frequencies. As a result, at lower frequencies, the electron hopping frequency between Fe²⁺ and Fe³⁺ is dramatically hampered. As a result, the observed conductivity of the materials at lower frequencies is reduced. However, when the frequency of the applied field increases, the conductive grains become more effective, stimulating Fe^{2+} and Fe^{3+} ion hopping on the octahedral B sites. As a result, as the frequency increases, the electrical conductivity increases as well [206, 207].

As demonstrated in table 4.18, the frequency dependent ac electrical conductivity of the cobalt ferrite nanocomposite materials is also clearly reliant on the SiO₂ mixing. Pure (CoFe₂O₄) and mixed at a ratio of 50% (CoFe₂O₄/SiO₂) conductivity values were found to be the highest and lowest respectively. Because the transfer of Fe³⁺ ions from the tetrahedral site (A site) to the octahedral site (B site) is restricted and weak, the number of Fe²⁺ and Fe³⁺ ion pairs decreases. As a result, the hopping of electrons involved in ferrites during the conduction process is lowered. The present study found that increasing the SiO₂ mixing reduces ac conductivity (σ_{ac}), which contradicts a previous study by K. Nadeem et al. [186].

Table 4.18 Values of dielectric constant (ε'), dielectric loss angle ($tan \delta$), dielectric
loss factor (ϵ'') and ac conductivity (σ_{ac}) at 200 KHz, 500 KHz, 1 MHz, 1.5 MHz and
2 MHz for CoFe ₂ O ₄ /SiO ₂ nanocomposites.

Dielectric parameters	0%	35%	40%	45%	50%
ε' (200KHz)	13.2	13.3	13.3	13.4	13.7
tan δ (200KHz)	0.661	0.588	0.446	0.381	0.286
ε'' (200KHz)	8.80	7.83	5.96	5.15	3.96
$\sigma_{ac} (200 KHz) \times 10^{-5}$	9.75	8.66	6.58	5.70	4.37
ε' (500KHz)	10.3	10.5	11.2	11.5	12.1
tan δ (500KHz)	0.551	0.433	0.316	0.270	0.205
ε'' (500KHz)	5.74	4.61	3.57	3.17	2.54
$\sigma_{ac} (500 KHz) \times 10^{-5}$	15.8	12.7	9.80	8.70	6.93
ε' (1MHz)	8.94	9.18	10.0	10.5	11.2
tan δ (1MHz)	0.404	0.326	0.233	0.204	0.157
ε'' (1MHz)	3.66	3.05	2.39	2.18	1.80
$\sigma_{ac} (1MHz) \times 10^{-5}$	20.1	16.7	13.1	11.8	9.76
ε' (1.5MHz)	8.19	8.45	9.35	9.78	10.5
tan δ (1.5MHz)	0.319	0.275	0.195	0.172	0.133
ε'' (1.5MHz)	2.65	2.37	1.87	1.73	1.45
$\sigma_{ac} (1.5 MHz) \times 10^{-5}$	21.7	19.4	15.2	14.0	11.7
ε' (2MHz)	7.60	7.85	8.77	9.17	9.84
tan δ (2MHz)	0.266	0.244	0.171	0.152	0.119
$\varepsilon^{\prime\prime}(2MHz)$	2.07	1.96	1.55	1.45	1.22
$\sigma_{ac} (2MHz) \times 10^{-5}$	22.4	21.2	16.7	15.5	13.0

Chapter Five

Conclusions and Suggestions for

Future Work

5.1 Conclusions

Sol-gel auto combustion method was used for the synthesis $Co_xFe_{3-x}O_4$ ferrite nanoparticle specimens (x= 0.8, 0.9, 1, 1.1, and 1.2) respectively, for as-burnt and calcined specimens at various temperatures 500,600, and 700°C. The conventional ceramics method is then used to generate nanoparticles of SiO₂ mixed cobalt ferrite with concentrations of CoFe₂O₄/SiO₂ of (0%, 35%, 40%, 45%, and 50%), respectively. The structural, magnetic, and electrical properties were also investigated. The following sections explain some key findings from the research.

1- The XRD patterns of $Co_xFe_{3-x}O_4$ nanoferrites showed all the major peaks corresponding to the single phase of the spinel structure. When the calcination temperature increased, the diffraction peaks became sharper, narrower and more intense, and the crystallite size increases basically with increasing cobalt content when calcination temperature is the same, for except (x=1.2). The XRD patterns of CoFe₂O₄/SiO₂ nanocomposites, on the other hand, revealed all of the key peaks corresponding to the spinel structure's single phase. Due to its amorphous nature, there are no signatures for SiO₂ in XRD.

2- FT-IR spectra revealed two principal absorption bands in the range of 600- 400 cm⁻¹, indicating the formation of a cubic spinel structure. 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 (x) content. Also the FT-IR spectra of CoFe₂O₄/SiO₂ revealed principal absorption bands at 468.702, 800.459 and 1103.28 cm⁻¹, indicating the formation of amorphous SiO₂. In CoFe₂O₄/SiO₂ nanocomposites a similar trend in shifting of the (Fe-O) (v_1) and (Co-O)

 (v_2) bands towards the lower frequencies compared with that in pure CoFe₂O₄ have observed with the increase mixing SiO₂.

3- FE-SEM image showed the porous nature, nanosized and almost homogenous distribution of particle size. From these images, one can notice the micrographs of cobalt ferrite nanoparticles $Co_xFe_{3-x}O_4$ reveal that the microstructure of the ferrites were affected by the calcination temperatures and (x) content and displays agglomerated with homogenous spherical and polyhedral particles. While FE-SEM images for $CoFe_2O_4/SiO_2$ showed that all specimens contain a compact order of homogeneous nanoparticles with a spherical form and polyhedral particles. From these images, one can notice the micrographs of cobalt ferrite-silica nanocomposites $CoFe_2O_4/SiO_2$ reveal that the microstructure of the ferrites were affected by the SiO₂ mixed and shows an increase in grain size when compared to cobalt ferrite.

4- The saturation magnetization (M_s) and remanence magnetization (M_r) of Co_xFe_{3-x}O₄ nanoferrites are increasing with increasing the calcined temperature, when Co²⁺ content increases, also the saturation magnetion at calcined temperatures (600 and 700 °C) increases except for x=1.2. Whereas the remnant magnetization decreases with increasing content Co²⁺ of as-burnt and calcined specimens. While the saturation magnetization (M_s) and remanence magnetization (M_r) of CoFe₂O₄/SiO₂ nanocomposites decrease with the increase of the SiO₂ mixing ratio.

5- The magnetic moment (n_B) of $\text{Co}_x\text{Fe}_{3-x}\text{O}_4$ nanoferrites are growing up with increasing the calcined temperature, when the Co^{2+} content is increased, also magnetic moment increases for calcined specimens at (500,600 and 700 °C) except for x=1.2. While the magnetic moment (n_B) of $\text{CoFe}_2\text{O}_4/\text{SiO}_2$ nanocomposites decrease with the increase of the SiO_2 mixing ratio. 6- The dielectric constant (ε') of the Co_xFe_{3-x}O₄ nanoferrites and CoFe₂O₄/SiO₂ nanocomposites decrease by increasing the frequency at room temperature indicates the normal dielectric behavior for all the specimens, caused by the lagging of polarization behind the changing frequency. It was also found that the dielectric constant (ε') decreases with increasing temperature and increases with increasing of the SiO₂ mixing ratio.

7- The dielectric loss angle $(tan\delta)$ and dielectric loss factor (ε'') shows a decreasing trend with increasing frequency, it was also found that the dielectric loss angle and dielectric loss factor increases with increasing temperature and decreases with an increasing of the SiO₂ mixing ratio.

8- The AC conductivity (σ_{ac}) of Co_xFe_{3-x}O₄ nanoferrites and CoFe₂O₄/SiO₂ nanocomposites, gradually increases as the frequency increases, it was also found that the ac conductivity (σ_{ac}) increases with increasing temperature and decreases with an increasing of the SiO₂ mixing ratio.

5.2 Suggestions for Future Work

- 1- Preparing the same ferrite (cobalt ferrite) by hydrothermal method and comparing the results with the results of the present work.
- 2- Preparing of cobalt ferrite-silica thin film and studying their structural, magnetic and electrical properties.
- 3- Studying of the UV-Vis absorption spectra to calculate the band gap of $CoFe_2O_4/SiO_2$ in different temperatures.
- 4- Studying the effect of Laser on the porosity of $CoFe_2O_4$, or treatment by laser.
- 5-Magnetization and mössbauer study of cobalt ferrite-silica nanocomposites.

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الخسلاصة

تم تحضير فيرايت الكوبلت ($Co_xFe_{3-x}O_4$) النانوية بنسب مختلفة (x = 8.0، 0.9، 1، 1.1، و (1.2) باستخدام طريقة السول – جل ذات الاحتراق التلقائي. تم الكلسنة بدرجة حرارة (500 و 600 و (0.00) لمدة ثلاث ساعات لزيادة التجانس و إزالة الشوائب. تم اضافة اربع قطرات من مادة (PVA) المواد النانوية المحضرة كمادة رابطة وكبسها على شكل أقراص دائرية بقطر (13mm)، ثم تلبيدها عند درجات حرارة (0.00 و 0.00 و 0.00) على التوالي لمدة ثلاث ساعات لفحص خصائص العزل الكهربائي. كما تم تحضير فيرايت الكوبلت – سيليكا ($CoFe_2O_4/SiO_2$) النانوية بنسب مختلفة (0.5%، 0.5%، 0.5%) باستخدام الطريقة السيراميكية وتم الكلسنة عند (0.000)، تم اضافة أربع قطرات من مادة (0.0%) باستخدام الطريقة السيراميكية وتم الكلسنة عند (0.000)، تم اضافة أربع قطرات تم تلبيدها عند درجة حرارة (0.000) لمدة ثلاث ساعات الموسود المائية (0.000)، تم اضافة أربع قطرات من مادة (0.000) المواد النانوية المحضرة كمادة رابطة وكبسها على شكل أقراص دائرية بقطر (0.000)، تم اضافة أربع قطرات تم تلبيدها عند درجة حرارة (0.000) لمدة ثلاث ساعات الموسود بنسب مختلفة (0.000)، تم اضافة أربع قطرات

أظهرت نتائج حيود الاشعة السينية XRD ان جميع القمم الرئيسة لفيرايت الكوبلت النانوية كانت مطابقة للتركيب السبنل الاحادي. يزداد حجم البلورات المتكونة لعينات الفيرايت مع زيادة درجة حرارة الكلسنة ومحتوى الكوبلت.

أظهرت نتائج حيود الاشعة السينية (XRD) لمركبات CoFe₂O₄/SiO₂ النانوية عن ان جميع القمم الرئيسية مطابقة لتركيب السبنل. بينما لا توجد قمم لـ SiO₂ في XRD بسبب طبيعتها غير المتبلورة.

أظهرت مطيافية الأشعة تحت الحمراء (FT-IR) نطاقي امتصاص رئيسية في المدى ${\rm Co}_{\rm x}{\rm Fe}_{3-{\rm x}}{\rm O}_4$. ولا المكتبي في فيرايت الكوبلت ${\rm Co}_{\rm x}{\rm Fe}_{3-{\rm x}}{\rm O}_4$. ولا المكتبي في فيرايت الكوبلت ${\rm Co}_{\rm x}{\rm Fe}_{3-{\rm x}}{\rm O}_4$. ولا المكتبي في فيرايت الكوبلت ${\rm Co}_{\rm x}{\rm Fe}_{3-{\rm x}}{\rm O}_4$. والنانوية، كما لوحظ نمط مماثل من انتقال القمم للنطاقات v_1 بإتجاه الترددات العالية بينما لوحظ نمط مماثل من انتقال القمم للنطاقات من بالمكتبي في فيرايت الكوبلت العالية بينما لوحظ نمط مماثل من النانوية، كما لوحظ نمط مماثل من انتقال القمم للنطاقات مع زيادة درجة الحرارة الكلسنة ومحتوى الكوبلت. من انتقال قمم النطاقات المنخفضة مع زيادة درجة الحرارة الكلسنة ومحتوى الكوبلت. أظهرت أطياف (FT-IR) نطاقات المتصاص رئيسية عند 400.200 و 800.459 و 800.459 م

مما يشير إلى تكوين SiO_2 غير المتبلورة في المركبات $CoFe_2O_4/SiO_2$ النانوية، وقد لوحظ انتقال مماثل لكل من النطاقين (v_1) (v_2) و (v_2) (Co-O) بإتجاة الترددات المنخفضة مع زيادة خلط SiO_2.

وفقًا لصور المجهر الإلكتروني الماسح الباعث للمجال (FE-SEM) لجسيمات فيرايت الكوبات Co_xFe_{3-x}O₄ النانوية، لوحظ زيادة حجم الجسيمات مع ارتفاع درجة حرارة الكلسنة ومحتوى الكوبالت كما تظهر تكتلات مع جسيمات متجانسة كروية واخرى متعددة السطوح. تم استخدام طيف تشتت الطاقة (EDS) لتأكيد وجود CO و Fe و O في جميع العينات. ومن جانب آخر، وفقًا لصور المجهر الإلكتروني الماسح الباعث للمجال (FE-SEM) لمركبات فير ايت الكوبلت - سيليكا COFe₂O₄/SiO₂ النانوية، تبين زيادة حجم الجسيمات مع زيادة نسبة خلط سيليكا، ويظهر ذلك أن جميع العينات تحتوي على ترتيب مضغوط من الجسيمات النانوية المتجانسة الكروية واخرى متعددة السطوح. تم استخدام طيف تشتت الطاقة (EDS) لتأكيد وجود OO و Fe و O في جميع العينات. ومن جانب آخر، وفقًا لصور المجهر الإلكتروني زيادة حجم الجسيمات مع زيادة نسبة خلط سيليكا، ويظهر ذلك أن جميع العينات تحتوي على ترتيب

 تم قياس خصائص العزل الكهربائي باستخدام مقياس (LCR) في مدى تردد (SOHz-2MHz) عند درجة حرارة الغرفة. وجد ان ثابت العزل الكهربائي ('ع) وزاوية ظل الفقد العزلي ($\delta m\delta$) ومعامل الفقد العزلي (''ع) للفيرليتات O_xFe_3 -xO4 النانوية و المركبات OFe_2O_4/SiO_2 النانوية تتناقص مع زيادة التردد. وان هذا السلوك يعتبر سلوكاً نموذجًا للفيرايت كما هو موضح في نظرية (s' (Koop)). وجد ان ثابت العزل الكهربائي يتناقص مع زيادة درجة الحرارة ويزداد مع زيادة نسبة خلط SiO_2 . كما وجد أن زاوية ظل الفقد العزلي ومعامل الفقد العزلي يزدادان مع زيادة درجة الحرارة وينخفضان مع زيادة دسبة الخلط SiO2. الفقد العزلي ومعامل الفقد العزلي يزدادان مع زيادة درجة الحرارة وينخفضان مع زيادة نسبة الخلط SiO2. ان التوصيلية المتناوبة (σ_{ac}) تزداد تدريجياً مع زيادة التردد، كما وجد أن التوصيلية المتناوبة تزداد مع زبادة درجة الحرارة وتنخفض مع زيادة نسبة خلط SiO2.



الخصائص المغناطيسية الكهربائية لمركبات فيرايت الكوبلت – سيليكا المحضر باستخدام تقنية السول-جل

باشراف

أ.م.د.علي مصطفى محمد

أد تحسين حسين مبارك

2021 م

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