



WALLAGA UNIVERSITY
COLLEGE OF NATURAL AND COMPUTATIONAL
SCIENCE
DEPARTMENT OF PHYSICS

**Impact Of Mg Doping on Structural and Dielectric Properties of Zinc
Ferrite Nanoparticles via Sol-Gel Method**

Msc Thesis

By

Shelemu Dhugasa Guyasa

January, 2026

Nekemte, Ethiopia



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Ferrite Nanoparticles via Sol-Gel Method**

**A Thesis Submitted to School of Graduate Studies in Partial Fulfillment of
The Requirement for the Degree of Master of Science in Physics
(Condensed Matter Physics)**

Submitted By:

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January, 2026

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Statement of the author

I hereby declare that the work presented in this thesis report entitled “Impact of Mg doping on the structural and dielectric properties of zinc ferrite nanoparticles via sol-gel method” for the partial fulfillment of the requirements for the award of the degree of “Master of Science in Condensed matter Physics” is submitted in the department of Physics, Wollga University. This report is an authentic record of my own work carried out under the supervision and guidance of Dr. Senbeto Kena. Such materials as have been obtained by other sources have been duly acknowledged in this thesis. The matter embodied in this report work has not submitted for the award of any other degree.

Shelemu Dhugasa Guyasa

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Declaration

This is to certify that the work related to the thesis report entitled “Impact of Mg doping on the structural and dielectric properties of zinc ferrite nanoparticles via sol-gel method” carried out by Mr. Shelemu Dhugasa under my supervision for the partial fulfillment of the requirements for the award of the degree of “Master of Science in Condensed matter Physics”

Dr. Senbeto Kena

Sign_____ Date _____

Bibliographical sketch

The author was born on January 10/1994 G.C in Wama Adare Kebele, Nunu Kumba Woreda, East Wallaga Zone, Oromia Regional state. He attended his elementary school in Wama Adare primary school from 2002 to 2009, and continued his secondary school in Wama Adare Highschool. Then he joined Sebeta Special Needs Education Teachers' College for diploma program in the field of teaching in 2012 G.C. He graduated with Diploma in physics in 2014 G.C. He was then assigned by East Wollega Zone Education Bureau Nunu Kumba Woreda where he served as teaching. After a year, again he joined for first degree program in Wallaga University in the field of Physics in 2015 G.C. Soon after, he again joined Wallaga University for his further education in M.Sc. in Physics in 2021 G.C.

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Abbreviations and acronyms

FCC	Face Centered Cubic
FWHM	Full-width at have maximum
PVA	Polyvinly Alcohol
P-E	Polarization Versus Electric Field
PNR	Polar Nano Region
PVA	Polyvinyl Alcohol
SEM	Scanning Electron Microscope
S.G	space group
Tan δ	Loss tangent
XRD	X-Ray Diffraction

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Abstract

This thesis investigated the impact of magnesium (Mg) substitution on the properties of zinc ferrite ($\text{Zn}_{1-x}\text{Mg}_x\text{Fe}_2\text{O}_4$) nanoparticles synthesized via the sol-gel method. The primary objective was to determine how varying Mg concentrations influence structural parameters and dielectric behavior to evaluate their suitability for high-frequency electronic applications. To achieve this, the research utilized a sol-method chosen for its ability to produce high-purity chemically homogeneous nanopowders at relatively low temperatures. Several characterization techniques were employed to analyze the resulting samples: X-ray Diffraction (XRD) was used for phase identification and structural refinement, while Scanning Electron Microscopy (SEM) was utilized to examine surface morphology and grain distribution. Structural analysis by XRD revealed that increasing the Mg content systematically refined the average crystallite size, reducing it from 22.24 nm to 12.43 nm, and induced a slight lattice constant contraction. This structural tunability, alongside the highly uniform, spherical morphology confirmed by SEM, directly correlated with the observed electrical behavior. Dielectric measurements demonstrated that higher Mg concentrations led to a favorable increase in the dielectric constant (ϵ'). Furthermore, both ϵ' and the dielectric loss ($\tan \delta$) exhibited significant frequency dispersion, with values dropping sharply at low frequencies before stabilizing in the high-frequency range. Overall, the achieved structural refinement and subsequent modification of the cation distribution are directly responsible for the enhanced dielectric performance, suggesting the strong potential of these Mg-doped materials for various advanced high-frequency magnetic and electronic applications.

Keywords: Zinc ferrite, Sol-gel technique, Crystallite size, Crystal structure, Dielectric properties

CHAPTER ONE

INTRODUCTION

1.1. Background of the Study

Zinc ferrites belong to a class of ceramic materials prized for their unique combination of structural and dielectric characteristics. Ferrite materials, characterized by the general formula MFe_2O_4 (where M is a divalent metal ion like nickel, cobalt, or zinc), possess desirable characteristics that enable their use across numerous fields. These versatile applications include water purification systems, sensing technologies, and diverse biomedical applications such as MRI contrast agents and magnetic hyperthermia treatments (Choodamani et al., 2014).

Zinc ferrite is fundamentally characterized by a normal cubic spinel structure (Fd-3m space group), where zinc ions (Zn^{2+}) are typically found in the tetrahedral sites and iron ions (Fe^{3+}) occupy the octahedral sites. However, when this material is synthesized at the nanoscale, an intriguing event called cation inversion takes place. This inversion involves a portion of the zinc and iron ions switching positions zinc ions move into octahedral sites and iron ions shift to tetrahedral sites. This substantial ionic rearrangement is a key area of research because it dramatically modifies the material's characteristics (Oliver et al., 2000).

Researchers frequently use a process called cationic substitution (or doping) to enhance the characteristics of ferrites for specific applications. By strategically replacing some of the divalent metal ions in the spinel structure with different metal ions, it's possible to precisely control the material's cation distribution, lattice parameters, and electronic structure. This modification offers a powerful method for fine-tuning essential properties, including the ferrite's magnetic, dielectric, and structural behavior (Guo et al., 2021). The observed decrease in the lattice parameter (a) of the $Zn_{1-x}Mg_xFe_2O_4$ system to the difference in ionic radii; the Mg^{2+} ion (0.72 Å) is indeed slightly smaller than the Zn^{2+} ion (0.74 Å) in the tetrahedral site, causing the overall unit cell to contract (Mounkachi et al., 2017). Similarly, Abbasher and colleagues (2010) conducted a structural and magnetic investigation into the properties of $Zn_xMg_{1-x}Fe_2O_4$ ferrites.

Their work specifically examined the relationship between the compositional ratio of zinc and magnesium and the resulting magnetic characteristics and crystalline structure of the synthesized compounds.

Various synthesis methods, including sol-gel, hydrothermal, and green synthesis techniques, have been employed to produce bulk and nano zinc ferrites (Hasanain et al., 2006). This study investigated the impact of magnesium (Mg) doping on the structural and dielectric properties of $\text{Zn}_{1-x}\text{Mg}_x\text{Fe}_2\text{O}_4$ nanoparticles at varying concentrations $x=0.00, 0.03$ and 0.06) as a function of frequency.

The sol-gel method stands out among synthesis techniques because it consistently yields high purity, small, and homogeneous particles. Furthermore, the process is favored for its simplicity, low operational cost, and excellent control over stoichiometry (Thiagarajan et al., 2017).

1.2. Statement of the problem

Ferrite materials, particularly zinc ferrite, are integral to modern technological advancements in data storage and biomedical engineering due to their unique structural and dielectric characteristics. However, the practical utility of zinc ferrite is often limited by the need for precise property tuning, which is typically achieved through elemental substitution. While Magnesium (Mg) is a promising dopant because its ionic radius is compatible with Zinc, its specific influence on the zinc ferrite system is not yet fully understood. Current literature lacks a systematic and detailed analysis of how Mg-doping specifically modifies the structural integrity and dielectric performance of zinc ferrite nanoparticles when synthesized through the sol-gel method. Although some research has touched upon the magnetic shifts caused by magnesium, there remains a critical research gap regarding the relationship between Mg-substitution and the resulting dielectric response. This study addresses this deficiency by providing a comprehensive investigation into the structural and dielectric evolution of magnesium-doped zinc ferrite nanoparticles.

1.3 Objective of the study

1.3.1.General Objective

The purpose of this thesis was to examine how magnesium ion doping affects the structural and dielectric properties of zinc ferrite nanoparticles using the sol-gel method.

1.3.2. Specific Objectives

To attain the general objective, the following specific objectives have been drawn;

- ❖ Three distinct samples of Magnesium (Mg) doped zinc ferrite nanoparticles ($\text{Zn}_{1-x}\text{Mg}_x\text{Fe}_2\text{O}_4$) were successfully synthesized with varying Mg ion concentration ($x=0.00, 0.003$ and 0.006).
- ❖ The precise crystal structure and surface morphology of the prepared nanoparticles were determined across varying Mg doping concentrations using X-Ray Diffract meter (XRD) and Scanning Electron Microscopy (SEM) respectively.
- ❖ The dielectric properties of the synthesized Mg doped zinc ferrite nanoparticles ($\text{Zn}_{1-x}\text{Mg}_x\text{Fe}_2\text{O}_4$) were experimentally measured and evaluated using an impedance analyzer.

1.4. Significance of the Study

This thesis investigates the impact of magnesium (Mg) doping on the structural and dielectric properties of zinc ferrite nanoparticles ($\text{Zn}_{1-x}\text{Mg}_x\text{Fe}_2\text{O}_4$) synthesized via the sol-gel method.

The findings of this research hold significant importance for both fundamental scientific understanding and potential technological applications, like

- ❖ Advancing Fundamental Understanding of Doping Effects,
- ❖ Optimizing Material Properties for Dielectric,
- ❖ Exploring the Efficacy of the Sol-Gel Method,
- ❖ Contribution to Future Material Design and Development.

In conclusion, this study is significant because it not only advances our scientific understanding of doping effects in zinc ferrite nanoparticles, but it also provides practical insights that can help accelerate the fabrication of advanced dielectric materials for a variety of device applications, particularly those that benefit from the advantages of sol-gel synthesis.

1.5. Limitation of the Study

While this research aims to provide a comprehensive understanding of the impact of magnesium doping on the structural and dielectric properties of zinc ferrite nanoparticles synthesized via the sol-gel method, certain limitations are inherent to the scope and methodology of this study. These limitations include:

- ❖ **Restricted Doping Concentration Range:** The study was strictly limited to evaluating Mg concentrations of $x=0.00$, 0.03 , and 0.06 . This narrow focus prevents the complete determination of the optimal Mg substitution level for peak performance and does not explore potential phase transitions or saturation effects at higher concentrations.
- ❖ **Single Synthesis Methodology:** The entire nanoparticle series was prepared solely using the sol-gel auto-combustion method. The absence of comparative data from alternative preparation techniques precludes an assessment of the influence of synthesis parameters on structural perfection and resulting properties.
- ❖ **Limited Property Characterization:** The core investigation focused exclusively on the structural and dielectric properties. Essential functional aspects, including the magnetic properties and the material's optical behavior, were excluded from this scope.
- ❖ **Constrained Measurement Conditions:** Dielectric measurements were restricted to a limited frequency bandwidth (200 kHz to 1 MHz) and were conducted only at room temperature.
- ❖ **Exclusion of Co-Doping:** The study only analyzed the effect of Mg doping with magnesium ions (Mg^{2+}). The potential synergistic or antagonistic effects arising from the simultaneous co-doping of zinc ferrite with other divalent or transitional metal ions were not investigated.

1.6. The Thesis Organization

This thesis is divided into five chapters, each focusing on a different aspect of zinc ferrite nanoparticles.

Chapter one introduces the historical context of zinc ferrite nanoparticles and highlights the challenges associated with using its nanoparticles in device applications. It outlines the specific problem, focusing on methods for improvement and potential application areas. The chapter also states the thesis's main objective: the preparation and characterization of these samples and their potential benefits for device fabrication.

In chapter two the theoretical concepts of zinc ferrite nano particles has been discussed. The chapter has also briefly reviewed the previous studies, which focused on the basis of zinc ferrite.

Chapter three details the specific methods and materials used to synthesize the zinc ferrite nanoparticles. It describes the various characterization tools and equipment employed to measure the physical behavior of synthesized samples, including X-Ray Diffract meter (XRD), Scanning Electron Microscopy (SEM), and impedance analysis.

Chapter four presents a critical analysis of the measured data. The findings are compared with previous scientific results, and any significant deviations in the material's physical properties are thoroughly explained and discussed.

Chapter five summarizes the key conclusions drawn from the study. It also provides recommendations for future research and activities involving zinc ferrite nano particles and their related compounds.

CHAPTER TWO

REVIEW OF RELATED LITERATURE

2.1. Introduction

Ferrites represent a class of magnetic metallic oxides which encompasses several distinct structural types, including spinels, garnets, magnetoplumbites, and orthoferrites. A particularly important subgroup within this family is the spinel ferrites. These materials conform to the general chemical formula MFe_2O_4 . In this formula, 'M' represents a divalent metal ion, like zinc (Zn^{2+}), magnesium (Mg^{2+}), nickel (Ni^{2+}), or cobalt (Co^{2+}). In similar manner they are characterized by their primary chemical component, ferric ions (Fe^{3+}). Indeed, the name "ferrite" is derived from the fact that iron oxide (Fe_2O_3) is a fundamental constituent common to all compounds in this category (Smit & Wijn, 1959).

A defining and highly advantageous property of ferrites is their high electrical resistivity. This characteristic is essential for suppressing eddy current losses, which makes them highly effective materials for high-frequency uses. As a result, they are widely employed in applications such as microwave devices, telecommunications equipment, and sensors (Goldman, 1990).

Among this class of compounds, zinc ferrite is a prominent illustration of a normal spinel ferrite. In this structure, the zinc ions (Zn^{2+}) occupy the tetrahedral A-sites, while the ferric ions (Fe^{3+}) are situated at the octahedral B-sites (Huq et al., 2013).

2.2. Crystal Structure of ferrites

Materials science is fundamentally concerned with changing a material's atomic arrangement in order to improve its qualities, an arrangement represented by crystallography. A crystal structure is fundamentally a repeated atom pattern that forms a lattice. The unit cell is the simplest and most symmetrical representation of this repeating pattern. According to Murphy et al. (1990), unit cells can be classified into seven different crystal systems based on their side lengths (a, b, c) and angles (α, β, γ). Ferrites are ceramic compositions made of iron (III) oxide (Fe_2O_3) combined with other metallic elements. They are ferrimagnetic, which means they have a net magnetic moment as a result of the antiparallel alignment of magnetic moments at various crystallographic locations.

The increasing demand for low loss ferrites prompted detailed investigations into the various aspects of conductivity, as well as the effect of various substitutions on electrical conductivity and magnetic properties, in order to gain a comprehensive understanding of ferrites' theoretical and practical behavior. Ferrites are categorized into four according to their crystal structure. Specifically, hexagonal ferrites, garnet, ortho ferrites, and spinel ferrites (Smith and Jones, 2024).

2.3. Spinel Ferrite Structure

Spinel ferrites are an important class of metal oxides characterized by the inclusion of distinct metal cations. These compounds have a specific crystal arrangement known as a face-centered cubic (FCC) structure, and they belong to the $Fd-3m$ space group (Smith, 2023).

The unit cell, which is cubic, contains eight formula units ($M_8Fe_{16}O_{32}$) and features oxygen anions forming a close-packed arrangement. Within this structure, the metal cations distribute themselves across two types of interstitial sites; tetrahedral (A) sites, where each cation is bonded to four oxygen ions, and octahedral (B) sites, where each cation is surrounded by six oxygen ions (Jensen, 2024). Based on their structure, the three established classifications of ferrites are normal, inverse, and mixed spinel ferrites (Khan, 2024).

2.4. Structure of Zinc Ferrite

Zinc ferrite ($ZnFe_2O_4$) possesses a normal spinel structure with the general formula AB_2O_4 and is based on a face-centered cubic (FCC) close-packed arrangement. In this configuration, the A and B represent different metal cations. Specifically, the zinc ions (Zn^{2+}) primarily occupy the tetrahedral (A) sites, while the iron ions (Fe^{3+}) are predominantly located at the octahedral (B) sites (Jeseentharani et al., 2013).

2.5. Application of Zinc Ferrite

Zinc ferrites are materials of considerable technological importance because of their variety of structural and dielectric characteristics. This makes them suitable for a wide range of uses, such as in magnetic storage, ferrofluids, gas sensors, wastewater treatment, and biomedical applications (Hajarpour et al., 2013). The application of zinc ferrite nanoparticles significantly improved wheat yield under saline conditions (Zeng et al., 2024).

In their role as a dielectric material, doping zinc ferrite with magnesium ion (Mg^{2+}) effectively increases the storage capacity of capacitors. It achieves this by neutralizing electrical charges at the capacitor electrodes, which would otherwise contribute to the external electric field (Arthur et al., 1961).

2.6. Difficulties with Zinc Ferrite

Challenges associated with synthesizing zinc ferrite nanoparticles primarily stem from difficulty in controlling the synthesis conditions to ensure phase purity and precise stoichiometry. However, researchers have successfully addressed these issues by developing specific methods to manipulate factors like temperature, the reaction environment, and the synthesis technique itself. Among these, the sol-gel process is a particularly favored method, because it is known to produce highly homogeneous, phase pure zinc ferrite nanoparticles at relatively lower temperatures compared to conventional approaches (Gupta et al., 2017).

2.7. Doping of Zinc Ferrite

The structural and dielectric properties of ferrites are critically dependent on the way cations are distributed within the tetrahedral and octahedral lattice sites of the spinel structure. These properties can be precisely tuned by the strategic selection of the divalent cation (A^{2+}) used in the ferrite compound. A range of ions, including Mg^{2+} , Fe^{2+} , Ni^{2+} , Cu^{2+} , and Zn^{2+} , can be employed to achieve the desired material characteristics (Zhang, 2021). In their 2024 study, Abbas and colleagues successfully created a ZnFe_2O_4 nanocomposite modified with cobalt and/or nickel. Their investigation centered on the material's performance in the photocatalytic degradation of azo dyes, aiming to provide a solution for remediating these organic pollutants.

2.8. Surveying Previous Studies

The field of nanoparticle materials is highly active due to their distinct properties compared to bulk materials, with Mg-Zn based nanoparticles being particularly prominent for their potential in electromagnetic devices, microwave technology, magnetic recording, and biomedicine. The relevant studies are summarized below in sequential order.

Khan (2024) studied the categorization of spinel ferrites by examining how their inherent crystal structure dictates the distribution of cations within the lattice sites. This work provides a framework for understanding and classifying ferrites based on whether they exhibit a normal, inverse, or mixed spinel arrangement, which is crucial for predicting their magnetic and electrical properties.

Smith and Jones (2024) assert that ferrites "are ceramic compositions made of iron (III) oxide (Fe_2O_3) combined with other metallic elements" and are categorized into four groups based on their crystal structure: hexagonal, garnet, ortho, and spinel.

N. Abbas et al. (2024) investigated the creation and application of a novel nanocomposite material designed for environmental remediation. Their study focused on synthesizing a specific core-shell structure: a Zinc Ferrite (ZnFe_2O_4) core encapsulated by a Cobalt/Nickel Metal-Organic Framework (Co/Ni-MOF) shell. This research highlights the development of advanced hybrid materials for use in treating environmental contamination.

Y. Zeng et al. (2024) demonstrated that combining zinc ferrite nanoparticles with gallic acid effectively mitigated salinity stress in wheat.

Jensen (2024) explains that the metal cations are distributed among two types of interstitial sites within the oxygen lattice.

G. Ramírez et al. (2023) explained the distinctions and overlaps between impedance and dielectric spectroscopy when applied to the study of functional materials. Their critical evaluation covered the fundamental principles, experimental procedures, and data analysis methods for both techniques, ultimately clarifying how each method contributes uniquely to the comprehensive electrical characterization of materials and how they should be utilized effectively in research.

Smith (2023) studied the fundamental characteristics of spinel ferrites, establishing them as an important class of metal oxides defined by the inclusion of distinct metal cations and further detailed that these compounds possess a specific crystal arrangement known as a face-centered cubic (FCC) structure and are classified under the Fd-3m space group.

Y. Zhang et al. (2022) successfully synthesized pure Zinc Ferrite (ZnFe_2O_4) using a microwave-assisted ball milling technique for 30 hours. The primary result demonstrated the superiority of this method, as the resulting nanoparticles exhibited a high saturation magnetization (Ms) value of 82.23 emu/g, which was significantly greater than the 30 emu/g typically achieved by standard high-energy ball milling.

F. Guo et al. (2021) explained the fundamental technique of cationic substitution (or doping) as a primary method researchers use to tailor the properties of ferrites for specific applications.

Shirsath et al. (2018) highlighted the importance of a synthesis technique that permits precise command over the morphology and dimensions of nanoparticles by adjusting simple processing variables, such as pH and precursor concentration. Crucially, this method facilitates crystallization at much lower temperatures than conventional approaches, which is essential for preserving the nanoscale integrity and inhibiting undesirable particle aggregation.

S. K. Pendyala et al. (2018) detailed the sol-gel auto-combustion synthesis of Magnesium-doped Zinc Ferrite ($\text{Mg}_x\text{Zn}_{1-x}\text{Fe}_2\text{O}_4$) nanoparticles. Their study focused on explaining the fundamental effect of Mg doping on the material's physical properties, specifically examining how the substitution impacts the crystal structure and the resulting lattice parameter.

S. Thiagarajan et al. (2017) studied that the sol-gel synthesis is a facile methodology for producing metal oxide nanostructures, noting its distinct advantages over other techniques, such as yielding high-purity, small, and homogeneous particles along with providing good stoichiometric control, simple preparation steps, and low operational cost.

O. Mounkachi et al. (2017), and others like it, explicitly attributes the observed decrease in the lattice parameter (a) of the $\text{Zn}_{1-x}\text{Mg}_x\text{Fe}_2\text{O}_4$ system to the difference in ionic radii; the Mg^{2+} ion (0.72 Å) is indeed slightly smaller than the Zn^{2+} ion (0.74 Å) in the tetrahedral site, causing the overall unit cell to contract.

P. R. Choodamani et al. (2014) synthesized Gd-substituted Mg-Zn ferrite nanoparticles using a chemical method to specifically study their ability to generate heat under an external magnetic field. The key finding was the successful demonstration of their magnetic induction heating properties, highlighting their potential for use in advanced biomedical fields like cancer treatment (hyperthermia) and precise drug delivery.

S. Hajarpour et al. (2013) explained that zinc ferrites are materials of considerable technological importance because of their variety of structural and dielectric characteristics. This suitability makes them valuable for a wide range of uses, such as in magnetic storage, ferrofluids, gas sensors, wastewater treatment, and biomedical applications.

M. S. Huq et al. (2013) clarified the structural configuration of Zinc Ferrite (ZnFe_2O_4), confirming it adheres to the normal spinel structure. In this specific configuration, all divalent zinc cations (Zn^{2+}) are located on the tetrahedral sites (A-sites), while all the trivalent ferric ions (Fe^{3+}) are confined to the octahedral sites (B-sites).

M. Abbasher et al. (2010) prepared spinel Magnesium-Zinc ferrites ($\text{Zn}_x\text{Mg}_{1-x}\text{Fe}_2\text{O}_4$) using the standard double sintering ceramic method. Their investigation utilized X-ray diffraction (XRD) and Mössbauer spectroscopy measurements taken at both 295 and 78 K to analyze the material's structure, specifically focusing on the cation distribution and magnetic ordering as a function of temperature and composition.

Fultz and Howe (2007) explained the core concepts of both Transmission Electron Microscopy (TEM) and X-Ray Diffractometry (XRD), emphasizing their combined importance in the characterization of materials. Their textbook provides a detailed, unified theoretical and practical treatment of topics common to both techniques, such as wave-matter interactions, scattering from atoms, and the analysis of diffraction patterns, while also covering advanced methods unique to TEM, including high-resolution imaging and spectroscopy.

B. P. Rane et al. (2007) reported on the synthesis of Zinc Ferrite (ZnFe_2O_4) nanoparticles using a novel sol-gel combustion method. A key component of their study was providing a comparative analysis of the common synthesis techniques for ZnFe_2O_4 , explicitly evaluating how different methods stack up against each other in terms of achieving fine particle size, ensuring high purity, and considering the overall production cost.

A. A. Hasanain et al. (2006) investigated the structural and magnetic properties of copper-substituted zinc ferrites ($\text{Zn}_{1-x}\text{Cu}_x\text{Fe}_2\text{O}_4$) by preparing samples across a range of copper concentrations. Through analysis using X-ray diffraction and magnetic measurements, the authors conclusively found that substituting copper ions for zinc ions dramatically altered the material's fundamental properties, leading to significant changes in its crystal structure and overall magnetic performance.

S. A. Oliver et al. (2000) studied the structural and compositional changes in zinc ferrite (ZnFe_2O_4) powders that were subjected to mechanical activation.

Goldman (1990) highlighted that a defining and highly advantageous property of ferrites is their high electrical resistivity. This characteristic is essential for suppressing eddy current losses, which in turn makes them exceptionally effective materials for high-frequency

J. Smit and H. P. J. Wijn (1959) explained that ferrites represent a class of magnetic metallic oxides and provided a comprehensive survey of their physical properties within ceramic materials, detailing the distinct structural types like spinels and garnets, and outlining the general chemical formula (MFe_2O_4) that defines these materials.

CHAPTER THREE

MATERIALS AND METHODS

3.1. Introduction

The practical section of this research, detailed in this chapter, describes the synthesis of magnesium-doped zinc ferrite with the chemical formula ($\text{Zn}_{1-x}\text{Mg}_x\text{Fe}_2\text{O}_4$) at specific concentrations of $x = 0.00, 0.03, \text{ and } 0.06$. The sol-gel auto-combustion method was the chosen technique, which is advantageous as it allows for the preparation of highly homogeneous products rapidly while successfully circumventing the use of the typically high processing temperatures associated with conventional methods. Following the chemical synthesis, the resulting nanoparticles were systematically subjected to thermal treatment across a range of temperatures, specifically between 220°C and 850°C , to precisely control their final structural and morphological characteristics.

3.2. Chemicals used

For the fabrication of the magnesium-substituted zinc ferrite nanoparticles ($\text{Zn}_{1-x}\text{Mg}_x\text{Fe}_2\text{O}_4$), a set of essential precursors and reagents was utilized. The main starting materials included zinc nitrate hexahydrate ($\text{Zn}(\text{NO}_3)_2 \cdot 6\text{H}_2\text{O}$) and ferric nitrate nonahydrate ($\text{Fe}(\text{NO}_3)_3 \cdot 9\text{H}_2\text{O}$) as the metal ion sources, alongside magnesium nitrate hexahydrate ($\text{Mg}(\text{NO}_3)_2 \cdot 6\text{H}_2\text{O}$) for the doping element. Citric acid ($\text{C}_6\text{H}_8\text{O}_7$) served a critical function as the chelating agent, helping to stabilize the metal ions and ensure homogeneity. The entire reaction was conducted in a solution phase using deionized water as the primary solvent. Finally, ammonia solution (NH_4OH) was meticulously added as the neutralizing agent to adjust the acidity of the mixture precisely to a pH of 7.

3.3. Apparatus used

- ❖ Electric balance: Weighing of precursors.
- ❖ Beakers (250ml, 150ml): Containers for dissolving and mixing solutions.
- ❖ Spoons/Spatulas: Transferring solid chemicals.
- ❖ Magnetic stirrer: Providing rotational force for mixing.
- ❖ Magnetic bead: Stirring element inside the solution.
- ❖ Litmus paper: Measuring and adjusting pH.
- ❖ Evaporating dish: Concentrating solution/forming gel by evaporating solvent.
- ❖ Hot oven: Drying the precursor material.
- ❖ Alumina crucible: High-temperature container for calcination.
- ❖ Agate mortar with Pestle: Grinding and fine-milling the synthesized powder.
- ❖ Hydraulic press: Compressing powder to make a pellet.
- ❖ Alumina plate: Support for compression or high-temperature handling.
- ❖ Cotton: Cleaning or wiping purposes.

3.4. Synthesis Methods

The synthesis of nanoparticles is broadly classified into two main categories: the top-down and the bottom-up approaches, based on the size of the starting material and the methodology used. The top-down approach is a subtractive method that involves starting with a large, bulk material and physically or mechanically breaking it down into nanosized particles while the bottom-up approach is an additive method that involves assembling individual atoms or molecules into larger clusters, which then grow into the desired nanoparticles.

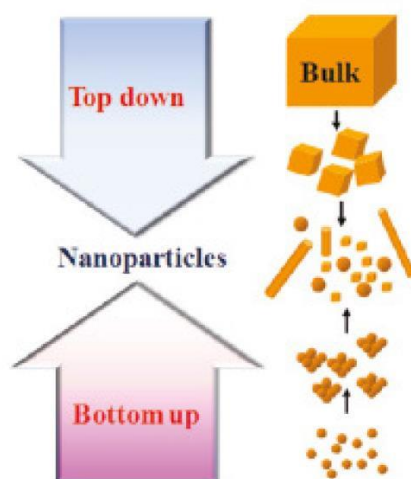


Figure 3.1: Top-down versus bottom-up approach

3.5. Sol–gel method

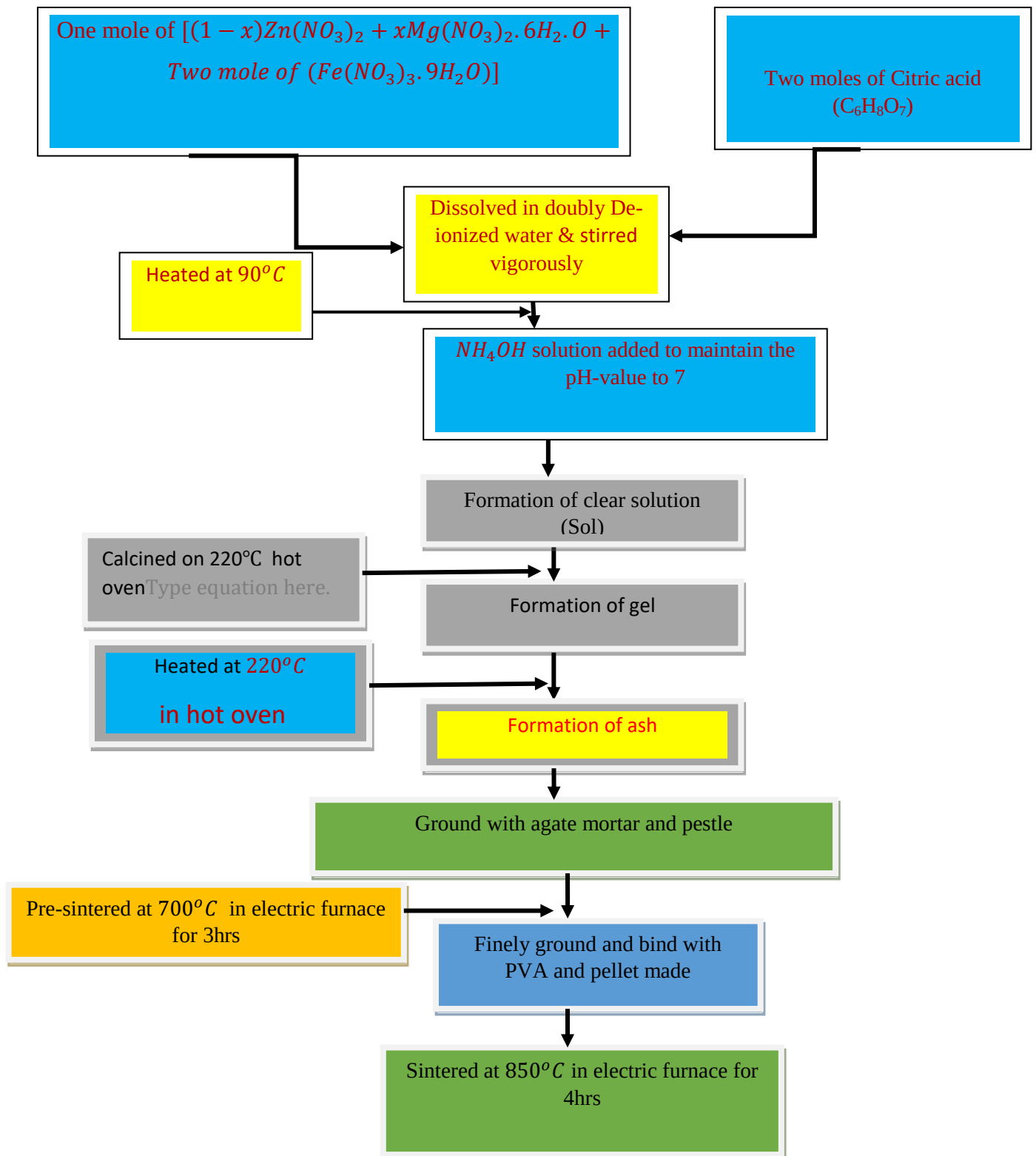
The sol-gel method was chosen to synthesize magnesium doped zinc ferrite nanoparticles because it is a bottom-up approach that assures extraordinary chemical homogeneity through atomic-level mixing in the liquid phase, leading to a highly uniform final product (Rane et al., 2007). This technique allows for precise control of nanoparticle size and morphology by adjusting synthesis parameters such as pH and concentration, while also allowing crystallization at significantly lower temperatures than traditional methods, preserving the desired nanoscale structure and preventing excessive particle (Shirsath et al., 2018).

3.6. Experimental Procedure

To synthesize magnesium doped zinc ferrite nanoparticles, the following experimental procedures were employed:

- ❖ Stoichiometric amounts of zinc nitrate hexahydrate $[Zn(NO_3)_2 \cdot 6H_2O]$, ferric nitrate nonahydrate $[Fe(NO_3)_3 \cdot 9H_2O]$, and magnesium nitrate hexahydrate $[Mg(NO_3)_2 \cdot 6H_2O]$ are accurately weighed according to the desired x value in the formula $Zn_{1-x}Mg_xFe_2O_4$.
- ❖ The metal nitrates are dissolved in a beaker together in a minimum volume of double deionized water.
- ❖ In the solution, equal mole of anhydrous citric acid with that of the total mole of metal nitrates was added acting as a chelating agent to bind the metal ions, preventing premature precipitation.
- ❖ As the solution is acidic because of addition of acidic solution, ammonia solution was added drop wise to maintain the solution neutral, pH-7
- ❖ The final solution on hot plate was vigorously mixed using a magnetic stirrer to ensure perfectly homogeneous sol, and the solution gradually thickens, becoming highly viscous, and eventually transforms into a sticky, brown or dark red gel.
- ❖ The gel was held in evaporating dish and loaded in hot oven at 200°C to remove moisture, and finally fluffy ash was formed.
- ❖ The fluffy ash was ground finely several times by agate mortar, and the ground powder was held in alumina crucible, then transferred to a temperature programmed furnace at 650°C to remove impurities and organic substances for 2 hrs.
- ❖ After the temperature of calcined powder was dropped to room temperature, it was again finely ground several times.
- ❖ Part of the powder was mixed with Polyvinyl Alcohol (PVA) as a binder and pressed into pellet using hydraulic press.
- ❖ Samples in powder and pelleted form were loaded into a temperature programmed furnace at 700°C for 3hrs for more densification.
- ❖ Finally, the samples were ready to transform for characterization.

3.7. Chemical synthesis route of sol-gel method



3.8. Characterization and measuring Techniques

To understand the properties of the synthesized magnesium-doped zinc ferrite nanoparticles, the sample was examined using several characterization techniques. Specifically, X-ray Diffraction (XRD), Scanning Electron Microscopy (SEM) were employed to determine the structural properties, while an impedance analyzer was utilized to investigate the dielectric properties of the material.

3.8.1. X-Ray Diffract meter (XRD)

The core function of X-Ray Diffraction (XRD) technology is to determine a material's phase composition and reveal its underlying crystallographic structure. In addition to these primary uses, XRD analysis is a powerful tool for obtaining precise, quantifiable structural information, such as the exact lattice parameters (the size and angles of the unit cell) and the average size of the crystallites within a sample (Fultz and Howe, 2007).

The crystal plane spacing corresponding to the Bragg's angle of reflection can be estimated using equ. (3.1).

$$2d\sin\theta = n\lambda \quad 3.1$$

In this formula, n stands for the order of planes, d represents the distance separating the crystal planes, λ is the X-ray wavelength, and θ is the glancing angle between the incident beam and crystal plane.

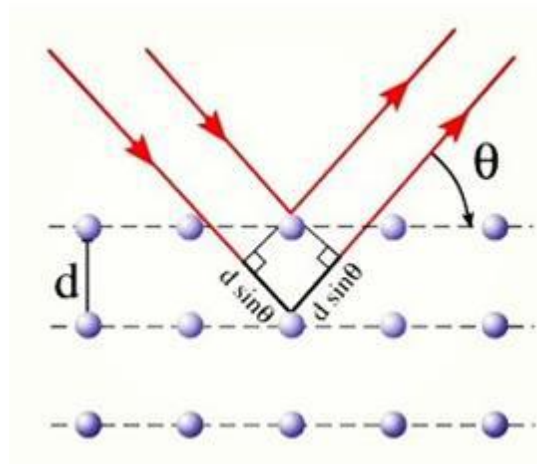


Figure 3.2: Bragg's diffraction for X-ray

The average crystallite size (D) for all samples was computed using the Scherrer formula, equ. (3.2):

$$D = \frac{k\lambda}{\beta \cos \theta} \quad 3.2$$

In this equation, D is the crystallite size, k is the Scherrer constant (k=0.89), λ is the X-ray wavelength, β represents the Full Width at Half Maximum (FWHM) of the sharp peak in radians, and θ is the diffraction angle.

The crystal lattice constant (a), d-spacing (d_{hkl}), miller indices (h,k,l) and the volume of a unit cell (V) for cubic crystal can be related as:

$$a = d_{hkl} \sqrt{h^2 + k^2 + l^2} \quad 3.3$$

$$V = a^3 \quad 3.4$$

Here, d is the interplanar distance between successive planes (defined by Miller indices h, k, l), and V is the unit cell volume.

The actual X-ray density (ρ_x) is calculated using the formula:

$$\rho_{x-ray} = \frac{ZM}{N_a V} \quad 3.5$$

where Z represents the number of formula per unit cell, M is the molecular weight (g/mol), N_a is Avogadro's number (6.023×10^{23} atoms/mol), and V is the volume of a unit cell in nm^3 .

3.8.2. Scanning Electron Microscopy

Scanning Electron Microscopy (SEM) is a powerful type of electron microscope used primarily to study the surface topography and compositions of solid samples at high magnifications up to (300,000) and high resolution (sub-nanometer scale). It surpasses the capabilities of traditional optical microscopes by using a focused beam of electrons instead of light, allowing for much greater detail and a large depth of field, which results in a characteristic three dimensional appearance of the sample surface.

3.8.3. Impedance Analyzer

An Impedance Analyzer is an electronic instrument used to measure the complex impedance of electrical components, materials, or circuits over a wide range of frequencies. This measurement provides crucial information about a material's dielectric, electrical, and magnetic properties by characterizing its response to an applied alternating current (AC) signal (Ramírez-Gonzalez et al., 2023).

$$Z^* = Z' - jZ''(\omega) = \frac{1}{j\omega C^*} \quad 3.6$$

$$\varepsilon^* = \varepsilon' - j\varepsilon'' = \frac{1}{j\omega C_o Z^*} \quad 3.7$$

The parameters Z' and Z'' represented as the real and imaginary parts of impedance, where as, ε' and ε'' are real and imaginary dielectric constants respectively.

The tangent loss is the ratio of imaginary part to the real part of dielectric constants as explained in equ. (3.8):

$$\text{Tan loss} = \text{Tan } \delta = \frac{\varepsilon''}{\varepsilon'} \quad 3.8$$

Where $\text{Tan } \delta$ represents the tangent loss and ε' and ε'' represents real and imaginary dielectric constants.

CHAPTER FOUR

RESULTS AND DISCUSSIONS

4.1. Introduction

The structural and dielectric properties of the $\text{Zn}_{1-x}\text{Mg}_x\text{Fe}_2\text{O}_4$ nanoparticles, fabricated through the sol-gel technique, are fully detailed and discussed in this chapter. Structural investigation via

X-ray diffraction (XRD) revealed that all samples ($x=0.00$, 0.03 , and 0.06) possess a desirable single-phase cubic spinel crystal structure. Further characterization involved a Scanning Electron Microscope (SEM) to analyze the surface morphology, and an impedance analyzer was employed for the systematic measurement of the material's dielectric response.

4.2. Structural Analysis

4.2.1. XRD Analysis

Figure 4.1 shows the XRD pattern of $\text{Zn}_{1-x}\text{Mg}_x\text{Fe}_2\text{O}_4$ ($x = 0.00$, 0.03 and 0.06) nanoparticles prepared by sol-gel method. The characterization confirmed a well indexed in cubic crystal structure with space group Fd-3m . Most prominent peaks are observed around the Bragg's angles $2\theta = 19^\circ$, 29° , 35° , 35.25° , 43° , 56° , 62° and 68° corresponding to plane of reflection (111), (220), (311), (222), (400), (422), (511), and (440), respectively.

The structural characterization involved a two-step analysis of the X-ray Diffraction (XRD) data. First, the entire diffraction pattern was subjected to Rietveld refinement using the FullProf program, which models the crystal structure to achieve highly accurate primary parameters. This sophisticated fitting process, validated by the prominent (311) characteristic peak of the spinel structure, precisely estimates the fundamental crystal parameters, including the lattice constants and the crucial distribution of metal ions within the unit cell.

Second, the highly accurate lattice parameters derived from the Rietveld refinement were used as the basis for calculating several important derived structural parameters via specific crystallographic equations (3.1-3.5). These calculations yielded the crystallite size (D), d-spacing, volume of the unit cell, and X-ray density ($\rho_{x\text{-ray}}$). This comprehensive approach combines the rigor of whole-pattern fitting with precise analytical calculations, providing a complete structural profile of the magnesium-doped zinc ferrite nanoparticles, with all results conveniently summarized in Table 4.1.

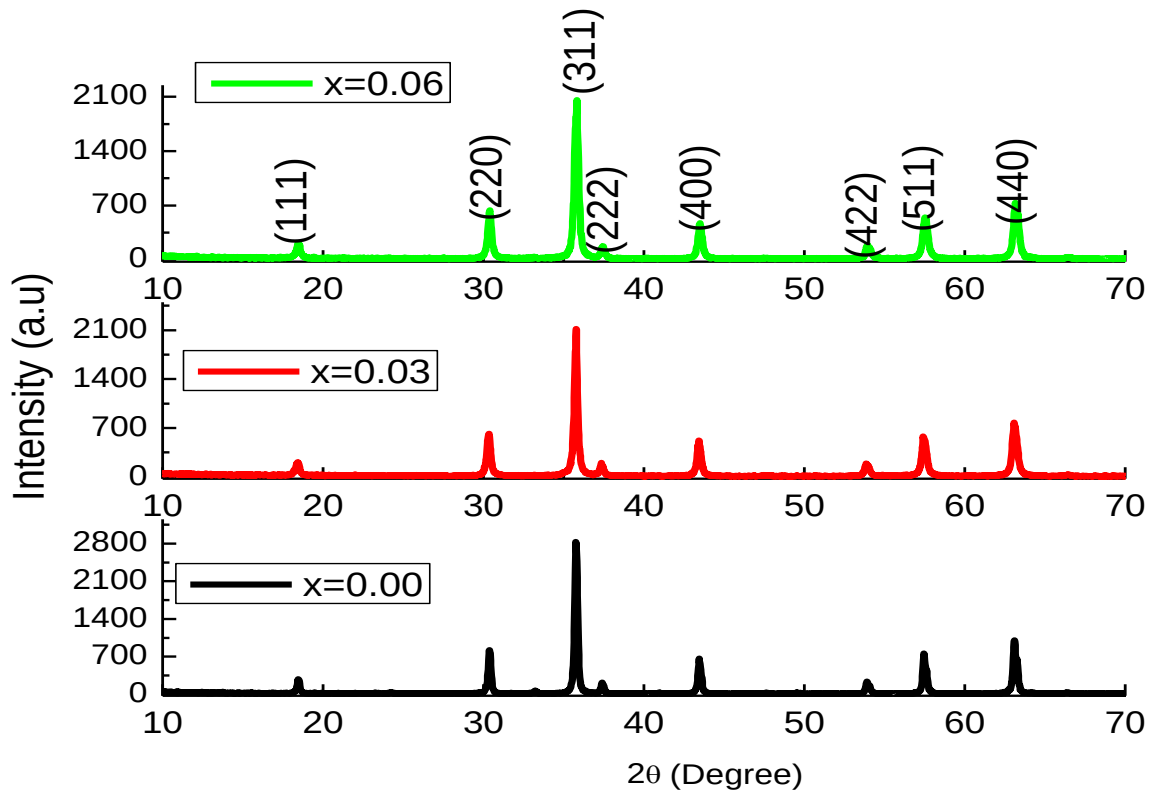


Figure 4.1: XRD spectra of $\text{Zn}_{1-x}\text{Mg}_x\text{Fe}_2\text{O}_4$ samples for $x=0.00$, 0.03 , and 0.06

Table 4.1: Structural parameters (D , d_{hkl} , a , V and X-ray density) for Mg-doped ZnFe_2O_4

Sample	Crystallite size $D_{311}(\text{nm})$	d-spacing (nm)	Lattice constants $a=b=c$ (nm)	Volume of unit cell $(\text{nm})^3$	x-ray density (g/cm^3)
ZnFe_2O_4	22.24	0.252	0.833	0.577	5.55
$\text{Zn}_{0.97}\text{Mg}_{0.03}\text{Fe}_2\text{O}_4$	15.67	0.251	0.832	0.576	5.52
$\text{Zn}_{0.94}\text{Mg}_{0.06}\text{Fe}_2\text{O}_4$	12.43	0.250	0.831	0.574	5.51

Table 4.1 Reveals that the decreasing in lattice constant attributed to the substitution of the larger Zn^{2+} ion (ionic radius ≈ 0.06 nm in tetrahedral coordination) with the slightly smaller Mg^{2+} ion (ionic radius ≈ 0.057 nm in tetrahedral coordination) at the A-site. Replacing the larger ion with a smaller one creates a structural strain that causes the entire unit cell to contract, thus reducing the lattice constant (a).

This is a direct consequence of Vegard's law applied to the doped ferrite system. From mathematical relation between lattice constant and d-spacing, the decreased value with Mg ion concentration confirmed. For the case of reduction of crystallite size, is primarily a kinetic effect of the synthesis. The addition of the Mg^{2+} dopant introduces a structural impurity during the material's crystallization process. This finding aligns with similar results reported in previous research, such as the work conducted by Pendyala and colleagues in 2018.

The table also shows that the X-ray density decreases with increasing concentration in Mg because the reduction in the formula weight (M) outweighs the contraction in the unit cell volume (V). The substitution of the heavier Zn^{2+} ion (65.38 g/mol) by the significantly lighter Mg^{2+} ion (24.31 g/mol) causes the overall molecular weight (M) of the compound to fall, while the smaller ionic radius of Mg^{2+} causes a small, opposing decrease in V; since density is proportional to the M/V ratio, the dominant reduction in mass leads to the observed decrease in the theoretical X-ray density.

4.2.2. Morphological studies

The SEM micrograph (Figure 4.2) of the Mg^{2+} ion-doped zinc ferrite nanoparticles ($\text{Zn}_{1-x}\text{Mg}_x\text{Fe}_2\text{O}_4$) synthesized via the sol-gel method at room temperature typically reveals a morphology characterized by homogeneous, spherical nanoparticles arranged in a compact or agglomerated fashion. This uniform, spherical shape is a direct result of the sol-gel process's ability to ensure atomic-level mixing of precursors, leading to uniform nucleation and isotropic growth. The subsequent compact arrangement is primarily due to the high surface energy of the fine nanoparticles, causing them to weakly agglomerate during the necessary high-temperature calcination step used to achieve the final crystalline ferrite phase.

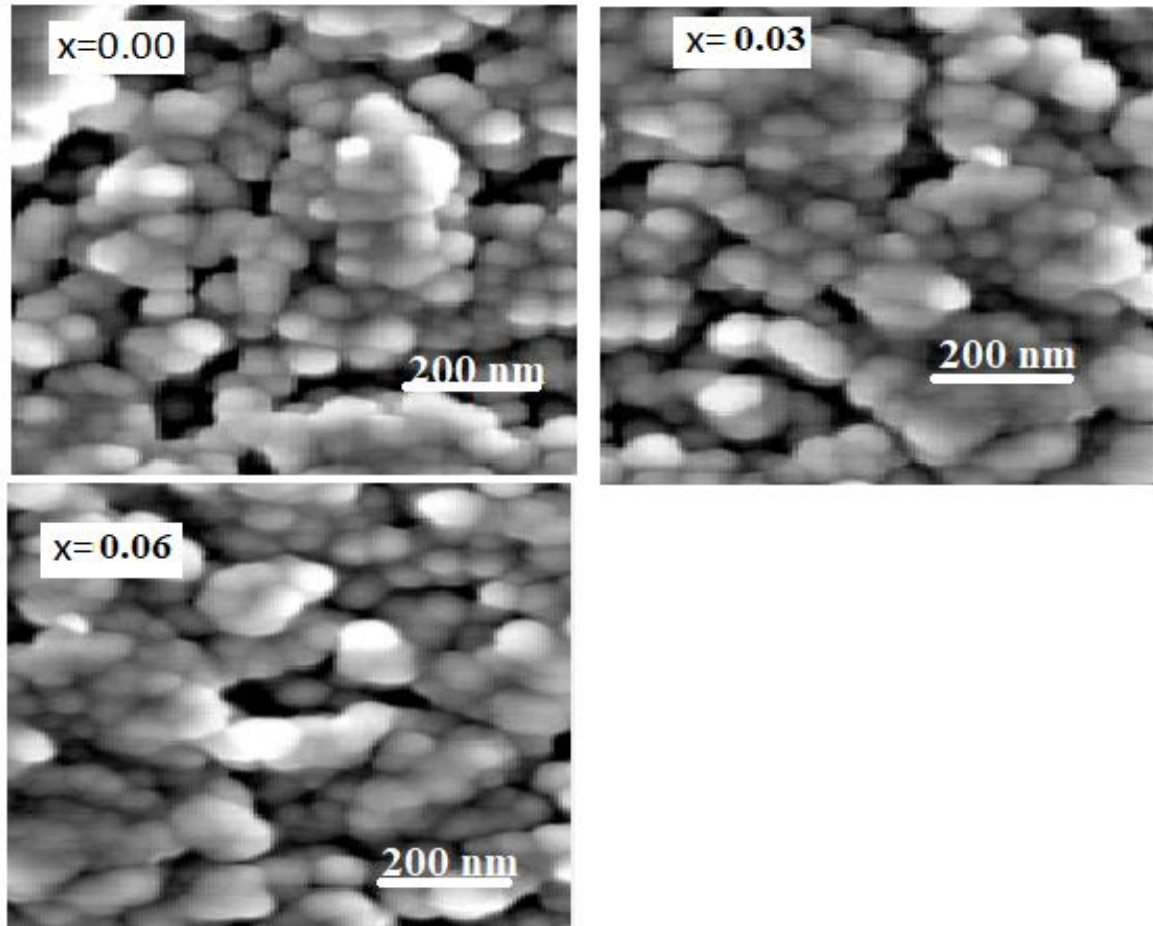


Figure 4.2: SEM micrograph of $\text{Zn}_{1-x}\text{Mg}_x\text{Fe}_2\text{O}_4$ samples for $x=0.00$, 0.03 and 0.06

4.3. Dielectric studies

Impedance spectroscopy is a powerful method for characterizing the dielectric properties of a material. It works by applying an alternating current (AC) voltage over a broad range of frequencies to analyze the material's response. An impedance analyzer is the key instrument used to perform this analysis.

In this study, an impedance analyzer was used to investigate how both frequency and magnesium (Mg) doping affect the dielectric properties of the samples. The results showed that the dielectric constant (ϵ'), dielectric loss ($\tan\delta$), and AC conductivity are highly sensitive to both the Mg doping concentration and the applied frequency. The dielectric properties of the ferrites were measured within a frequency range of 200 kHz to 1 MHz, and their specific behavior varied depending on the concentration of magnesium.

4.3.1. Dielectric Constant (ϵ')

The dielectric constant which is a material's ability to store electrical energy in an electric field exhibits a dispersive behavior, being high at lower frequencies and decreasing significantly as the frequency increases, because at low frequencies, charge carriers and dipoles have enough time to align with the applied electric field.

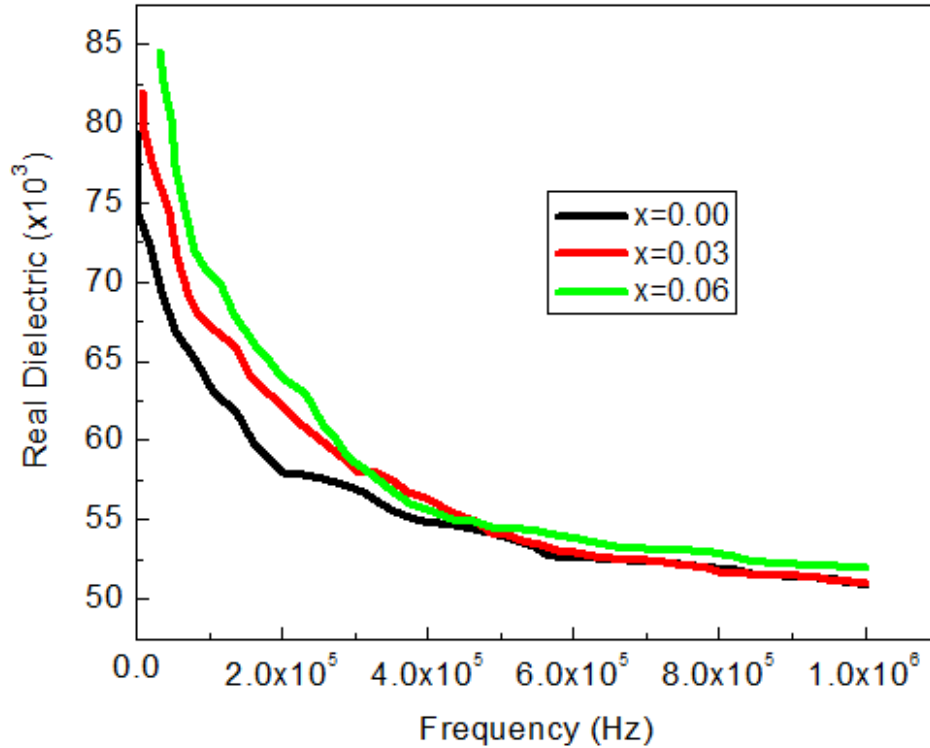


Figure 4.3: Real dielectric versus frequency for $\text{Zn}_{1-x}\text{Mg}_x\text{Fe}_2\text{O}_4$ ($x=0.00, 0.03$ and 0.06)

Figure 4.3 depicts the frequency dependence of the real dielectric constant for $\text{Zn}_{1-x}\text{Mg}_x\text{Fe}_2\text{O}_4$, ($x=0.0, 0.03$ and 0.06) sample measured at room temperature (300K) in the frequency range (200kHz to 1MHz). As the frequency increases, the charge carriers can not keep pace with the rapidly changing electric field. This leads to a decrease in the polarization and, consequently, a sharp drop in the dielectric constant. Also, Mg doping affects the dielectric constant for all samples.

The real dielectric constant (ϵ') increases with Mg doping due to a mechanism known as Maxwell-Wagner interfacial polarization. This substitution significantly affect the cation

distribution in the tetrahedral (A) and octahedral (B) sites of the spinel structure as ionic radius of Mg^{2+} (0.72 Å) is smaller than that of Zn^{2+} (0.74 Å).

This difference lead to a change in lattice parameters and a rearrangement of the cations, which in turn influences the hopping mechanism of the electrons and thus the dielectric properties as shown in Fig.4.3. The obtained result confirmed that, with $x=0.00$ to have the lowest ϵ' , followed by $x=0.03$, and then $x=0.06$ having the highest.

The introduction of Mg^{2+} ions into the zinc ferrite lattice influences the electrical properties of both the grains and the grain boundaries. Specifically, Mg substitution can modify the overall resistivity of the grain boundaries. With increasing Mg concentration, the resistivity of the grain boundaries decrease, allowing for a more effective buildup of charge, which enhances the overall Maxwell-Wagner polarization and, consequently, increases the dielectric constant.

4.3.2. Dielectric loss tangent ($\tan\delta$)

Dielectric loss tangent is the parameter that quantifies the energy lost as heat within the material under an AC field which is a crucial indicator of the material's insulating quality. Similar to the dielectric constant, the dielectric loss tangent also tends to be high at low frequencies and decreases with increasing frequency. Furthermore, when magnesium (Mg^{2+}) is substituted for zinc (Zn^{2+}) in zinc ferrite, it changes the distribution of cations at the tetrahedral (A) and octahedral (B) lattice sites. Since Mg^{2+} ions are known to occupy octahedral sites, their presence disrupt the continuous path for electron hopping between iron ions on those same sites.

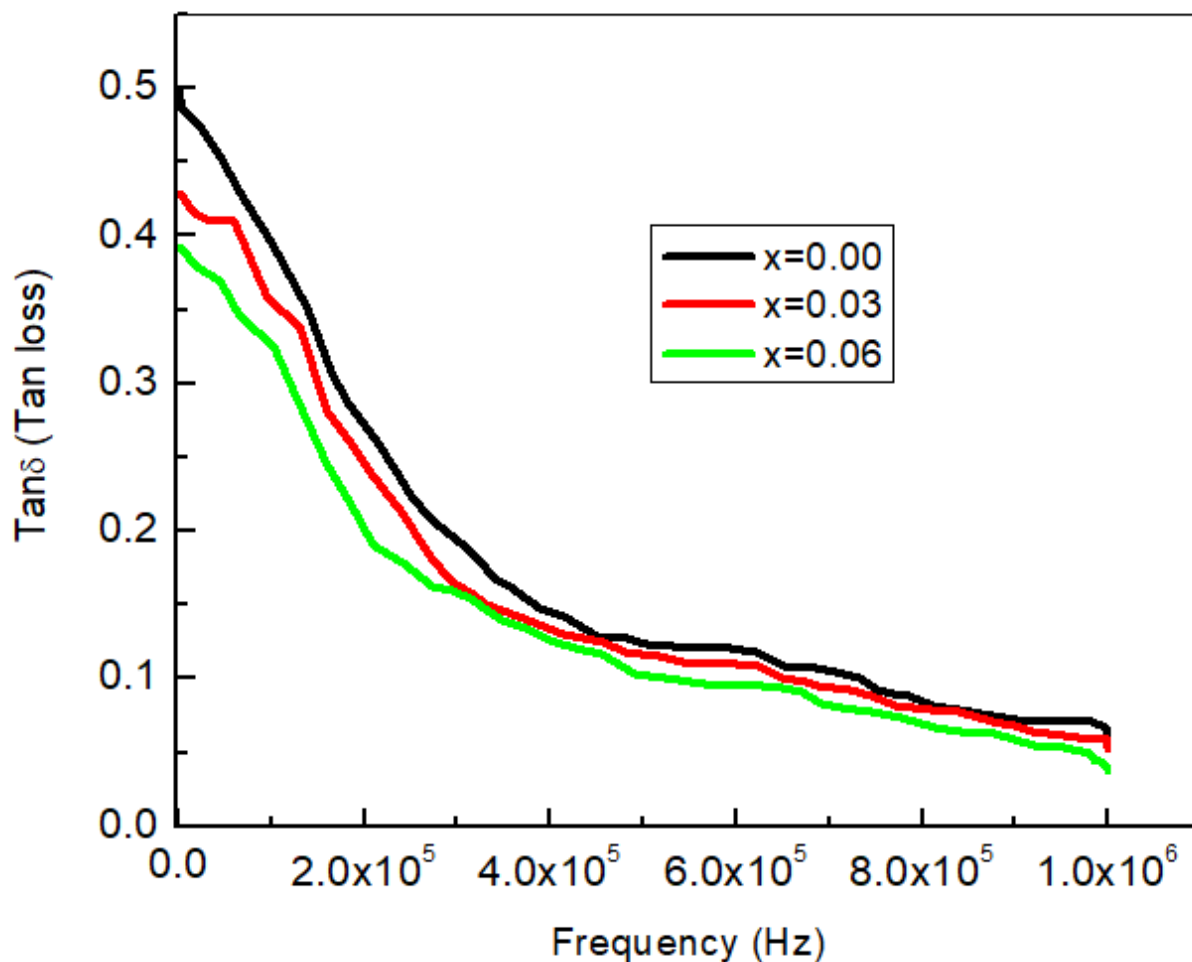


Figure 4.4: Tan loss versus frequency for $\text{Zn}_{1-x}\text{Mg}_x\text{Fe}_2\text{O}_4$ ($x=0.00, 0.03$ and 0.06)

In fig.4.4, it was also observed that the dielectric loss is frequency dependent and optimized Mg concentrations can lead to lower dielectric loss at high frequencies, making the materials suitable for high-frequency applications where minimal energy dissipation is desired. The reduction of dielectric loss with the increased percentage amount of magnesium substituted in zinc ferrite while preserving the improvement of dielectric constant and stabilizing the structure of zinc ferrite nanoparticles.

CHAPTER FIVE

CONCLUSION AND RECOMMENDATIONS

5.1. Conclusion

This thesis successfully investigated the synthesis and characterization of $\text{Zn}_{1-x}\text{Mg}_x\text{Fe}_2\text{O}_4$ nanoparticles ($x = 0.00, 0.03$ and 0.06), definitively establishing the profound impact of magnesium (Mg) substitution on their structural and dielectric properties. The materials were reliably synthesized using the sol-gel auto-combustion method and thoroughly analyzed using X-ray Diffraction (XRD) and Scanning Electron Microscopy (SEM). Structural analysis confirmed the formation of a pure, single-phase cubic spinel structure across all doping concentrations. Crucially, the substitution of the larger Zn^{2+} ion ($0.74 \text{ \AA}$) with the smaller Mg^{2+} ion ($0.72 \text{ \AA}$) induced a systematic lattice contraction. This was evidenced by a linear decrease in the lattice constant (a), unit cell volume (V), and a notable reduction in crystallite size from 22.24 nm to 12.43 nm at $x=0.06$. Concurrently, a decrease in X-ray density correlated precisely with the lower molecular weight of the Mg substituent, while SEM confirmed a desired uniform, spherical morphology with minimal inter-particle agglomeration.

The investigation of dielectric properties via Impedance Spectroscopy revealed a significant and favorable enhancement in the electrical response directly proportional to the Mg content. All compositions exhibited classic frequency-dispersive behavior-high permittivity (ϵ') and loss tangent ($\tan \delta$) at low frequencies primarily attributed to space charge polarization. Critically, the Mg doping strategy successfully resulted in an increase in ϵ' coupled with a decrease in $\tan \delta$ in the technologically relevant mid-to-high frequency range. This optimized performance stems from a dual electronic mechanism: first, Mg^{2+} introduction modifies the cation distribution (particularly the $\text{Fe}^{3+}/\text{Fe}^{2+}$ ratio) across the tetrahedral and octahedral sites, thereby enhancing the Maxwell-Wagner interfacial polarization at grain boundaries. Second, the Mg^{2+} ions strategically occupying the octahedral sites efficiently disrupt the electron hopping path between the iron ions, which is key to mitigating energy dissipation and thus achieving lower dielectric loss.

In conclusion, this research unequivocally demonstrates that Mg doping is an effective, precise strategy for tuning the electromagnetic functionality of zinc ferrite nanoparticles. The composition $\text{Zn}_{0.94}\text{Mg}_{0.06}\text{Fe}_2\text{O}_4$ represents the optimal material, achieving an exceptional balance of structural stability, significantly enhanced permittivity, and drastically reduced dissipation. These engineered properties confirm the material's high suitability for integration into advanced high-frequency devices.

5.2. Recommendations for Future Work

Based on the findings of this thesis, the following recommendations are suggested for further exploration of this material system:

- Exploration of alternative synthesis routes: Evaluate the influence of material preparation techniques such as the solid-state reaction, co-precipitation, or hydrothermal methods on the resulting structural, morphological, and functional properties, and compare these outcomes with the sol-gel technique.
- Extended doping concentration study: Conduct further research at higher Mg doping concentrations ($x > 0.06$) to fully delineate the saturation point of the spinel lattice and identify the precise concentration that yields the ultimate optimization of the dielectric response.
- Temperature dependence of dielectric behavior: Investigate the dielectric properties as a function of temperature across a wide thermal range. This comprehensive understanding is crucial for assessing the material's thermal stability and suitability for practical operation in diverse environments.
- Optical characterization: Analyze the optical properties of the doped samples using UV-Visible spectroscopy. This would facilitate the calculation of the direct and indirect band gap energies, providing insights into the electronic structure and potential for optoelectronic applications.
- Magnetic and electrical property interrogation: Perform a detailed investigation of the material's magnetic and static electrical properties. The use of a Vibrating Sample Magnetometer (VSM) is recommended to fully characterize parameters like saturation magnetization and coercivity, which is essential for evaluating the material's full potential in magneto-electronic devices.
- Substitution with other divalent and transitional metals: Expand the scope of the study by exploring the impact of substituting Zn^{2+} with other divalent or transitional metal ions.

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