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Effect of La^{3+} Substitution on the Structure and Functional Properties of Mg-Co Ferrites for High-Frequency Technology

Aqsa Saleem
Faculty of Sciences, The Superior University, Lahore, Pakistan.

Hafiz Safdar Ali
Faculty of Sciences, The Superior University, Lahore, Pakistan.

Muhammad Arslan
Faculty of Sciences, The Superior University, Lahore, Pakistan.

Kanwal Akhtar*
Department of Physics, Government College Women University, Faisalabad, Pakistan

Ayesha Younus
Department of Physics, Government College Women University, Faisalabad, Pakistan

Yasir Javed
Department of Physics, University of Agriculture, Faisalabad, Pakistan

ABSTRACT

The rich magnetic/electrical properties of spinel ferrites lead to multiple applications in modern electronic and high-frequency domains, making them one of the most commonly used magnetic ceramic materials. Such materials display high electrical resistivity, low eddy current losses, sufficient permeability and good thermal stability, making them ideal candidates for such components such as transformers/inductors/sensors/microwave devices/data storage systems. As nanotechnology develops, nanoscale ferrites (1–100 nm) have improved physical properties due to a large number of atoms existing at grain boundaries that yield increased surface energy and diffusion rates. Based on the building blocks of crystal structures, ferrites can be classified as spinel, garnet, orthoferrites and hexagonal [1]. Spinel ferrites are one of the most explored classes, with their compositional flexibility and tunable physical properties. Rare-earth elements, like Lanthanum (La^{3+}), are frequently employed as dopants to improve certain properties of Mg–Co ferrite composition, resulting in interesting materials for advanced electronic applications.

Keywords: Nanotechnology, Spinel Ferrites, Mg–Co Ferrites, Nano Hardness, Lanthanum Doping

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Introduction

Spinel ferrites have become one of the most important functional materials in modern electronic and high-frequency technology, as they provide beneficial magnetic and electrical properties. Ferrites were discovered in 1930 and developed as an important class of magnetic ceramic materials [1]. They have high electrical resistivity, low eddy current losses, good magnetic permeability and strong thermal stability [2]. The electrical and magnetic behavior they exhibit at nanoscale is more pronounced, further propelling their use in miniaturized electronic devices[3]. Nanotechnology deals with the branch of materials whose structural dimensions are in between 1–100 nm. At this scale, significant number of atoms are at grain boundaries (e.g. boundaries between different grains) which increases the surface energy and diffusion rates. Nanomaterials find applications in diverse domains due to their modified mechanical, thermal, optical and electrical properties at reduced scale. Miniaturization and improved functionality are achieved through nanoscale engineering in electronics and thin-film devices. Due to these properties, ferrites are commonly used in transformers, inductors, permanent magnets, microwave devices, magnetic shielding and sensors etc. Ferrites have variety of crystal structures. Based on their crystal structures, ferrites can be classified in four types, i.e., spinel, garnet, ortho, and hexagonal ferrites [4].

Lanthanum (La) is a soft, silvery-white rare-earth metal belonging to the lanthanide series. It exhibits notable structural transformations under high pressure, retaining a face-centered cubic (Fcc) structure above 100 GPa and transforming into a body-centered tetragonal phase beyond approximately 180 GPa. In addition to fundamental research interest, lanthanum has wide industrial and medical applications. It is used in high-refractive-index optical glasses, improves the mechanical properties of steel, enhances plant growth in fertilizers, and is employed as lanthanum carbonate in the treatment of chronic kidney disease. Although the element is quite abundant in the crust of the Earth, its extraction and processing continue to be of economic significance because of intricate separation processes 5. In terms of mechanics, both double high-pressure phases of lanthanum are compressible and rather brittle. The electronic structure of lanthanum is well explained using the full potential of linearized augmented plane wave methods, and the data generated have been satisfactorily related to experimental results on phase stability and superconductivity 6. Thus, in-depth analysis of La 3+ -substituted Mg–Co ferrites is crucial for identifying the connection of the structural alteration with high-frequency performance.

Lanthanum is investigated in high-pressure superconducting hydrides within advanced materials, and it has applications as a dielectric ceramic and in hydrogen storage systems. In medicine, lanthanum carbonate is used to regulate phosphate in patients with chronic kidney disease. Moreover, La^{3+} is utilized as dopant for the enhancement of structural, magnetic, and electrical properties in ferrites. Besides fundamental research interest, lanthanum has various industrial and medical applications. It is employed in high-refractive-index optical glasses, enhances the mechanical properties of steel, accelerates plant growth (in fertilizers), and is used as lanthanum carbonate for the treatment of chronic kidney disease. This is due to Lanthanum's ability to increase refractive index and improve the image quality, so lanthanum is widely used in optical glasses and camera lenses. It serves as an important catalyst component in petroleum

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refining and is also a key material in nickel–metal hydride batteries for hybrid vehicles. It is also used as an alloying additive to improve the strength and oxidation resistance of steels. Lanthanum is widely used in optical glasses and camera lenses because it increases refractive index and improves image clarity. It plays an important role in petroleum refining as a catalyst component and is a key material in nickel–metal hydride batteries used in hybrid vehicles. Lanthanum is also applied as an alloying additive to enhance the strength and oxidation resistance of steels.

In advanced materials, lanthanum is studied for its role in high-pressure superconducting hydrides and is used in ceramic dielectrics and hydrogen storage systems. In medicine, lanthanum carbonate is prescribed to control phosphate levels in patients with chronic kidney disease. Additionally, La^{3+} is used as a dopant in ferrites to modify structural, magnetic, and electrical properties.

Literature review: Ferrites, and especially spinel-type Mg–Co ferrites, are known to have significant magnetic, electrical and structural properties but limited microwave applications as such materials will continue to be sought out for high-frequency technology interface. The substitution of lanthanum (La^{3+}) for various rare-earth elements is a simple yet effective approach to adjust some magnetic and dielectric properties as well as characteristics of Mg–Co ferrites. an overview of literature that show detailed information regarding Mg–Co ferrites and the enhancement that results upon substitution with La^{3+}. It critically discusses earlier experimental results, synthesis methods, characterization techniques, and the underlying mechanisms behind property enhancement.

Hassan et al. synthesized $Mg_{0.5}Zn_{0.5}Ce_xLa_xFe_{2-2x}O_4$ spinel-ferrite nanoparticles via conventional co-precipitation method and investigated the influence of Ce and La substitution on their microstructural, optical, electrical and dielectric characteristics. The XRD analysis concluded that the formation of a cubic spinel structure, with an increase in lattice constant and a decrease in crystallite size observed with increasing dopant concentration. Adjustments in their electronic configuration were shown by doping-tunable optical bandgap. The DC resistivity increased sharply with doping level, and the Curie temperature decreased upon substitution (suggesting weaker magnetic interactions) through electrical characterization. Regarding dielectric constant and dielectric loss, we found that they decreased with frequency and dopant concentration, whereas impedance rose. The work illustrates that rare-earth doping is effective in tuning the structural as well as the functional properties of MgZn ferrites, which can lead to further high-frequency applications with a better performance [7].

Singh et al. co-precipitated nanosized Mg–Cd–La ferrite powders using systematical oxalate co-precipitation method and investigated the effect of La^{3+} substitution on the structural (XRDA, XPS, SEM and TEM) and magnetic performance of as-synthesized NPs. Discriminating crystalline phases based on the XRD analysis showed a biphasic structure, and several parameters (lattice constant, density, porosity and crystallite size) have also been significantly altered after La^{3+} incorporation. The average crystallite size was determined to be between 25–31 nm while FTIR spectra confirmed the presence of characteristic tetrahedral and octahedral Fe–O vibrations analogous to spinel ferrites. However, the addition of La^{3+} led to decreased grain size and abnormal grain growth.

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The substitution also enhanced saturation magnetization, remanent magnetization and coercivity with size influenced magnetic behavior.

It was proposed that the increased magnetic performance of La^{3+} doped Mg–Cd ferrites are appropriate for high-frequency conditions [8]. Rashid et al. synthesized La^{3+}-substituted manganese ferrites using the sol–gel auto-combustion technique and investigated the effects of different La^{3+} concentrations on structural, optical, and dielectric properties. XRD analysis indicated that the material has a cubic spinel crystal structure and structural parameters were adjusted to increase La content. The decrease in Optical Band Gap with higher La^{3+} substitution suggests alteration in the electronic structure, which has been communicated via DRS results. The morphology and elemental composition of the nano-complex were examined by FE-SEM, EDAX analysis showed that Fe_3O_4 has a spinel structure validated from FTIR as well as Raman spectra analysis. The dielectric constant increased with temperature and La^{3+} compositional X, and AC conductivity showed a frequency dependent behavior which was found to increase as the temperature increases. The study showed that La^{3+} substitution leads to an impressive improvement in dielectric behavior of Mn–Co ferrites, therefore making them potential candidates for high-frequency applications [9].

Zhang et al. synthesized La^{3+}-doped magnesium–cobalt nanoferrites of the composition $Mg_{0.2}Co_{0.8}Fe_2{-}xLa_xO_4$ ($x = 0.0–0.1$) by the sol–gel method and studied their structural and magnetic properties. XRD results further confirmed the formation of a cubic spinel structure and successful incorporation of La^{3+} ions into the lattice. The FTIR spectra indicated that the chemical reaction was completed and demonstrated the characteristic vibrational absorption bands of spinel ferrites. The SEM images indicated that the acquired particles had a consistent spherical shape and a small size dispersion, while energy dispersive spectrums (EDS) confirmed that abundant Mg, Co, Fe and La could be detected but no other impurities were observed.

Magnetic measurements through VSM showed that the substitution of La^{3+} affects both the crystal and magnetic properties of the samples, with coercivity ranging from 611.24 to 779.43 Oe classifying it as a hard magnetic material. They concluded that La^{3+} doping modifies the structural and magnetic properties of Mg–Co nanoferrites, thus making them suitable candidates for some magnetic and high frequency devices [10]. Khan *et al.* synthesized La^{3+}-doped cobalt ferrites with the composition $CoLa_xFe_{2-x}O_4$ ($0.00 \leq x \leq 0.20$) using the co-precipitation method and confirmed the formation of a single-phase spinel structure by XRD analysis. The study focused on the dielectric and electrical properties and reported that dielectric constant and dielectric loss decreased with increasing frequency, while AC conductivity increased and followed Jonscher's power law. Impedance analysis revealed the contribution of both grain and grain boundary effects, and DC resistivity decreased with increasing temperature, indicating a hopping conduction mechanism. With increasing La^{3+} substitution, DC resistivity and drift mobility decreased, while activation energy increased. Current–voltage measurements showed improved nonlinear hysteresis behavior in doped samples, explained by space charge limited conduction and oxygen vacancy migration. The study concluded that La^{3+}-doped cobalt ferrites exhibit promising electrical characteristics for non-volatile memory and neuromorphic device applications [11].

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Material and Methods:

This research article explains the methods used to make and study the Mg-Co ferrites. It first gives a detailed explanation of how the material was prepared. Then it describes the various tools used to analyze the structure of a sample, how it responds to light, its vibrations and surface. They utilize Scanning Electron microscopy (SEM), Ultraviolet-Visible (UV-Vis) Spectroscopy, Fourier Transform Infrared (FTIR) Spectroscopy and X-ray Diffraction (XRD). The text identifies the advantages and disadvantages of each method, explains how it works, and indicates why this method was selected for this study. Making nanoparticles can be done in two primary ways: top-down methods and bottom-up methods. These two approaches rely on different concepts to create nanoscale materials.

Top-Down Approach: The top-down approach involves the fragmentation of larger particles into nanoparticles. Various mechanical, physical and chemical methods can be employed for this objective.

Bottom-Up Approach: While the bottom-up method constructs nanoparticles atom by atom or molecule by molecules, similar to a structure constructed brick by brick. This allows for improved size, morphology and composition control making it the method of choice to synthesize nanomaterials with homogenous and reproducible properties.

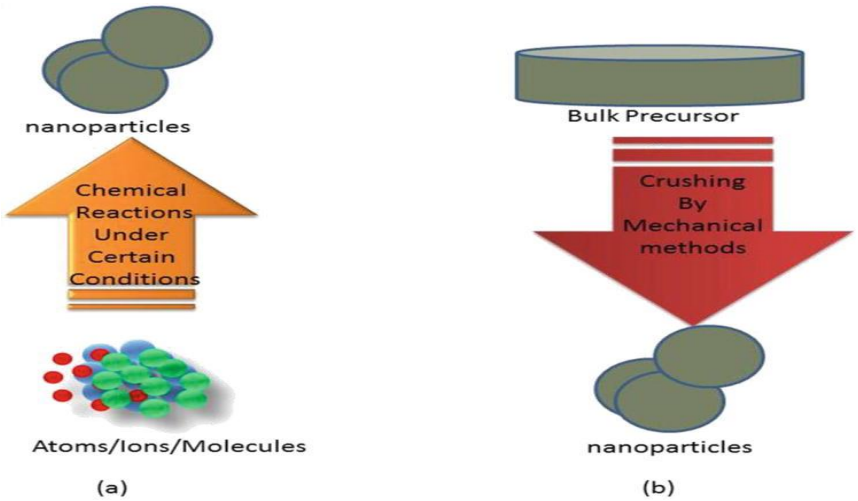


Figure 2: Top-Down and Bottom-Up approach

Common methods of bottom-up synthesis include sol gel, hydrothermal, chemical vapor deposition and chemical reduction [12].

Co-Precipitation Synthesis: The co-precipitation method is one of the easiest and inexpensive methods among the various synthesis techniques to obtain nanostructured ferrites. This is an example of the bottom-up class of synthesis strategies, in which nanoparticles are constructed from molecular or ionic precursors.

In this process, two or more metal ions are simultaneously precipitated from a homogeneous aqueous solution by the addition of a precipitating agent, generally a base

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such as sodium hydroxide (NaOH) or ammonium hydroxide (NH_4OH). The reaction produces metal hydroxide intermediates that subsequently convert to the corresponding oxide or ferrite phase after drying and calcination [13,14]

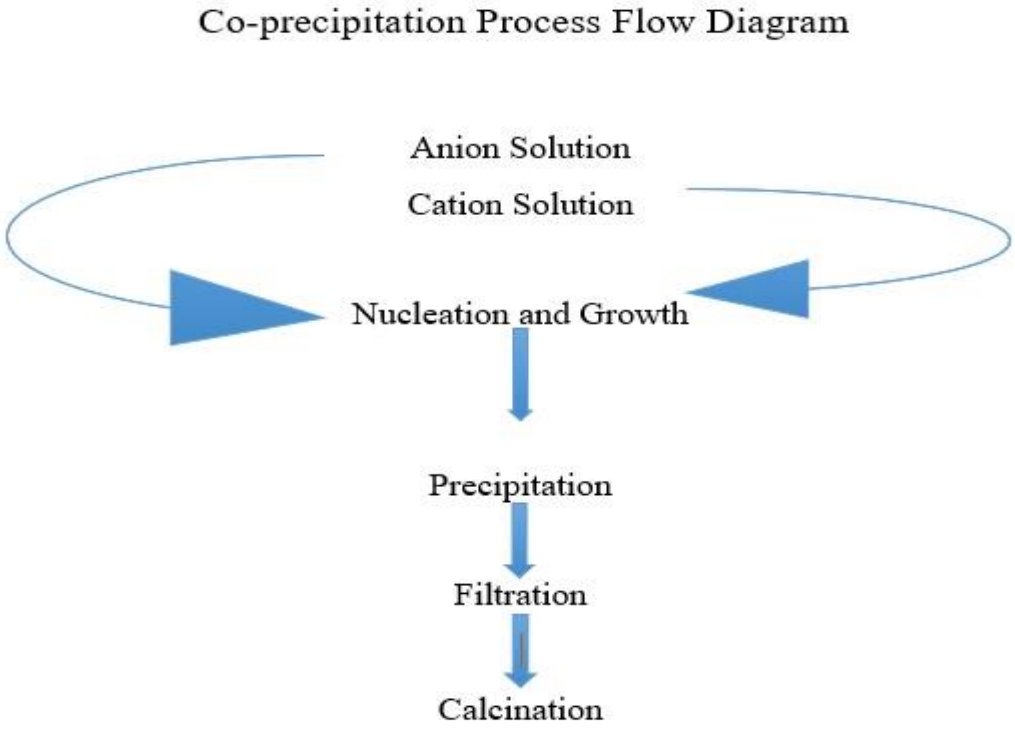


Figure 3: Co-precipitation Process Flow Diagram

The process is rapid enough to be employed for large-scale synthesis.
Produces nanosized powders with high surface area and uniform morphology.
The equipment setup is minimal, and precursors are inexpensive.
Phase purity and particle size are sensitive to slight conditions, such as pH, temperature, or concentration.
Crystallization is hindered due to particle growth from required drying and calcination.
Size is readily controllable, but specific morphology is difficult to achieve compared to sol-gel or hydrothermal methods.

Synthesis of La-substituted Mg–Co ferrites: The samples were synthesized by means of co-precipitation method. In the case of lanthanum-doped samples, the nitrates of the precursors such as $Mg(NO_3)$, $Co(NO_3)$ and $Fe(NO_3)$, were dissolved in distilled water and stirred at 100 ^oC for 6h. In the doped samples, the addition of $La(N\ O_3)_3$ was also done. At the same time, NaOH was placed on the magnetic stirring in 100 ml of distilled water, stirred at the same time 60-90 minutes. The precursor solution was then stirreded continuously with dropwise addition of the NaOH solution keeping the pH constant (10-12). Using full precipitation, the solution was filtered and washed well with distilled water. The resulting precipitate was dried at at room temperature and thereafter calcined to increase crystal purity. The resulting product was finely ground using the calcined material. To analyze structural, morphological and optical properties they characterized the synthesized samples by XRD, SEM, FTIR and UV- absorbance.

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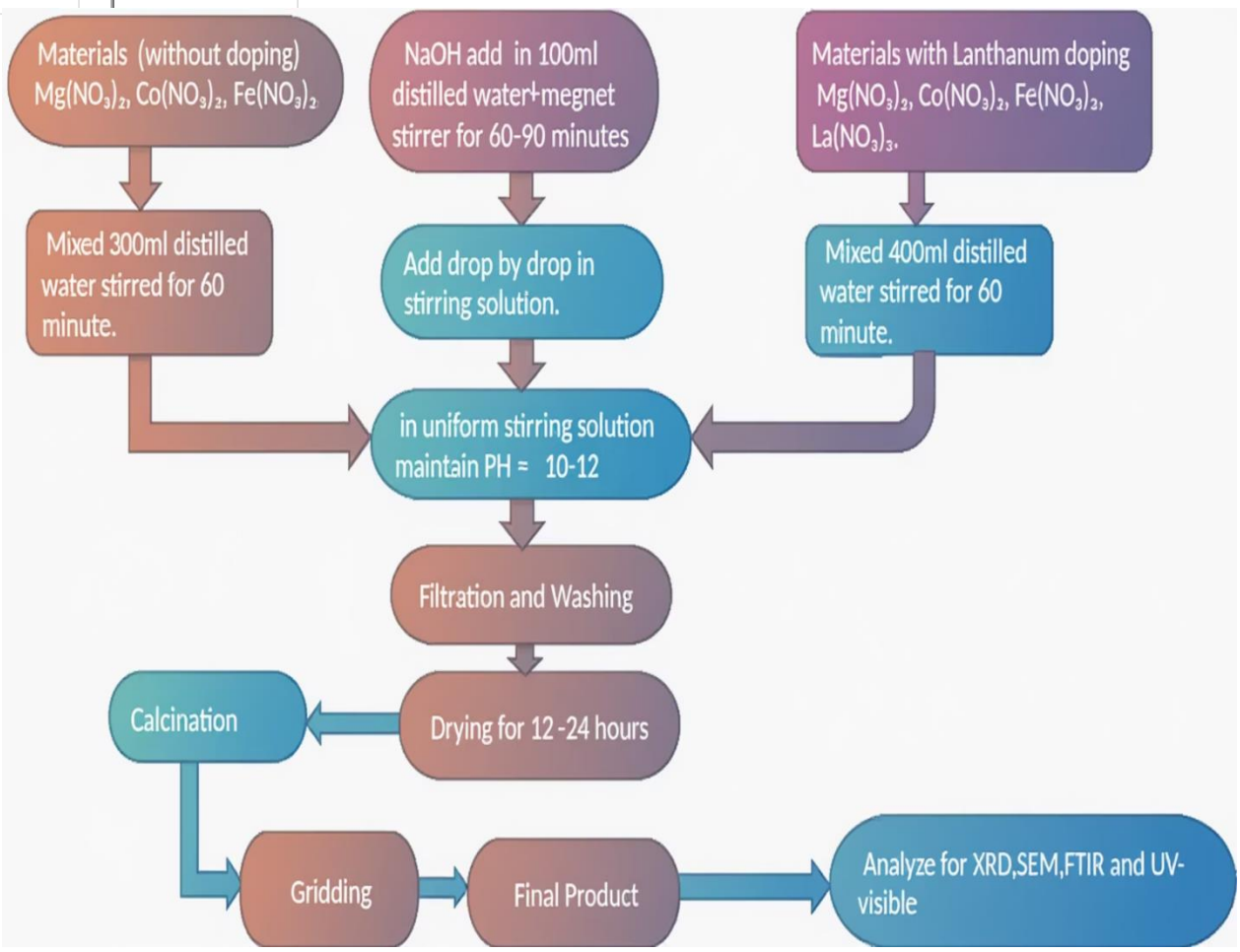


Figure 4: Synthesis of **La-substituted Mg–Co ferrites**.

Results and discussion:

Fourier Transform Infrared (FTIR) spectroscopy is a powerful technique for characterization that can provide information on the identification of chemical bonds and functional groups in materials. It detects the absorption of infrared radiation, which corresponds to molecular vibrations like stretching and bending. Since every bond absorbs at a certain frequency, FTIR gives a distinctive molecular fingerprint for the sample [15]. It was recorded in the range of **4000–400 cm^{-1}** to identify the functional groups present in the sample. The broad absorption band observed at **3435 cm^{-1}** corresponds to the **O–H stretching vibration** of adsorbed water molecules, while the peak at **1644 cm^{-1}** is attributed to the **bending vibration of H–O–H**, confirming the presence of moisture on the sample surface. The bands appearing at **2366 cm^{-1} and 2070 cm^{-1}** may be due to **atmospheric CO_2 absorption** during the measurement. Peaks at **1446 cm^{-1} and 1340 cm^{-1}** are associated with **C–O stretching vibrations** of residual organic compounds. The characteristic absorption bands observed at **1051 cm^{-1} and 874 cm^{-1}** correspond to **metal–oxygen (M–O) stretching vibrations**, which confirm the formation of the **spinel ferrite structure of $CoMgFe_2O_4$**.

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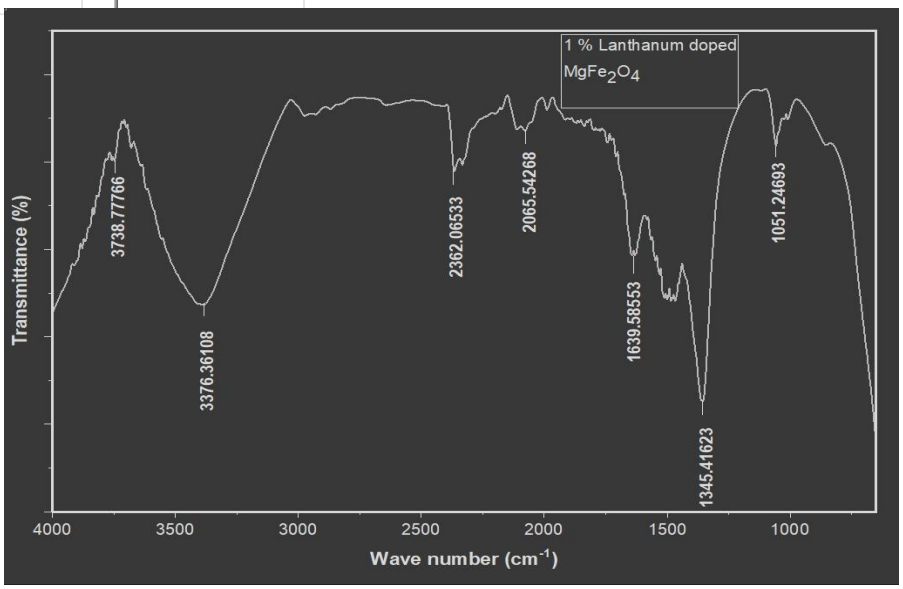


Figure 5: 1% Lanthanum doped $MgFe_2O_4$

The FTIR spectra of 1% Lanthanum-doped $MgFe_2O_4$ were recorded in the range between 4000–400 cm^{-1} to determine certain functional groups present in the sample. The broad absorption band between 3376 cm^{-1} is associated with O–H stretching vibrations of H_2O molecules adsorbed on the particle surface; The band around 1639 cm^{-1} is assigned to H–O–H bending vibrations, suggesting moisture content in the sample. The other features seen at ~2362 cm^{-1} and 2065 cm^{-1} can be interference from CO_2 in the atmosphere during measurement. The band in the range of 1345 $cm^{-1\prime}$ is attributed to C–O stretching vibrations resulting from leftover organic groups. The peak at 1051 cm^{-1} is attributed to metal–oxygen (M–O) stretching vibrations, which confirms the formation of the spinel ferrite structure of lanthanum-doped $MgFe_2O_4$.

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Ferrite materials and their magnetic behavior have been widely studied for device and nanoparticle applications (Adam et al., 2002; Dairy et al., 2019).

The co-precipitation method is commonly used for preparing ferrite nanoparticles because it supports good control over structural and magnetic properties (Shaikh et al., 2010; Petcharoen & Sirivat, 2012).

Lanthanum substitution has a significant effect on the structural, dielectric, and magnetic properties of ferrites (Gadkari et al., 2011; Huang et al., 2021; Kamran & Anis-ur-Rehman, 2023).

Recent studies have shown that La3+ doping can improve dielectric and electronic performance in ferrite-based materials for advanced applications (Haq et al., 2022; Khatoon et al., 2024).

Ferrite nanoparticles are also important beyond magnetism, including catalytic and multifunctional material applications (Kharisov et al., 2019; Moustafa et al., 2022).