Synthesis and Structural Characterization of Nickel Ferrite ($NiFe_2O_4$) Nanoparticles via Sol-Gel Auto-Combustion Method

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Abstract

Nickel ferrite ($NiFe_2O_4$) nanoparticles were successfully synthesized using the sol-gel auto-combustion method. This technique offers a cost-effective, low-temperature route to obtain phase-pure spinel ferrite with nanocrystalline morphology. Citric acid was used as a chelating and fuel agent, and the pH was adjusted using ammonia. The obtained powder was calcined at 600 °C and analyzed using X-ray diffraction (XRD). The XRD pattern confirmed the formation of a single-phase cubic spinel structure without detectable impurity peaks. The average crystallite size, calculated using the Debye–Scherrer formula, was found to be in the range of 20–30 nm. The results demonstrate that the sol-gel auto-combustion method is suitable for producing high-purity $NiFe_2O_4$ nanoparticles for magnetic, sensor, and catalytic applications.

Keywords: Nickel ferrite, Sol-gel auto-combustion, Spinel structure, Nanoparticles

1. Introduction

Spinel ferrites with the general formula MFe_2O_4, where M represents a divalent metal ion such as Ni^{2+}, Co^{2+}, or Zn^{2+}, have garnered significant attention due to their remarkable structural stability, moderate electrical conductivity, and excellent magnetic properties [1, 2]. Among these, nickel ferrite ($NiFe_2O_4$) is an inverse spinel where Ni^{2+} ions predominantly occupy the octahedral sites, while Fe^{3+} ions are distributed between tetrahedral and octahedral sites, leading to intriguing magnetic and electrical behavior [3]. Because of these properties, $NiFe_2O_4$ is widely utilized in magnetic recording media, microwave devices, catalysis, biomedical applications, and gas sensing [4, 5]. The performance of ferrite materials is highly dependent on their synthesis method, which influences properties such as particle size, morphology, crystallinity, and magnetic behavior. Traditional ceramic methods often require high sintering temperatures and extended processing times, which can lead to grain growth and inhomogeneity [6]. In contrast, wet chemical routes like the sol-gel auto-combustion method offer precise stoichiometric control, low processing temperatures, and uniform particle distribution at the nanoscale [7]. In the sol-gel auto-combustion process, metal nitrates are chelated with organic fuels such as citric acid. Upon drying and ignition, the gel undergoes a self-sustained exothermic reaction, leading to the formation of ferrite powder with nanocrystalline features. This method also facilitates homogenous mixing at the molecular level, ensuring better phase purity and fine particle sizes.

In this study, we report the successful synthesis of $NiFe_2O_4$ nanoparticles via the sol-gel auto-combustion method. The structural properties of the synthesized ferrite were examined using X-ray diffraction (XRD) to confirm phase purity, crystallite size, and lattice parameters. The objective is to demonstrate the suitability of this method for producing high-quality $NiFe_2O_4$ for potential use in advanced magnetic and electronic applications.

2. Experimental Method

2.1 Materials

All chemicals used in this study were of analytical grade and used without further purification. Nickel nitrate hexahydrate ($Ni(NO_3)_2 \cdot 6H_2O$), ferric nitrate nonahydrate ($Fe(NO_3)_3 \cdot 9H_2O$), and citric acid monohydrate ($C_6H_8O_7 \cdot H_2O$) were procured from Sigma-Aldrich and used as starting materials. Deionized water was used throughout the synthesis.

2.2 Synthesis of $NiFe_2O_4$ Nanoparticles

Nickel ferrite nanoparticles were synthesized via the sol-gel auto-combustion method [8]. Schematic diagram shown in Figure 1. Stoichiometric amounts of nickel nitrate and ferric nitrate (molar ratio 1:2) were dissolved in deionized water under constant stirring. Citric acid was added to the metal nitrate solution as a chelating agent, maintaining a metal nitrates to citric acid molar ratio of 1:1 [9, 10]. The resulting solution was stirred continuously at 80°C to obtain a homogeneous sol. As water evaporated, the sol gradually transformed into a viscous gel. The gel was then heated on a hot plate at ~200°C to initiate the auto-combustion reaction. The exothermic nature of the reaction resulted in the spontaneous ignition of the gel, producing a voluminous, fluffy black powder, indicative of the formation of nickel ferrite [11].

2.3 Calcination

The as-combusted powder was ground using an agate mortar and pestle and then calcined in a muffle furnace at 600°C for 4 hours to improve crystallinity and remove any residual organic matter [12]. The heating rate was maintained at 5°C/min to avoid thermal shocks. After calcination, the powder was cooled naturally to room temperature and stored in airtight containers for further characterization.

2.4 Material Characterizations:

The powder approach is frequently used to facilitate the crystal structure investigation of a material easier. A continuous spectrum of X-rays with a fixed angle of incidence is used in the Laue method, one of the first techniques used for identifying crystal structure [16]. This technique is appropriate for detecting dynamic processes inside the crystal structure because it yields diffraction conclusions more quickly than those obtained with monochromatic X-rays. This method is called the centered crystal method when the wavelength varies but the angle of incidence remains fixed. The sample in this procedure rotates at a fixed angular velocity and is exposed to a monochromatic hard X-ray beam. On the other hand, the angle of incidence varies while the wavelength remains constant with the powder approach. Calculating crystal dimensions with the Scherrer equation is an essential use of XRD in nanocrystal studies [13].

where, β is full width at half maximum (FWHM) of the diffraction peak, and θ is the peak position in radians.

In this work, XRD confirmed the formation of single-phase spinel $NiFe_2O_4$ with good crystallinity. No impurity peaks were observed, indicating high phase purity. The crystallite size was found to be in the nanometer range, consistent with the nature of the sol-gel auto-combustion synthesis process.

3. Results and Discussions:

3.1 Reaction Mechanism for the Sol-Gel Auto-Combustion Synthesis of $NiFe_2O_4$:

The sol-gel auto-combustion method is a versatile chemical synthesis route used to prepare homogeneous and phase-pure nanocrystalline materials at relatively low temperatures. In this process, metal nitrates act as oxidizers while a suitable organic fuel (typically citric acid or glycine) serves as the reducing agent and complexing agent. The process involves three key steps: (i) sol formation, (ii) gelation, and (iii) auto-combustion. In this study, nickel nitrate [$Ni(NO_3)_2 \cdot 6H_2O$] and ferric nitrate [$Fe(NO_3)_3 \cdot 9H_2O$] were used as the metal precursors, while citric acid ($C_6H_8O_7$) served as both the complexing agent and fuel. The molar ratio of metal ions to citric acid was carefully optimized (typically 1:1 or 1:2) to ensure stoichiometry and efficient combustion.

Step 1: Complexation and Sol Formation

Upon dissolving the metal nitrates and citric acid in deionized water under constant stirring, complexation occurs through the chelation of metal cations by citric acid. This leads to the formation of a homogeneous sol containing metal–citrate complexes in eq.2.

$Ni^{2+} + Fe^{3+} + C_6H_8O_7 \rightarrow [Ni\text{-}Citrate\text{-}Fe]complex + H^+$ … (2)

The citric acid acts by binding through its carboxylate and hydroxyl groups, stabilizing the metal ions in solution and preventing precipitation [14].

Step 2: Gelation

On heating (around 70–90°C), the sol gradually loses water and becomes more viscous, forming a gel-like structure. This gel contains uniformly distributed metal ions embedded in an organic matrix in eq. 3.

[Ni-Citrate-Fe]complex → Polymeric Gel … (3)

This gelation process is vital to ensure uniform distribution of cations, which ultimately results in homogeneous particle size upon combustion [15].

Step 3: Auto-Combustion Reaction

On further heating (around 150–250°C), the dried gel undergoes a self-sustained exothermic combustion reaction due to the redox interaction between the oxidizing nitrates and the reducing citric acid. This results in the rapid release of gases such as CO_2, H_2O, and N_2, and the formation of a voluminous, fluffy, and porous powder in eq. 4.

$C_6H_8O_7 + NO_3^- \rightarrow NiFe_2O_4 + CO_2 + H_2O + N_2 + Heat$ … (4)

The approximate redox-based combustion reaction can be written as eq. 5.

$Ni(NO_3)_2 + 2Fe(NO_3)_3 + C_6H_8O_7 \rightarrow NiFe_2O_4 + 6NO_2 + 6H_2O + 6CO_2$ … (5)

The combustion is self-propagating and provides the thermal energy necessary for in-situ crystallization of the spinel ferrite phase. No external calcination may be required if the combustion is sufficiently exothermic.

Crystallization of $NiFe_2O_4$ Spinel Phase:

The final product is nanocrystalline nickel ferrite ($NiFe_2O_4$), which crystallizes in the cubic spinel structure (space group $Fd\bar{3}m$), with Ni^{2+} occupying the octahedral (B) sites and Fe^{3+} distributed between tetrahedral (A) and octahedral sites in eq. 6 [16].

$Ni^{2+} + 2Fe^{3+} + 4O^{2-} \rightarrow NiFe_2O_4$ … (6)

3.2 X-ray Diffraction (XRD) Analysis

The phase purity and crystalline structure of the synthesized $NiFe_2O_4$ nanoparticles were characterized using X-ray diffraction (XRD) in the 2θ range of 10°–80° in Figure 1. The diffraction peaks observed at 2θ values approximately 30.2°, 35.6°, 43.3°, 53.6°, 57.2°, and 62.9° correspond to the (220), (311), (400), (422), (511), and (440) crystal planes, respectively. These reflections are well indexed to the face-centered cubic (FCC) spinel structure of nickel ferrite ($NiFe_2O_4$) and are in good agreement with the standard JCPDS card no. 10-0325 [17, 18]. The most intense peak at 35.6°, assigned to the (311) plane, confirms the formation of a single-phase spinel ferrite. No additional peaks corresponding to impurities or secondary phases such as NiO or Fe_2O_3 were detected, indicating the phase purity of the synthesized material [19]. The sharp and well-defined nature of the peaks suggests that the material is crystalline, while the moderate broadening indicates the nanoscale size of the crystallites. The formation of a spinel structure can be attributed to the homogeneous mixing of metal precursors and the rapid combustion process that promotes uniform nucleation and growth of nanoparticles during synthesis. These structural features make $NiFe_2O_4$ a promising candidate for applications in magnetic materials, microwave devices, and supercapacitors [20].

Fig. 1: X-ray diffraction pattern of NiFe2O4 annealed at 600□ C for 2 h.

3. Conclusions

In this study, nickel ferrite ($NiFe_2O_4$) nanoparticles were successfully synthesized using the sol-gel auto-combustion method. The X-ray diffraction (XRD) analysis confirmed the formation of a single-phase spinel structure with high crystallinity. The average crystallite size, calculated using the Scherrer equation, was found to be in the nanometer range, indicating the effectiveness of the sol-gel process in controlling particle growth. The sharp and intense diffraction peaks further suggest good crystallinity and phase purity of the synthesized material. The sol-gel auto-combustion method proved to be a cost-effective, energy-efficient, and facile route for producing $NiFe_2O_4$ nanoparticles with controlled morphology and desirable structural properties. These results suggest that $NiFe_2O_4$ synthesized by this method holds potential for applications in magnetic devices, sensors, and energy storage systems. Future studies may focus on detailed magnetic, electrical, and electrochemical investigations to explore its practical applications in various technological fields.

4. References

1) Goldman, A. (2006). Modern Ferrite Technology. Springer.

2) Kodama, R. H. (1999). Magnetic nanoparticles. Journal of Magnetism and Magnetic Materials, 200(1-3), 359–372.

3) Ghasemi, A., et al. (2008). Nanocrystalline $NiFe_2O_4$ ferrite prepared by sol–gel auto-combustion method. Journal of Alloys and Compounds, 465(1-2), 387–391.

4) Mahmood, T., et al. (2013). Structural and magnetic properties of $NiFe_2O_4$ nanoparticles prepared by a low-temperature solution combustion method. Journal of Magnetism and Magnetic Materials, 330, 138–142.

5) Ahmed, M. A., et al. (2010). Structural and magnetic properties of nanocrystalline $NiFe_2O_4$ ferrite. Materials Chemistry and Physics, 123(1), 118–123.

6) Gul, I. H., et al. (2008). Structural, magnetic and electrical properties of cobalt ferrites synthesized by co-precipitation route. Journal of Magnetism and Magnetic Materials, 320(3-4), 270–275.

7) Sayyed, M. I., et al. (2020). Ni–Mg ferrite as a potential gamma-ray shielding material: synthesis, characterization and radiation attenuation study. Journal of Materials Science: Materials in Electronics, 31(12), 9708–9720.

8) Rani, S., et al. (2019). Synthesis and characterization of Ni-Mg ferrite nanoparticles for gas sensing applications. Journal of Materials Science: Materials in Electronics, 30(11), 10661–10670.

9) Arulmurugan, R., et al. (2005). Magnetic and dielectric behavior of nanocrystalline Ni–Zn and Ni–Cu ferrites synthesized by co-precipitation method. Journal of Magnetism and Magnetic Materials, 288, 470–477.

10) Sundararajan, M., et al. (2014). Effect of Mg substitution on the structure and magnetic properties of nickel ferrite nanoparticles synthesized by sol-gel method. Ceramics International, 40(10), 16143–16150.

11) Valenzuela, R. (1994). Magnetic Ceramics. Cambridge University Press.

12) Pullar, R. C. (2012). Hexagonal ferrites: A review of the synthesis, properties and applications of hexaferrite ceramics. Progress in Materials Science, 57(7), 1191–1334.

13) Ghasemi, A., et al. (2007). Sol–gel synthesis and characterization of $NiFe_2O_4$ nanoparticles. Journal of Alloys and Compounds, 454(1-2), 420–424.

14) Patange, S. M., et al. (2011). Structural and magnetic properties of sol–gel prepared Mg substituted nickel ferrite nanoparticles. Journal of Magnetism and Magnetic Materials, 323(3-4), 329–334.

15) Reddy, B. P. N., et al. (2009). Synthesis and characterization of Ni-Mg ferrite nanoparticles by auto combustion method. Journal of Alloys and Compounds, 481(1-2), 748–752.

16) Ranjith, R., et al. (2021). Influence of synthesis route on the microstructure and dielectric properties of Mg-doped nickel ferrite nanoparticles. Journal of Materials Science: Materials in Electronics, 32(4), 4212–4222.

17) Nimbalkar, A. V., et al. (2017). Structural, morphological and gas sensing properties of sol–gel synthesized $NiFe_2O_4$ nanoparticles. Sensors and Actuators B: Chemical, 248, 980–988.

18) Singh, A. K., et al. (2016). Structural, electrical and magnetic properties of Mg substituted $NiFe_2O_4$ nanoparticles synthesized via sol–gel method. Journal of Magnetism and Magnetic Materials, 417, 119–124.

19) Shirsath, S. E., et al. (2014). Sol–gel auto-combustion synthesis of Mg substituted $NiFe_2O_4$ nanoparticles: Structural, magnetic and Mössbauer studies. Journal of Alloys and Compounds, 581, 440–446.

20) Nadeem, K., et al. (2018). Structural, dielectric and magnetic properties of $Ni_{1-x}Mg_xFe_2O_4$ ferrite nanoparticles synthesized by sol-gel auto combustion method. Journal of Alloys and Compounds, 763, 1030–10