Review

Not peer-reviewed version

Sol-Gel Auto-Combustion Synthesis of Magnetic Nanomaterials as an Efficient Route for Advanced Functional Materials

Usama Nazir *

Posted Date: 16 December 2025

doi: 10.20944/preprints202512.1270.v1

Keywords: sol-gel auto-combustion; magnetic nanoparticles; spinel ferrites; citric acid; combustion synthesis; nanocrystalline materials

Review

Sol-Gel Auto-Combustion Synthesis of Magnetic Nanomaterials as an Efficient Route for Advanced Functional Materials

Usama Nazir

Department of Physics, COMSATS University Islamabad, Lahore Campus, Pakistan; usamanazir13@gmail.com

Abstract

The sol-gel auto-combustion method has emerged as a versatile and efficient technique for synthesizing magnetic nanomaterials with controlled structural and functional properties. This review comprehensively examines the fundamental principles, reaction mechanisms, and process parameters governing the sol-gel auto-combustion synthesis of magnetic nanoparticles, with particular emphasis on ferrite materials. The method exploits the exothermic redox reaction between metal nitrates (oxidizers) and organic fuels such as citric acid, glycine, and urea to produce nanocrystalline powders at relatively low temperatures. Critical parameters including fuel-to-oxidizer ratio, pH, complexing agents, calcination temperature, and heating rate are systematically analyzed for their influence on phase purity, crystallite size, morphology, and magnetic properties. The review covers the synthesis of various magnetic materials including spinel ferrites (MFe_2O_4, where M = Co, Ni, Zn, Mn), hexagonal ferrites ($BaFe_{12}O_{19}$, $SrFe_{12}O_{19}$), and mixed ferrite systems. Characterization techniques commonly employed for product evaluation, including X-ray diffraction, electron microscopy, and vibrating sample magnetometry, are discussed. Applications of sol-gel derived magnetic nanomaterials in data storage, biomedical fields, catalysis, environmental remediation, and electromagnetic interference shielding are highlighted. Finally, current challenges and future perspectives for advancing this synthesis methodology are outlined.

Keywords: sol-gel auto-combustion; magnetic nanoparticles; spinel ferrites; citric acid; combustion synthesis; nanocrystalline materials

---

1. Introduction

Magnetic nanomaterials have attracted significant research interest over the past three decades due to their unique size-dependent properties and diverse technological applications [1,2]. Among magnetic materials, ferrites occupy a prominent position owing to their exceptional combination of magnetic properties, chemical stability, and cost-effectiveness [3]. Ferrite nanoparticles find applications in high-density magnetic recording media, microwave absorption devices, biomedical imaging and drug delivery, catalysis, environmental remediation, and electromagnetic interference shielding [4,5].

The synthesis of ferrite nanoparticles with controlled composition, phase purity, particle size, and morphology remains a critical challenge that determines their functional performance. Common synthesis methods include conventional solid-state reaction, co-precipitation, hydrothermal synthesis, microemulsion, and mechanical alloying [6]. While each method offers specific advantages, the sol-gel auto-combustion technique has gained widespread popularity as a simple, rapid, energy-efficient, and cost-effective approach for producing nanocrystalline ferrite powders [1].

The sol-gel auto-combustion method, also termed solution combustion synthesis (SCS) or gel combustion synthesis, combines the advantages of sol-gel processing (molecular-level homogeneity) with combustion synthesis (rapid exothermic reaction) [7]. The process involves dissolving metal

 doi:10.20944/preprints202512.1270.v1

nitrate precursors along with organic fuel(s) in an aqueous medium, followed by gel formation upon heating and subsequent self-propagating combustion to yield nanocrystalline oxide powders. The exothermic reaction between the oxidizing metal nitrates and reducing organic fuel generates high temperatures locally, facilitating rapid crystallization while the large volume of evolved gases prevents particle agglomeration [8].

Since its introduction by Kingsley and Patil in 1988 for the synthesis of alumina powders [9], the solution combustion method has been applied to synthesize oxide materials including ferrites, perovskites, garnets, and mixed metal oxides [10]. The method offers the following key advantages: (i) low processing temperature compared to conventional ceramic routes; (ii) molecular-level mixing ensuring compositional homogeneity; (iii) rapid synthesis time (minutes vs. hours); (iv) simple equipment requirements; (v) ability to produce metastable phases; and (vi) easy scalability [11].

This review aims to provide a comprehensive overview of the sol-gel auto-combustion synthesis of magnetic nanomaterials, with emphasis on ferrite systems. The fundamental principles and reaction mechanisms are first discussed, followed by detailed analysis of the critical process parameters that control product characteristics. The synthesis of various ferrite systems including spinels and hexaferrites is reviewed. Finally, applications and future perspectives are outlined.

2. Fundamental Principles and Reaction Mechanism

2.1. Basic Concepts of Combustion Synthesis

Combustion synthesis relies on highly exothermic self-sustaining reactions between reactants to produce the desired product [12]. In the context of sol-gel auto-combustion, the reaction occurs between metal nitrates (acting as oxidizers) and organic compounds (acting as fuels) in an aqueous solution. The combustion reaction can be thermodynamically described using concepts from propellant chemistry, where the stoichiometric fuel-to-oxidizer ratio determines the maximum energy release [13].

The combustion reaction can be represented generically as:

$$\text{Metal Nitrates} + \text{Fuel} \rightarrow \text{Metal Oxide} + \text{Gaseous Products} \tag{1}$$

For example, the synthesis of nickel ferrite using citric acid as fuel proceeds as:

$$Ni(NO_3)_2 + 2Fe(NO_3)_3 + C_6H_8O_7 \rightarrow NiFe_2O_4 + CO_2\uparrow + H_2O\uparrow + N_2\uparrow \tag{2}$$

2.2. Role of Organic Fuels

Organic fuels serve dual functions in the sol-gel auto-combustion process: (i) as chelating agents that complex with metal ions to ensure homogeneous mixing, and (ii) as fuels that provide the reducing power for the combustion reaction [1]. The most commonly employed fuels include:

Table 1. Common fuels used in sol-gel auto-combustion synthesis.

Fuel	Formula	Reducing Valence	Characteristics
Citric acid	$C_6H_8O_7$	+18	Strong chelating, moderate flame
Glycine	$C_2H_5NO_2$	+9	High flame temperature
Urea	CH_4N_2O	+6	Vigorous combustion
Ethylene glycol	$C_2H_6O_2$	+10	Good complexing ability
Tartaric acid	$C_4H_6O_6$	+10	Moderate chelating

Citric acid is the most widely used fuel due to its excellent chelating ability with most metal ions and its moderate combustion characteristics that allow better control over particle properties [14,15].

2.3. Fuel-to-Oxidizer Ratio

The fuel-to-oxidizer ratio (ϕ) is a critical parameter that determines the thermodynamics of the combustion reaction. Based on propellant chemistry concepts, ϕ is defined as:

$$\phi = \frac{\text{Sum of reducing valences of fuel}}{\text{Sum of oxidizing valences of oxidizer}} \tag{3}$$

For $\phi = 1$, the reaction is stoichiometric, theoretically producing maximum flame temperature and complete combustion. When $\phi < 1$, the mixture is fuel-lean (oxidizer excess), while $\phi > 1$ indicates a fuel-rich condition [13]. Studies have shown that the optimal ϕ for synthesizing pure-phase ferrites often deviates from unity due to practical considerations such as heat losses and incomplete combustion [1,16].

2.4. Combustion Modes

The auto-combustion process can proceed through different modes depending on the reaction conditions:

- **Volume Combustion Synthesis (VCS):** The entire gel mass ignites uniformly, resulting in rapid heat release throughout the sample.
- **Self-Propagating High-Temperature Synthesis (SHS):** Combustion initiates at one point and propagates as a wave through the sample.
- **Smoldering Combustion:** Slow, flameless reaction that proceeds without visible flame.

The combustion mode affects the heat generation rate and product characteristics. Generally, smoldering combustion produces finer particles due to lower peak temperatures [10].

3. Critical Process Parameters

3.1. pH of the Precursor Solution

The pH of the precursor solution significantly influences the chelation efficiency and gel formation behavior. For citric acid-based synthesis, the pH is typically adjusted to 7 using ammonia solution to ensure complete complexation of metal ions [16]. At low pH, carboxylic groups of citric acid are protonated, reducing their chelating ability. At high pH (>10), hydroxide precipitation may occur preferentially. Studies have demonstrated that pH values between 6-8 generally yield optimal results for ferrite synthesis [8].

3.2. Fuel Type and Concentration

Different fuels produce distinct combustion characteristics:

- **Citric acid:** Produces moderate flame temperatures (typically 200-400°C), yielding nanocrystalline products with good phase purity. The citrate anion effectively chelates trivalent metal ions through its carboxyl and hydroxyl groups [15].
- **Glycine:** Generates higher flame temperatures (>600°C), resulting in larger crystallite sizes but excellent crystallinity. Glycine's zwitterionic nature allows it to effectively complex both acidic and basic metal ions [9].
- **Urea:** Produces very high flame temperatures with vigorous gas evolution. The combustion tends to be more violent, which can compromise particle size control [11].

The fuel concentration relative to metal nitrates influences the combustion characteristics. Excess fuel leads to incomplete oxidation and carbon contamination, while fuel deficiency results in unreacted nitrates and poor crystallinity [1].

3.3. Calcination Temperature and Time

While the as-burnt product often exhibits the desired phase, post-combustion calcination is frequently employed to:

- Improve crystallinity by allowing atomic rearrangement
- Remove residual carbon and organic residues
- Eliminate secondary phases
- Control final particle size

Calcination temperatures typically range from 500-1000°C for ferrites, with higher temperatures promoting grain growth through Ostwald ripening [1]. The crystallite size generally follows an Arrhenius-type relationship with temperature:

$$D = D_0 \exp\left(-\frac{E_a}{RT}\right) \tag{4}$$

where D is crystallite size, E_a is activation energy, R is the gas constant, and T is temperature.

3.4. Heating Rate

The heating rate during gel combustion affects the nature of combustion:

- **Slow heating:** Gradual decomposition, lower peak temperature, smaller particles
- **Rapid heating:** Violent combustion, higher temperature, larger particles

Controlled heating rates (typically 5-10°C/min) are often preferred for reproducible synthesis [8].

4. Synthesis of Magnetic Nanomaterials

4.1. Spinel Ferrites (MFe_2O_4)

Spinel ferrites with the general formula MFe_2O_4 (where M = Co, Ni, Zn, Mn, Cu, Mg) represent the most extensively studied magnetic materials synthesized via sol-gel auto-combustion [1].

4.1.1. Cobalt Ferrite ($CoFe_2O_4$)

Cobalt ferrite exhibits high magnetocrystalline anisotropy ($K_1 = 2.65 \times 10^6$ erg/cm^3) and moderate saturation magnetization, making it attractive for magnetic recording applications [2]. Synthesis typically involves:

1. Dissolving $Co(NO_3)_2 \cdot 6H_2O$ and $Fe(NO_3)_3 \cdot 9H_2O$ in water
2. Adding citric acid in 1:1 molar ratio to total metal ions
3. Adjusting pH to 7 with NH_4OH
4. Heating at 80°C to form gel
5. Auto-combustion to produce powder
6. Calcination at 500-800°C

Typical results show crystallite sizes of 20-60 nm and saturation magnetization of 60-80 emu/g depending on synthesis parameters [17].

4.1.2. Nickel Ferrite ($NiFe_2O_4$)

Nickel ferrite is a typical inverse spinel with soft magnetic properties, widely used in high-frequency applications [3]. The sol-gel auto-combustion synthesis follows similar procedures as cobalt ferrite. Key findings include:

- Crystallite size: 15-45 nm (as-burnt), 30-80 nm (calcined at 700°C)
- Saturation magnetization: 35-50 emu/g
- Coercivity: 50-200 Oe (soft magnetic behavior)

4.1.3. Zinc Ferrite ($ZnFe_2O_4$)

Zinc ferrite is a normal spinel with paramagnetic behavior in bulk but exhibits ferrimagnetic properties at the nanoscale due to cation redistribution [6]. This size-dependent magnetic transition makes it particularly interesting for nanoscale applications.

Preprints.org (www.preprints.org) | NOT PEER-REVIEWED | Posted: 16 December 2025 doi:10.20944/preprints202512.1270.v1

4.1.4. Mixed Spinel Ferrites

The sol-gel auto-combustion method readily allows synthesis of mixed ferrites such as $Ni_{1-x}Zn_xFe_2O_4$, $Co_{1-x}Zn_xFe_2O_4$, and $Mn_{1-x}Zn_xFe_2O_4$ by adjusting the precursor stoichiometry [18]. These mixed systems allow tuning of magnetic properties between end-member compositions.

4.2. Hexagonal Ferrites

4.2.1. M-Type Hexaferrites ($BaFe_{12}O_{19}$, $SrFe_{12}O_{19}$)

M-type hexaferrites are hard magnetic materials with the magnetoplumbite structure, widely used as permanent magnets [5]. The sol-gel auto-combustion synthesis of these materials requires careful control of stoichiometry due to the large Fe:Ba(Sr) ratio of 12:1.

Synthesis considerations include:

- Higher calcination temperatures (850-1100°C) required
- Fe/Ba(Sr) ratio slightly above stoichiometry (12.2:1) to compensate for Fe losses
- Phase purity sensitive to pH and fuel ratio

Typical magnetic properties achieved: Ms = 50-70 emu/g, Hc = 3000-6000 Oe [14].

4.2.2. Rare Earth Doped Hexaferrites

The incorporation of rare earth elements (La, Nd, Sm, Gd, Ho, Dy) into hexaferrites has been extensively studied to modify their magnetic and optical properties [19]. Sol-gel auto-combustion is particularly suitable for synthesizing doped hexaferrites due to its molecular-level mixing capability that ensures uniform dopant distribution.

5. Characterization Techniques

5.1. X-Ray Diffraction (XRD)

XRD is the primary technique for:

- Phase identification by matching diffraction patterns with ICDD database
- Lattice parameter determination using Bragg's law
- Crystallite size estimation using Scherrer equation:

$$D = \frac{K\lambda}{\beta \cos\theta} \tag{5}$$

where D is crystallite size, K is shape factor (0.9), λ is X-ray wavelength, β is FWHM, and θ is Bragg angle [21].

5.2. Electron Microscopy

- **SEM:** Surface morphology, particle shape, agglomeration
- **TEM:** Particle size distribution, crystallinity, selected area diffraction
- **HRTEM:** Lattice fringes, defects, core-shell structures

5.3. Vibrating Sample Magnetometry (VSM)

VSM measures magnetic properties including:

- Saturation magnetization (M_s)
- Coercivity (H_c)
- Remanence (M_r)
- Squareness ratio (M_r/M_s)

5.4. Fourier Transform Infrared Spectroscopy (FTIR)

FTIR identifies:

- Metal-oxygen stretching vibrations (400-600 cm^{-1})

 doi:10.20944/preprints202512.1270.v1

- Residual organic species
- Tetrahedral and octahedral site occupancy

6. Applications

6.1. Data Storage and Magnetic Recording

Magnetic nanoparticles with high coercivity and squareness ratio are essential for high-density data storage media. Sol-gel derived $CoFe_2O_4$ and barium hexaferrite nanoparticles have been investigated as potential recording media materials [4].

6.2. Biomedical Applications

Magnetic nanoparticles synthesized via sol-gel auto-combustion find applications in:

- Magnetic resonance imaging (MRI) contrast agents
- Magnetic hyperthermia for cancer treatment
- Targeted drug delivery
- Bioseparation

Biocompatibility and superparamagnetic behavior at appropriate sizes are critical requirements [2].

6.3. Environmental Remediation

Magnetic ferrite nanoparticles serve as efficient adsorbents for:

- Heavy metal removal from wastewater
- Dye degradation (photocatalysis, Fenton-like reactions)
- Oil spill cleanup

The magnetic property enables easy separation and recovery of the adsorbent material [20].

6.4. Electromagnetic Interference (EMI) Shielding

Ferrite nanomaterials, particularly hexaferrites and composite systems, are employed in EMI shielding and microwave absorption applications due to their magnetic and dielectric losses in the GHz frequency range [5].

6.5. Catalysis

Spinel ferrites have demonstrated catalytic activity in various reactions including:

- Water-gas shift reaction
- Hydrogen peroxide decomposition
- Organic pollutant degradation

7. Challenges and Future Perspectives

Despite its numerous advantages, the sol-gel auto-combustion method faces several challenges:

- **Process control:** The violent nature of combustion makes precise temperature and atmosphere control difficult.
- **Reproducibility:** Batch-to-batch variations in combustion behavior can affect product consistency.
- **Scale-up:** Large-scale synthesis faces heat dissipation and gas evolution challenges.
- **Environmental concerns:** Evolution of NOx gases during combustion requires appropriate handling.

Future research directions include:

- Development of green fuels derived from natural sources
- Microwave-assisted combustion for improved control
- In-situ monitoring of combustion process
- Integration with 3D printing for structured materials

- Computational modeling for process optimization

8. Conclusion

The sol-gel auto-combustion method has established itself as a powerful and versatile technique for synthesizing magnetic nanomaterials with controlled properties. The method successfully combines the homogeneity advantages of sol-gel processing with the rapid kinetics of combustion synthesis to produce nanocrystalline ferrites at relatively low temperatures. Critical parameters including fuel type, fuel-to-oxidizer ratio, pH, and calcination conditions significantly influence the structural and magnetic properties of the products. The technique has been successfully applied to synthesize a wide range of magnetic materials including spinel ferrites, hexaferrites, and their doped variants. With continued research addressing current challenges, the sol-gel auto-combustion method will remain a key synthesis route for advanced magnetic nanomaterials serving diverse technological applications.

Author Contributions: Conceptualization, U.N.; literature survey and analysis, U.N.; writing—original draft preparation, U.N.; writing—review and editing, U.N. The author has read and agreed to the published version of the manuscript.

Funding: This research received no external funding.

Data Availability Statement: Not applicable. This is a review article and no new datasets were generated or analyzed.

Acknowledgments: The authors acknowledge the Department of Physics, COMSATS University Islamabad, Lahore Campus, for providing the academic environment for this work.

Conflicts of Interest: The authors declare no conflict of interest.

References

1. Sutka, A., & Mezinskis, G. (2012). Sol-gel auto-combustion synthesis of spinel-type ferrite nanomaterials. *Frontiers of Materials Science*, 6(2), 128-141.
2. Puli, V.S., Adireddy, S., & Ramana, C.V. (2015). Chemical bonding and magnetic properties of gadolinium (Gd) substituted cobalt ferrite. *Journal of Alloys and Compounds*, 644, 470-475.
3. Smit, J., & Wijn, H.P.J. (1959). *Ferrites: Physical Properties of Ferrimagnetic Oxides in Relation to Their Technical Applications*. Philips Technical Library.
4. Harris, V.G. (2012). Modern microwave ferrites. *IEEE Transactions on Magnetics*, 48(3), 1075-1104.
5. 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.
6. Valenzuela, R. (2012). Novel applications of ferrites. *Physics Research International*, 2012, Article ID 591839.
7. Aruna, S.T., & Mukasyan, A.S. (2008). Combustion synthesis and nanomaterials. *Current Opinion in Solid State and Materials Science*, 12(3-4), 44-50.
8. Costa, A.C.F.M., Tortella, E., Morelli, M.R., & Kiminami, R.H.G.A. (2003). Synthesis, microstructure and magnetic properties of Ni-Zn ferrites. *Journal of Magnetism and Magnetic Materials*, 256(1-3), 174-182.
9. Kingsley, J.J., & Patil, K.C. (1988). A novel combustion process for the synthesis of fine particle alpha-alumina and related oxide materials. *Materials Letters*, 6(11-12), 427-432.
10. Mukasyan, A.S., Epstein, P., & Dinka, P. (2007). Solution combustion synthesis of nanomaterials. *Proceedings of the Combustion Institute*, 31(2), 1789-1795.
11. Varma, A., Mukasyan, A.S., Rogachev, A.S., & Manukyan, K.V. (2016). Solution combustion synthesis of nanoscale materials. *Chemical Reviews*, 116(23), 14493-14586.
12. Moore, J.J., & Feng, H.J. (1995). Combustion synthesis of advanced materials: Part I. Reaction parameters. *Progress in Materials Science*, 39(4-5), 243-273.
13. Jain, S.R., Adiga, K.C., & Pai Verneker, V.R. (1981). A new approach to thermochemical calculations of condensed fuel-oxidizer mixtures. *Combustion and Flame*, 40, 71-79.
14. Ataie, A., & Mali, A. (2008). Characteristics of barium hexaferrite nanocrystalline powders prepared by a sol-gel combustion method using inorganic agent. *Journal of Electroceramics*, 21, 357-360.
15. Pechini, M.P. (1967). Method of preparing lead and alkaline earth titanates and niobates and coating method using the same to form a capacitor. U.S. Patent 3,330,697.

16. Junliang, L., Yanwei, Z., Cuijing, G., Wei, Z., & Xiaowei, Y. (2010). One-step synthesis of barium hexaferrite nano-powders via microwave-assisted sol-gel auto-combustion. *Journal of the European Ceramic Society*, 30(4), 993-997.
17. Pradeep, A., Priyadharsini, P., & Chandrasekaran, G. (2008). Sol-gel route of synthesis of nanoparticles of $MgFe_2O_4$ and XRD, FTIR and VSM study. *Journal of Magnetism and Magnetic Materials*, 320(21), 2774-2779.
18. Mazaleyrat, F., & Varga, L.K. (2000). Ferromagnetic nanocomposites. *Journal of Magnetism and Magnetic Materials*, 215-216, 253-259.
19. Sharma, V., Kumari, S., & Kuanr, B.K. (2018). Rare earth doped M-Type hexaferrites: Ferromagnetic resonance and magnetization dynamics. *AIP Advances*, 8(5), 056232.
20. Kharisov, B.I., Dias, H.V.R., & Kharissova, O.V. (2019). Mini-review: Ferrite nanoparticles in the catalysis. *Arabian Journal of Chemistry*, 12(7), 1234-1246.
21. Cullity, B.D., & Stock, S.R. (2001). *Elements of X-ray Diffraction* (3rd ed.). Prentice Hall.