SYNTHESIS OF COBALT AND COPPER NANOFERRITES BY A SOL-GEL METHOD USING PLANTS AND THEIR CATALYTIC ACTIVITY

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Abstract. *This brief review presents an analysis of data from the past five years on the "green" synthesis and catalytic activity of cobalt and copper nanoferrites, which are characterized by a wide range of applications. Due to their large reserves, availability, and abundance, plants are the best bioagents for the sol-gel synthesis of ferrites, which utilizes plant extracts along with precursors. Extracts can be obtained from leaves, fruits, roots, seeds, and flowers of plants, which contain polysaccharides, carbohydrates, phenols, flavonoids, terpenoids, and amino acids, which can act as chelating, reducing, and stabilizing agents for ferrite nanoparticles. Existing literature reviews examine the synthesis and application of green nanoferrites for water purification, heavy metal removal, their antibacterial activity, photocatalytic activity in the degradation of various dyes, and their potential use in biological processes. In contrast, this brief review focuses on a comparative analysis of methods for obtaining extracts from various plants, as well as the processing conditions of precursor and extract mixtures for ferrite synthesis. The influence of extract concentration and heat treatment method on the properties of the synthesized ferrites and nanoparticle size is examined. Examples of the few catalytic reactions involving nanoferrites synthesized by the sol-gel method using plant extracts are provided. The development of new nanoferrite-based catalysts using green chemistry methods, using plants, is a promising direction for the synthesis of various catalytic systems, both from an environmental and economic perspective.*

Keywords: *green synthesis, cobalt and copper nanoferrites, sol-gel method*
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1. Introduction

Green chemistry is a new and important scientific field in chemistry that involves conducting chemical processes with minimal harm to the environment. One branch of green chemistry is green synthesis, in which plant extracts are used alongside precursors rather than conventional toxic reagents. Since catalysts play an extremely important role in the chemical industry, and 90% of modern chemical production is based on catalytic processes, the application of green chemistry principles to catalyst production is highly relevant.

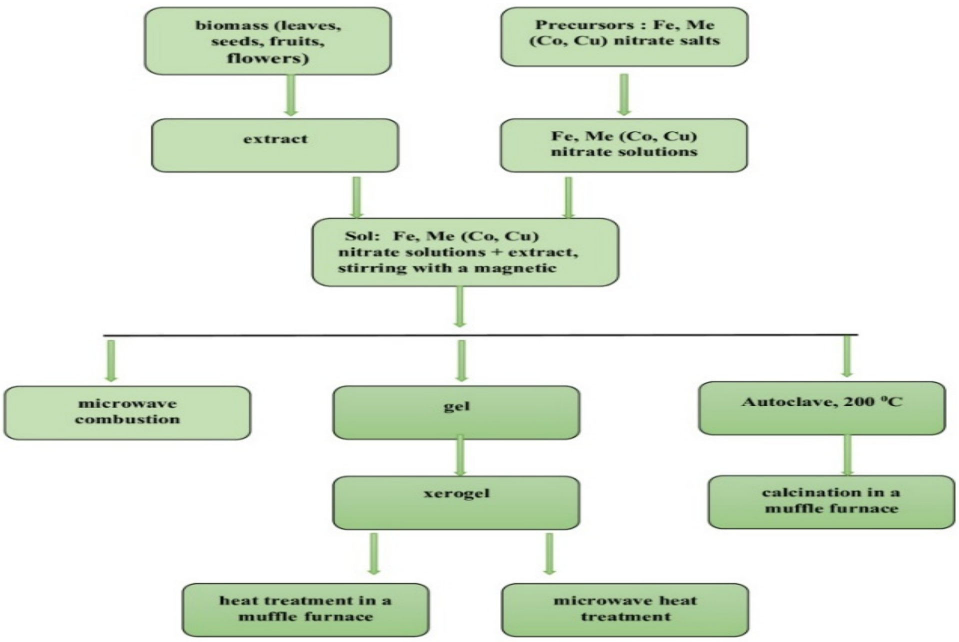
One of the promising green chemistry approaches for catalyst synthesis is the sol–gel method, including its autocatalytic variant, which exploits the energy released by exothermic reactions. The main advantages of this method include low energy consumption, short synthesis time, and the possibility of simultaneous conversion of precursors into the final product using the energy of the chemical reaction. Various compounds, such as organic acids and alcohols, are used as complexing agents and fuels in this method. Given that plant seeds, leaves, and extracts contain flavonoids, terpenoids, alcohols, carbohydrates, sugars, polyphenols, and amino acids that act as reducing and stabilizing agents, they can serve as alternatives to synthetic organic reagents, thereby expanding the potential of the green sol–gel method for catalyst preparation.

This article provides a brief overview of the literature data from the last 5 years on the green sol-gel synthesis of cobalt and copper ferrites using various plants. The interest in ferrites is due to their wide range of applications. They are widely used in radio engineering, electronics, and computing for the purification of wastewater from heavy metals and organic dyes, as well as in medicine. Therefore, it is no coincidence that researchers are turning to green synthesis of ferrites. The paper [1] provides an overview of current trends in the field of environmentally friendly synthesis of ferrite nanoparticles and their promising applications. Review [2] discusses biomedical applications of green spinel ferrites. Works [3-4] also consider green cobalt ferrite nanoparticles as a new material for biomedical and environmental applications. The review [5] examines the possibilities of using green nanoferrites for water purification, removal of heavy metals, their antibacterial activity, and potential use in biological processes.

As for the catalytic properties of ferrites, ferrite-based catalytic systems are active in processes such as oxidative dehydration of hydrocarbons, aldol reactions, alkylation and dehydrogenation reactions, synthesis of various organic compounds, decomposition of alcohols and hydrogen peroxide, purification of automobile exhaust gases, oxidation of various compounds, including alkenes, etc. [6]. Cobalt, copper, nickel, and zinc ferrites, as well as their combinations with Cr, Cd, Mn, and some lanthanides, are mainly used as catalysts. Green synthesis of nanoferrites further broadens their application potential in the aforementioned reactions from an environmental perspective.

This brief review discusses reactions catalyzed by cobalt and copper nanoferrites obtained via green sol–gel synthesis. The development of new nanoferrite-based catalysts using plant-mediated green chemistry approaches represents a promising direction for creating diverse catalytic systems, offering both environmental and economic advantages due to the renewable nature of these resources.

2. General method for obtaining nanoferrites using the green sol-gel method. The general methodology for obtaining ferrites using the green sol-gel method consists of selecting plant material (seeds, leaves, fruits, flowers, plant resins, etc.), obtaining an extract, and using it in various concentrations as a stabilizing and reducing agent. To synthesize ferrite, the extract is mixed with aqueous solutions of iron nitrates and the corresponding metal. There are then two options: centrifuging the extract and placing the precipitate in a muffle furnace for calcination at a high temperature for several hours, or slowly evaporating it to obtain a gel, which ignites when dried in an oven, and due to the heat of the exothermic reaction, ferritization occurs in a very short time and at a lower temperature, after which additional heat treatment is carried out at 500-700 °C. Microwave technology can also be used for gel combustion and heat treatment of the residue. Below is a general scheme for the synthesis of ferrites using plants:



2.1. Preparation of plant extracts. Analysis of the literature showed that authors mainly perform extraction with distilled water, but there is no single methodology (amount of plants, water, temperature), and the temperatures of subsequent heat treatment also vary. Given the complex phytochemical composition of plants, the conditions for extracting organic components from them are important, since, as noted above, these components are complexing and stabilizing agents for nanoparticles. Therefore, we decided to examine these issues in detail.

Various authors have used *Eucalyptus leaves* [7], Olive leaves [8], *Hibiscus rosa-*sinensis leaves [9], *Áloë véra* leaves [10], *Barleria lupulina leaves* [11], *Erythrina variegata* leaves [12], *Dillenia indica fruit* [13], *passion fruit* [14], *grape skin and pulp* [15], *coconut water* [16], *cashew gum* [17], and, for the synthesis of copper ferrite, *asturtium leaves* [18], *Corchorus Olitorius* [19], *chlorella green microalgae biomass* [20], *Moringa oleifera* [21], *Morus alba L* [22], and *Zingiber officinale and cardamom seed* [23].

In study [**7**], *eucalyptus leaf* extract was prepared by heating dry eucalyptus leaves (2.5 g) with 100 ml of distilled water at 80 °C on a magnetic stirrer with stirring for 30 minutes, cooling, and subsequent filtration.

The *olive leaf* extract was prepared by adding 200 ml of ethanol and 200 ml of distilled water to 5 g of leaves, stirring for 3 hours at 60 °C, and filtering [8].

To obtain 100 mg of hibiscus *rosa-sinensis* leaf extract, the leaves were mixed with 100 ml of distilled water and then stirred while heating and filtered, and the filtrate was used for ferrite synthesis [9].

Aloe vera leaf extract was prepared from air-dried leaves of the plant. To 20 g of finely chopped leaves, 200 ml of distilled water was added, and the mixture was boiled for 15 minutes at 80 °C. After cooling to 37 °C, the mixture was centrifuged for 20 minutes, and then the filtrate was further filtered [10].

Barleria Lupulina leaf extract was prepared by heating 200 ml of distilled water with 10 g of crushed leaves at a temperature of 40 °C with stirring for 1 hour and subsequent filtration of the solution [11].

The authors [12**]** used *Erythrina variegata* leaf extract to prepare cobalt ferrite. To obtain the extract, 20 g of leaves were boiled in 200 ml of distilled water and then filtered.

In study [13], *Dillenia indica* fruit was used to prepare the extract. The extraction was performed in an Erlenmeyer flask using 10 g of finely chopped fruit and pulp mixed with 200 mL of distilled water. The mixture was boiled for 10 min and subsequently filtered.

Cobalt ferrite nanoparticles were obtained using grape skin and pulp extracts in work [15]. The extracts were prepared by boiling grape skins/pulp for 15 minutes, leaving the mixture overnight, and filtering it the next day.

In work [18**]**, copper ferrite nanostructures were synthesized using *nasturtium* extract. For each gram of dry plant powder, 20 ml of distilled water was added and mixed at 27 °C, then left overnight. The next day, the extract was separated by centrifugation and filtration.

To prepare the *Moringa oleifera* leaf extract, 10 g of crushed leaves were mixed with 100 ml of distilled water, and then the aqueous extract was boiled for 10 minutes at a temperature of 80ºC. The plant extract was left to cool at room temperature and then filtered [21].

When using *cashew gum* (Anacardium occidentale, Brazil) for the green synthesis of cobalt ferrite, a starting solution of 43.8 g of gum in 200 ml of distilled water was prepared. The solution was homogenized with intensive mechanical stirring for 24 hours [17].

When using *ginger root* extract and *cardamom seeds* for the synthesis of copper-substituted cobalt ferrites, 10 g of ginger roots were crushed, 50 ml of water was added, and the mixture was boiled for 10 minutes, after which it was cooled and filtered. Five grams of cardamom seeds were ground into powder, 50 ml of distilled water was added, and the mixture was stirred while heating for 4 hours and then filtered [23].

In addition to leaves and seeds, some authors use fruit waste for green synthesis. For example, in work [14], the authors use citric acid isolated from rotten *passion fruit* as a complexing reagent and “fuel” in the synthesis of cobalt ferrite.

2.2. Green synthesis of cobalt nanoferrite: synthesis conditions and heat treatment. The synthesis of cobalt ferrite nanoparticles using *eucalyptus* leaf extract was carried out by adding a calculated amount of iron (III) and cobalt (II) nitrates to it while stirring and adding a 25% NH_4OH solution to pH = 10 [7]. The resulting precipitate was separated, washed, dried, and calcined at 800 °C for 1 hour. Thermal analysis of the sample dried before calcination showed that the carbon residue of the eucalyptus extract is removed between 320 and 700 °C, with a weight loss of ~2.5%. The results of the X-ray diffraction method (XRD) analysis of the sample calcined at 800 °C indicated the formation of a $CoFe_2O_4$ spinel structure.

When different concentrations of olive leaf extract were used for the synthesis of cobalt ferrite [8], sucrose was added to the mixture of metal nitrates and extract as a gelling agent. The resulting gel was dried and calcined at 500, 600, and 700 °C for 3 h. X-ray diffraction (XRD) analysis showed that the extract concentration did not affect the crystal structure of the obtained cobalt ferrite samples and that no impurity phases were detected. All diffraction patterns corresponded to the cubic cobalt ferrite structure with the space group *Fd*-3*m*. However, the amount of extract influenced the crystallite size: higher extract concentrations led to the formation of smaller crystallites.

To synthesize $CoFe_2O_4$ nanoparticles using *Hibiscus rosa-sinensis* leaf extract, the authors [9] took a molar ratio (1:1) of $Co(NO_3)_2{\cdot}9H_20$ and $Fe(NO_3)_3{\cdot}6H_20$ salts. Ten milliliters of extract were added to the mixed nitrate solution and stirred for 1 hour at a temperature of 60°C. The solution was then transferred to an autoclave and heated to 200 °C for 6 hours, after which the samples were filtered, washed, dried, and calcined at 300 °C for 2 hours. The crystallite size of the obtained spinel nanoparticles was 24 nm.

In [11], 30 ml of *Barleria Lupulina* leaf extract were added to 20 ml of a mixture of 10 ml of 0.05 M $Co(NO_3)_2{\cdot}6H_2O$ and 10 ml of 0.1 M $Fe(NO_3)_3{\cdot}9H_2O$ solutions with vigorous stirring for 2 hours. The temperature was then raised to 80 °C and heated until the water was completely removed. The resulting residue was calcined at 600 °C for 4 hours in a muffle furnace. XRD analysis of the sample showed the formation of cobalt ferrite nanoparticles with a nanoparticle size of 30.07 nm.

For the green synthesis of cobalt ferrite, various concentrations (10, 20, 30, and 40 ml) of the extract of *Erythrina variegata* leaves [12] were added dropwise to a solution of metal nitrates (0.1 mol Co^{+2} and 0.2 mol Fe $^{3+}$) with constant stirring. The solution was dried for 3 h at 100 ◦C, and the resulting powder was placed in a muffle furnace at 900 ◦C for 3 h for calcination. The formation of the crystal structure of cubic spinel $CoFe_2O_4$ nanoparticles was determined using Powder X-ray Diffraction (PXRD) analysis.

In the work [13], $CoFe_2O_4$ was synthesized using the extract of the fruits and pulp of *Dillenia indica*, adding 30 ml of 0.16 M $CoCl_2$ and 30 ml of 0.23 M $FeSO_4{\cdot}7H_2O$ to 50 ml of the extract and stirring at 40°C for 1 h. Then 15 ml of 30% NH_4OH solution was added to the solution. The resulting solution with the precipitate was centrifuged, and the precipitate was separated, which, after drying, was annealed at 300 °C for 2 h. The formation of ferrite nanoparticles was confirmed by XRD, transmission electron microscopy (TEM), and X-ray photoelectron spectroscopy (XPS) analyses. In the scanning electron microscopy (SEM) images of the samples obtained without using the extract, nanoparticles with an irregular shape and slight agglomeration were observed, and in the images of the samples synthesized with the extract, spherical particles with slight agglomeration were observed.

Cobalt ferrite nanoparticles were synthesized using *grape skin* and *pulp* extracts by a sol-gel combustion method [15] An aqueous solution of Co(+2) and Fe(+3) nitrate salts was heated to 65°C, and the extract was added dropwise. The solution was heated for 25 min to evaporate and cause autoignition. After the autocombustion stage, the resulting powders were washed with hot distilled water and annealed at 400°C for 2 h. X-ray diffraction showed that all samples contained pure $CoFe_2O_4$ with a cubic spinel structure and the Fd3m space group. The size of nanoparticles synthesized using grape skin extract was ~5 nm, and the size of nanoparticles synthesized with grape pulp extract was 25 nm.

In work [16], 47.90 g of *coconut water* powder dissolved in 500 ml of water was used for the synthesis of cobalt ferrite. Cobalt and iron nitrates were taken in a ratio of Fe:Co = 1:1. The nitrate and coconut water solutions were stirred on a magnetic stirrer at t=80°C for 2 hours and t=100°C for 1 hour to obtain a high-viscosity gel. The gel was then heat treated at 350 °C in an oven for one hour to remove solvents and obtain a dry powder. X-ray diffraction and Raman spectroscopy confirmed the presence of a pure $CoFe_2O_4$ phase in the sample treated at 1100 °C, with an average grain size of 542 nm.

In the work [17], nitrates of these metals and 20 ml of natural *cashew gum* extract were used as precursors to prepare cobalt and nickel ferrites with a molar ratio of Fe:Me(Co, Ni)=2:1. After homogenization of the mixtures for 2 h at 300 K, a gel was formed, which was subjected to heat treatment for 1 h at 773 K. X-ray diffraction results showed the formation of single-phase nanoparticles with the Fd-3m space group and a crystallite size of 7.4 and 6.0 nm for cobalt ferrite and nickel ferrite, respectively.

In the synthesis of cobalt ferrite using citric acid isolated from *passion fruit*, cobalt-iron citrate was first obtained by coprecipitation, followed by pyrolysis in air at 850 °C. X-ray diffraction revealed the formation of $CoFe_2O_4$.

In the work [23], cobalt ferrite nanoparticles $Cu_xCo_{1-x}Fe_2O_4$ (x = 0.0; 0.3; 0.6; 0.9) were synthesized using extracts of *ginge*r and *cardamom seeds*. For this purpose, an NaOH solution was added dropwise to a mixed solution of nitrates with ginger root and cardamom extracts until pH = 12 was established with stirring. The solution was then placed in an autoclave and heated in an oven at 160 °C for 12 hours. After cooling and filtering the precipitate, it was washed with distilled water and dried at 80 °C for 1 hour. The resulting ferrite nanoparticles had a cubic spinel structure with a size of 7-45 nm (according to the Debye-Scherrer formula). Scanning electron microscopy (SEM) analysis revealed that the spherical nanoparticles with a diameter of 13 nm for unsubstituted cobalt ferrite transform into a hexagonal structure with a size of 46 nm at a Cu concentration of (x = 0.9).

2.3. Green synthesis of copper ferrite. In the work [18], spinel nanostructures of copper ferrite ($CuFe_2O_4$) were synthesized using an extract of *Nasturtium officinale*. For this purpose, 3.2 g $FeCl_3 \cdot 6H_2O$ and 1.6 g $CuCl_2 \cdot 2H_2O$ were added to 200 ml of an aqueous extract of nasturtium at 60 °C with stirring, and the pH of the mixture was increased to 10 by dropwise adding 1 M NaOH solution. The final solution was stirred for 2 h at 60 °C. The resulting nanoparticles were washed with deionized water, dried at 60 °C, and calcined at 500 °C for 8 h.

In the article [20], the green synthesis of $CuFe_2O_4$ nanoparticles was carried out by a hydrothermal method using the biomass extract of *green microalgae Chlorella*. The results of X-ray diffraction analysis showed that copper ferrite nanoparticles synthesized with the microalgae biomass extract and NaOH at pH 12 and 180 °C for 8 hours formed face-centered cubic structures with the space group Fd3m, with a nanoparticle size of 24 nm.

When using *Moringa oleifera* extract as a reducing and stabilizing agent for the synthesis of $Cu_xZn_{1-x}Fe_2O_4$ ferrite nanoparticles (where x = 0.6 and 0.7), precursors ($CuSO_4 \cdot 5H_2O$) (0.1N), ($ZnSO_4 \cdot 7H_2O$) (0.1N), and ($FeSO_4 \cdot 7H_2O$) (0.1N) were used by stirring at 80ºC for 20-30 minutes [20-21]. After this, the extract was added to the solution, followed by a dropwise addition of 20 ml of NaOH (0.1 N). The resulting precipitate was filtered, washed with distilled water, and air-dried. The samples were then heated in a furnace at 150 °C for 4 hours and sintered at 400 °C for 4 hours. X-ray diffraction analysis revealed the crystalline nature of the resulting ferrites. The particle size of the $Cu_{0.6}Zn_{0.4}Fe_2O_4$ sample ranged from 9.11 nm to 20.96 nm, while for the $Cu_{0.7}Zn_{0.3}Fe_2O_4$ sample, the particle size was 10.92 nm to 15.41 nm.

In work [22], $CuFe_2O_4$ nanoparticles were obtained using green synthesis with an extract of Morus *Alba L.* leaves. X-ray diffraction data confirm the formation of a CuFeO phase with a cubic space group Fd-3m. The nanoparticles have a spherical shape with a distributed particle size in the range of 20–70 nm.

The authors of Ref. [19] reported a green synthesis of copper and cobalt ferrites using *Corchorus olitorius* (jute mallow) via a so-called “dry method.” In this approach, powdered dried leaves of *Corchorus olitorius* served as a fuel for the combustion process. The metal precursor composition corresponded to a stoichiometric Fe/M ratio of 2 (M = Cu, Co). Equimolar amounts of metal nitrates were mixed with 0.5 g of the plant powder, thoroughly ground in a crucible, and heated at 60 °C to remove residual moisture and increase the viscosity of the precursor gel. Subsequent heating at 300 °C initiated self-combustion of the gel, leading to the formation of the ferrite products. Additional calcination at 700 °C significantly enhanced the crystallinity of both Co and Cu ferrite nanoparticles. Surface morphological analysis by SEM/EDS and transmission electron microscopy (TEM) confirmed the formation of uniform, nanosized ferrite particles.

Copper ferrite stabilized on clinoptilolite was prepared using green tea (Camellia sinensis) leaf extract [24]. After fixation on clinoptilolite, the copper ferrite crystallite size was confirmed by Field Emission Scanning electron microscopy (FESEM) and TEM images to be 48.7 nm, the average pore diameter of the catalyst was 22.2 nm, and the BET surface area of the catalyst was estimated to be 43.7 m^2/g.

Green sol-gel synthesis has also been used to produce nanocomposites, such as $Fe_2O_3/CuFe_2O_4$/chitosan [25]. Initially, nanostructures of copper ferrite $CuFe_2O_4$ and iron oxide Fe_2O_3 were prepared by the sol-gel method using autocombustion, using onion as a green reducing agent. Chitosan was then added to the nanostructures dispersed in water. Chitosan was used to improve surface properties and enhance metal ion adsorption.

2.4. Use of animal products for the synthesis of ferrites. In addition to plants, some studies use animal products as fuel for the synthesis of ferrites using the "dry" method. Thus, in work [26], *cottage cheese* was used to obtain cobalt ferrite nanoparticles. Metal nitrates were ground with cottage cheese until a homogeneous mixture was obtained. The mixture was held in a preheated muffle furnace at 500°C for approximately 5 minutes. The resulting powder was calcined at 650°C for 5 hours. The X-ray diffraction pattern of samples calcined at 650°C shows the formation of a cubic spinel structure of $CoFe_2O_4$ with the Fd3m space group. No secondary phase was observed in the sample.

In [27], egg white was used to synthesize copper ferrite. Egg white and distilled water were mixed in a 3:1 ratio at room temperature for an hour to obtain a homogeneous solution. $Cu(NO_3)_2{\bullet}6H_2O$ and $Fe(NO_3)_3{\bullet}9H_2O$ salts were added to the solution in a 1:2 molar ratio and stirred at room temperature for four hours. The solution was then dried by heating at 80°C. The resulting powder was calcined for three hours at 600°C to obtain the final product. According to X-ray diffraction data, the $CuFe_2O_4$ nanoparticles have a cubic spinel structure with a particle size of 37.3 nm.

An analysis of the literature on the green synthesis of ferrites indicates *several common features and methodological variations*:

- First, during the preparation of plant extracts, different ratios of plant material to water or alcohol are employed, along with varying extraction times and temperatures. According to published data, extraction is typically carried out at temperatures between 40 and 80 °C, with extraction times ranging from 10–15 min to 3 h. In some studies, the obtained extracts were left to stand overnight prior to filtration.
- Second, ferrite synthesis is generally performed by vigorously mixing the appropriate metal salts with the plant extract under heating at 60–80 °C. Subsequently, several synthesis routes are reported in the literature:
  (a) continuous stirring of the reaction mixture until gel formation, followed by thermal treatment; in some cases, gelling agents such as sucrose are added to promote gel formation;
  (b) gradual evaporation of the solvent by slow heating until complete water removal, followed by heat treatment of the resulting solid residue;
  (c) addition of a NaOH solution to the mixture of metal precursors and plant extract to induce precipitation, followed by filtration, washing with distilled water, drying, and subsequent heat treatment;
  (d) hydrothermal treatment of the precursor–extract mixture in an autoclave, followed by filtration and final heat treatment.
- The heat-treatment conditions reported in the literature vary widely in both temperature and duration. Calcination temperatures range from 300 to 1100 °C, while the treatment time typically spans from 1 to 8 h, depending on the synthesis route and the desired phase purity and

crystallinity. In addition to extract-based approaches, ferrites can also be synthesized via a dry method, in which plant biomass is directly used as a fuel without prior extraction. In this case, the combustion of a mixture of metal precursors and plant material is followed by an additional heat treatment, usually performed at temperatures of 500–650 °C.

- The combustion of gels or precursor solutions can be achieved either by conventional thermal heating or by microwave irradiation. For example, cobalt ferrite was synthesized using okra extract by placing a solution containing metal nitrates and the plant extract in a quartz crucible and exposing it to microwave radiation at a frequency of 2.45 GHz and a magnetron output power of 850 W for 15 min [28]. X-ray diffraction analysis demonstrated that both conventional and microwave heating led to the formation of single-phase cobalt ferrite, with average crystallite sizes of 47 and 55 nm, respectively. A similar green microwave-assisted synthesis of cobalt ferrite using *Opuntia dillenii* extract was reported in Ref. [29], where X-ray diffraction confirmed the formation of single-phase crystalline $CoFe_2O_4$ nanoparticles. Scanning electron microscopy revealed predominantly spherical particles with sizes ranging from 40 to 200 nm. The relatively large particle size observed in some samples (up to 200 nm) is likely associated with the microwave processing parameters, particularly prolonged irradiation times and high magnetron power. Such conditions promote particle sintering, leading to a reduction in specific surface area, in agreement with previously reported results [30, 31].
- In many studies, authors report the phytochemical composition of the plants employed, often supported by relevant literature data. However, owing to the complex and variable chemical nature of plant materials, comprehensive phytochemical analyses are not always performed. In this context, Ref. [12] is noteworthy, as it presents a detailed phytochemical characterization of *Erythrina variegata* leaf extract used for the green synthesis of cobalt ferrite. The extract was analyzed for the presence of alkaloids, flavonoids, steroids, terpenoids, saponins, anthraquinones, coumarins, glycosides, tannins, and phenolic compounds. Similarly, a phytochemical analysis of *Morus alba* L. leaves revealed the presence of alkaloids, flavonoids, and saponins [22].
- Furthermore, thermal analysis aimed at identifying residual plant-derived species and determining ferritization temperatures is not systematically carried out in all studies. This omission is particularly critical for ferrites synthesized and calcined at relatively low temperatures (300–500 °C), where incomplete phase formation or residual organic matter may persist. From this standpoint, studies reported in Refs. [7, 12, 16] are of particular interest, as they include thermal analysis data that provide insight into decomposition processes and ferrite formation mechanisms.
- Nevertheless, in the vast majority of studies, irrespective of the phytochemical composition of the plant source, extraction conditions, synthesis route, or subsequent heat-treatment parameters, X-ray diffraction analysis consistently indicates the formation of cubic nanoferrites with the spinel structure (space group $Fd\overline{3}m$), without the detection of secondary phases.
- At the same time, it should be emphasized that the formation of single-phase ferrites is thermodynamically feasible only when the initial metal ratio strictly corresponds to the stoichiometric composition, i.e., Fe:Me=2:1(Me = Co, Cu). In the case of non-stoichiometric metal ratios, the formation of secondary oxide phases is unavoidable. From this perspective, the results reported in Refs. [9,16], where cobalt and copper ferrites were synthesized using an initial metal ratio of

Fe:Co(Cu) = 1:1 and the authors claimed, based solely on X-ray diffraction data, the formation of pure ferrite phases, should be interpreted with caution. Under such conditions, the coexistence of cobalt or copper oxides is expected, even if these phases are not clearly resolved in the XRD patterns.

3. Catalytic Application of Ferrites. However, as noted above, given the important role catalysts play in the chemical industry, it is important to extend the principles of green chemistry to the production of catalysts for various processes. Currently, the use of green ferrites in catalytic processes is limited. Let's review some recent studies devoted to catalysis using green ferrites.

Thus, in the work [32], the authors investigated green $CuFe_2O_4$ nanoparticles (particle size 60-75 nm) obtained using gum tragacanth as a catalyst for the selective oxidation of alcohols to aldehydes using oxone (potassium hydropersulfate) as an oxidizing agent in the presence of acetonitrile as a solvent at 40 ºC. The oxidation of benzyl alcohol and its halogen derivatives, as well as aliphatic alcohols (2-butanol and isobutanol), was studied. The optimal catalyst loading was 7 mol% per 1 mmol of alcohol. The oxidation time for aliphatic alcohols was about 120 minutes with an aldehyde yield of 98.6-99%. The oxidation time for benzyl alcohol and its halogen derivatives was slightly shorter, at 90-95 minutes, with a corresponding aldehyde yield of 81-89%. The catalyst was easily separated from the products by applying an external magnet to the reaction vessel. The authors reused the catalyst in five subsequent cycles, with yields of 86, 86, 85, 84, and 84%, respectively, indicating stable catalyst performance without a decrease in activity.

In Ref. [13], cobalt ferrite nanoparticles synthesized using *Dillenia indica* fruit extract were used for the first time for the facile ipso-nitration and ipso-hydroxylation of arylboronic acids for the cheaper and more efficient production of nitroarenes and phenols of pharmaceutical and industrial importance. The authors note that the catalyst is easily recovered and can be reused for up to five catalytic cycles without loss of activity.

The study of cobalt ferrite synthesized using grape skin extract showed that cobalt ferrite with a 5 nm nanoparticle size is an active catalyst for the decomposition of hydrogen peroxide [15]. H_2O_2 decomposes by 97.4% on this catalyst. A study of the catalyst surface morphology after catalytic decomposition of H_2O_2 showed that it remains unchanged. The authors believe that cobalt nanoferrites synthesized using green methods can be used in Fenton processes as catalysts. As is well known, the Fenton reaction is a reaction between hydrogen peroxide (H_2O_2) and iron ions (Fe^{2+}) to form hydroxyl radicals (•OH), which are highly reactive and can oxidize various organic compounds. Given the important practical application of this reaction for the purification of wastewater and groundwater, as well as soil, from organic pollutants (various dyes, solvents, phenols, pesticides, etc.), the development of green nanoferrites opens up the prospect of making the entire process, from catalyst synthesis to the purification of the listed objects, completely environmentally friendly.

In study [33], extracts from tomato fruit, persimmon, flax seeds, and ginkgo biloba leaves were used to compare the effect of the nature of the plant material used in the synthesis of cobalt ferrite on its catalytic activity in the decomposition of hydrogen peroxide. The catalysts were obtained by the sol-gel method with combustion. Testing of the catalysts showed that cobalt ferrite obtained using persimmon and tomato extract exhibits higher catalytic activity than cobalt ferrite obtained with flax extract. In the presence of catalysts synthesized with tomato and persimmon extract, H_2O_2 with an initial concentration of 100 mM decomposes by 90% in 1 hour. Based on experimental data, the authors believe that the type of extract used affects the structure and surface characteristics of $CoFe_2O_4$ samples and, thus, their catalytic properties.

The work [24] investigates the catalytic activity of copper nanoferrite on clinoptilolite in the production of benzodiazepine (the reaction of o-phenylenediamine with ketones). Copper nanoferrite was obtained using green tea leaf extract (Camellia sinensis). After fixation on clinoptilolite, the size of copper ferrite crystallites was determined to be 48.7 nm using the Debye-Sherrer equation. The average pore diameter of the catalyst was 22.2 nm, and the surface area was 43.7 m²/g. The experimental results showed that the reaction of o-phenylenediamine with ketones on the catalyst proceeds under mild conditions and is easily separated from the reaction product, benzodiazepine, using a magnet, and can also be reused without a significant decrease in its catalytic activity.

In the study [22], the catalytic properties of copper nanoferrite $CuFe_2O_4$, synthesized using mulberry leaves, were investigated in the Mannich reaction. The green copper ferrite demonstrated good catalytic ability, with the yield of the naphthoquinone derivative being 83.83%.

As can be seen from the examples provided, the use of green ferrites for catalytic purposes is limited. However, these studies open up new prospects for the green synthesis of various nanoscale catalysts and their application in catalytic processes.

Conclusions

The vast diversity of plant species—numbering in the tens to hundreds of thousands—and their renewable nature provide promising opportunities for advancing environmentally benign approaches in chemical synthesis. The present literature review demonstrates that leaf extracts, as well as plant fruits, roots, and seeds, can be effectively employed in the green sol–gel synthesis of cobalt and copper ferrites. Irrespective of extraction conditions or the specific phytochemical composition of the plant precursors, the reported studies consistently indicate the formation of nanoferrites with a cubic spinel structure. This observation significantly broadens the prospects for applying green sol-gel routes not only to ferrite materials but also to other functional and catalytic systems. Moreover, the integration of green sol-gel synthesis with microwave-assisted processing enables a substantial reduction in thermal treatment time, further supporting compliance with the principles of green chemistry.

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