

Approaches on Ferrofluid Synthesis and Applications: Current Status and Future Perspectives

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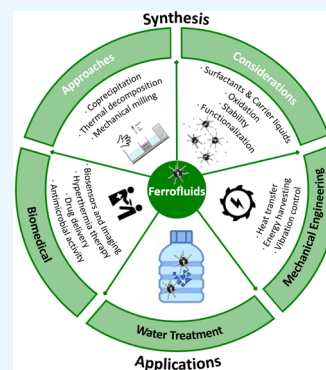
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ABSTRACT: Ferrofluids are colloidal suspensions of iron oxide nanoparticles (IONPs) within aqueous or nonaqueous liquids that exhibit strong magnetic properties. These magnetic properties allow ferrofluids to be manipulated and controlled when exposed to magnetic fields. This review aims to provide the current scope and research opportunities regarding the methods of synthesis of nanoparticles, surfactants, and carrier liquids for ferrofluid production, along with the rheology and applications of ferrofluids within the fields of medicine, water treatment, and mechanical engineering. A ferrofluid is composed of IONPs, a surfactant that coats the magnetic IONPs to prevent agglomeration, and a carrier liquid that suspends the IONPs. Coprecipitation and thermal decomposition are the main methods used for the synthesis of IONPs. Despite the fact that thermal decomposition provides precise control on the nanoparticle size, coprecipitation is the most used method, even when the oxidation of iron can occur. This oxidation alters the ratio of maghemite/magnetite, influencing the magnetic properties of ferrofluids. Strategies to overcome iron oxidation have been proposed, such as the use of an inert atmosphere, adjusting the Fe(II) and Fe(III) ratio to 1:2, and the exploration of other metals with the oxidation state +2. Surfactants and carrier liquids are chosen according to the ferrofluid application to ensure stability. Hence, a compatible carrier liquid (polar or nonpolar) is selected, and then, a surfactant, mainly a polymer, is embedded in the IONPs, providing a steric barrier. Due to the variety of surfactants and carrier liquids, the rheological properties of ferrofluids are an important response variable evaluated when synthesizing ferrofluids. There are many reported applications of ferrofluids, including biosensing, medical imaging, medicinal therapy, magnetic nanoemulsions, and magnetic impedance. Other applications include water treatment, energy harvesting and transfer, and vibration control. To progress from synthesis to applications, research is still ongoing to ensure control of the ferrofluids' properties.



1. INTRODUCTION

Ferrofluids are colloidal liquids possessing strong magnetic features whose physical properties can be altered or controlled when exposed to a magnetic field.^{1,2} They are fascinating materials with a wide range of applications, including magnetic resonance imaging (MRI),^{3,4} dynamic loudspeakers,⁵ magneto-optic sensors,⁶ heat transfer/dissipation,⁷ and more. The diverse applications of ferrofluids arise from the unique properties modulated by the nanoparticle (NP) characteristics and the composition of the carrier liquids and surfactants.⁸ This article aims to provide a general perspective on the commonly reported methods of synthesis of ferrofluids, an overview of the future challenges and components that comprise ferrofluids, and the most reported and potential ferrofluid applications.

Magnetic fluids are suspensions of fine particles of a ferromagnetic material in a carrier liquid.⁹ Fluids with magnetic properties are composed of two fluid types, magnetorheological fluids (MR fluids) and ferrofluids. Both are composed of a magnetic solid such as magnetite (Fe_3O_4), a surfactant that prevents agglomeration, and a carrier liquid that suspends the particles. The primary distinction between these two types of magnetic fluids is the size of the magnetic particles that are

suspended within the carrier liquid. MR fluids incorporate micron-sized particles for suspension,¹⁰ while ferrofluids integrate particles in the lower nanometer range.¹¹ The smaller particle size in ferrofluids results in higher suspension stability, which produces an extended suspension time.¹² The enhanced stability makes ferrofluids ideal for many applications and opens up opportunities that could not be achieved with MR fluids. Furthermore, magnetic fluids can be distinguished based on visual examination when placed in a magnetic field. When MR fluids are in the presence of a magnetic field, the ferromagnetic particles undergo dipole interactions and form chains along the flux paths.¹⁰ The formation of chains within MR fluids restricts the movement of the carrier liquid, which can result in a viscosity increase of up to 10^3 times,¹³ as shown in Figure 1a.

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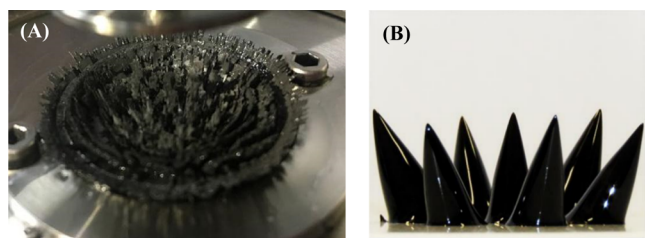


Figure 1. Characteristic pattern of spikes of a (A) MR fluid and (B) ferrofluid under a magnetic field. Photograph A: courtesy of Wang et al.¹⁴ (free domain). Copyright 2021, Wang, H.; Bi, C.; Zhang, Y.; Zhang, L.; Zhou, F. Photograph B: courtesy of Priyananda, et al.¹⁵ Copyright 2018, American Chemical Society.

Alternatively, ferrofluids experience a Rosensweig instability pattern of peaks and valleys (spikes). These spikes are highly ordered self-organized structures. Furthermore, it has been reported that a ferrofluid placed under a magnetic field for over 35 years has kept its liquid state with no sign of sedimentation, showing the particular pattern of spikes and meaningful stability (Figure 1b).¹⁴

Research in ferrofluid synthesis has considerably increased in the past 50 years. While a large number of research articles published in the early years might not be available online, there has been a large increase in publications in the last 20 years. Figure 2 shows the results of a simple search in Google Scholar using the words “synthesis of ferrofluids”, which reflects the growing interest in the synthesis and applications of these magnetic fluids. There is a record of the publication of 1500% more articles in the synthesis of ferrofluids during the years 2001–2021 compared to those during 1981–2000. Moreover, according to the best of the authors’ knowledge, there are no current reviews that summarize the methods of synthesis and describe the applications in a straightforward approach that facilitates building a general idea of how ferrofluids are produced and highlights how research in ferrofluids has evolved over the years.

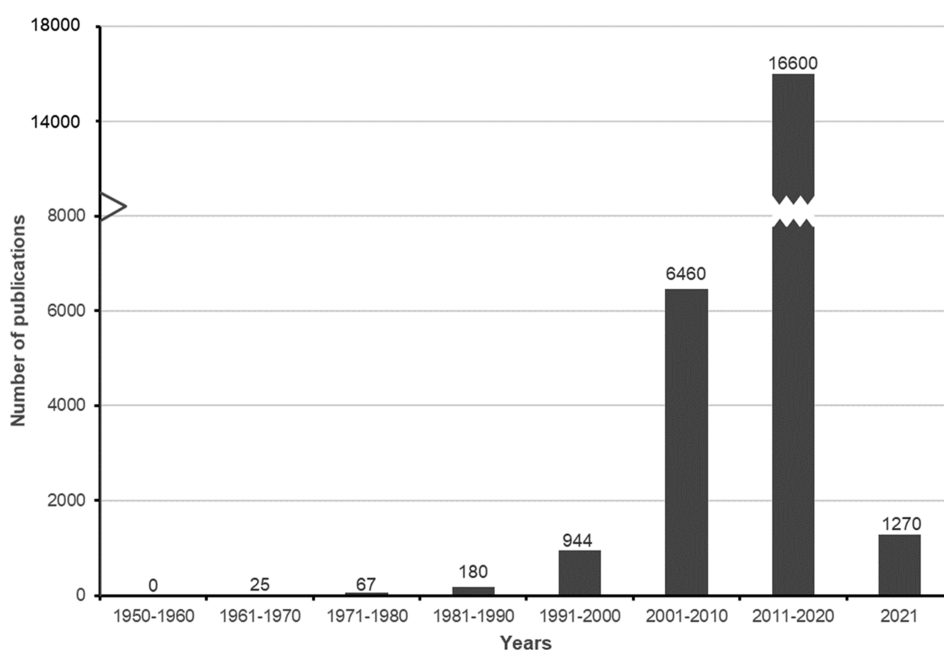


Figure 2. Number of scientific publications in the synthesis of ferrofluids over the years.

2. SYNTHESIS OF FERROFLUIDS

Ferrofluids are composed of three primary elements (Figure 3), which include a ferromagnetic compound in the lower

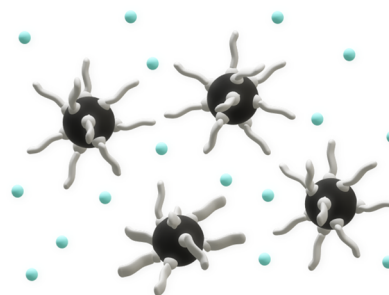


Figure 3. Components of a ferrofluid: magnetic NPs (black) coated with surfactant molecules (gray) suspended in a liquid carrier (blue). Adapted from Raj and Chorney.³⁴

nanometer range,¹¹ a surfactant that coats the magnetic compound to prevent agglomeration, and a carrier liquid that works in tandem with the surfactant to suspend the magnetic compound. The combination of the three components results in a stable colloidal suspension of ferromagnetic NPs.^{16–18} The magnetic phase of a ferrofluid is most commonly a mix of magnetite (Fe_3O_4) and maghemite (Fe_2O_3).^{19–21} Magnetite has higher magnetic properties²¹ and is the preferred compound obtained from synthesis. In addition to magnetite and maghemite, ferromagnetic compounds composed of a mix of iron and oxidation state +2 metals such as cobalt,^{22–27} zinc,^{28,29} nickel, and manganese^{22,23,30–33} are also used.

Due to the large variety of applications of ferrofluids, different combinations of NPs, surfactants, and carrier liquids are required. In general, high levels of magnetism and long-term stability are coveted. Additional research on surfactants, carrier liquids, and NP composition is needed to increase the stability and magnetism of ferrofluids.

2.1. Methods of Synthesis. The synthesis of ferrofluids begins with the production of iron oxide NPs (IONPs).³⁵ The size and magnetic properties of IONPs determine the quality of a ferrofluid by affecting the magnetism and stability of a ferrofluid, respectively.

There are various methods of synthesis used in the creation of IONPs, including sol–gel pyrolysis, mechanical alloying, the hydrothermal technique, mechanochemical synthesis,¹¹ flame synthesis, thermal decomposition, coprecipitation, mechanical milling, the sonochemical method, and one-pot synthesis. Out of these, the most reported methods for synthesis are thermal decomposition and coprecipitation (Figure 4), mainly due to

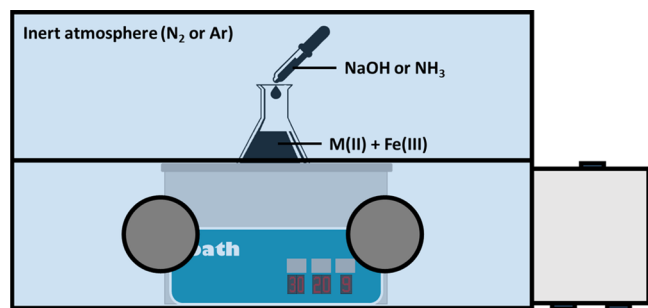
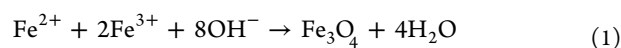


Figure 4. Representation of the synthesis of superparamagnetic IONPs via coprecipitation within an Ar or N atmosphere.

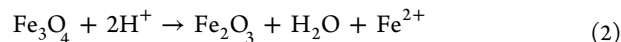
their ease and reliability.³⁶ Moreover, these methods either require less equipment, provide more consistent results, or, to the best of the authors' knowledge, are simply more thoroughly documented compared to other methods.

2.1.1. Coprecipitation. Coprecipitation is widely used in IONP synthesis due to its simplicity and large volume capacity.²¹ Coprecipitation of IONPs relies on the reaction between iron(II) and iron(III) sources inside of a water bath after the addition of a base. However, this form of synthesis can substitute iron(II) with various metallic(II) [M(II)] sources as long as they form magnetic compounds when bonded to iron(III). Some common examples of M(II) sources include Mohr's salt [di-ammonium iron(II) sulfate hexahydrate], ferrous chloride, ferrous sulfate, and some salts of oxidation state II of the first series of transition metals (Table 1). The synthesis of IONPs via coprecipitation under ideal inert conditions, such as within an argon or nitrogen atmosphere, involves the combination of one part M(II) source with two parts iron(III) source in a water bath (Figure 4).^{21,37}

After the addition of a M(II) source and an iron(III) source to a water bath, the pH must be increased slowly by the dropwise addition of a base, for example, sodium hydroxide (NaOH) or ammonia (NH₃)^{19,21,49,63–68} The increase in pH results in the formation of magnetite (Fe₃O₄) as follows (eq 1)^{21,49,63–68}



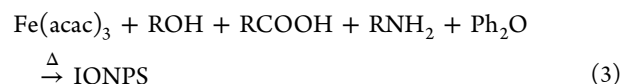
The complete precipitation of magnetite can be achieved in an inert atmosphere when using a nonoxidizing alkaline medium. However, the oxidation of magnetite into maghemite occurs by natural weathering or other processes that convert Fe²⁺ ions into Fe³⁺ ions, creating a final product containing both magnetite and maghemite (eq 2)⁶⁹



When the addition of basic NaOH or NH₃ is carried out while the solution undergoes vigorous stirring, the maghemite and magnetite precipitate has an average diameter of 12.6 ± 0.2 nm.^{1,17,70} The ideal size distribution for a ferrofluid is ~10 nm because the magnetization is reduced below 10 nm and because smaller particles are more stable in a suspension.^{12,58}

While coprecipitation is a desirable synthesis method due to its low cost and simple methodology, the lack of control over the NP size range and high likelihood of oxidation due to oxidizing environments make coprecipitation unreliable. When a higher control over the size distribution and magnetic activity is required, synthesis via thermal decomposition is usually the chosen method.

2.1.2. Thermal Decomposition. Thermal decomposition is a method for preparing IONPs via the decomposition of iron precursors (Table 1), commonly iron carbonyls^{38–45} and organometallics.^{42,57} The most common precursor is iron pentacarbonyl [Fe(CO)₅], a relatively toxic compound that when decomposed in a reducing environment, such as an inert gas flow, will produce magnetite.^{38,41} Another common iron precursor is Fe(C₅H₇O₂)₃, which is often abbreviated Fe(acac)₃.^{57,62,71,72} Thermal decomposition is chosen over coprecipitation when a greater control over IONP size is required.⁷³ Size is controlled in thermal decomposition by a multitude of factors but primarily by the molar ratio of the iron precursor and surfactant. The larger the ratio of the iron precursor is, the larger the size of the synthesized particles is.⁷³ To the best of the authors' knowledge, the most cited method for producing IONPs via thermal decomposition is Sun's method (eq 3)⁷²



According to Sun (2002),⁷² 4 nm Fe₃O₄ NPs are produced by mixing 2 mmol Fe(acac)₃ in 20 mL of phenyl ether with 10 mmol 1,2-hexadecanediol, 6 mmol oleic acid, and 6 mmol oleylamine under nitrogen and heated to reflux for 30 min. After cooling to room temperature, the dark-brown mixture is treated with ethanol under air, and a dark-brown material is precipitated from the solution. The precipitate is then dissolved in hexane in

Table 1. Common Sources of Iron(III), M(II), Surfactants, and Carrier Liquids for Ferrofluid Synthesis

decomposition sources	iron(III) sources		M(II) sources	surfactants	nonpolar liquids	polar liquids
iron pentacarbonyl ^{38–45}	ferric alum ⁴⁶	Mohr's salt ⁴⁶		oleic acid ^{45,47–50}	kerosene ^{8,51}	water ⁵¹
triiron dodecacarbonyl ³⁸	ferric chloride ^{46,52,53}	ferrous chloride ^{21,53}		silica ^{24,54–56}	mineral oil ⁵⁰	
Fe(acac) ₃ ⁵⁷	ferric nitrate ⁴⁶	ferrous sulfate ^{8,21,49,50}		chitosan ^{11,33,35}	silicon oil ^{13,58,59}	
		hydrosoluble salts of oxidation state II of the first series of transition metals ⁵²		acrylic acid ^{60–62}		

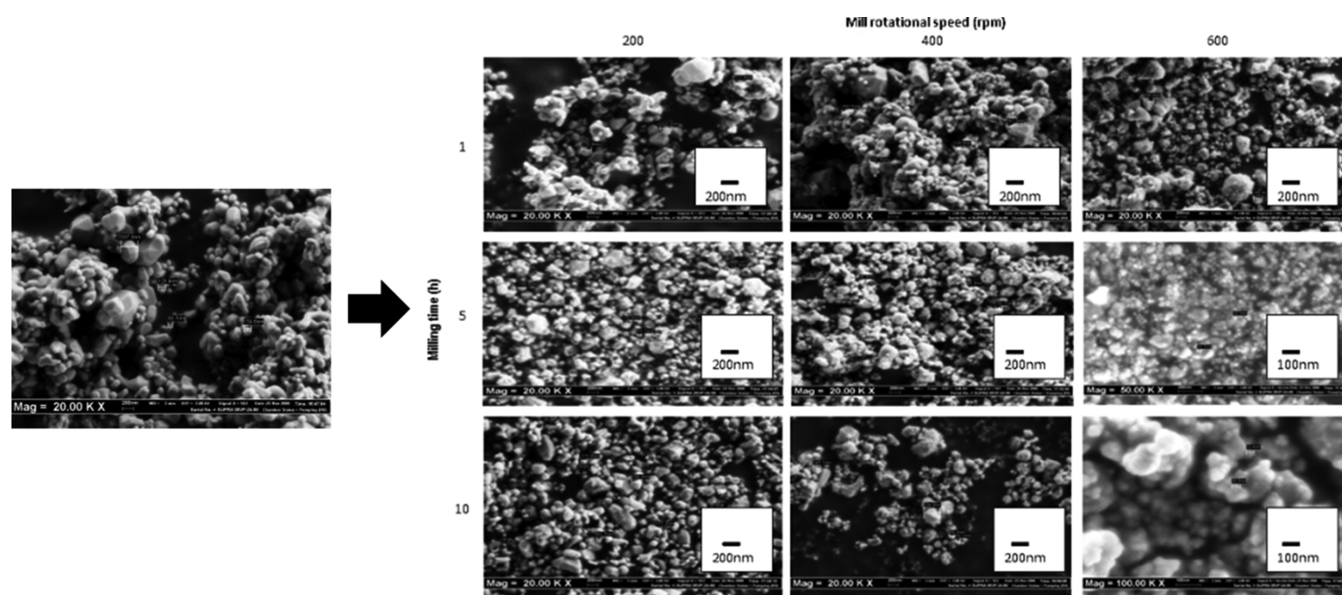


Figure 5. (Left) Photomicrograph of hematite before mill grinding and (right) ground product at rotational speeds of 200, 400, and 600 rpm during 1, 5, and 10 h. Reproduced with the authorization from R. Arbain, et al.⁷⁸ Copyright 2010, Elsevier.

the presence of oleic acid and oleylamine and reprecipitated with ethanol to give 4 nm Fe_3O_4 NPs.⁷²

Thermal decomposition has many advantages over other synthesis methods, primarily including its control over the size distribution in addition to the availability of documentation, which make it a commonly used method.⁷³ Thermal decomposition is superior to coprecipitation when creating isolated and monodispersed Fe_3O_4 NPs, which is important when considering biomedical applications.⁴² However, thermal decomposition requires more pieces of equipment and more expensive chemicals than other methods. Moreover, the reaction yield is lower when compared to that of reactions such as coprecipitation or mechanical milling.

2.1.3. Mechanical Milling. Mechanical milling is a simple reaction that utilizes milling iron powders such as hematite (Fe_2O_3) alongside other metal oxides to produce IONPs. Welding, fracturing, deformation, and rewelding of the iron powder during the high-pressure grinding process induces chemical reactions between the powder phases.⁷⁴ The fracturing results in a large surface area to react with, and the welding builds interfaces with the reactant phase.¹¹ The process of mechanical milling is described as a self-sustaining reaction of exothermic powder mixtures. The reaction begins with an activation period that involves the fracturing and deformation of the powders. When the powders reach a defined critical state, the reaction begins and is propagated throughout the powder as a combustion reaction.⁷⁵ Mechanical milling is economically useful since it can produce large quantities of IONPs at a cheaper cost than other methods of synthesis. However, this method involves less control over the NP size distribution and higher IONP aggregation.^{76,77} To obtain a size-controlled product and to minimize the agglomeration of IONPs in the ferrofluid, it is recommended to sonicate the product powder and centrifuge it to separate the larger particles from the NPs.¹¹

In mechanical milling, the speed of the reaction and output volume vary with the mechanism employed during the milling process.⁷⁷ Because mechanical milling has a low chance of oxidation, molar ratios can be used when determining the amount of reactants and effectively predict the final product.

Moreover, increasing the milling time (to a maximum threshold) results in a higher product yield.¹¹ However, it should be noted that as the milling speed and time increase above 400 rpm and 10 h, respectively, particles become larger as smaller particles begin to aggregate (Figure 5).⁷⁸

2.1.4. Synthesis of IONPs, Further Research, and Critical Insights. The properties of IONPs are based on the application they are synthesized for. Depending on which properties are more critical, the most appropriate method of synthesis must be chosen, which explains the current use of different methods. Common issues that need further research in all synthesis methods include controlling the size distribution and reducing the production costs. Greater control over IONP size has been made in synthesis methods such as thermal decomposition, but the higher cost and lower product yield are issues that could still be improved.

2.2. Oxidation during IONPs Synthesis. A common issue during coprecipitation (see Section 2.1.1), which is sometimes overlooked in the methodology, is the oxidation of iron(II) into iron(III). While exposed to an oxygen atmosphere, iron(II) is oxidized to iron(III).²¹ It is worth noting that this oxidation reaction does not occur for other M(II) sources because they are not iron-based. The oxidation process disrupts the ideal ratio of compounds for IONP synthesis. If an excess of iron(III) exists, maghemite will form as a dominant species. Maghemite is less magnetic than magnetite and will reduce the magnetic properties of the ferrofluid.

A proposed solution to the oxidation problem that has been reported is to perform the synthesis in an inert atmosphere. An oxygen-free atmosphere will inhibit the oxidation of iron(II) and allow for simple molarity calculations of iron(II) and iron(III) solutions to achieve a 1:2 ratio. However, performing the synthesis in an inert atmosphere is expensive as it requires additional equipment and spare gases such as argon or nitrogen. Another solution to this oxidation issue is to adjust the ratio of iron(II) and iron(III). This fix has been attempted by multiple researchers to varying degrees of success.^{37,79–84} The adjusted ratio that has seen the most success between these studies is a

1:~1.7 ratio, where an expected 5–10% of iron(II) will oxidize into iron(III).^{37,79}

However, a study performed by Maity and Agrawal (2007)²¹ contradicts the idea of adjusting the reactants ratios to reduce oxidation. Using X-ray diffraction, infrared spectroscopy, chemical analysis, magnetic measurements, and Mossbauer spectroscopy, the authors concluded that contrary to the popular assumption, the primary compound produced is maghemite with a small quantity of magnetite within the phase because a 1:2 ratio is never maintained during the precipitation. They propose that the oxidation of iron(II) to iron(III) is rapid and that adjusting the ratio results in a similar distribution to that obtained using a traditional 1:2 ratio.

2.3. Surfactants. During the synthesis of ferrofluids, surfactants (sometimes called dispersants)⁴⁴ are added to the mixture mainly to prevent the IONPs from agglomerating. Surfactants form layers around the magnetic NPs, allowing the NPs to remain suspended in the fluid. This is achieved through the formation of bonds with specific functional groups,⁸⁵ hence preventing agglomeration. Surfactants are an integral component leading to the stability of the colloidal suspension within a ferrofluid. This is important as the stability of the suspension determines the ferrofluid longevity. Moreover, for specific applications, surfactants can determine the biocompatibility of core magnetic NPs and act as protective layers on the NPs.⁵² It is important to mention that the dielectric properties of the surfactant must match those of the carrier liquid. Therefore, the surfactants are chosen based on the carrier liquid properties. Frequently used surfactants for aqueous media include oleic acid, silica, chitosan, polyvinyl alcohol (PVA), and acrylic acid (Table 1). The most commonly used surfactant, oleic acid, has a hydrophobic tail that allows for suspension within nonpolar carrier liquids such as kerosene. When the suspension in polar liquids is desired, for instance, in biomedical research, surfactants that contain hydrophilic tails such as acrylic acid or chitosan are used.^{86,87}

The surfactant role involves the formation of an external hydrophobic layer if the NPs are dispersed in a nonpolar carrier. Polar heads are pointed toward the surface of the magnetic NPs, and the alkyl chains are aligned to the nonpolar medium. However, when magnetic NPs are dispersed in a polar medium, a double layer of the surfactant is necessary to create an external hydrophilic layer. In this way, the polar groups are oriented to the polar medium.^{88,89} Amphiphilic surfactants such as oleic acid, oleylamine, fatty acids, and hexadecylamine allow the adjustment of the nucleation and growth kinetics of the NPs. Dioctyl sodium sulfosuccinate, cetyltrimethylammonium bromide, sodium dodecylsulfate (SDS) and polyethoxylates are employed for the formation of micellar microemulsion systems.⁹⁰ Among other materials used as surfactants are sodium citrate, polyacrylic acid (PAA), poly(maleic anhydride-*alt*-1-octadecene) (PMAO), oleylamine, and poly(ethylene glycol) (PEG).^{91,92}

2.4. Carrier Liquids. A carrier liquid is the third component of a ferrofluid, additional to the magnetic NPs and the surfactant. Carrier liquids are nonmagnetic liquids in which the NPs are dispersed. These liquids are able to tune the properties of ferrofluids such as viscosity, surface tension, vapor pressure, and stability at high or low temperatures.^{88,93}

Carrier liquids vary in properties such as the reactivity, viscosity, boiling point, and freezing point. These factors help determine what applications certain carrier liquids are best suited for. The list of reported carrier liquids includes water,

mineral oil,⁵⁰ ionic liquid, ester, hydrocarbons,⁹⁴ chitosan (considered a proper carrier for biological applications),⁹⁵ kerosene oil,⁸⁵ olive oil,⁹⁶ synthetic and semi-synthetic oils, and lubricating oil. Among these carrier liquids, the most commonly used are water and kerosene.^{21,28} However, synthetic oil and semi-synthetic oil are also used.¹⁶ When selecting a carrier liquid, the application of the ferrofluid to be synthesized needs to be taken into account. For instance, a carrier liquid should not be reactive with other materials in the application. Hence, after a carrier liquid is selected, the surfactant can be chosen. An example of this selection process is seen in biomedical applications of ferrofluids. In such applications, water is the most commonly used carrier liquid due to its nontoxic nature. Water then lends itself to surfactants such as chitosan, which also further reduces the toxicity of the IONPs. The combination results in ideal ferrofluids for biomedical usage.^{11,35}

2.5. Stability of Ferrofluids. The stability of ferrofluids is highly dependent on the affinity between the surfactant and carrier liquid, but it also depends on the synthesis method and NP size. As mentioned in Section 2.1, IONPs precursors include iron salts and organoiron materials, which impact the surface composition of the IONPs. However, the surface of the IONPs can be also modified during or after the synthesis process with the addition of compatible coatings in order to provide stability for a specific application. For example, for biomedical applications, IONPs are dispersed in biological media to prevent them from oxidation, improve their biocompatibility, enhance their stability, and to attach functional molecules. The most common materials to coat IONPs are polymers such as polyvinylpyrrolidone (PVP) or polyethyleneimine (PEI).^{90,97,98}

The stability of ferrofluids results from the balance between attractive forces, dipole–dipole interactions, and repulsive forces. When particles in a ferrofluid do not settle for a long period and do not experience a phase separation phenomenon when a strong magnetic field is applied, it can be considered stable. Measurement parameters and techniques to evaluate the stability of ferrofluids include zeta potential, spectral absorbance (via UV–vis spectroscopy), and sedimentation (via the centrifugation method).⁸⁷

The generation of repulsive interactions among the NPs (with the aim of stabilizing ferrofluids) can be performed by coating the surface or by creating a charge on the NPs. Surfacted ferrofluids use steric interactions to avoid agglomeration, while ionic ferrofluids generate electrostatic repulsions. In surfactated ferrofluids, the coating allows magnetic NPs to have long-chain molecules, avoiding agglomeration. The mixture of thermal motion, steric repulsions, and electrostatic repulsions favor the obtention of highly stable ferrofluids. A balance of attractive and repulsive interactions among the magnetic particles is key for the stability of ferrofluids.⁸⁸ Surfactants are the most commonly used materials to stabilize ferrofluids because of their high molecular weight. This characteristic favors the establishment of steric repulsions. Specifically, ionic surfactants generate ionic repulsion.⁹⁹ Additionally, the magnetic NPs can be stabilized in a carrier liquid through van der Waals forces, dipolar attractive interactions, and magnetostatic interactions.¹⁰⁰ In summary, stability results from the balance of repulsive (Brownian motions, steric forces, and electrostatic forces) and attractive forces (van der Waals and dipolar attractive forces).⁹⁴

Since IONPs are extensively used in biomedicine, water treatment, and engineering applications⁹⁸ (see Section 4), stability must be assessed in their respective environment. To obtain stable ferrofluids is important to reduce the agglomer-

ation. This can be achieved by using a proper synthesis technique or by adding surfactants to obtain a uniform dispersion of NPs. In general terms, stability is higher in surfactant-coated IONPs than that in bare IONPs. Surfactants such as oleic acid, citric acid, silica, PVA, and chitosan improve the NP dispersibility in an aqueous medium.^{87,101,102} The size of IONPs in ferrofluids is important as well since the thermal motion of smaller particles imparts better stability. However, if the particles are too small (1–2 nm), their magnetic properties could be diminished.⁸⁷

For instance, IONPs of size 83.54 nm synthesized by thermal decomposition (298 °C), using iron(III) acetylacetonate and $\text{Fe}(\text{acac})_3$ coated with PMAO, produced a steric barrier that can limit the precipitation of IONPs. These NPs have a polydispersity index (PDI) of 0.19 and a zeta potential of -40.9 mV 6 months after being synthesized.⁶² As reported, PMAO provides ferrofluids great stability for half a year. In comparison, IONPs of size 8.4 nm synthesized using $\text{FeCl}_2 \cdot 4\text{H}_2\text{O}$ as the precursor and stabilized with PAA proved to have a highly homogeneous size distribution with a zeta potential of -34.1 mV after 6 months of storage.¹⁰³

Although some polymers such as PMAO and PAA have been demonstrated to provide prolonged stability of IONPs in ferrofluids, further research is necessary to elucidate the relevant conditions and surfactants that preserve the stability of ferrofluids' in different applications, such as drug delivery, magnetic fluid hyperthermia, and some other applications discussed in Section 4.

2.6. Functionalization and Physicochemical Properties of Ferrofluids. As mentioned in Section 2.5, the stability of ferrofluids is crucial during their storage and applications. Bare NPs experience particle agglomeration and a large particle diameter because of the van der Waals and electrostatic forces. Modification of the NPs' surface (functionalization) is a way to improve NP stability.^{104,105} Furthermore, in some applications in the biomedical field, NPs could be functionalized to target certain cells and increase biocompatibility.⁸⁵ In general, surface functionalization can be achieved by using (a) organic materials, (b) polymers, (c) biomolecules, (d) metal oxides, (e) metal sulfides, (f) metals, and (g) carbon-based coatings.¹⁰⁴

Apart from preventing agglomeration and improving stability, the functionalization of NPs allows compatibility with certain molecules or materials and enhances the magnetic controllability. Functionalization is also a strategy to protect magnetic NPs from undesired changes. For instance, metal shells are used as coating agents to prevent NP oxidation.¹⁰⁴ In the biomedical field, NP functionalization is performed to target specific cells or tissues and to increase biocompatibility.⁸⁵ The surface modification can provide reactive handles that allow the interaction with biologically active substances, nonantigenicity, and protection from opsonization by plasma proteins, all important processes for biological applications.¹⁰⁶

Nontoxic silica has shown to be a good surface coating in the biomedical field. Silica forms cross-linking bonds and an inert external shielding layer to protect the magnetic NPs. Silica can be further activated to add additional functional groups in other applications, such as catalysis, adsorption, and magnetic separation.¹⁰⁴

Among the physicochemical properties of ferrofluids, magnetic and hydrodynamic properties are of relevance to the applications later discussed in this review (Section 4). Under the presence of a magnetic field, the magnetic torque of the NPs increases to resist the viscous torque, which provokes the rise of

friction force between the NPs and the carrier liquid. This leads to an increase in the viscosity of the ferrofluid, a phenomenon that is known as magnetoviscous characteristic.

Additionally, ferrofluids possess superparamagnetic properties.¹⁰⁷ The NPs in the ferrofluids are superparamagnetic at room temperature. Magnetic NPs behave as individual magnetic monodomains. Magnetic NPs are able to stay randomly in a carrier fluid (Figure 1) because of the Brownian motion, and the ferrofluid has no net magnetization. When there is an external magnetic field, the dipole interaction energy increases, overcoming the thermal energy. This allows the NPs to get oriented to the direction of the field, forming chain-like structures. The phenomenon promotes the enhancement of the thermal conductivity of ferrofluids.⁸⁵ Specifically, when Fe_3O_4 is formed as crystals of size 20 nm or less, it loses its permanent magnetism, becoming superparamagnetic. Under the action of a magnetic field, superparamagnetic Fe_3O_4 nanocrystals are strongly magnetized as each nanocrystal acts like a single magnetic moment. Superparamagnetism is important for biomedical applications because the action of NPs (interactions between NPs) is controlled through the magnetic field. Additionally, the superparamagnetism property allows the minimization of the risk of the embolization of the capillary vessels in the body. In general, the properties of Fe_3O_4 NPs have shown to be successful for biomedical applications because of their biodegradability, biocompatibility, minimal toxicity, high magnetic susceptibility, superparamagnetism, and high coercivity and physicochemical properties.¹⁰⁸

3. RHEOLOGY OF FERROFLUIDS

Rheology focuses on understanding how the fluids, especially Newtonian and non-Newtonian fluids, react to a force applied to them. One of the most known forces of fluids is shear stress, which is defined as the force that acts parallel to a surface and causes the deformation of a fluid when sliding in a certain area. The shear stress and deformation rate of the fluid in specific conditions and environments are the main rheological responses of ferrofluids.^{8,87} These responses are thoroughly examined in ferrofluids to maximize their properties, such as magnetism and adsorption. Besides, the generation of the theoretical and experimental knowledge about rheological responses of ferrofluids can overcome a particular problem of efficiency during their applications in biomedicine, MRI, and energy harvesting, and as agents for hyperthermia therapy.

Understanding the changes in viscosity is the typical outcome of rheological studies about fluids under different conditions. However, for ferrofluids, the rheological responses are conditioned to a magnetic field, resulting in the magnetoviscous effect,^{109,110} which may vary in several human matrices and environmental media.

The rheological responses of ferrofluids have been approximated via viscosity models developed by Carson, Einstein, Brinkman, Batchelor, and Bicerano,¹⁶ among others. All of them consider a ferrofluid as a suspension of particles in a carrier fluid. The approximation of rheologic models is the ratio between both viscosities: (1) the viscosity of the mixture of the particles and carrier fluid (η) and (2) the carrier liquid's viscosity itself (η_0) as a function of the volume of particles in the solid phase (ϕ), as given in eq 4. When a magnetic field is applied to a ferrofluid, η can be replaced by the viscosity of the suspension at that magnetic field (η_H), which is defined as a function of the magnetic field (M) and the viscosity of the carrier liquid in eq 5.

$$\frac{\eta}{\eta_0} = f(\phi) \quad (4)$$

$$M = \frac{\eta_H - \eta_0}{\eta_0} \quad (5)$$

The rheology of ferrofluids can vary according to the physicochemical characteristics that provide stability to the particles in the suspension¹¹⁰ and to those intrinsic characteristics of the carrier liquid that influences its viscosity. Figure 6

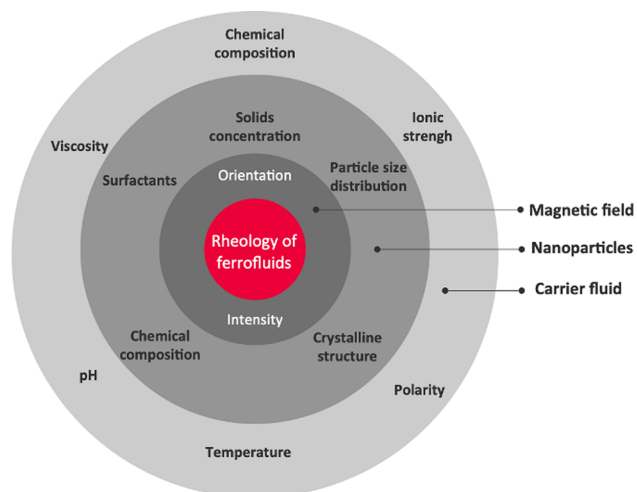


Figure 6. Factors influencing the rheology of ferrofluids as a function of the particle, carrier fluid, and magnetic field attributes.

shows the main properties of the particles and carrier fluids that influence the rheology of ferrofluids. While for particles, the attributes include the size distribution (5–10 nm), chemical composition (e.g., Mn–Zn ferrites, and iron oxide, among others), crystalline structure, concentration of solids and surfactants, for carrier liquids, the pH, temperature, ionic strength, and polarity of the substance are considered. Since ferrofluids are colloidal suspensions of NPs with a single magnetic momentum, their plastic behavior and the changes of viscosity are usually below 500 mPa·s.¹⁶ However, this behavior might also depend on the concentration of particles and carrier liquid, as well as the orientation and intensity of the magnetic field.¹¹¹

The elucidation of rheological responses for ferrofluids in different matrices either by modeling or experimentation is a topic that needs to be researched and developed in the near future.

4. APPLICATIONS OF FERROFLUIDS

4.1. Biomedical Applications. In the biomedical field, ferrofluids are recognized for their unique properties, including bioaffinity, cytocompatibility, small size, superparamagnetism, and a rapid response to magnetic fields. In order to enhance these properties, research on ferrofluids has focused on improving their behavior in three main areas: biosensors, medical imaging techniques, and therapy (Figure 7).¹¹²

4.1.1. Biosensors. Magnetic NPs composed of metals such as iron, nickel, and cobalt and their corresponding oxides with the size in the range between 5 and 10 nm possess a magnetic behavior in a dielectric matrix, and when they are below the Curie temperature, such NPs are considered superparamagnetic particles. Superparamagnetic particles are magnetized when

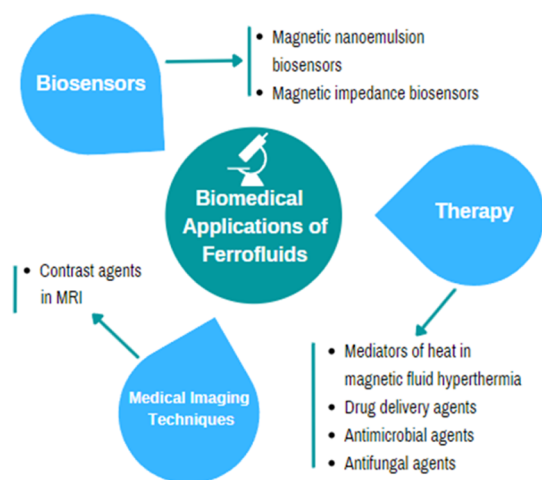


Figure 7. Biomedical applications of ferrofluids.

there is an external magnetic field. It is known that as living systems can produce biomagnetic fields, magnetic sensors can detect biological tissues or organs (biosensors). For example, superparamagnetic NPs are able to identify the biological activity in living systems. Moreover, either bare NPs or NPs with a surfactant can be functionalized with biochemical compounds to capture molecules such as deoxyribonucleic acid, proteins, enzymes, and cells.⁵⁹ In general, there are two strategies for biosensing when ferrofluids are applied (Figure 8). The first

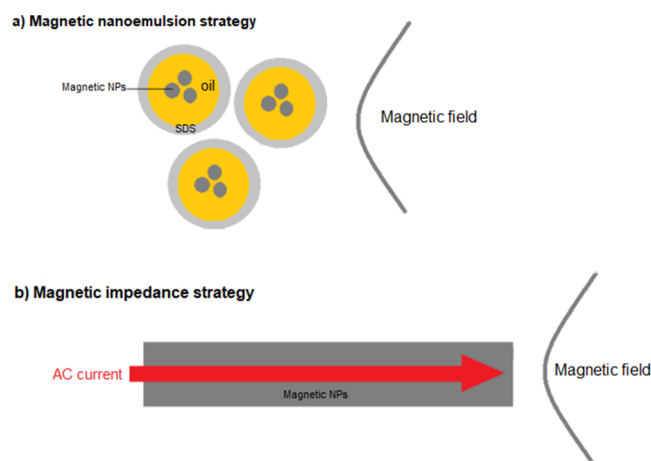


Figure 8. Biosensing strategies when ferrofluids are applied: (a) magnetic nanoemulsion strategy and (b) magnetic impedance strategy.

strategy is called magnetic nanoemulsions. In this approach, ferromagnetic particles of 10 nm are encapsulated in a 200 nm oil droplet and stabilized by an anionic surfactant of SDS. If there is an external magnetic field present, the magnetic nanoemulsions would assemble into an orderly array that causes a specific diffraction peak.¹¹² In the second strategy, known as magnetic impedance biosensors, the biosensors follow the giant magnetoimpedance (GMI) effect. The GMI effect is a physical effect in which the current flowing through a magnetic material suffers variations in the electrical impedance when an external magnetic field is applied.^{59,113,114} For magnetic impedance biosensors, the use of Fe₃O₄ IONPs of size up to 10 nm has been reported.^{113,115}

4.1.2. Medical Imaging Technique. It is well-known that ferrofluids have the ability to improve MRI, a widely used

medical imaging technique. In the clinical field, using magnetic NPs in MRI results in a noninvasive strategy for molecular imaging where IONPs are used as contrast agents.^{3,4} The MRI has magnets that induce a magnetic field that make the protons present in the water of the tissues to align with the field. When a radiofrequency current is pulsed through a body, the protons are stimulated and spin out of the equilibrium. Once the radiofrequency is turned off, the equipment detects the energy formed when the protons realign with the magnetic field. Contrast agents are needed to increase the speed at which the protons realign with the magnetic field, producing a brighter image. The transverse relaxation time is the time that the spinning protons take to reach the equilibrium. IONPs are able to shorten the transverse relaxation time of water protons, creating a local magnetic field that forces the surrounding protons to undergo faster spin–spin relaxation time, enhancing the relaxation time-weighted contrast degree, which improves the quality of MRI images.^{60,103,116} It is also known that IONPs have great potential as MRI contrast agents because of their efficacy and safety. Moreover, IONPs have been approved for use as liver MRI contrast agents by the United States Food and Drug Administration. The small size and easy penetration of IONPs into the cells enhance the contrast during stem cell imaging, magnetofection, and cancer imaging.¹¹⁷

The surface of IONPs can be modified with a biocompatible coating in order to increase their biocompatibility and target specificity and to prevent agglomeration, oxidation, corrosion, and toxicity. The materials used for surface modification include polymers, fatty acids, metals, and oxides. However, polymers such as dextran, gelatin, alginate, chitosan, starch, albumin, casein, PEG, PVP, PVA, polydopamine, poly lactic-*co*-glycolic acid (PLGA), and dendrimers are the most popular coating materials.^{90,92,97}

Particles of Fe₃O₄ (8 nm), synthesized from FeCl₂·4H₂O as the precursor and stabilized with PAA ligand, are potential contrast agents for MRI applications.¹⁰³ Xiao et al. (2018)¹¹⁶ reported that PAA (20 nm size)-coated nanomagnetic iron oxide shows better signal sensitivity for liver imaging in vivo than those coated with PVP or PEI. Furthermore, Yusefi et al. (2021)¹¹⁸ suggested that IONPs of size 14.38 nm stabilized with the *Punica granatum* fruit peel extract showed good MRI signals due to diminished iron aggregation. The extract is a green stabilizer that increased the water permeability of the IONPs, which improved the relaxivity of MRI and showed effective biodistribution. Furthermore, Vangijzegem et al. (2020)⁹¹ proved that oleylamine and oleic acid used as surfactants of IONPs yielded to stable and concentrated colloidal suspensions at a ratio of 4:1 (surfactants to the iron precursor), suggesting that there is an optimum ratio at which the organic surfactants provide better stability of IONPs for MRI purposes. Shelat et al. (2019)¹¹⁷ used L-arginine and L-histidine as coating agents for IONPs. The size of the coated IONPs was between 15 and 25 nm, and they were nontoxic for tissues or organs (IONPs coated with L-arginine and L-histidine are highly biocompatible). It was also proved that IONPs coated with L-arginine were better as a tracer material compared to L-histidine for in vivo MRI due to their increased magnetization, high iron content, and better tissue contrast. Moreover, Cha et al. (2017)¹¹⁹ used IONPs (22 nm) with cyclodextrin-containing star-shaped poly(2-(dimethylamino) ethyl methacrylate) and demonstrated that the coated particles have low cytotoxicity and are suitable as an MRI contrast agent as they enhance the signal sensitivity. In addition to IONPs, ferrofluids based on cobalt ferrite NPs have all the

characteristics and properties that are needed for their application in MRI diagnosis.¹²⁰ Besides, ferrofluids based on manganese ferrite NPs coated with polysaccharide chitosan have also shown promising results for MRI.³³

4.1.3. Hyperthermia Therapy. In the field of cancer therapy, magnetic fluid hyperthermia has been studied. In magnetic fluid hyperthermia, magnetic NPs act as mediators of heat into a tumor exposed to an external alternating magnetic field. In the process, the temperature of the tumor increases because of the generation of heat from internalized magnetic NPs that are being subjected to a high-frequency alternating magnetic field, inducing dissipation of the magnetic energy.¹²¹ During this method, the magnetic NP suspensions are delivered to target organs (or near) with cancerous sites, where they generate local heat, increasing the temperature to a value ranging between 41 and 46 °C and inducing processes that are able to kill cancer cells such as necrosis, protein denaturation, protein folding, aggregation, and DNA cross-linking.¹²² For these abilities and because of their high saturation, magnetization, and biocompatibility, superparamagnetic ferrofluids are extensively studied as potential candidates in cancer therapy.⁶² Although most hyperthermia techniques are developed for cancer treatment, hyperthermia is also used to treat restenosis, remove plaques, ablate nerves, and alleviate pain by increasing the regional blood flow.¹²³ The combination of ferrofluid hyperthermia and MRI is recently considered to be one of the most promising noninvasive cancer treatments.

IONPs are the most studied materials for hyperthermia therapy. Among the commonly used coating materials for IONPs are polymers such as dextran, PEG, and PVA. Less common materials such as copolymers, silica, gold, and fatty acids have also been applied.^{123,124} A recent investigation conducted by Yusefi et al. (2021)¹¹⁸ proved that IONPs (14.38 nm) coated with the fruit peel extract from *Punica granatum* provided sufficient potential to control the medium temperature increase within the secure hyperthermia range, a good performance for hyperthermia treatment. Moreover, Mazario et al. (2017)¹²⁵ showed that IONPs (10 nm) coated with human serum albumin have the potential for magnetic hyperthermia between 41 and 46 °C.

In addition, Fotukian et al. (2020)¹²⁶ demonstrated that CuFe₂O₄ (19.9 nm) coated with triethylene glycol shows better results for hyperthermia therapy than IONPs. Nevertheless, the magnetic properties of IONPs can be altered by changing the synthesis procedure. For example, the synthesis of other NPs such as cobalt-doped magnetite NPs by the partial replacement of Fe²⁺ with Co²⁺ species is an option to improve hyperthermia properties due to the increase in the magnetocrystalline anisotropy. Cobalt doping may improve the magnetic hyperthermia performance of 7–8 nm IONPs for applications in multimodal cancer therapy.¹²⁷

4.1.4. Drug Delivery. Regarding drug delivery, magnetic NPs in ferrofluids have a greater reactive area and ability to cross biological barriers (e.g., cell membrane) than their micrometric counterparts, which make them efficient in this field. The most common materials for coating are polymers, chitosan, oleic acid, trisodium citrate, lipids, proteins, and peptides. Various classes of drugs can be directly bound to IONPs or to core–shell nanosystems by adsorption, dispersion in the polymer matrix, encapsulation in the nucleus, electrostatic interactions, and covalent attachment to the surface. IONPs have been used as carriers of anticancer, immunosuppressive, anticonvulsant, anti-inflammatory, antibiotic, and antifungal agents.¹²⁸ In general

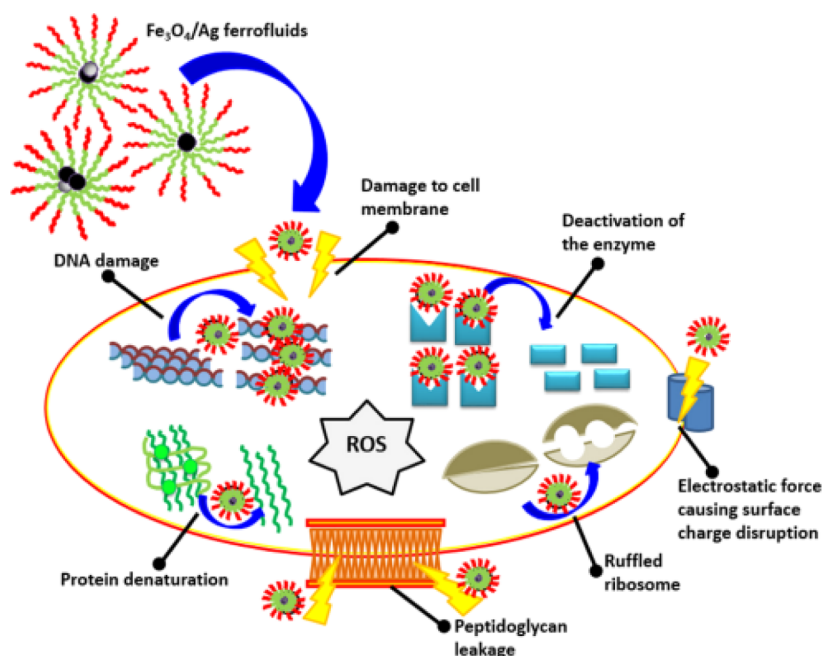


Figure 9. Mechanism of bacterial destruction by the $\text{Fe}_3\text{O}_4/\text{Ag}$ ferrofluids. Image reprinted with permission from Taufiq, et al.⁹⁶ Copyright 2020, Elsevier.

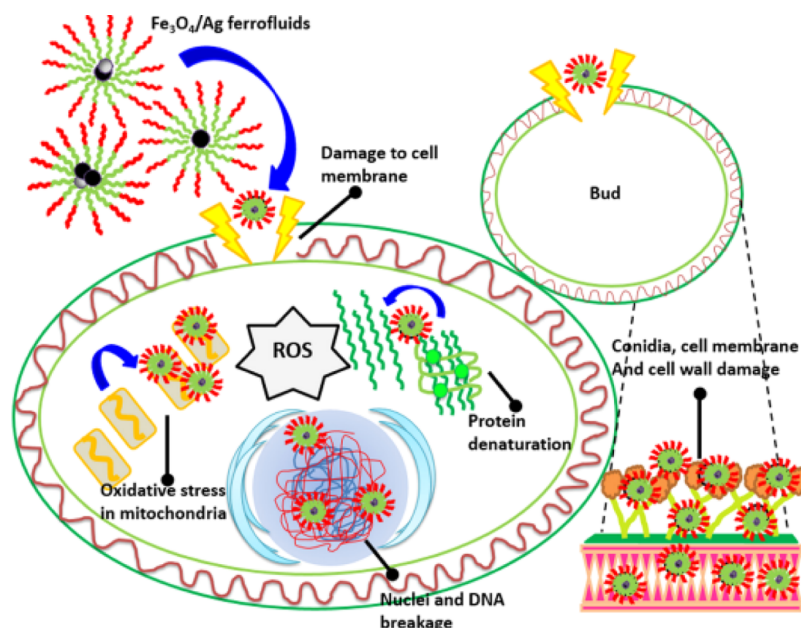


Figure 10. Mechanism of fungal destruction by the $\text{Fe}_3\text{O}_4/\text{Ag}$ ferrofluids. Image reprinted with permission from Taufiq, et al.⁹⁶ Copyright 2020, Elsevier.

terms, magnetic drug targeting is used to enhance the accumulation and the efficacy of IONPs loaded with chemotherapeutic drugs. However, advanced agents such as DNA, siRNA, and radionuclides have been combined with IONPs and magnetic drug targeting.⁹⁰

IONPs are the most studied NPs for the magnetically guided drug delivery in cancer therapeutics due to their chemical stability, low toxicity, biodegradability, and superparamagnetism.^{60,129–131} For cancer treatment, IONPs loaded with doxorubicin have been coated with hydroxyapatite (HA).¹³² Similarly, IONPs (10 nm) with a shell of aqueous stable PEG conjugated with doxorubicin have shown a great capacity for

tumor chemotherapy.¹³³ Citric acid-coated IONPs functionalized with PEG have also demonstrated potential for drug delivery to treat cancer.¹³⁴ Furthermore, highly water-dispersible surfactant-stabilized IONPs prepared by the self-assembly of SDS on hydrophobic (oleic acid-coated) NPs (10 nm) loaded with doxorubicin and curcumin demonstrated to be effective for cancer therapy.¹³⁵ Other anticancer drugs that have been attached to IONPs include β -cyclodextrin, carmustine, cetuximab, cytarabine, daunomycin, docetaxel, epirubicin, 5-fluorouracil, gemcitabine, methotrexate, mitoxantrone, and paclitaxel.¹²⁸

4.1.5. Antimicrobial and Antifungal Activity. Ferrofluids have also proved to be effective as antimicrobial and antifungal agents. Drug resistance has accelerated the development of novel antimicrobial agents such as IONPs, which are effective antimicrobial agents against *Staphylococcus aureus*, *Staphylococcus epidermidis*, *Escherichia coli*, *Xanthomonas*, and *Proteus vulgaris*.¹³⁶ Fe₃O₄/Ag nanohybrid ferrofluids (12.1 nm) coated with oleic acid and dimethyl sulfoxide (DMSO) have also showed antimicrobial activity against *Bacillus subtilis*, *S. aureus*, *E. coli*, and *Candida albicans*. Besides, this nanohybrid ferrofluid has been effective in decreasing the progression of liver fibrosis-related inflammation and fibrogenic activity on hepatic stellate cells.⁹⁶

Figure 9 shows the proposed mechanism of destruction of the bacteria by Fe₃O₄/Ag ferrofluids, while Figure 10 illustrates the antifungal mechanism proposed for *C. albicans*. Metal oxides are thought to inhibit the growth of bacteria by producing oxidative stress and reactive oxygen species (ROS),^{96,137} which limits the synthesis of amino acids, lipid peroxidation, and DNA replication. Additionally, oxidative stress changes the cell membrane permeability and causes irreversible membrane damage.¹³⁸ The surface charge of NPs contains ROS in the form of a superoxide, which in the presence of surfactants becomes positive. As the surface charge of microbes is negative, an electrostatic attraction between the NPs and microbes occurs. The ROS penetrate cells through microbial pores, resulting in the destruction of the microbial cell nucleus along with the pathogenic DNA. In summary, the Fe₃O₄/Ag nanohybrid ferrofluids provoke cell death by dissolving the outer envelope of the cell walls and accessing the microbial nucleus and DNA.⁹⁶

4.1.6. Perspectives about Biomedical Applications. Research in biomedical applications of ferrofluids has continuously advanced in the last years. However, there are still some challenges to overcome. In general terms, one of the most relevant challenges is the size control of NPs. Size must be selected according to the needs of the application. Thus, an appropriate synthesis method must be developed or adapted depending on the ferrofluid purpose.¹⁰⁴

Despite the ongoing development of the methods for the synthesis of ferrofluids, there is still room for improvement, so the final product possesses proper stability and biocompatibility. Besides, problems related to toxicity have to be solved.⁹⁵ Toxic effects are due to ROS generated by the NPs, which provokes oxidative stress, damaging functions inside the cell. Thus, there is a need to study the possible toxicity from the coexposure of biological systems to two or more different types of NPs.¹³⁹

Another important challenge is the accurate evaluation of temperature when using ferrofluids during hyperthermia. The development of numerical models that correlate the magnetic NP hyperthermia parameters to the thermal response is relevant. When a ferrofluid solution is evaluated on different parameters and the temperature distribution of the system is calculated in space and time, the optimum therapeutic conditions can be found.¹⁴⁰

Regarding MRI, research has shown that IONPs contrast agents are less toxic than other materials such as gadolinium-based contrast agents. Nevertheless, biocompatibility, biodistribution, and pharmacokinetics should be studied for the future applications of IONPs in clinical stages. Furthermore, IONPs may lead to the possibility of integrating MRI with supplementary imaging techniques such as positron emission, tomography, and computed tomography.¹⁴¹

Specifically, new IONPs synthesis methods will help find better ways to apply IONPs as theranostic agents in the biomedical field including disease diagnosis, early detection, imaging, and drug and gene delivery, along with multifunctional therapeutics. Additionally, multifunctional IONPs are attractive materials that may change the traditional model of the pharmaceutical industry.¹⁴²

Finally, as most of the studies in the biomedical field are conducted in vitro, it is necessary to perform in vivo studies to satisfy clinical requirements. In order to use ferrofluids for in vivo studies, novel nanofabrication techniques and a deeper understanding of the ferrofluid–biological interactions are required.⁸⁷ Among the most important requirements for the in vivo applications of ferrofluids are the biodegradability of NPs and/or their capacity of being excreted from the body, along with the fact that they must not induce a not desirable immune response.¹³⁹

4.2. Water Treatment. Iron oxides, magnetite, and maghemite are part of IONPs with high surface area and adsorption capacity that are able to form agglomerates. Since magnetic NPs in ferrofluids have a large surface area that can easily bind to many molecules, they can remove contaminants from aqueous effluents such as color, turbidity, metals, organic matter, and bacteria from waters and then can be separated from the bulk solution with an external magnetic field.^{63,143–145} Recently, IONPs have caught the attention of researchers in water treatment as they are known to be effective adsorbents to remove contaminants from water and wastewater.^{112,146,147}

Impurity-free IONPs of Fe₃O₄ NPs (23 to 32 nm) are able to absorb and remove the water turbidity, hence clarifying the water.¹⁴⁴ Besides, Hatamie et al. (2016)⁶³ synthesized IONPs without a surfactant and found that the magnetic fluid had the ability to decrease the turbidity of the water and to remove the color, hazardous cations and anions, and fecal coliforms, while simultaneously reducing the chemical oxygen demand (COD), without affecting the water alkalinity. Moreover, IONPs can remove oil in oily micropolluted water.¹⁴⁸ Furthermore, IONPs with tetramethylammonium hydroxide as surfactant (30 nm) are potential adsorbents for aqueous Cr⁶⁺ and Pb²⁺ removal,¹⁴⁹ while IONPs of Fe₃O₄ functionalized with EDTA (35 nm) were able to remove heavy metals such as Ag(I), Hg(II), Mn(II), Zn(II), Pb(II), and Cd(II) from water and soil.^{150,151} The absorption of pharmaceuticals atenolol, ciprofloxacin, and gemfibrozil in water has also been performed using Fe₃O₄ coated with a polymer clay composite.¹⁵² Additionally, Fe₃O₄ NPs (11.7 nm) coated with activated carbon removed rhodamine B and methyl orange from wastewater.¹⁵³

Additionally, it has been shown that a Fe₃O₄/bentonite magnetic nanocomposite (40 nm) is efficient in reducing the concentrations of nitrate, biochemical oxygen demand (BOD), and COD in industrial water.¹⁴⁶ Similarly, ferrofluids based on the Fe₃O₄/bentonite nanocomposite showed a good adsorption capacity and a fast adsorption rate of the cationic dye methylene blue removal from water.¹⁵⁴ A study on Fe₃O₄/Au NPs loaded with activated carbon showed the ability to remove disulfine blue and rhodamine 123 present in water,¹⁵⁵ while Fe₃O₄/CuO NPs assembled on graphene oxide (pore size = 27.62 nm) enhanced the absorption of As(III) and As(V) from water.¹⁵⁶ In addition, Fe₃O₄-loaded sawdust carbon and EDTA-modified (30 nm) NPs are a promising adsorbent for Cd(II) removal from aqueous media.¹⁵⁷ Moreover, Fe₃O₄/Ag (32 nm) NPs have catalytic activity toward the reduction of organic dyes.¹⁵⁸ Finally, Mn_{1-x}Zn_xFe₂O₄ ferrofluids from natural sand have also

been utilized as radar-absorbing materials and as magnetic sensors with high stability.¹⁵⁹

Further research is required to elucidate the behavior of NPs in realistic environmental conditions considering the origin and composition of treated water. Also, the determination of the optimum conditions for successive treatment cycles without affecting the absorption efficiency and stability of ferrofluids⁹³ is an area of opportunity for ferrofluids.

4.3. Mechanical Engineering. Apart from the biomedical and water treatment areas, ferrofluids have been utilized in heat transfer, energy harvesting, and vibration control (see Figure 11).

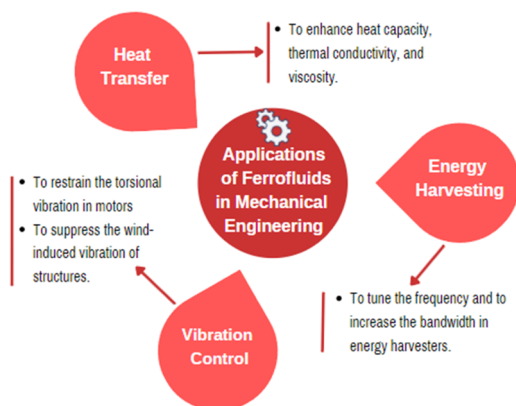


Figure 11. Applications of ferrofluids in mechanical engineering.

4.3.1. Heat Transfer. Ferrofluids are fluids with great potential for heat transfer applications. They have proved to be effective in improving the heat capacity, thermal conductivity, and viscosity to control the transfer of heat and the movement of particles by applying external magnetic fields. Because of their characteristics, ferrofluids are promising in bioengineering, thermal engineering, electronics, and energy harvesting. The thermal conductivity of magnetic nanofluids increases when a magnetic field parallel to the temperature gradient is applied. Consequently, an enhancement in the heat transfer coefficient is expected. Fe_3O_4 , $\gamma\text{-Fe}_2\text{O}_3$, and spinel-type ferrite NPs are the most commonly used due to their chemical stability.⁴⁷ It has been demonstrated that platelet-shaped IONPs can also enhance the thermal conductivity.^{160–162} Furthermore, the hybrid ferrofluid Fe_3O_4 /carbon nanotubes (CNT, 30 nm) is one of the most studied for enhancing the thermal conductivity in an oscillating magnetic field.¹⁶³ Multiwalled carbon nanotubes coupled to Fe_3O_4 NPs (f-MWCNTs) dispersed in ethylene glycol (30 nm) showed high thermal conductivity.¹⁶⁴ Lastly, reduced graphene oxide/ Fe_3O_4 (between 10 and 20 nm) have shown a high heat transfer coefficient due to the increased thermal conductivity, decreased thickness of the boundary layer, and the viscosity, which enhanced the convective heat transfer coefficient.¹⁶⁵

The use of ferrofluids in heat transfer has allowed the development of new practical devices and the opportunity to look for new methods of synthesis and to better understand the heat transfer improvement mechanisms. The control of the particle morphology and size may be studied since it will allow the production of a ferrofluid with desired thermal properties along with long-term stability, which remains a challenge.⁴⁷

4.3.2. Energy Harvesting. In the field of energy, magnetic nanofluids are potential fluids for vibrational energy harvesting

applications due to their fluidity and magnetic properties serving as soft ferromagnetic substances. Challenges that are associated with vibrational energy harvesters such as widening, bandwidth, and tunability are solved using suspensions of magnetic NPs. Hybrid ferrofluid-embedded energy harvesters have demonstrated a positive impact by collecting subtle and irregular vibrations as they feature a low threshold amplitude and a wide operating frequency range. In this field, Fe_3O_4 and $\gamma\text{-Fe}_2\text{O}_3$ are the materials most commonly used.^{166–168} Embedded ferrofluids have been applied to tune the frequency and to increase the bandwidth in energy harvesters. Magnetic actuation mechanisms employed to control the ferrofluid to a specific location along the length of cantilevers have shown promising results for energy harvesting devices.¹⁶⁶ In a similar way, nonlinear resonance electromagnetic energy harvesters that utilize the energy from the human motion to power the wearable devices using magnetic springs and ferrofluids to minimize the friction losses and maximize the output power have been developed.¹⁶⁹

4.3.3. Vibration Control. Ferrofluids are used as damping agents because of their magnetoviscous characteristics.¹⁰⁷ Moreover, ferrofluids have been utilized for vibration control in motors and structures as they do not precipitate under ambient conditions. Applications in vibration control include restraining the torsional vibration in high-speed motors, step motors, and hydraulic servo torque motors as a damping material, suppressing the wind-induced vibration of civil structures as vibration isolators, and solving some vibration problems in the space microgravity environment.¹⁷⁰

Additionally, ferrofluids are used in high-power loudspeakers. In the loudspeaker operation, a strong magnetic field has to be in a small air gap. When that gap is filled using a ferrofluid, the loudspeaker improves its performance. The advantages of using ferrofluids in loudspeakers include voice coil cooling, voice coil centering, reduction of power compression, and damping. This is because when ferrofluids have a proper thermal conductivity and viscosity, they are able to dissipate the excess heat from the coil and damp excessive resonances.⁸⁵

In this section, some applications of ferrofluids are discussed. The most commonly used magnetic NPs and coating materials in the biomedical field are presented for their use in biosensors, MRI, magnetic fluid hyperthermia, and drug delivery and as antiseptics. Moreover, other applications of ferrofluids in the fields of water treatment, energy harvesters, heat transfer, and vibration control are explained. Ferrofluids are promising in diverse areas because of their chemical stability, low toxicity, biodegradability, high surface area, adsorption capacity, fluidity, and magnetic properties. Research in magnetic NPs and surface modifications has improved the traditional techniques, processes, and technologies, allowing different disciplines and areas to continue developing and overcoming modern challenges.

5. CONCLUSIONS AND FUTURE PERSPECTIVES

This article reports the synthesis of IONPs for ferrofluids via coprecipitation, thermal decomposition, and mechanical milling methods. Surfactant and carrier liquid properties and their uses are briefly discussed. The rheology of ferrofluids and the functional components that contribute to their stability are described. Finally, the applications of ferrofluids within the medical and nonmedical fields are highlighted.

It should be noted that during IONP synthesis, a common issue for noninert environments is oxidation. The common

methods of correction for this oxidation involve adjusting the reactant concentration. Nevertheless, a recent study contradicts this idea and proposes that oxidation is rapid and that regardless of the reactants' concentrations, the product will be unchanged.

From the authors' perspective, future research should pay attention to IONPs characteristics (size, chemical composition, and crystalline structure), properties of the carrier liquid and the surfactant, and the affinity between the surfactant and carrier liquid. Since these properties determine the stability and rheology of ferrofluids, small changes could have unintended effects on how the synthesized fluid behaves.

As is highlighted in Section 4, the most promising areas for ferrofluid applications are in the biomedical field and water treatment, importantly, as contrast agents, drug delivery agents, and antimicrobial agents. Additionally, ferrofluid uses in magnetic nanoemulsion biosensors and magnetic impedance biosensors and as mediators of heat in magnetic fluid hyperthermia have been reported. It can be noted that when incorporated as contrast agents in MRI, ferrite compounds including cobalt and manganese have shown encouraging results and have characteristics and properties that make them promising for applications within the MRI diagnosis. Studies involving IONPs, specifically magnetite, for water treatment have shown that they have a high capacity for the absorption of impurities and contaminants such as color, turbidity, metals, organic matter, hazardous cations and anions, oils, and bacteria.

However, as ferrofluids applications continue expanding, additional research needs to be performed to overcome current challenges, including the following:

- (i) Oxidation during coprecipitation in noninert environments and production rate of maghemite or magnetite during synthesis;
- (ii) Influence of the addition of oxidation state +2 metals on the oxidation of iron and magnetic properties of IONPs;
- (iii) Long-term ferrofluid stability for drug delivery applications;
- (iv) Changes in rheology properties for relevant biological and environmental matrices;
- (v) Size control of NPs during synthesis;
- (vi) Improvement of the stability and biocompatibility;
- (vii) Toxicity from the coexposure of biological systems to two or more different types of NPs;
- (viii) Accurate evaluation of temperature during the use of ferrofluids in hyperthermia by developing numerical models;
- (ix) Integrated research of biocompatibility, biodistribution, and pharmacokinetics for future applications in the clinic;
- (x) The integration of MRI with other imaging techniques using ferrofluids;
- (xi) In vivo studies of the ferrofluid applications to satisfy clinical requirements;
- (xii) Understanding the behavior of the NPs in realistic environments for water treatment;
- (xiii) Determining the conditions for conducting successive water treatment cycles without affecting the ferrofluid; and
- (xiv) In terms of energy, the control of the particle morphology and size should be improved during the synthesis to obtain ferrofluids with specific thermal properties and long-term stability.

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Notes

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