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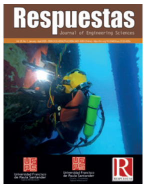

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Magnetorheological fluids: synthesis, properties and applications

Fluidos magnetorreológicos: síntesis, propiedades y aplicaciones

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ABSTRACT

Keywords:

Magnetorheological fluids,
viscosity,
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Smart materials,
magnetite.

The magnetorheological fluids have the ability to modify their viscosity quickly by subjecting it to a magnetic field, a quality that classifies them in the category of intelligent materials. Such fluids include three main components; the base fluid that is generally mineral or synthetic oil, magnetizable particles which are dispersed in the base fluid and the additives or stabilizers that prevent agglomeration and sedimentation of the particles, as well as the degradation induced by the carrier medium. The main challenge of these fluids is to maintain the rheological response to the magnetic field, as well as avoid the chemical and microstructural instability of the magnetoreological fluids. In the present investigation an exhaustive bibliographic review was carried out to understand the main aspects of these types of materials, their properties as well as the main applications in the different branches of industry and medicine. The most recent contributions focused on the chemical stability of magnetic particles through the application of surface coatings are discussed.

RESUMEN

Palabras clave:

Fluidos
magnetorreológicos,
viscosidad,
campo magnético,
materiales inteligentes,
magnetita.

Los fluidos magnetorreológicos tienen la capacidad de modificar su viscosidad rápidamente al someterlos a un campo magnético, cualidad que los clasifica en la categoría de los materiales inteligentes. Dichos fluidos incluyen tres componentes principales; el fluido base que generalmente es aceite mineral o sintético, partículas magnetizables las cuales se dispersan en el fluido base y los aditivos o estabilizadores que evitan la aglomeración y sedimentación de las partículas, así como la degradación inducida por el medio portador. El reto principal de estos fluidos es mantener la respuesta reológica al campo magnético, así como evitar la inestabilidad química y microestructural del fluido magnetorreológico. En la presente investigación se realizó una revisión bibliográfica exhaustiva para comprender las principales características de estos tipos de materiales, sus propiedades en términos de viscosidad, así como las principales aplicaciones en las distintas ramas de la industria y la medicina. Se discuten las aportaciones más recientes enfocadas a la estabilidad química de las partículas magnéticas mediante la aplicación de recubrimientos superficiales.

Introduction

The study of new materials is currently oriented towards the development of intelligent materials, i.e. materials whose response depends on external stimulation. Magnetorheological fluids present these characteristics since their viscosity changes considerably in the presence of a magnetic field, since it induces the formation of a dipole in each of the magnetic particles, which then join together in the form of chains that restrict the movement of the fluid. Hence its ability to provide a simple, silent and fast response [1], [2].

Magnetorheological fluids (FMR) are colloidal suspensions of magnetizable particles that are on the order of a few microns (0.5-10 m) in size. The initial discovery and development of magnetorheological devices can be credited to Jacob Rabinow at the US

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National Bureau of Standards in 1948 [3].

The particles in FMR are multidomain magnetic, so that the application of an external magnetic field induces a dipole on each particle and produces very strong interactions between them and/or agglomerates in the suspension. The FMR reversibly changes from a liquid state to a semi-solid state in the presence of a magnetic field. In the absence of a magnetic field, the viscosity of the fluid is a function of the properties of the carrier fluid, the suspending agents and the fraction of magnetizable particles. The rheological behavior of fluids is analogous to that of non-Newtonian fluids, which have yield stresses dependent on the applied magnetic field, and these characteristics can be controlled by manipulating the intensity of the field [4], [5].

Since the discovery of magnetorheological fluids, a great deal of research has been done [6], [8] to improve their properties and optimize their applications. To observe the behaviour and effects of external forces on the particles inside the carrier fluid, the finite element method has been used with the ANSYS [6] computer package. Also, Y.B Kazakov et al [7] used the finite element method to carry out the analysis of a magnetorheological damper taking into account the mutual influence of electromagnetic, hydrodynamic and thermal fields. The results of the finite element simulation are validated by tying with analytical solutions, which has allowed the use of this tool in the design of devices with this technology. O. Arteaga et al [8] implemented an FMR in prostheses for patients with lower limb amputations, which provides comfort and autonomy for proper functioning that depends on the activity performed or the environment in which it is moved. The effectiveness of the prosthesis is demonstrated experimentally. An MR damper was used to measure its magnetorheological characteristics, such as the damping force, where it was determined that to generate optimal damping, a lower magnetic field induction is necessary to perform activities such as walking.

One of the main challenges for those who study FMR is to obtain the best magnetorheological effect since problems such as sedimentation and aggregation of magnetizable particles commonly occur, producing the detriment of such effect. This article reviews the specialized and updated literature describing the main parameters related to the behavior and properties of MRFs. The understanding of all the aspects involved in FMR allows the reader to have a broad view of the technological innovations through the optimized applications of these intelligent materials.

Properties of magnetorheological fluids

The magnetorheological effect is characterized by a reversible increase in the viscosity of a fluid and shows an upper limit of creep due to the introduction of a magnetic field that can be explained by the formation of chains of particles. The magnetorheological effect can be controlled by the strength of the magnetic field acting on the rheological characteristics of the FMR constituents, in other words, the controllable rheological characteristics of an FMR are attributed to the polarity, of the suspended particles, induced by the magnetic field [2].

In the presence of a magnetic field, each magnetic particle is transformed into a dipole and forms a chain with its adjacent particles that can resist deformation at a certain shear rate and consequently provides a semi-solid structure. The interactions between these induced dipoles cause the particles to align along the applied field and form a columnar structure. The chain-like structure will resist the movement of the fluid and therefore increase the viscosity of the suspension. The mechanical energy required to overcome this chain-like structure is increased proportionally to the applied magnetic field [4].

When the shear rate exceeds an extreme value, the chain structure will break and the fluid will flow, the stress that supports an FMR at this intense shear rate is called the apparent fluid yield stress. The yield stress is a crucial factor in industrial applications of FMR ranging from 10 to 100 kPa in a specific magnetic field range. This factor depends on the shape, the size distribution, the volumetric fraction of the particles, the applied magnetic field strength, the interactions of the particles and the formation of agglomerates. Table 1 summarizes the main properties of FMR [2], [4].

Table 1. Typical properties of FMR, taken from Kumdhar et al [9]

Property	Typical value
Initial viscosity	0.2-0.5 [Pa s](a 25 °C)
Density	3-4 [g/cm^3]
Magnetic field strength	150-250 [kA/m]
Elasticity limit	50-100 [kPa]
Reaction time	15-25 ms
Working temperature	-50 a 150 °C
Supply voltage and current	2-25 V, 1-2 A

Creep shear is one of the main characteristics of FMR and is derived from its non-Newtonian behavior. An FMR behaves according to Bingham's law, which means that it exhibits a different shear value, behaving more like a solid than a liquid. The shear value without shear rate is called yield stress in an FMR and is controlled by the magnetic field applied to it. The higher the elastic limit, the higher the stress that the material can withstand without flowing [10] - [2].

The angle of the magnetic field applied to the FMR has a clear influence on its properties. The properties and the friction between the fluid and the surrounding walls have been investigated experimentally [13].

Composition of magnetorheological fluids

After choosing the fraction and the type of materials to be used for its composition, the FMR are prepared by mixing the three main components: base fluid, magnetizable particles (dispersed phase) and stabilizing additives. A descriptive scheme of the components of an FMR is shown in figure 1. The base fluid behaves like a vehicle containing the magnetic particles in suspension, which perform the essential function on the magnetorheological effect. Stabilizing additives are used to combat the problem of sedimentation and aggregation of the magnetizable particles. One of the ways to obtain an FMR is to add the stabilizing additives before adding the magnetic particles, with this an effective preparation of the FMR can be achieved [4].

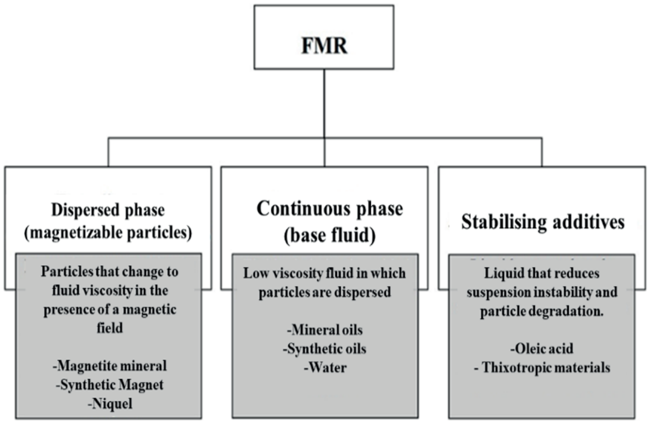

Figura 1. Principales componentes de un FMR [4].

Continuous phase (base fluid)

The base fluid keeps the magnetic particles in suspension and as its viscosity is lower the MR effect is improved. Generally, mineral oils, synthetic oils, water, glycol and others are used. Another very important characteristic of the carrier fluid is its low vapor pressure, since it does not vaporize easily, therefore, it can be used in a wide range of temperatures [4], [14].

C. Mesquida et al [15] made a comparison between an FMR containing water and oil and another one prepared only with oil as carrier fluid, and concluded that FMR without water content present better rheological properties than those containing it. They also found that oils with low viscosity generate less energy expenditure for the generation of the magnetic field.

Dispersed phase (magnetizable particles)

There are several materials that can be used as dispersed phase, it is sought that these present the lowest coercitivity and the highest saturation magnetization, so that as soon as the field is removed, the particles return to their initial state of demagnetization, table 2 shows the characteristics of some materials used as dispersed phase in an MR fluid.

Table 2. Materials and their characteristics used as a dispersed phase in an FMR

Particles	Size	Magnetic field	Carrier fluid	Reference
Nickel-Carbonyl	Diameter 30 nm	Up to 0.6 T	Silicone oil	Bell and col. [17]
	Length 5-25 µm			
Carbonyl Iron	7 µm	0-343 kA/m	Silicone oil	Esmaeilnezhad and col. [18]
Magnetite	100 nm			
Magnetite ore	44.5 µm	0-1200 Gauss	Automotive oil	Rojas and col. [5]
Magnetite ore	0.025- 0.045 µm	0-200 Gauss	Automotive oil	Molina and col. [6]
Synthetic magnetite	13-17 nm	2100 Am^{-1}	Water	Dyachenko and col.[19]

Nickel has been little researched for application in RFM but has an advantage over iron particles as it is less prone to oxidation [16]. Bell et al [17] investigated the influence of particle shape by considering nickel carbonyl spheres with diameters between 1 and 10 µm and nickel nanowires of 300 nm diameter and 5 to 25 µm length and found that both FMRs are composed of different particles in shape and size, but with the same volumetric concentration (7. 6%) have a similar behavior with respect to the yield stress measured in the presence of a 0.6 T magnetic field. It was concluded that the elongated particle FMR has a better response against sedimentation than the one composed of spherical particles. Another material that has been used as a dispersed phase is iron carbonyl as described by Esmaeilnezhad et al [18], they synthesized an FMR with such material as a dispersed phase, reported that magnetite nanoparticles adhere to carbonyl particles filling the spaces between them, increasing the stability of the fluid and consequently decreasing the sedimentation rate.

Another material commonly used as a dispersed phase in FMR is magnetite (Fe3O4), which can be obtained in a mineral and/or synthetic state. Rojas et al [5] reported the development of an FMR with magnetite obtained from mining waste with a particle size of 44.5 µm, in concentrations of 5, 10, and 15 % in volume and with variations in the magnetic field strength of 0, 50, 100, 300, 600 and 1200 Gauss, concluded that for the same magnetic field strength, the higher the concentration of mineral magnetite particles, the higher their viscosity. Likewise, Molina et al [6] used mineral magnetite as a dispersed phase, however, they used a particle size ranging from 0.025 to 0.045 µm in concentrations of 15 and 20% by volume, as well as Rojas et al [5] inferred that the fluid with the highest volumetric concentration and the smallest particle size presents the best magnetorheological behavior in the presence of the magnetic field. One of the ways to obtain synthetic magnetite is by chemical coprecipitation as shown in Dyachenko et al [19], they synthesized an FMR using magnetite as a dispersed phase, the magnetite synthesis was carried out in a temperature range from 25 to 80 oC obtaining a crystallite size between 11.6 and 19.9 nm, it should be noted that the size is proportional to the increase of the synthesis temperature.

Additives (stabilizers)

The additives are necessary to control the viscosity of the fluid, the friction between particles, besides avoiding the thickening of the fluid after several cycles of use, they can be classified as stabilizers and surfactants. The additives give the system properties such as good adhesion, particularly to particles, anti-corrosive effect and stability at high temperature (~ 150 oC). Additionally, high viscosity additives such as fat or other thixotropic materials are added to the base fluid to improve the stability of the particles against sedimentation [4], [20].

A wide range of materials can be used for FMR stabilisation effects, e.g. Jinaga et al [20] implemented lithium grease as a stabiliser for mixtures of silicone oil, iron carbonyl particles and the commercial additive Triton X-100, finding a clear influence on viscosity and sedimentation rate [20]. Mesquida and Lässing [15] studied the effect of two surfactant additives, the sodium acid dodecylbenzene sulfate salt and versamul (in solution state), obtaining better performance with versamul in terms of viscosity and sedimentation. Oleic acid and sodium oleate show good properties as stabilizers, Dyachenko et al [19] compared the properties of an FMR with both stabilizers, added to the carrier fluid and the magnetic particles were added, with permanent agitation of 600 rpm for 30 minutes to homogenize, finding that oleic acid delays more the sedimentation of the particles. Proaño [21] used oleic acid in the FMR in volumetric percentages of 40 and 50 %. The fluid used in the suspension system of a buggy vehicle showed good performance since this type of vehicle generally operates in extremely irregular terrain.

Stabilisation of magnetisable particles by means of a coating

In recent years research has focused on stabilizing magnetic particles by means of a coating, obtaining a nucleus-core structure to overcome sedimentation problems in FMR. The shell-core structure is widely used as they often have improved physical and chemical characteristics. The shell can efficiently prevent the aggregation of particles and protect them from the effects of external degradation [4].

R. Gu et al. [22] present in their research an iron particle structure covered by a silica layer as shown in figure 2, the dark particles encapsulated in the silica layer are observed. The synthesis of the armoured particles was carried out by the hydrogen reduction method using the iron particles in spherical form, the thickness of the silica shell is approximately 10 nm and this is directly proportional to the concentration of tetraethyl orthosilicate (TEOS). The resulting hollow structures give the composite particles a low density, which is beneficial for addressing sedimentation problems in FMR. The Fe-SiO2 composite particles had a saturation magnetization value of 48.56 emu/g making them suitable for application as a dispersed phase in FMR.

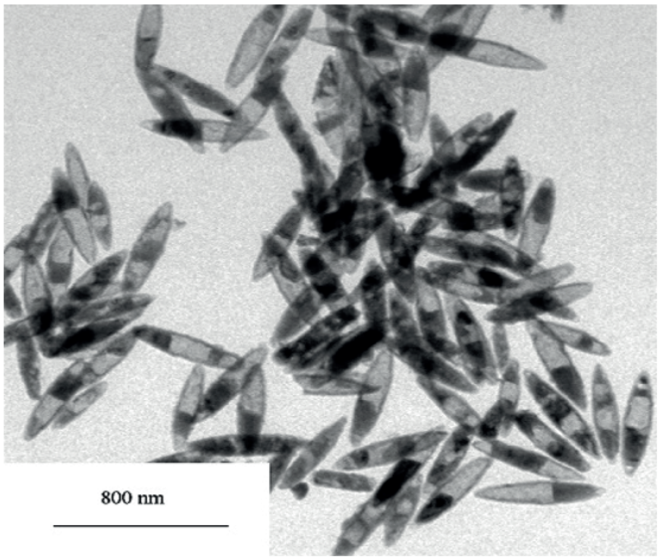

Figure 2. TEM image of FeSiO2 particles with the nucleus-hell structure, taken from R. Gu et al [22].

Andrade et al. [23] reported a silica coating on magnetic iron nanoparticles, as did Gu et al. [22] used tetraethyl orthosilicate (TEOS) as a precursor deposited by the sol-gel method. The research shows through zeta potential measurements that the coating on the nanoparticles was effective and that it did not affect the magnetic capacity of the nanoparticles, in addition, the particle size decreased by approximately 30 % after the coating, probably due to the expected effect of the silica coating that prevents the agglomeration of nanoparticles.

More recent studies [24], [25] agree that tetraethyl orthosilicate (TEOS) is widely used as a precursor for the coating of magnetic particles. As in previous investigations, it is possible to obtain a shell-core coating and the thickness of the outer layer is proportional to the concentration of the TEOS, and the superparamagnetic effect of the particles is not affected since the silica layer keeps them isolated from the base fluid, preventing oxidation and degradation.

Due to their silica content, river sands are one more option to coat iron particles. Rasheed and Meera [26] compared a biological method with the traditional sol-gel method for the coating of iron particles, concluding that biological methods are a better way to coat iron particles since the highest percentages of coated surface are obtained.

Silva et al. [27] synthesized magnetite (Fe3O4) nanoparticles with polysaccharides, and in the investigation they describe that the coating was done by the chemical coprecipitation method. The particle size obtained fluctuates around 10 nm with a superparamagnetic behavior with zero coercitivity and remanence. In addition, they found that in the particles coated with polysaccharides the magnetization magnitude is higher than those that are not coated, this is mainly due to the fact that the coating weakens the interaction between the particles, which in turn reduces the disorder of the spines on the surface, thus increasing magnetization.

Recently, Divandari et al [28] reported a citric acid coating on magnetite nanoparticles, the synthesis was performed by a simple and economic process obtaining nanoparticles with a citric acid shell with spherical morphology, in figure 3 is shown the schematic representation of the magnetite coating. The researchers conclude that the citric acid coating has a positive impact on sedimentation effects and on magnetic induction.

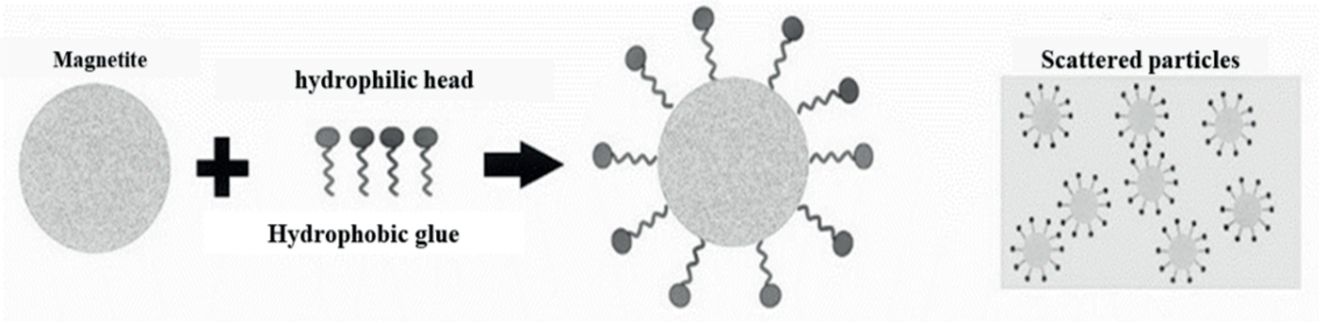

Figure 3. Diagram of citric acid coating on magnetite, taken and modified from Divandari et al [28].

Magnetorheological fluids applications

In recent years, efforts have been multiplied in the study of magnetorheological fluids and their application ranging from the automotive industry to medicine. Researchers have focused on optimizing the properties of these fluids to generate the best performance when they are put into service.

Figure 4 shows a schematic representation of the main applications of FMR.

Automotive industry

Suspension system

The automotive industry is one of the branches that has benefited the most since the invention of these fluids since they have been able to be applied in several automotive components improving the performance of the car, an example is the car suspension, this system is connected from the chassis to the wheels and consists of a shock absorber, a spring and links. The suspension systems have the purpose of contributing to the handling and braking on the road for a good operational safety and the driving pleasure and comfort of the occupants and, consequently, to keep them isolated from the shocks and vibrations generated by the road. It is important that the suspension keeps the tyre in contact with the road as much as possible for better traction. Damping is the reduction of movement or oscillation with the use of hydraulic valves and dampers on a vehicle's shock absorber. With proper damping the car will return to its normal condition in a minimum time [29], [30].

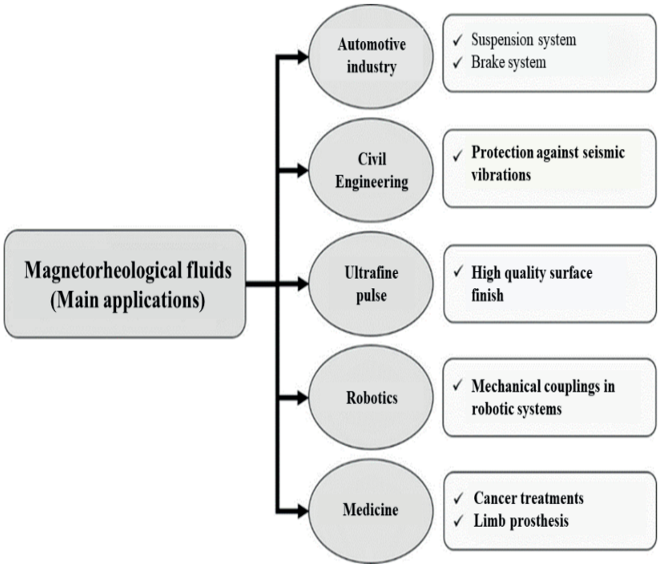

Figure 4. Main applications of magnetorheological fluids

Vibration control systems have been proposed using the semi-active mode, i.e. an on/off control where the desired asymmetric and variable damping characteristics of a magnetorheological damper used to counteract vibrations generated in moving cars are obtained [31]. On the other hand, there are permanent magnet dampers that are placed symmetrically in an external cavity of the cylinder. These generate the magnetic field that interacts with the FMR, keeping it permanently active and thus improving the damping in the presence of vibrations [32]. Some cars in the market already have magnetorheological technology, brands such as Cadillac, Ferrari, Audi and GM's Corvette use this type of device in their suspension systems [33].

Braking system

A magnetorheological brake consists of a rotating disc in an FMR enclosed in an electromagnet, the elastic limit of the fluid varies according to the applied field. MR brakes have recently been studied as an alternative to conventional hydraulic brakes for road vehicle applications. Kumbar et al. [9] synthesised several samples comparing their behaviour, under different percentages in volume of the magnetic particles and the carrier fluid, finding that the carbonyl iron particles produce a considerably higher saturation magnetisation, making them more suitable for use in MR brakes. For this reason the silicone oil fluid with iron-carbonyl particles is the most suitable for this application [9].

Medicine

In the case of biomechanical prostheses to replace amputated limbs, both aesthetic and functional functionality are sought, which is why Arteaga et al [8] have recently taken on the task of designing robotic prostheses with FMR in the cushioning system. The purpose of this prosthesis is to allow the patient to walk properly according to the environment in which they find themselves and the activity they carry out, providing them with comfort and autonomy. Derived from the experimental tests, these researchers conclude that for walking the magnetic field induction should be low, while for running a higher induction is required. Another application of FMR in medicine is cancer therapies and controlled drug release. Flores et al [34] reported that it is possible to use water-based RM fluids with relatively low magnetic particle fractions ranging from 0.25 to 5% which are induced through 3.2 mm diameter tubes with a magnetic field intensity of 60 mT through tumor necrosis.

Civil engineering

The proof of the increased growth in magnetorheological device technology is that shock absorbers of a size suitable for civil engineering applications have been designed and built for protection against natural vibrations and seismic shocks [33], [35]. Lin et al. studied the performance of a structure supported by a hybrid base isolation system that includes a rolling pendulum system and a 20kN MR damper. The system was tested on a shaking table monitored by motion sensors and data was collected on a controller. The study showed favourable results for the application of the system on a large scale [36].

Robotics

To ensure safety during human-robot collaboration, safety methods are required for the tools. Since standard industrial robots are capable of moving large objects at high speeds and the mechanical structure of the robot involves large moving masses, it represents a significant danger to human operators. Lämmle evaluated the development of a mechanical safety coupling using MR fluids. This acts as an inherent safety joint substance, and it was shown that decoupling the effective mass of the robot significantly reduces the energy transmitted to the affected parts of the human body, thus causing less damage in case of a collision [37].

Ultrafine polishing

The FMR-based surface finishing process is a process that is applied to a wide variety of materials, ranging from optical glass to steels. Under the influence of a magnetic field, magnetic particles and non-magnetic polishing particles remove the material from the surface being polished [38]. The process consists of the rotation of the workpiece on the surface to be polished, the imposition of a magnetic field on the FMR helps to improve the surface finish [39]. Recently Singh et al. reported that it is possible to use an FMR for finishing cold rolled steel, as the surface of the rolls must be as smooth as possible because the surface quality of the finished products being processed will depend on it [40].

Conclusions

Today, FMR has taken on great importance in a wide variety of industrial applications, as well as in medicine. Researchers have focused on optimizing their properties for better performance and utilization. However, there are still challenges to be overcome, as problems such as sedimentation and aggregation of magnetizable particles are the main current problems with these materials. The choice of the carrier fluid, as well as the type, size, morphology and fraction of magnetizable particles are also an important part of FMR preparation since the response of the fluid in the presence of the applied magnetic field strongly depends on it. According to the studied literature, the iron carbonyl particles are one of the most used as dispersed phase since they present excellent saturation magnetization, good stabilization, but above all relatively low synthesis costs.

Another important challenge for this discipline is the stabilization of the magnetizable particles, one of the means to achieve it is to make a coating on them. The most recent researches point out that silica is the most profitable material to carry out this procedure since a film is generated with a thickness dependent on the concentration of the precursor and the time of synthesis. This film keeps the nucleus isolated from the medium in which it is found, providing it with stability and avoiding degradation. However, as well as silica there are other materials that can also be used for the same purpose, such as: citric acid, polysaccharides and some polymers. The formulation of FMR implies having an optimal balance for the different applications it is given, this means that a component for the automotive suspension system will need different carrier fluid, as well as a fraction of magnetizable particles than a shock absorber used in a prosthesis for people with lower limb amputations. Researchers will need to take these aspects into account to obtain an FMR with the best properties to meet the needs of the application.

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