Ultrasonics 61 (2015) 10–14

Contents lists available at ScienceDirect

Ultrasonics

journal homepage: www.elsevier.com/locate/ultras

Short Communication

# Study of the effect of particle volume fraction on the microstructure of magnetorheological fluids using ultrasound: Transition between the strong-link to the weak-link regimes

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ARTICLE INFO

*Article history:*
Received 17 January 2015
Received in revised form 25 March 2015
Accepted 27 March 2015
Available online 8 April 2015

*Keywords:*
Magnetorheological fluids
Ultrasonic characterization
Strong-link regimen
Weak-link regimen
Magnetic field

ABSTRACT

The effect of particle volume fraction on the microstructure of magnetorheological (MR) fluids has been studied using ultrasonic techniques. When no magnetic field is applied, they behave as slurry. However, when magnetic field is applied, important features regarding the change of the microstructure have been found with the help of ultrasonic waves propagating in the direction of the magnetic field. As the volume fraction increases, a rearrangement of particles which decrease the compressibility of the system is detected; nevertheless, the material behaves as a non-consolidated material. Three different particle volume fraction regions are found identifying a critical particle volume fraction predicted in the literature. Ultrasounds are confirmed as an interesting tool to study MR fluids in static conditions.

## 1. Introduction

Magnetorheological (MR) fluids are smart suspensions with tunable properties by applying an external magnetic stimulus. When no magnetic excitation is applied, they are isotropic suspensions of magnetic particles in a carrier fluid; however, when they are subjected to a magnetic field, the particles interact among them and the MR fluid becomes a structured and anisotropic material. The rearrangement of the particles into the MR fluid is the main mechanism responsible of the change of its mechanical properties [1–4].

In these kinds of smart systems, particles play the main active role. Particles which can be oriented by external magnetic fields can be used in these formulations; however, carbonyl iron powder (CIP) is the most widely used due to the reversibility and reproducibility of the magnetorheological effect [5]. Although CIP shows very good performance, it also presents some drawbacks regarding long term practical applications like sedimentation, abrasion or oxidation issues and difficulty to be redispersed. Nevertheless, these problems have been overcome coating them with, for example, organic materials [6], silica based coatings [7], cholesteryl functional groups [8], graphene oxide [9] or Zinc oxide [10] among others.

The microstructure formed under the effect of the magnetic field depends on many factors: the shape, the nature and concentration of the particles, the properties of the carrier oil, the presence of surfactants and thickening agents, the uniformity and intensity of the magnetic field applied and the size of the cell where the fluid is studied [11–16].

These fluids are used in different industrial applications as it is the case of the automotive industry where they are components of high tech breaks and clutches. The MR fluids are also used as seals or earthquake absorbers [3,17–19]. As the properties of the MR fluids are strongly dependent on their inner structure, a better knowledge of the microstructure as a function of the external magnetic field will permit a better understanding of the response of these fluids and, therefore, an improvement of their industrial applications. To this end, different scientific works have been made to infer the microstructure of MR fluids from their optical, rheological or acoustical properties, both in static and dynamic conditions [16,19–25]. Different models have been proposed to explain the structure formation and the yielding process related to the application of a shear stress in MR fluids. Models have been usually applied successfully for very low particle volume fractions [26–28], while for higher volume fractions, simulations are compulsory [15,29,30]. The yielding of MR fluids of iron particles suspended in a low viscosity oil was studied as a function of the particle volume fraction by Segovia-Gutiérrez et al. [23]. In that work, a two-step behavior was observed when they were subjected

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http://dx.doi.org/10.1016/j.ultras.2015.03.011

to intermediate magnetic fields. This feature was explained as a transition from a strong-link to a weak-link particle regime when particle concentration was increased. Considering the situation on which the magnetic field energy is higher than the thermal energy, the rearrangement of the particles is induced. For low particle volume fraction, the fluid will be in the strong regime: the links between chain aggregates (transversal direction) are stronger than the links which keep particles forming the chain-like structures – longitudinal direction. Consequently, if any breakage under shear stress takes place, the breaking of bonds will occur within a chain-like aggregate. However, when particle volume fraction is high, the chains are thicker and there are lots of transversal connections between them. The chain aggregates are more resilient than the interlinks between different structures so two different breaking mechanisms are found; this is the weak-link regime. The particle volume fraction which gives way from one regime to another is a critical or threshold particle volume fraction which depends on the features of the suspension [23].

Ultrasonic techniques are very convenient to study MR fluids. They are non invasive, universal and give results on real time [31,32]. As ultrasonic propagation is sensitive to the inner structure of the fluid, changes in the mechanical stiffness and density in one direction will be accompanied by changes in the ultrasonic velocity and attenuation along this direction [33–35]. Due to attenuation mechanisms, acoustic filters or active matching layers associated to changes in the velocity have been studied using ultrasound [24,36]. Sedimentation processes, thermal behavior, hysteretic effects, yielding processes and evolution of the inner microstructure of MR fluids have been studied with these techniques [25,37,38].

In the present paper, a through-transmission ultrasonic technique has been used to measure the variations of the velocity of sound as a function of particle volume fraction in static conditions. Three different regions regarding the kind of structure have been found, identifying the particle volume threshold between the strong-link and the weak-link regimes. Pictures of the inner structure at the three regions have been taken.

## 2. Materials and methods

Ten MR fluids of different particle volume fractions – from 0.01 to 0.35 – of carbonyl iron powder, grade CC (BASF), suspended in an epoxy resin with a viscosity of 1000 mPa s have been made. Due to the high viscosity of the host fluid, the suspensions are stable for at least 3 h as can be deduced from the velocity of sound measurement which remains constant. The carbonyl iron powder used has a magnetization saturation of 190 emu/g at 9 kOe at room temperature and a diameter between 3.8 μm and 5.3 μm. This particular grade has been chosen as particles show good ferromagnetic response and they are easily dispersed in the carrier fluid.

Two piezoceramics – PZ 27 Ferroperm – with a frequency resonance of 1 MHz were used as the emitter (A) and the receiver (B). An electromagnet with 212-mT magnetic field (C) was used applying the field parallel to the ultrasonic propagation (D) as it is shown in Fig. 1b. The fluids were introduced in a $33 \times 33 \times 6$ mm$^3$-cell, made of methacrylate, being the ultrasonic path 6 mm.

The cell (1) was placed between the two poles of the electromagnet (5). The electromagnet, controlled by a current source (6), provides a very uniform magnetic field which affects the whole measuring cell.

The ultrasonic emitter (A) was excited by a function generator – Agilent 33250A – (2). The ultrasonic received signal was acquired by an oscilloscope – Tektronics 1020 – (3) connected to a PC (4). The phase velocity of the ultrasonic waves was obtained from their spectrum phase using a FFT algorithm [39].

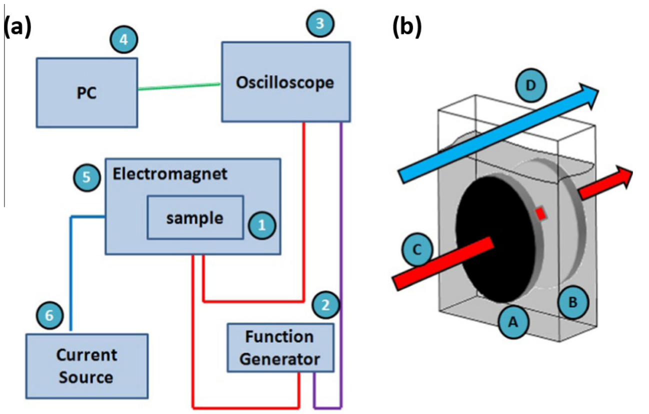

**Fig. 1.** (a) Experimental set up. (b) Detail of the experimental set up where the cell is shown. The blue arrow (D) indicates the ultrasonic direction of propagation, while the red (C) one shows the direction of the magnetic field applied. Both fields are parallel. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

The fluid fabrication procedure consists in mixing thoroughly the particles with the carrier fluid with a stirrer for about 10 min at 100 rpm till the mixture becomes homogenous slurry. After this step, the sample is introduced in a vacuum chamber at 1 kPa for 15 min to remove all the air bubbles. Next, the sample is poured into the cell and kept at 30 °C for 1 h until the sample reaches the thermal and mechanical equilibrium. After this step, the experiments are carried out, maintaining a constant temperature, in order to avoid fluctuations of the velocity induced by thermal variations.

## 3. Results

In Fig. 2, it is shown the longitudinal velocity of sound as a function of the particle volume fraction ($c^{\phi}$) when no magnetic field is applied (subscript $X_0$, dark squares) and when the samples are subjected to a 212-mT parallel magnetic field (subscript $X_B$, light circles).

When no magnetic field is applied, the typical behavior of a suspension of high dense solid particles in a fluid is obtained. As a general trend, the velocity of sound depends inversely on the square root of the compressibility ($\kappa^{\phi}$) and density ($\rho^{\phi}$), $c^{\phi} = (\kappa^{\phi}\rho)^{-1/2}$. Measuring the velocity and the density, the compressibility $\kappa_0^{\phi}$ is calculated. As it is shown in Table 1, the higher the particle volume fraction, the lower the compressibility and the higher the density;

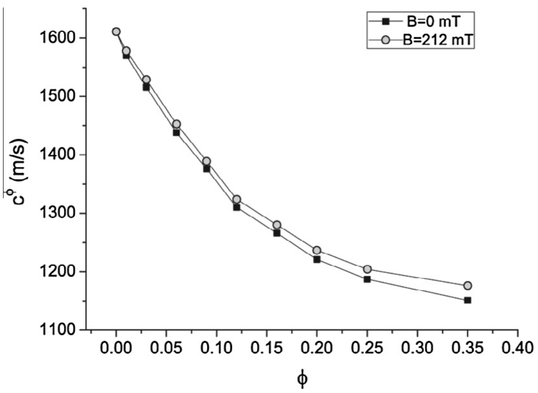

**Fig. 2.** Velocity of sound as a function of the particle volume fraction ($c^{\phi}$), when no magnetic field is applied (dark squares) and when the sample is subjected to a 212-mT magnetic field parallel to the ultrasonic waves (light circles).