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## Structure and physical properties of magneto-rheological slurries under perturbation

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Dispersions of micrometric magnetic particles in inert liquids acquire a complex multifractal structure when they are exposed to a static magnetic field. If in addition to the static field it is applied on the dispersion an oscillatory magnetic perturbation, the structure formed by the particles, and consequently, some of the physical properties of the system, can be strongly affected. Under certain conditions, by the influence of the perturbation, the structure in the dispersion can be reconstructed becoming an ordered one. In this work, these phenomena are discussed addressing the transformation of the structure in the scheme of a glass transition. For low particle concentration, the pattern formation induced by the fields is quite different. The relationship between the pattern formation and the rheological properties of dilute dispersions are also discussed.

**Keywords:** Glass transition; magneto rheological dispersions; reconstruction processes; aggregation under perturbation.

Las dispersiones magneto reológicas son partículas micrométricas de magnetita dispersadas en líquidos inertes, las cuales forman una estructura compleja multifractal cuando son expuestas a un campo magnético estático. Si en adición a este campo estático se aplica a la dispersión, una perturbación magnética oscilatoria, la estructura formada por las partículas, y de manera consecuente, algunas de sus propiedades físicas del sistema, se ven afectadas de forma considerable. Bajo ciertas condiciones experimentales, por efecto de la perturbación, la estructura en la dispersión puede ser reconstruida llegando a ser una estructura ordenada. En esta investigación, este proceso es discutido con atención en la transformación de la estructura bajo el esquema teórico de una transición vítrea. Para bajas concentraciones de partículas, la dinámica de formación de estructuras inducido por el campo magnético es ligeramente diferente. La relación entre el patrón de formación y las propiedades reológicas de dispersiones diluidas también se discute en este trabajo.

**Descriptores:** Transición vítrea; dispersión magneto reológica; proceso de reconstrucción; agregación bajo perturbaciones.

PACS: 83.80.Gv; 45.70.Qj; 83.60.Np

### 1. Introduction

Micrometric magnetic particles dispersed in inert liquids exhibit complex aggregation processes when they are exposed to a static magnetic field. The most distinctive characteristic of a moderately concentrated ( $\approx 0.1$  in volume fraction) magneto rheological dispersion under this conditions, is the multifractal structure formed by the particles [1]. It is due to the sequential formation of clusters of different generations which posse a well defined average size and mass fractal dimension. This is a consequence of the predominance of the different interactions during the aggregation process, which starts with the application of the field. A characteristic time of formation of the clusters for these generations can be easily estimated [1], and some of the physical properties of these composites can be calculated on the basis of the knowledge of the fractal structure of the dispersions [2]. If in addition to the static field is applied a transversal magnetic perturbation, important changes can be induced in the structure of the dispersion [3]. For dilute dispersions ( $\approx 0.02$  in volume fraction), in the presence of both fields, the pattern formation exhibits important differences [2,4]. The static field induces the formation of chain-shaped clusters and the perturbation enhances the formation of larger and thicker chains [5]. Here,

for moderate particle concentrations, we address the study of the time evolution of the structure due to the application of both fields, in analogy to the transformations of the structure in the glass transition suffered by a supercooled liquid [7].

#### 1.1. Experimental

For low and medium concentrations, we observe by optical microscopy the patten formation. The rheological dispersions were prepared as follows: Magnetita's particles of average size  $30\ \mu$  were dispersed in mineral oil (Dextron, 76 cP). The dispersion was poured into a circular glass cell of 5 cm diameter, designed to fit on the microscope stage. This cell allows water circulation, in order to control the sample temperature. The magnetic fields were applied by means of two pairs of Helmholtz coils allocated in such a way that the static and perturbation fields become transversal each other. The perturbation in all the experimental series was kept about 10% of the intensity of the static field. Additionally to the observations by means of the optical light polarized microscope, we conduct experiments to observe the effect of the perturbation on the viscosity of the sample under different conditions. The effect of the perturbation on the rheological properties of the dispersion is particularly important for the low particle concentration. For dilute dispersions these rheological mea-

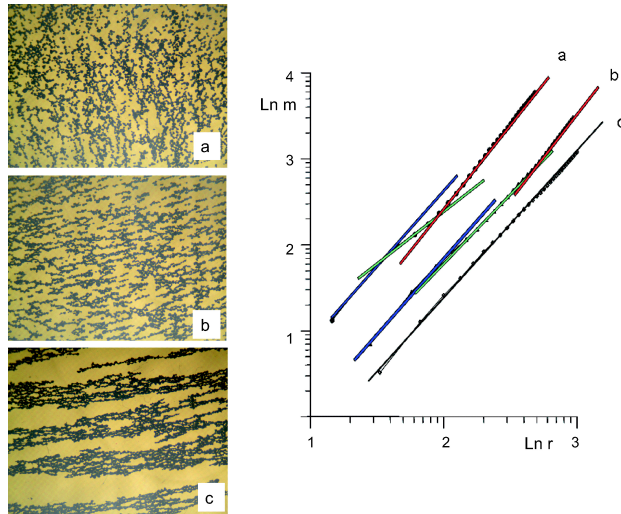


FIGURE 1. Photographs corresponding to three stages of the structure time evolution with its corresponding graph of the scaling relation  $\log_{10}(m) - \log_{10}(r)$ .

measurements were performed by means a Brookfield rheometer LVDV-III.

## 1.2. Fractal structure

To analyze the fractal characteristics of the structure formed by the particles in the dispersion, the time evolution caused by the application of the static and oscillatory fields, is recorded by means a digital camera coupled to the microscope, from this film some photographs are obtained at different stages and a multifractal analysis is performed on them. Figure 1 shows photographs of three different stages of the reconstruction process. The photograph a) corresponds to the initial condition when only the perturbation field, 12 G amplitude and 4 Hz frequency, is applied. On the right side appears the graph  $\log_{10}(m) - \log_{10}(r)$  obtained by a box counting method. The slope of the right portions is the so called mass fractal dimension.

Curve and image a) show the characteristic three straight segments related to the formation of three generations of clusters. Photograph b) shows the structure in a stage few seconds after the instant which also the static field was turned on. The corresponding graph shows the structure modification in terms of the changes in the scaling relations at different ranges of the radial distance. The segment corresponding to the second generation clusters exhibits a clear reduction as a consequence of the perturbation. Curve and image c) show the situation some time after the perturbation and the static field were acting on the dispersion. The perturbation has led the system to an homogeneous ordered structure, with a slope in the graph  $\log_{10}(m) - \log_{10}(r)$   $D \simeq 2$ .

The multifractal characteristics of the magneto-rheological dispersion exposed to the static and perturbation fields, can be corroborated by means of a more detailed multifractal analysis. This analysis was made by using the plug-in FracLac for the free package ImageJ. Figure 2 shows

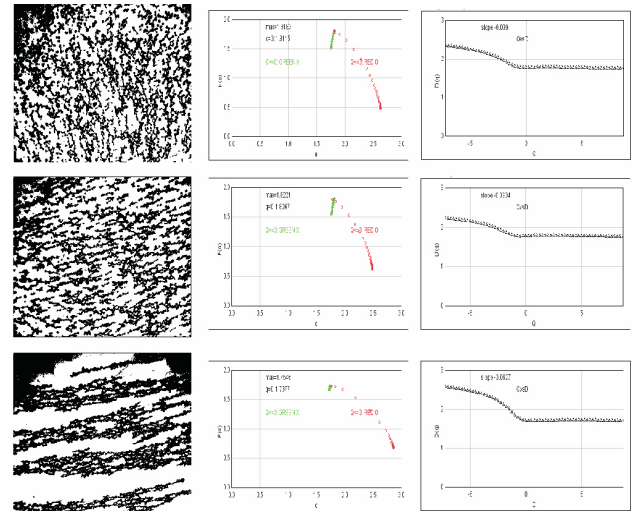


FIGURE 2. Three stages of the time evolution of the structure of a magneto-rheological dispersions under perturbation. At the right side there appears the corresponding multifractal characterization of the structure.

three different stages of the time evolution of a magneto-rheological dispersion exposed to both fields. The upper image correspond to an initial stage, immediately after the simultaneous application of the static and perturbation fields. At the right side of the photograph appears its fractal characterization in terms of the behavior of the spectrum  $f(\alpha) V_s \alpha$  and the behavior of  $D(Q) V_s Q$ . In this scheme if the graph is humped as in the upper figure, the structure is considered a multifractal object. If the graph converges as in the lowest figure the structure is considered non-fractal. Additionally, if the graph of  $D(Q) V_s Q$  is decreasing this is a characteristic of a multifractal object, but if the behavior of the curve is non-decreasing it indicates that the object is mono-fractal or non-fractal.

## 2. Glass transition analogy

A theoretical approach to understand the nature of the glass transition, is to address this phenomena as the result of a process, in which a molecular liquid is supercooled at a fast rate, in such a way that the crystallization is prevented. Usually, this glass transition is distinguished from the crystalline transition by using a molecular time of relaxation, which characterizes the threshold rate of cooling leading the liquid to a glass or a crystal. We wish to analyze the transformation which undergoes the structure formed by the particles in a magneto rheological dispersion, caused by the application of a static as well as an oscillatory perturbation, by making an analogy with the glass transition. We assume that in the structural transformation undergone in a magneto rheological dispersion due to the applied fields, the rate of the perturbation amplitude to the intensity of the static field, has the role that the temperature has driving the transition in a supercooled liquid. The experimental procedure to determine the analogous of the molecular time of relaxation  $\tau_m$  is the following:

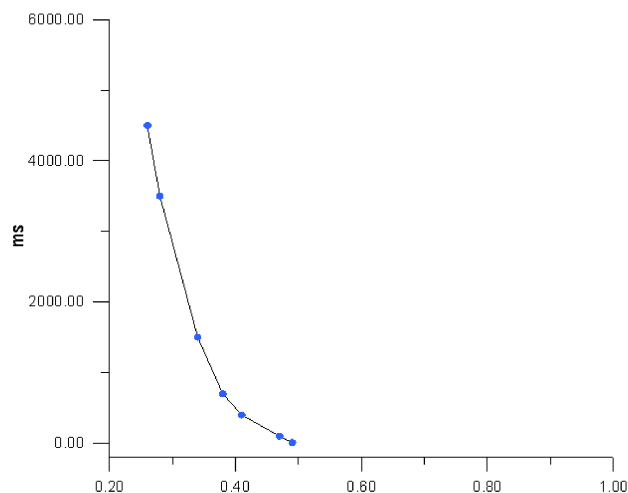


FIGURE 3. The measured relaxation time  $\tau_m$  as a function of the particle concentration. The oscillatory perturbation was kept to 12 G.

the dispersion is prepared at a given concentration of particles and the oscillatory perturbation 12 G is turned on, then the static field is applied by steps up to a final intensity of 120 G. The number of steps per unit time to reach the final intensity of the static field determines the height of the step. In this series of experiments one tries to determine the height of the steps, it is the characteristic time of relaxation, which distinguishes when the evolution of the structure ends with an ordered (crystalline) structure or a disordered one. A detailed analysis of the structural transformation in analogy with glass transition on the basis of the calculation of thermodynamic quantities is presently in progress [6].

### Effect of the perturbation on the viscosity

In the low particle concentration regime, the perturbation generates new aggregation mechanisms that produce a faster aggregation of chains [5]. In turn, it causes a larger and faster increment on the measured viscosity [5]. The behavior of the long time effective viscosity was discussed in detail in Ref. 5, here we focus our attention in the interval of time when the rheological changes occur abruptly. In the short time regime the application of the fields induces the formation of the chain shaped clusters. The length and thickness of these chains influence the effective viscosity of the dispersion. Figure 4 shows the effective viscosity of the dispersion for three different conditions. The upper curve shows in a logarithmic graph, the short time behavior of the viscosity when a static (90 G) and perturbation (12 G rms, 2 Hz) fields are applied simultaneously. In the lower curve appears the viscosity when only a static of 102 G field is applied. Notice that when both fields are applied simultaneously, the value reached by the effective viscosity after some few dozens of

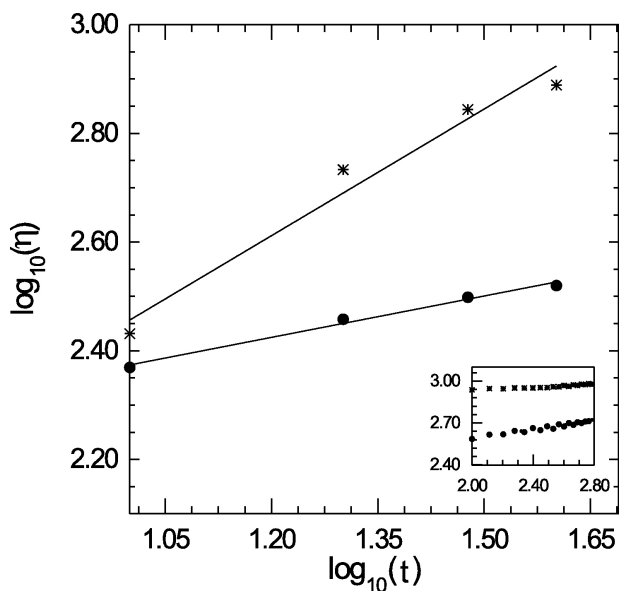


FIGURE 4. The short and long (inset) time behavior of the viscosity as a function of time showing the relative increment caused by the simultaneous application of both fields. In the lower curve the perturbation was applied with a delay of 800 seconds.

seconds, is several times larger than that reached by the application of a static field, this is true even if the intensity of the static field is larger than that applied simultaneously with the perturbation. Obviously this is due to the comparatively larger average length of the chains formed when both fields are present.

The long time behavior of the effective viscosity is shown in the inset. The upper curve of the log-log graph shows the effective viscosity when both fields are applied simultaneously. In the lower curve the perturbation was applied after a delay of 800 seconds. Two aspects of the Fig. 4 must be emphasized: the first one is that both viscosity curves exhibit a clear scaling behavior with time. This power law behavior is also observed in the effective viscosity as a function of other external variables of the system, such as particle concentration, static field intensity, frequency and amplitude of the perturbation, etc. It makes possible to construct easily general rate equations to describe the rheological behavior or dilute magneto-rheological dispersions under perturbations [5]. The second aspect worthy of remark is the tremendous effect, even in the long time effective viscosity, of applying simultaneously the static and the perturbation fields. Notice that at times as long as  $10^3$  seconds, still the effective viscosity value is several times larger for the sample that was exposed from the beginning to both fields.

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