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Kinetics of internal structures growth in magnetic suspensions

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ABSTRACT

The kinetics of aggregation of non Brownian magnetizable particles in the presence of a magnetic field is studied both theoretically and by means of computer simulations. A theoretical approach is based on a system of Smoluchowski equations for the distribution function of the number of particles in linear chain-like aggregates. Results obtained in the two dimensional (2D) and three dimensional (3D) models are analyzed in relation with the size of the cell, containing the particles, and the particle volume fraction φ. The theoretical model reproduces the change of the aggregation kinetics with the size of the cell and with the particle volume fraction as long as the lateral aggregation of chains is negligible.

The simulations show that lateral aggregation takes place when, roughly, $\varphi_{2D} > 5\%$ and $\varphi_{3D} > 1.5\%$.

Dependence of the average size of the chains with time can be described by a power law; the corresponding exponent decreases with the particle volume fraction in relation with the lateral aggregation.

In the 3D simulations, dense labyrinthine-like structures, aligned along the applied field, are observed when the particle concentration is high enough ($\varphi_{3D} > 5\%$).

1. Introduction

Suspensions of micron-sized magnetizable particles in non magnetic liquids (magnetorheological suspensions, MRS) attract considerable interest from researchers and engineers due to a rich set of unique physical properties, valuable for many modern technologies. An overview of the physics of these systems as well as their practical applications can be found in Ref. [1].

Without a magnetic field MRSs behave like ordinary suspensions of solid particles. When an MRS is subjected to an external magnetic field, the particles are magnetized and, under the action of the magnetic forces, form heterogeneous aggregates—linear chains and dense bulk clusters. The formation of these aggregates changes dramatically the macroscopical properties of MRS. For example, under an applied magnetic field the effective viscosity of MRS can increase up to several orders of magnitude. When the aggregates entirely fill the chamber (channel) containing the suspension and bond its opposite boundaries, the rheological behavior of the MRS changes from viscous to quasi elastic as long as the applied stress remains lower than the so called yield stress. Simultaneously the electrical conductivity of MRS increases significantly.

Practical applications of magnetic suspensions depend on the rate of change of their properties after application of the magnetic field. This is why the study of the growing rate of the aggregates is important for the prediction of the response time of MRS. To the best of our knowledge, the first model of the kinetics of evolution of the chain lengths in the systems of

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magnetizable particles has been developed by M. Doi et al. [2]. In this hierarchical theory all the aggregates have the same size at a given time, which is a quite crude approximation. Nevertheless this model takes into account that the force between two aggregates depends on their respective size and also that the average distance between aggregates increases during the aggregation process. Several numerical simulations have focused on the exponent of the growth of the chain average size l with time: $l(t) \propto t^{\alpha}$ and have found α between 0.5 and 0.7; the last limit corresponds to the case where dipolar forces dominate Brownian forces [3,4]. A detailed analysis of the power law exponents was carried out both by Brownian dynamics simulation and experimentally, for large box sizes with energy of the dipole–dipole interaction of the particles being between two and three order of magnitude larger than the thermal energy kT [5]. No clear evolution was seen in this range of dipolar forces; on the other hand the exponent α increased with the volume fraction. The analysis of the continuum version of the Smoluchowski equations in the presence of the dipolar forces has been investigated in Ref. [6] with a chain diffusion coefficient inversely proportional to the length of the chains; some analytical form for the size distribution function was obtained in this work. A model of aggregation of Brownian magnetic particles in linear chains has been developed in Ref. [7] on the basis of the Smoluchowski equations for the distribution function of the number of particles in the chain. The results of this model are in good agreement with experiments. Nevertheless the theoretical results were determined through the assumption that the Brownian motion strongly affects the kinetics of the chain formation. At the same time, in many cases the effect of Brownian motion in MRS is negligible because the energy of magnetic interaction between the micron-sized particles is usually several orders of magnitude larger than the thermal energy kT.

Recently an analytical model of the kinetics of the chain formation in a system of magnetizable non Brownian particles has been developed. [8]. This model is based on the usual system of the Smoluchowski equations for the density of chains of a given length. The size distribution functions obtained from the numerical solution of these equations reproduces well the distribution obtained from the computer simulations.

In this work we present results of computer simulations and of analytical modeling of aggregation in MRS. Both, 2D and 3D systems are considered. The effect of the size of the cell, containing the MRS, on the aggregation kinetics is studied.

The organization of this work is the following: in the next part we present the main features of the analytical model. In Section 3 we present the model used for the computer simulations. The comparison between the analytical and computer results is presented in Section 4 for 2D simulations and in Section 5 for 3D simulations. Computer simulations show that the particles form linear chains when their volume concentration is low enough (about 1%–2%). When the concentration of the particles exceeds some threshold magnitude, dense bulk structures appear instead of the linear chains. The evolution of this system of thick clusters is studied in Section 6

2. Analytical model of the chain growth

The details of the analytical model were described in Ref. [8]. Here we will briefly recall the main points of this model. We consider a suspension of non Brownian magnetizable particles in a flat gap. The system is subjected to a homogeneous magnetic field H perpendicular to the gap boundary. We suppose that, under the field action, the particles coalesce uniquely in linear chains aligned along the field. It should be stressed that the appearance of any branched or bulk aggregates is ignored in this model. That is why it is restricted to suspensions of relatively low volume fraction.

Let us denote the number of n-particle chains per unit volume of the system as g_n. Our aim is to determine the evolution of this function with time.

In the framework of the Smoluchowski equations, the evolution of the chain population can be represented by the following system of kinetics equations:

$$\frac{\mathrm{d}g_n}{\mathrm{d}t} = \frac{1}{2}\sum_{k=1}^{n-1}\alpha_{n-k,k}g_{n-k}g_k - g_n\sum_{l=1}^{N-n}\alpha_{n,l}g_l. \tag{1}$$

Here α_{nl}-is a matrix of kinetics coefficient for the transformation of two chains of n- and l-particles into a chain with $(n+l)$ particles, N is the maximal number of particles that a chain can contain; N is determined by the gap thickness. According to their physical meaning, the coefficient α_{nl} must be symmetrical with respect to the indexes n and l. Taking this into account one can show that Eq. (1) automatically satisfies the condition of the particle number conservation:

$$\sum_{n=1}^{N} ng_n = \mathrm{const} = \phi/\mathrm{V}$$

where V is the volume of a particle, and φ is the total volume fraction of the particles. For the kinetic coefficients α_{mn} we will use the following estimate, suggested in Ref. [8]:

$$\alpha_{kn} = \int_{S^*_{attr}} \mathrm{v}^*_r \mathrm{d}S, \tag{2}$$

where

$$\mathrm{v}^*_r = \begin{cases} \mathrm{v_r}, & \mathrm{v_r} \le 0 \\ 0, & \mathrm{v_r} > 0 \end{cases}$$