



On the theory of transport phenomena in ferrofluids. Effect of chain-like aggregates



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ABSTRACT

The paper deals with the theoretical study of the effect of chain-like aggregates on diffusion and magnetophoretic transport in ferrofluids. Analysis shows that the appearance of the chains leads to a strong anisotropy of the diffusion transport – the coefficient of diffusion in the direction of applied magnetic field is significantly more than that in the perpendicular direction. The presence of the chains in a ferrofluid strongly affects the coefficient of the particle magnetophoresis.

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1. Introduction

Ferrofluids are colloidal suspensions of single-domain ferromagnetic particles in a carrier liquid. The typical size of the particles in modern ferrofluids is about 10–20 nm. These systems are attracting considerable interest from researchers and engineers due to a rich set of unique physical properties, which find active applications in various modern industrial, biological and medical technologies. Diffusion and magnetophoretic phenomena in ferrofluids determine many important properties of these systems and their behavior in various technological applications. That is why a detailed study of transport phenomena in ferrofluids is important both from the point of view of the fundamental theory of magnetic fluids and from the viewpoint of the development of a scientific background for their practical usage.

In the case of very dilute systems, where any interactions between particles are negligible, the coefficient of the ferroparticle diffusion can be determined by the classical Einstein formula for the diffusion coefficient of a Brownian particle. When the interparticle interactions are significant, this coefficient depends on the particle concentration and applied magnetic field as well. Theoretical investigations of the diffusion and magnetophoretic transport in magnetic fluids with an account of magnetic, hydrodynamic and steric interactions between particles are presented in Refs. [1–4]. It is shown that in a magnetic field the diffusion coefficient is anisotropic—its magnitude along the field differs from that in the perpendicular direction.

The models [1–4] deal with the case of moderate and weak interparticle interactions, when the particles cannot form any heterogeneous structures. However observations demonstrate that linear chains, bulk dense “drops” and other internal structures occur in many modern ferrofluids. An applied magnetic field stimulates the appearance of these structural transformations (see, for example, Refs. [5–9]).

The typical size of the bulk drops in ferrofluids is about several microns. Usually they are clearly seen in optical microscopes [5–7]. Since the wavelength of visible light is much more than the size of the particles in ferrofluids, the linear chains can not be detected optically, but they have been observed with the help of electron microscopes [8,9].

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The appearance of the internal heterogeneous structures significantly affects the magnetic, rheological, optical and other properties of ferrofluids [10,11]. One can expect an influence of these structures on the transport properties and phenomena in these systems.

Many modern ferrofluids are polydisperse, often with a wide distribution over the particle size [10]. Particles of mean size in them are too small to form any heterogeneous aggregates and structures. Usually the chain-like and drop-like aggregates consist of the largest particles, whose concentration in ferrofluids, as a rule, is small. That is why the effect of the largest particles and heterogeneous structures on the transport phenomena in traditional ferrofluids is rather weak.

However, various ferrofluids, consisting of strongly interacting particles, which are able to form chains and other structures, have been synthesized for the past decade. Ferrofluids consisting of cobalt [11] as well as of cluster particles [12,13] are examples of fluids with a strong magnetic interparticle interaction.

In these and similar ferrofluids, a significant fraction of the particles can be united into chains or other heterogeneous aggregates. That is why the effect of the aggregates on the transport phenomena in these systems can be very strong. Analysis of the magnetodiffusion and magnetophoretic transport in ferrofluids with chains is important also in connection with the problem of magnetic separation of polydisperse systems with the aim to remove the largest particles, which can form heterogeneous aggregates.

The goal of this work is the analysis of the effect of the chain-like aggregates on the diffusion and magnetophoretic (i.e. under the action of spatially inhomogeneous magnetic field) transport in ferrofluids with strongly interacting particles.

2. Physical model and the main approximations

We will use a simple model of the chain-like aggregates, developed in Ref. [14]. In spite of simplifications, this model has allowed us to describe rheological properties of various magnetic fluids with the single-domain and cluster particles as well [15–18].

In the framework of the model [14] the ferrofluid particles are considered as identical ferromagnetic spheres with diameter d and magnetic moment m , fixed in the particle body. It is supposed that the particles can form only linear chain-like structures. Any interactions between the chains are ignored.

Following Refs. [14–18] we will suppose that the energy of the dipole–dipole interaction between the nearest particles in the chain is significantly more than the thermal energy kT . This condition is necessary for the appearance of any heterogeneous aggregates in a ferrofluid. We neglect the thermal fluctuations of the chain shape and orientations of the particle moments in the chain. In other words, the chain is modeled by a straight rigid rod-like aggregate; the particle magnetic moments are aligned along the aggregate axis. The criterion for applicability of this approximation is discussed in Ref. [14]. In part, it is shown that this approximation is justified when the inequalities $\varepsilon > \kappa$, $\varepsilon \gg 1$ are fulfilled. Here $\varepsilon = \frac{\mu_0 m^2}{2\pi d^3 kT}$, $\kappa = \mu_0 \frac{mH}{kT}$, H is the local magnetic field in the sample, μ_0 is the vacuum magnetic permeability. The first inequality means that the energy of magnetic interaction between the particles is more than the energy of their interaction with the magnetic field.

Next, we will take into account interaction only between the nearest particles in the chain.

Approximation of the rigid rod-like chains, of course, is a very strong simplification. A model of the flexible chains in an arbitrary magnetic field has been developed in Ref. [19]. However, this model leads to complicated and cumbersome calculations. At the same time results [19] show that the model of the rod-like aggregates allows one to get quite reasonable estimates for ferrofluid equilibrium magnetization. It has been noted already, that this model allows a description of rheological phenomena in various ferrofluids. It gives us a background to consider this model as a robust foundation for the description of the transport phenomena in ferrofluids.

3. Thermodynamical functions of magnetic fluid

Let us consider a unit representative volume of the ferrofluid. We will suppose that this volume contains a tremendous number of the particles; its size is much less than the characteristic distance of change of the particle concentration. As is well known, introduction of the small representative volume is necessary for the usage of continuous methods of description of transport and other nonequilibrium phenomena.

We will denote the number of n -particle chains in this volume as g_n . Let us suppose that at each moment of time the thermodynamical state of every elementary volume of the fluid can be considered as in equilibrium. In the framework of the chosen approximations, the free energy F of the unit volume of the fluid can be presented as [14]:

$$F = kT \sum_{n=1}^{\infty} \left[g_n \ln \frac{g_n v}{e} - g_n \varepsilon (n-1) - g_n \ln \frac{\sinh(\kappa n)}{\kappa n} \right]. \quad (1)$$

The first term in the square brackets (1) corresponds to the entropy of an ideal gas of the non-interacting chains. The second term describes the energy of interaction of the neighbor particles in the chains. The third term presents the Langevin free energy of the chain interaction with the local magnetic field H .

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