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Influence of interparticle interactions on diffusion processes in magnetic fluids

Pshenichnikov A. F.[a], Elfimova E. A.[b,*]

[a]Institute of Continuous Media Mechanics, Russian Academy of Science Ural Branch, Perm 614013, Russia
[b]Urals State University, Ekaterinburg 620000, Russia

Abstract

The diffusion coefficient is calculated for a concentrated magnetic fluid in the absence of a magnetic field, taking into account the many-body correlations. The ferroparticle density distribution in a gravitational field is determined using both Monte Carlo simulations and theoretical modeling. Numerical and theoretical results are in good agreement.

Keywords: magnetic fluids; gradient diffusion; interparticle correlations

1. Introduction

Gravitational sedimentation and magnetophoresis in an initially homogeneous magnetic fluid in a cavity result in spatial heterogeneity over time. In the absence of convection, gradient diffusion leads to a variation in the concentration profile. The concentration profile in a cavity can be found from the boundary problem consisting of Maxwell's equations for the magnetic field, and a dynamic diffusion equation taking into account particle sedimentation and magnetophoresis. This problem has only been solved for dilute magnetic fluids [1,2]. In this case the magnetic and diffusion parts of the boundary problem are separable, can be considered independently. Unfortunately this method cannot be used for concentrated magnetic fluids. If the ferroparticle concentration is high, then the magnetic-field problem is connected with the diffusion problem, and hence the concentration profile will depend on steric, magnetic dipolar, and hydrodynamic interactions between the particles. The taking into account all of these interactions is a very complicated problem.

The coefficient of gradient diffusion D was calculated in [3] using the Carnahan-Starling formula for the free energy of hard spheres, plus a modification that accounts for the effective dipole-dipole attraction. The latter term was treated to leading order in the volume ferroparticle concentration φ :

* Corresponding author Tel.: +7-343-350-7541; fax:+7-343-350-7401.
E-mail address: Ekaterina.Elfimova@usu.ru

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$$D = D_0 K(\varphi)\left[1 + 2\varphi\frac{4-\varphi}{(1-\varphi)^4} - \frac{8}{3}\lambda^2\varphi\right] \tag{1}$$

where $K(\varphi)$ is the ferroparticle mobility, D_0 is the diffusion coefficient for solutions at infinite dilution, and λ is the dipolar coupling constant relating the strength of the interaction energy between two particles at contact to the thermal energy. Equation (1) can be used only for magnetic fluids with low values of the volume concentration φ. For concentrated magnetic fluids, the diffusion coefficient should take into account many-particle correlations and as a result additional nonlinear terms will appear in (1). This paper considers the influence of interparticle correlations on the gradient diffusion of ferroparticles in concentrated magnetic fluids. The theoretical results are tested on data from Monte Carlo computer simulations.

2. Diffusion coefficient

The theoretical model of the magnetic fluid consists of a monodisperse system of N dipolar hard spheres with diameter d and constant magnetic moment m. The Gibbs free energy of the ferroparticle system may be represented as:

$$\Phi = N_0\mu_0 + N\mu^0 + NkT\ln(\varphi)) - kT\ln(Q_s) - kTN\varphi G(\lambda,\varphi) \tag{2}$$

where kT is the thermal energy, N_0 is the number of molecules in the carrier fluid, μ_0, μ^0 are the chemical potentials of the carrier fluid and a single ferroparticle, respectively, Q_s is the configuration integral of the hard-sphere fluid, $\lambda = m^2 / d^3kT$ is the dipolar coupling constant; and $G(\lambda,\varphi)$ is the contribution of the dipole-dipole interactions to total free energy. To obtain $G(\lambda,\varphi)$ we use a virial expansion in terms of the ferroparticle volume concentration φ. Each virial coefficient is calculated using the diagram method [4]. The coefficient of φ^{i-2} describe the mutual interactions of i particles. In this paper, $G(\lambda,\varphi)$ was calculated up to φ^2:

$$G(\lambda,\varphi) = \frac{4}{3}\lambda^2 + \frac{4}{75}\lambda^4 + \varphi\left[\left(2\ln 2 + \frac{1}{3}\right)\lambda^2 - \frac{10}{9}\lambda^3 - 0.34194\lambda^4\right] + \varphi^2 0.96724\lambda^2 \tag{3}$$

Computer simulation data [5] allow a determination of the range of validity of (3): this yields an upper limit of $\lambda \sim 1.5$, and a range of $\varphi < 0.3$. In order to extend this region we have developed a modified expression for the dipolar contribution to the free energy:

$$\tilde{G}(\lambda,\varphi) = \frac{4\lambda^2}{3} \times \left(\frac{1 + a_{20}\lambda^2}{1 - a_{01}\varphi + a_{02}\varphi^2 + a_{11}\lambda\varphi + a_{21}\lambda^2\varphi - a_{12}\lambda\varphi^2}\right) \tag{4}$$

where a_{20}=0.04, a_{01}=1.28972, a_{02}=0.93795, a_{11}=0.83333, a_{21}=0.30804, and a_{12}=1.01149535. The leading-order terms of the Taylor series of $\tilde{G}(\lambda,\varphi)$ in φ are identical to those of $G(\lambda,\varphi)$. The modified expression $\tilde{G}(\lambda,\varphi)$ is in good agreement with data from computer simulations [5] up to $\lambda \sim 3$ and $\varphi < 0.35$. Using the Batchelor formula and standard thermodynamic equations for the calculation of the chemical potential μ:

$$D = D_0 K(\varphi)\frac{\varphi}{1-\varphi}\left(\frac{\partial\mu}{\partial\varphi}\right)_{p,T}, \qquad \mu = \left.\frac{\partial\Phi}{\partial N}\right|_{No,p,T}$$

the diffusion coefficient for concentrated magnetic fluid takes the form:

$$D = D_0 K(\varphi)\left(1+\varphi\frac{8-2\varphi}{(1-\varphi)^4}-2\varphi\widetilde{G}-4\varphi^2\frac{\partial\widetilde{G}}{\partial\varphi}-\varphi^3\frac{\partial^2\widetilde{G}}{\partial\varphi^2}\right) \qquad (5)$$

Figure 1 shows the dimensionless coefficient of gradient diffusion as a function of volume concentration, for different values of the dipolar coupling constant.

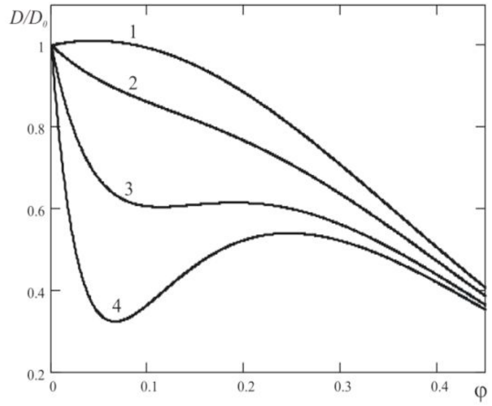

Fig.1. Dimensionless coefficient of gradient diffusion as a function of volume ferroparticle concentration φ for different values of the dipolar coupling constant λ : curve 1 is $\lambda = 0$; curve 2 - $\lambda = 1$; curve 3 - $\lambda = 2$; curve 4 - $\lambda = 3$.

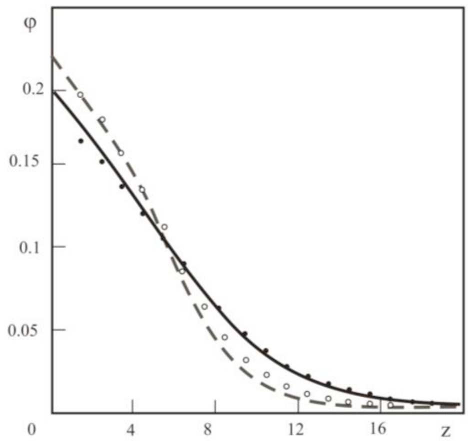

Fig. 2. The ferroparticle distribution in a gravitational field for a round vertical cylinder along height *z*. The lines are theoretical predictions and the points are from Monte Carlo simulations: $\lambda = 2$ (dashed line and open circles) $\lambda = 3$ (solid line and filled circles).

3. Ferroparticle distribution over gravitational field

Dipole-dipole interaction is a reason of effective attraction between ferroparticles, which leads to additional drift of ferroparticle in heterogeneous over concentration magnetic fluids. The corresponding additional term to total ferroparticle flux is proportional to a gradient of the number concentration *n*. Thus the flux density of the ferroparticle can be written as:

$$\mathrm{J} = D_0 K(\varphi)\left\{ nL(\xi_\varepsilon)\nabla(\xi_\varepsilon) + ng\mathrm{e} - \left(1 + \varphi\frac{8-2\varphi}{(1-\varphi)^4} - 2\varphi\widetilde{G} - 4\varphi^2\frac{\partial\widetilde{G}}{\partial\varphi} - \varphi^3\frac{\partial^2\widetilde{G}}{\partial\varphi^2}\right)\nabla n \right\} \tag{6}$$

where g is a gravitational parameter, being the inverse of the barometric height, $L(\xi)$ is the Langevin function, $\xi_\varepsilon = m(H + 4\pi M_L/3)/kT$ is the Langevin parameter found with the help of mean-field theory, and $M_L = mnL(\xi)$ is the Langevin magnetization. The validity of the effective-field approach was has proved in [6]. The derivation of (6) takes into consideration the quasiequilibrium character of the magnetisation within a modified mean-field theory [7]. The right-hand side of (6) equals zero in static conditions; the stationary solution yields the spatial distribution of ferroparticles in a cavity. Under a magnetic field the solution of the problem should be solved together with Maxwell's equations and a constitutive relation. Figure 2 shows a comparison of (6) with data from Monte Carlo simulations for the distribution of ferroparticles in a circular vertical cylinder at height z.

The method of computer simulation is similar to that described in [8]. A colloidal particle is modeled as a sphere with a constant value of the magnetic moment. The system contains 10^3 particles. The energy of the i^{th} particle is a sum of the dipolar interactions and the gravitational potential:

$$\frac{U_i}{kT} = gz_i - \xi_0 \cos\theta_i - \lambda\sum_{\substack{j=1 \\ j\neq i}}^{N}\left[\frac{3(e_i \cdot R_{ij})(e_j \cdot R_{ij})}{R_{ij}^5} - \frac{(e_i \cdot e_j)}{R_{ij}^3}\right] \tag{7}$$

Here ξ_0 is the Langevin parameter, R_{ij} is the distance between the centers of the i^{th} and j^{th} particles, and θ_i is the angle between the external magnetic field and the magnetic moment of a particle. Steric interactions were taken into account by forbidding the hard spheres to overlap with each other or with the cylinder wall. To obtain the stationary particle distribution profile, the cylinder is divided in to 20 horizontal layers. The average concentration of the magnetic particles in each layer is determined at each MC step. After the establishment of thermodynamic equilibrium, the local concentration profile is averaged over 10^5 MC steps. Data from the top and bottom layers are not take into consideration because of well-known boundary effects. Figure 2 shows reasonable agreement between theory and simulation.

4. Conclusions

Analytical expressions of free energy and gradient diffusion coefficient were calculated taking into account many body correlations. Theoretical predictions demonstrate a good agreement with data of computer simulations (MC) for concentrated magnetic fluids with moderate and strong intensity of dipole-dipole interactions.

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