MAGNETICALLY INDUCED MASS TRANSFER THROUGH A GRID IN NONISOTHERMAL FERROFLUIDS

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Introduction. Thermodiffusion experiments in ferrofluids in the presence of a magnetic field often are disturbed by an uncontrollable thermo-and solute magnetic convection. One possibility to eliminate the parasitic convection is performing experiments in porous media. However, there appear new problems at interpretation of measurement results. In porous layers, the thermophoresis can be accompanied by particle thermoosmosis [1]. Moreover, recently has been found a strong influence of the magnetic field on the particle transfer through a thin non-isothermal ferrofluid layer between two nonmagnetic permeable walls [2]. The authors explain those results as a magnetic Soret effect. The aim of this paper is to clarify whether the observed effect can be interpreted as a specific microconvective mass transfer. Due to difference in magnetic susceptibilities and thermal conductivities of the liquid and the porous matrix, the external magnetic field under non-isothermal conditions can induce a magnetic microconvection inside the pores. Formally, the particle transfer by magnetic microconvection is similar to that of thermoosmosis. The only difference is that instead of the short-range surface driving force, now the convection is induced by a long-range volumetric magnetic force. Due to this analogy, the magnetic particle transfer induced by filter grains in [3] is called "thermomagnetoosmosis".

1. Basic principle of magnetoconvective particle transfer. According to the concept of thermomagnetoosmosis, under non-isothermal conditions the local perturbations of the magnetic field near the grid element induce both a concentration difference due to magnetophoresis and a thermomagnetic convection. As a result, there should appear a transient convective mass transfer directed along the temperature gradient.

Let us consider the membrane as a grid consisting of single spherical grains of magnetic permeability μ_i different from that of a surrounding liquid μ. An external uniform magnetic field $\mathbf{H}$ is directed parallel to the gradient of permeability $\nabla\mu = -\alpha_T \nabla T$ (α_T is the fluid pyromagnetic coefficient) towards the azimuthal coordinate $\vartheta = 0$. Under the Stokes approximation (the Reynolds number for micro-size grains is very small) the convection velocity around the sphere $\mathbf{u}$ is governed by a vorticity equation ($\Omega = \mathrm{rot}\,\mathbf{u}$)

$$\nabla \times \left(\nabla \times \mathbf{\Omega}\right) = \frac{\mu_0}{2\eta} \nabla\mu \times \nabla H^2 \tag{1}$$

with η being the fluid viscosity. If the heat conductivities of the liquid (λ) and the sphere (λ_i) are the same, $\nabla\mu$ in (11) is equal to the macroscopic one. The mathematical problem is similar to that of particle thermophoresis [4, 5], but in the frame of linear approximation considered here, the sphere-induced perturbations of the magnetic field can be calculated by introducing the scalar magnetic potential

$$\psi = \frac{K}{\rho^2} H_0 \cos\vartheta \tag{2}$$

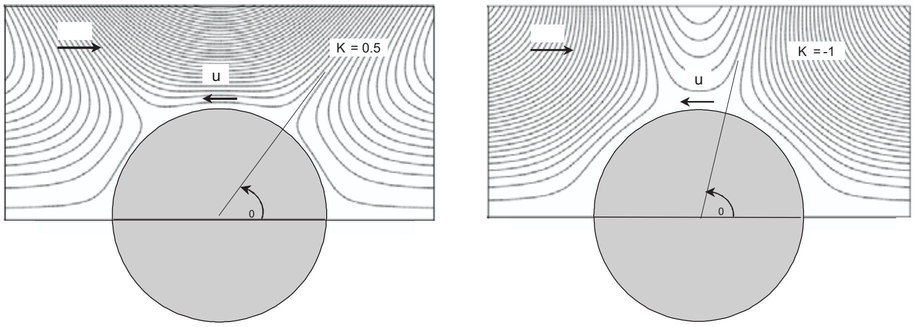

Fig. 1. Flow pattern around a sphere, $\mathbf{H} \parallel \nabla T$, $K = 0.2$.

with no account for smaller terms with $\nabla\mu$ [6]. Here ρ is the non-dimensional radial coordinate scaled by the grain radius R_0 and the coefficient K represents the difference in magnetic permeabilities, $K = (\mu - \mu_i)/(\mu_i + 2\mu)$. If $\lambda_i \neq \lambda$, the permeability gradient $\nabla\nu$ in (1) should account for local temperature perturbations induced by the sphere. The convection is very slow (the thermal Peclet number $\mathrm{Pe} \ll 1$), therefore, the expression for local temperature distribution can be taken equal to (2) but with another coefficient $K_\lambda = (\lambda - \lambda_i)/(2\lambda + \lambda_i)$.

Fig. 1 illustrates the flow pattern around a nonmagnetic sphere immersed in a magnetic fluid (the streamlines are calculated from the analytic solution of equations (1) and (2) given in Refs. [5] and [6]. The external magnetic field induces six extended convection vortices. Four pared asymmetric vortices cause only a volumetric fluid mixing, whereas two other vortices, located in the region $\vartheta \pm \pi/2$, induce a macroscopic translation flow ("thermomagnetoosmosis"). The non-magnetic spheres induce a resulting flow directed towards the decreasing fluid permeability. On the contrary, the magnetic sphere (negative K) causes a convective motion in an opposite direction, towards increasing μ. The convection around the thermally nonconducting spheres ($K_\lambda = 0.5$) is significantly stronger than that around the spheres of high λ_i ($K_\lambda = -1$).

The local magnetic field gradients induce not only the convection but also a magnetophoretic transfer of particles. Their velocity in diluted colloids is equal to

$$\mathbf{u}_m = \frac{2\mu_0 a^2}{9\eta} M_p \nabla H$$

(here a is the particle radius and M_p is the longitudinal component of particle magnetization, for Brownian particles it is proportional to the Langevin function). The magnetophoresis causes a redistribution of particle concentration near the sphere, the excess concentration $\Delta c = c - c_0$ in diffusion boundary layer in the plane $\vartheta = \pm\pi/2$ is proportional to the coefficient K. As a result, there appears a convective mass flux $\mathbf{j} = 2\pi \int \mathbf{u}\Delta c R \mathrm{d}R$, which is directed towards the increasing temperature independently on the sign of the difference in magnetic permeabilities for the grid and the surrounding liquid. The microconvective mass flux can be evaluated introducing the approximation of concentration boundary layer, which starts to develop at the frontal hydrodynamic attack angle ϑ_0. If the excess concentration Δc in the boundary layer is approximated by a polynomial, the mass transfer problem can be treated analytically [3]. For approximate analysis the resultant "magnetoosmotic" nanoparticle flux in a thin porous filter sheet of low planar density n of grains can be assumed in proportion to the cross-sectional packing coefficient $p = n\pi R_0^2$. In Fig. 2 the microconvective mass transfer through one-layer sheet is shown in the form of equivalent thermophoretic mobility St.

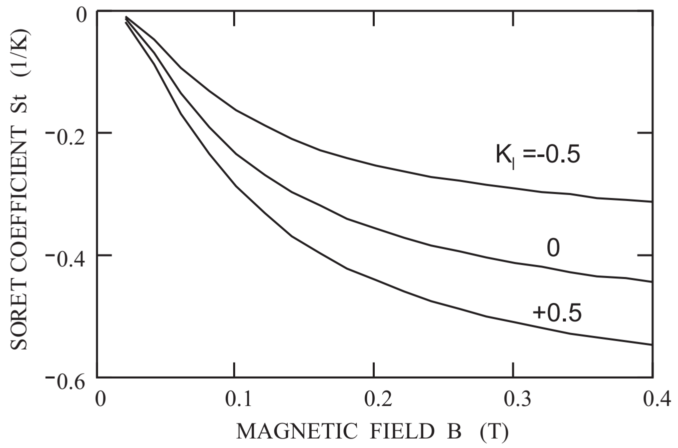

Fig. 2. Effective Soret coefficient, one-layer sheet, $R_0 = 10$ μm, $p = 0.1$.

The curves correspond to a magnetite based ferrofluid, volumetric concentration of particles $\varphi = 0.1$, $a = 5$ nm, $\eta = 0.002$ N $\cdot$ s/m^2, $\nabla T = 10^4$ K/m. One can see that negative values of the thermomagneto-osmotic coefficient St can exceed the positive zero-field values of the particle Soret coefficient (they are close to +0.1). At $\mathbf{B} \perp \nabla T$, the resulting flow induced by a membrane grain has an opposite direction, but due to a complicated vortical structure of convection; the integral mass flux should be found numerically.

It should be noted that irrespective of very low diffusion coefficients of the ferrofluid nanoparticles, the diffusion Peclet number for μm-size spheres and $\nabla T = 10^4$ K/m even in strong magnetic fields does not exceed the value $\mathrm{Pe}_D = 100$. Thus, the validity of the boundary layer approximation in mass transfer calculations is questionable. For exact analysis it is necessary to perform additional numerical simulation of the convective diffusion problem.

2. Experimental results. To verify the proposed concept, a series of separation measurements through a one-layer filter sheet was performed. The filter represents a thin (thickness 1 μm) squared copper grid of period 100 μm and cross-sectional porosity (transparency) $P = 0.32$. Together with a plastic gaskin (thickness $d = 1$ mm) of low thermal conductivity the grid is cramped between two identical copper cylinders (diameter 15 mm, length $L = 15$ mm), which are kept using two thermostats at different temperatures T_c and T_h. The cell is oriented vertically (a colder chamber with T_c at the bottom) and placed inside a solenoid. Experiments are performed employing a well-examined ferrofluid sample TD5: magnetite in tetradecane stabilized by oleic acid, saturation magnetization $M_{\mathrm{s}} = 10.6$ kA/m, volume fraction of magnetic phase $\varphi_0 = 0.023$, mean "magnetic" diameter of particles $d_{\mathrm{m}} = 8.5$ nm, diffusion coefficient (at zero field) $D_0 = 1.18 \cdot 10^{-11}$ m^2/s, Soret coefficient $\mathrm{S_T} = 0.16$ 1/K.

Thermophoretic transfer of particles through the grid is determined from the growth rate of concentration difference in both separation chambers $\nabla\varphi = \varphi_h - \varphi_c$. During experiments in the separation chambers a convection can develop (especially, if the magnetic field is applied). Nevertheless, due to a low porosity the convective vortices cannot penetrate the grid, they equalize the temperature in each chamber. Thus, we can guess that the measured mass flux corresponds to the temperature gradient $\nabla T = (T_h - T_c)/d$ and the separation parameter St

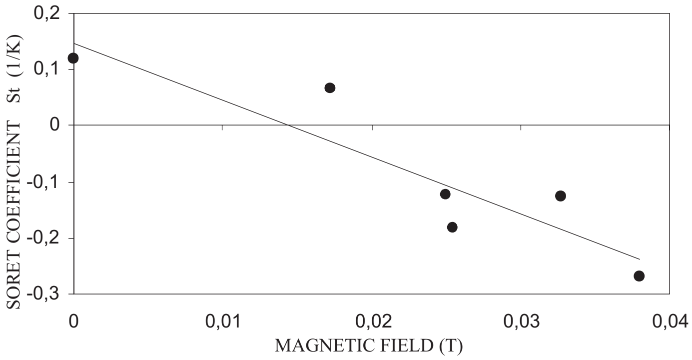

Fig. 3. Effective Soret coefficient of the grid, $\mathbf{B} \parallel \nabla T$.

(effective Soret coefficient) can be calculated using the expression

$$\mathrm{St} = \frac{\Delta\varphi}{\varphi_0} \frac{L}{2PD_0 t \nabla T} \,. \tag{3}$$

Fig. 3 represents the measurement results. Duration of the experiment $t = 24$ hours, the difference in particle concentration $\Delta\varphi$ is determined from magnetization measurements of colloid samples taken from the both chambers after finishing the experiment. At zero magnetic field the particles are transferred to the cold chamber, $S = +0.13$. This value agrees relatively well with the Soret coefficient $\mathrm{S_T} = +0.16$ which was defined separately using the particle optical grating technique [7]. A parallel magnetic field $\mathbf{B} \parallel \nabla T$ induces a transfer of particles in opposite direction, toward increasing temperatures. Even at small fields (about 20 mT) we observe a redirection of the summary mass flux (S becomes negative). The optical grating experiments indicate that the thermophoretic mobility (thermodiffusion coefficient $D_T = \mathrm{S_T} D$) of ferroparticles does not depend on the magnetic field [7]. Thus, the observed field effect cannot be interpreted as a magnetic Soret effect (note – the points of Fig. 3 are calculated introducing in (3) the zero-field value D_0). The measurement results agree qualitatively well with estimations made under the concept of a grain-induced microconvective mass transfer. In the presence of a transverse field $\mathbf{B} \perp \nabla T$, visible changes in thermophoretic mass transfer are not observed.

3. Conclusions. The strong influence of magnetic field on thermophoretic transfer of ferrofluid particles through a micro-scale grid is observed. The results are interpreted as a microconvective mass transfer induced by filter grains.

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