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Non-isothermal separation of ferrofluid particles through grids: Abnormal magnetic Soret effect

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ABSTRACT

Nanoparticle transport through thin non-isothermal ferrofluid layer between permeable walls is investigated. The transient mass flux is determined from measurements of particle concentration changes in two fluid chambers of different temperatures which are attached on both sides of the layer. Experiments are performed employing fluid samples of small ordinary magnetic Soret effect, which is detected by thermal grating technique. The separation measurements say that a magnetic field, aligned along a temperature gradient, causes a remarkable increase in the mass diffusion coefficient and a simultaneous decrease in particle thermodiffusion mobility. It is proposed that the observed effects may be evoked by specific microconvective mass transfer induced by nonmagnetic grid elements of the permeable walls.

1. Introduction

Recently, a strong anisotropic magnetic Soret effect was established. Theory says [1] that a magnetic field, oriented transversally to the temperature gradient, accelerates the mass transfer, whereas a longitudinal field causes a decrease in the particles' Soret coefficient. Though, direct influence of a field on nanoparticle thermophoretic motion is not very high, the main reason of magnetic Soret effect is changes in mass diffusion under the effect of internal magnetic field gradients due to the development of non-uniformity of particle concentration. The main conclusions of the theory of the magnetic Soret effect are confirmed experimentally. Two principally different techniques, relaxation measurements of optically induced thermal grating in thin ferrofluid layers [2,3] and direct measurements of particle dynamic separation in thermodiffusion columns [4–6], both testify the theoretically predicted anisotropy of nanoparticle Soret coefficient in the presence of a magnetic field. Unfortunately, both measurement techniques allow obtaining quantitatively safe results only in case of relatively small magnetic fields [7]. Under stronger fields the separation process usually is disturbed by the appearance of parasitic thermomagnetic convection. To eliminate a possibility to originate the convection, authors of Ref. [8] have performed experiments by measuring the particle flux through a thin non-isothermal ferrofluid layer between two permeable walls. An unexpectedly strong influence of the magnetic field on the effective Soret coefficient was established. One reason of that might be a microconvective particle transfer induced by wall grid elements [9]. The present paper deals with experiments aimed to examine this theoretical prediction.

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2. Theoretic background of experiments

2.1. Magnetic Soret effect

Considering stable colloids without chemical reactions, the mass conservation equation for two component systems (particles of mass concentration ρ_i and a carrier liquid) is the following:

$$\frac{\mathrm{d}\rho_i}{\mathrm{d}t} = -\operatorname{div}\mathbf{j}_i \tag{1}$$

The mass flux $\mathbf{j}_i$ includes stochastic particle transfer as well as various slip processes of phoretic origin. Ultra-fine particles dispersed in fluids obey an intensive Brownian motion. Therefore, mass transfer in colloids can be considered similar to that in molecular liquids by involving the concept of gradient diffusion. The mass diffusion coefficient of nanoparticles is determined by relation $D = kT/f_v$, where f_v is the hydrodynamic drag force (for spherical particles of radius r $f_v = 6\pi\eta r$, η is the carrier liquid viscosity). The mass flux, besides terms of gradient diffusion and thermodiffusion, contains a new term of magnetic sedimentation (for stable nanocolloids, we assume the ordinary barodiffusion being negligible small) [1]:

$$\mathbf{j}_i = -D\nabla\rho_i - \rho_i(1-n_i)DS_T\nabla T + \frac{m_g}{f_v}\big[\mu_0(\bar{M}_i - \bar{M}_0)\nabla H\big]\rho_i(1-n_i) \tag{2}$$

Here S_T is the Soret coefficient, m_g is the particle mass, $\bar{M}_i$ and $\bar{M}_0$ is the specific magnetization of particles and that of a carrier liquid, n_i is the mass fraction of the solid phase.

Let us consider a homogeneous external magnetic field $\mathbf{B} = const$ directed along the temperature gradient. From the equation $\operatorname{div}\mathbf{H} = -\operatorname{div}\mathbf{M}$ it follows that in colloids of non-uniform temperature and non-uniform particle concentration there appears an internal magnetic field gradient (we assume $M_0 = 0$):

$$\nabla H = -\nabla M = -\frac{\partial M}{\partial\rho_i}\nabla\rho_i - \frac{\partial M}{\partial T}\nabla T - \frac{\partial M}{\partial H}\nabla H \tag{3}$$

For diluted colloids ($n_i \ll 1$) the mass flux (2) with (3) can be rewritten in the form of:

$$\mathbf{j}_i = -D(1+k_\mathrm{m})\nabla\rho_i - S_T D\left(1 - k_\mathrm{m}\frac{\alpha_T}{S_T}\right)\rho_i\nabla T \tag{4}$$

Here $\alpha_T = -M^{-1}\partial M/\partial T$ is the fluid pyromagnetic coefficient. The magnetic parameter k_m for colloids of Langevin-type magnetization ($M = \varphi M_s L(\xi)$ with $L(\xi) = cth\xi - \xi^{-1}$) obeys the form of:

$$k_\mathrm{m} = \frac{\gamma L^2(\xi)}{1+\gamma L'(\xi)} \tag{5}$$

where $\xi = \mu_0 mH/kT$, m is the magnetic moment of particles (for subdomain particles, m is proportional to their volume V, $m = M_\mathrm{s}V$), M_s is the saturation magnetization of particle material, φ is the volumetric part of particles in the dispersion and $\gamma = \varphi\xi M_\mathrm{s}/H$ is the magnetic interaction parameter of particles in the dispersion. Pyromagnetic coefficient α_T slightly depends on magnetic field: $\alpha_T = \beta_\mathrm{m}(1 + \xi L'(\xi)L(\xi)^{-1}(1 + (\beta_\mathrm{m}T)^{-1}))$ with β_m being the pyromagnetic coefficient of the particle material, $\beta_\mathrm{m} = -M_\mathrm{s}^{-1}\partial M_\mathrm{s}/\partial T$.

From (4) it follows that magnetic stratification of ferroparticles under the influence of internal field gradients can be considered as an increase in the mass diffusion coefficient and as a reduction of the Soret coefficient (in surfacted colloids usually $S_T > 0$ and the pyromagnetic coefficient α_T according to its definition is positive). Numerical estimations show that the parameter k_m in ferrofluids may reach values close or higher than 1. Therefore the increase in relative diffusion coefficient $d_\mathrm{m} = D(H)/D = (1 + k_\mathrm{m})$ is remarkable. On the contrary, the predicted reduction of the relative thermodiffusion coefficient $s_\mathrm{m} = D(H)S_T(H)/(DS_T) = (1 - k_\mathrm{m}\alpha_T/S_T)$ is very small, because in colloids $\alpha_T \ll S_T$.

The mass transport coefficients in colloids can be determined from unsteady particle separation measurements. In optical grating [2], as well in thermodiffusion column [4], experiments there develop a concentration difference inside the examined fluid layers due to zero mass flux at their passive borders. Eq. (1) with mass flux (4) predicts an exponential growth of the concentration difference. Its saturated level gives the value of magnetic Soret coefficient $S(H) = s_\mathrm{m}S_T/d_\mathrm{m}$, whereas the separation relaxation time allows determining the magnetic diffusion coefficient $D(H) = d_\mathrm{m}D$ [10].

2.2. Microconvective particle transfer induced by nonmagnetic bodies

In experiments with non-isothermal fluid layer having permeable walls [8], the situation is quite different. Now is measured a transient particle flux through the layer, which causes a growth of the concentration difference in volumes outside the layer. Therefore, the separation dynamics may be influenced not only by ordinary particle thermophoresis across the layer but also by specific transport processes inside the walls. Considering the permeable wall as a grid consisting of single spherical grains of magnetic permeability μ_i different from that of a surrounding liquid μ, in Ref. [9] it is shown that a

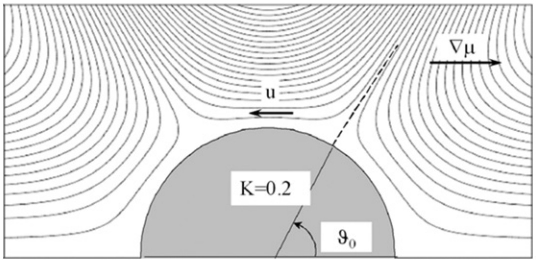

Fig. 1. Structure of magnetically induced microconvection near nonmagnetic sphere ($K = 0.2$) in a ferrofluid of non-uniform magnetization, $\mathbf{B}//\nabla\mu$.

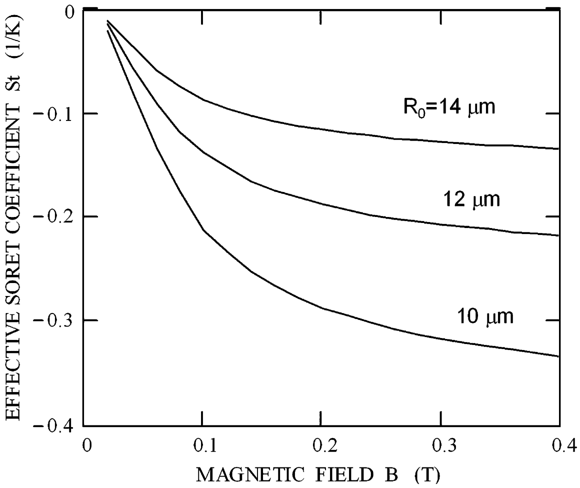

Fig. 2. Particle transfer in non-isothermal ferrocolloid induced by nonmagnetic spherical elements of one-layered grid of planar porosity 0.9 (R_0 is grain radius) [9].

magnetic field oriented along the gradient of permeability $\nabla\mu = -\alpha_T \nabla T$ induces a local microconvection near the grain surface. The convection velocity $\mathbf{v}$ is governed by vorticity equation ($\boldsymbol{\Omega} = \mathrm{rot}\,\mathbf{v}$)

$$\nabla \times (\nabla \times \boldsymbol{\Omega}) = \frac{\mu_0}{2\eta} \nabla\mu \times \nabla H^2 \tag{6}$$

with η being the fluid viscosity. For simplicity we assume that heat conductivities of the liquid and the sphere are identical, therefore $\nabla\mu$ in (6) is equal to the macroscopic one. The sphere-induced perturbations of external magnetic field ∇H^2 can be calculated introducing a scalar magnetic potential:

$$\psi = \frac{K}{R^2} H_0 \cos\vartheta \tag{7}$$

Here R is the non-dimensional radial coordinate scaled by grain radius R_0 and coefficient K represents the difference in magnetic permeabilities of the fluid μ and the grain μ_i, $K = (\mu - \mu_i)/(\mu_i + 2\mu)$.

Fig. 1 represents the 2D flow structure around nonmagnetic sphere immersed in a ferrofluid [9]. External magnetic field induces three extended toroidal convection vortices. The two antisymmetrical vortices cause only a fluid mixing near the particle, whereas the third one, located in the meridional plane perpendicular to $\nabla\mu$, induce a macroscopic translation flow ("thermomagnetoosmosis"). Besides, the local magnetic field gradients induce also a magnetophoretic transfer of particles toward the sphere surface. According to Eq. (2), their slip velocity in diluted colloids is equal to:

$$\mathbf{u}_\mathrm{p} = \frac{2\mu_0 r^2}{9\eta} m L(\xi) \nabla H \tag{8}$$

As a result, there appears a macroscopic mass flux which is always directed toward increasing temperatures, independently of the sign of difference in magnetic permeabilities $\mu - \mu_i$. Fig. 2 characterizes the summary microconvective mass transfer induced by one-layered filter sheet consisting of spherical grains. The results are represented in form of "effective" (convective) Soret coefficient *St*. For simplicity, the calculations are performed for a filter of high planar porosity $\alpha = 0.9$ when an additive impact of each filtering element in summary mass transfer may be proposed. Calculations say that even under small fields the microconvective separation parameter values may compensate and even exceed the positive thermophoretic Soret coefficient which for surfacted ferrocolloids is approximately equal to $+0.1\ \mathrm{K}^{-1}$. To distinguish both effects, the magnetic

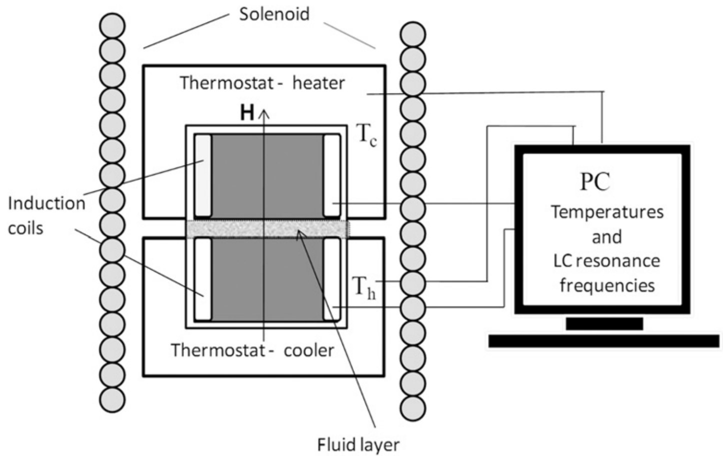

Fig. 3. Experimental setup. The ferrofluid layer between heating and cooling chambers is formed by two permeable grid-like nonmagnetic walls. Particle concentration and mean temperature in chambers are detected from measurements of inductance and of electric resistance of coils.

Soret effect and the microconvective nanoparticle transport, it is necessary to perform thermal mass transfer experiments through permeable walls employing a ferrofluid sample of independently measured magnetic Soret coefficient.

3. Experiments and analysis of results

3.1. Experimental setup

Mass transfer experiments were performed employing a setup (see Fig. 3) that consists of two equal cylindrical volumes $V_0 = 2.65\ \text{cm}^3$ kept at different temperatures and united by two permeable walls which form a flat fluid layer of thickness $\delta = 1$ mm. Both walls are made of equal plastic filaments of diameter 50 μm which form a square shaped grid of planar porosity $\alpha = 0.38$. The cross sectional area of the filter is $S_0 = 1.8\ \text{cm}^2$. Inside both volumes electric coils have been mounted. They are used as inductances of two LC oscillators. Measurements of their resonance frequencies (several kHz) allow determining the particle concentration in both volumes during mass transfer through the fluid layer between grids. The temperature regime in both volumes was controlled by measuring the electric resistance of the corresponding induction coils (they are made of copper wires). The cell upper volume was heated and the lower volume was cooled by two water thermostats. The heat exchange is organized in a manner that provides the homogeneity of the fluid temperature in both volumes. Transversal to the fluid layer, a magnetic field $\mathbf{H} /\!/ \nabla T$ was generated by external water cooled electric coils. The experiment and acquisition of measurement results is computerized. Setup can operate in autonomous regime for a long time (more than one week), which is necessary to complete the series of experiments.

3.2. Ferrofluid sample: physical properties and magnetic Soret effect

The examined two-component ferrofluid consists of magnetite nanoparticles coated with oleic acid and suspended in tetradecane. The volumetric concentration of magnetic phase $\phi_0 = 0.05$ and the mean "magnetic" radius of colloidal particles $r = 4.1$ nm. Transport properties of the sample (diluted to $\phi = 0.023$) were examined employing optically induced thermal grating technique [10]. Performing analysis of measurement results, special efforts were made to eliminate the influence of uncontrolled convective processes. Transport coefficients were calculated from the initial exponential part of the diffraction signal until the grating is not disturbed by parasitic double diffusive magnetic convection. At zero magnetic field, the measured diffusion and Soret coefficient values are the following: $D = 1.18 \cdot 10^{-11}\ \text{m}^2/\text{s}$ and $S_T = 0.16\ \text{K}^{-1}$. In the presence of a transversal magnetic field $\mathbf{H} /\!/ \nabla T$, mass transfer measurements agree well with theoretical predictions: a growing field causes an increase of $D(H)$ and a reduction of $S_T(H)$. Both experimentally detected field dependencies quantitatively agree well with theoretical predictions (4) and (5) (see Fig. 4). In the context of the present experiments on particle separation through a fluid layer, it is important to note that the fluid thermodiffusion coefficient $S_T(H)D(H)$ within the examined magnetic field interval 0–100 kA/m remains practically unchanged.

3.3. Particle separation through non-isothermal fluid layer

The transient thermal separation measurements were carried out employing the colloid sample DF5 of initial particle concentration $\phi_0 = 0.05$. Fig. 5 shows several curves of particle separation dynamics measured at various magnetic field strength values. Steady temperature gradient inside the layer $\Delta T/\delta$, which is due to temperatures difference in upper and lower cell volumes $\Delta T = T_\text{h} - T_\text{c}$, induces transient thermophoretic particle transfer through the fluid layer. Due to fluid

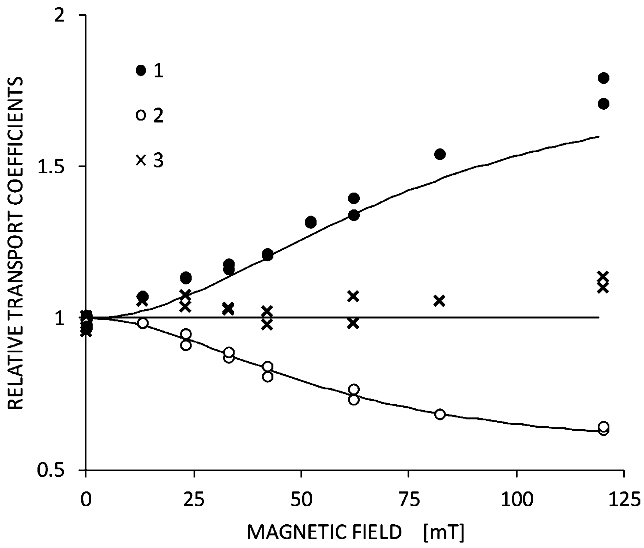

Fig. 4. Diffusion d_m (1), Soret s_m/d_m (2) and thermodiffusion s_m (3) coefficients of the ferrofluid [10]. Solid lines represent calculations according to (4) and (5).

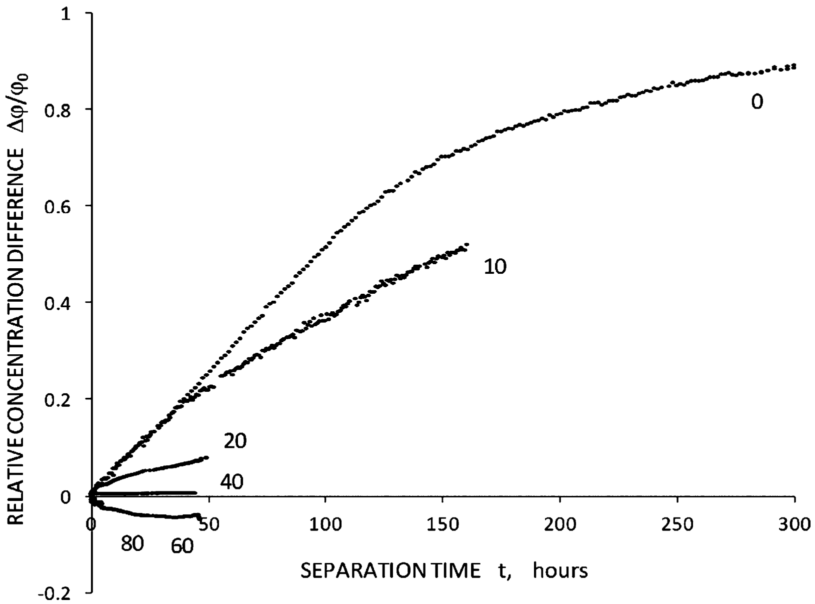

Fig. 5. Unsteady particle separation through the fluid layer, $\Delta T = 34$ K. Numbers near curves denote the magnetic field strength, mT.

convective mixing inside both volumes (not only the ends but also the cylindrical side walls are heated or cooled), we may assume that ΔT is equal to the temperature difference that is found from the measurements of the electric resistance of induction coils. The observed very slow relaxation of the separation process allows assuming also a quasi-steadiness of the concentration gradient across the filter layer $\nabla\phi = \Delta\phi/\delta$, with $\Delta\phi$ being the difference in mean concentrations between both volumes measured by LC oscillators (due to convective mixing, the concentrations in both volumes also are homogenized). Under such simplification, the solution of the particle conservation equation (1) with the particle flux (4) gives the following exponential law of development of the concentration difference in volumes on both sides of the layer [11]:

$$\frac{\Delta\varphi}{\varphi_0} = \sigma\left[1 - \exp\left(-\frac{t}{\tau}\right)\right] \tag{9}$$

The asymptotic level of particle separation σ depends of the effective Soret parameter S_{eff}:

$$\pm\sigma = \frac{2S_{\text{eff}}\Delta T}{2 + S_{\text{eff}}\Delta T} \tag{10}$$

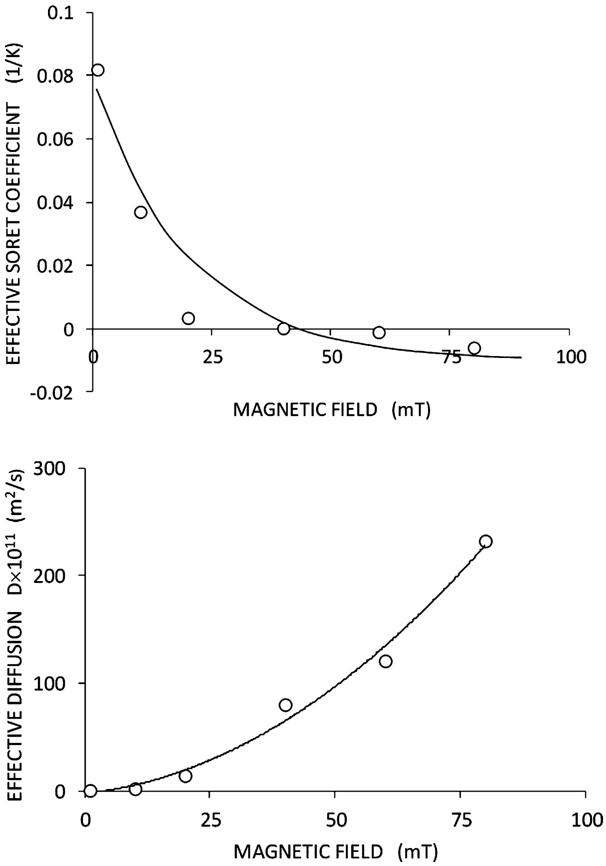

Fig. 6. Dependence of effective Soret coefficient (at the top) and effective diffusion coefficient (at the bottom) on uniform magnetic field aligned along the temperature gradient.

(the sign "+" correspond to a positive Soret effect and "−" to a negative one), whereas the relaxation time of separation dynamics includes both the Soret parameter as well as the effective diffusion coefficient D_{eff}:

$$\tau = \frac{\delta V_0}{(2 + S_{\text{eff}}\Delta T)\alpha S_0 D_{\text{eff}}} \tag{11}$$

The measured particle separation curves shown in Fig. 5 follow to the exponential law (9) very well. The corresponding values of the asymptotic particle separation level σ and of relaxation time τ allow the calculation of both the effective Soret coefficient and the particle mass diffusion coefficient:

$$S_{\text{eff}} = \frac{2\sigma}{(2-\sigma)\Delta T} \quad \text{and} \quad D_{\text{eff}} = \frac{(2-\sigma)\delta V_0}{4\alpha S_0 \tau} \tag{12}$$

The separation relaxation curve for $\mathbf{H} = 0$ gives the following zero-field values of transport coefficients: $D = 1.28 \cdot 10^{-11}\ \text{m}^2/\text{s}$ and $S_T = 0.082\ \text{K}^{-1}$. These values agree well with the thermal grating findings given above, especially if taking into account that thermal grating measurements were carried out employing a diluted TD5 sample of lower φ (according to Batchelor's theory, the particle diffusion coefficient increases as $D = D_0(1 + 1.45\varphi)$, whereas experiments [12] testify that the Soret coefficient decreases with growing particle concentration). On the contrary, in the presence of magnetic field our separation measurements give results qualitatively different from those of previous thermal grating experiments (compare the results presented in Fig. 4 with those of Fig. 6). The effective Soret coefficient S_{eff} now decreases very strongly. When the magnetic field reaches 35 kA/m, the thermophoretic transfer even changes of direction; conversely, the particles start to move toward increasing temperatures. Simultaneously, a very fast increase of the effective magnetic diffusion coefficient D_{eff} is observed. Contrarily to the results of magnetic Soret effect presented in Fig. 4, the separation measurements testify to a strong reduction of thermophoretic mobility $S_{\text{eff}} \cdot D_{\text{eff}}$ up to reaching its negative values with increasing the magnetic field.

It is interesting to note, that after reversal of the particle's transfer direction, the influence of the magnetic field on particle separation gradually vanishes. Obviously, the reason for that is a reduction of the externally applied thermomagnetic driving force by originating a contrarily oriented solute-magnetic driving force. From Eqs. (3) and (4), it follows that after reaching a certain negative concentration difference:

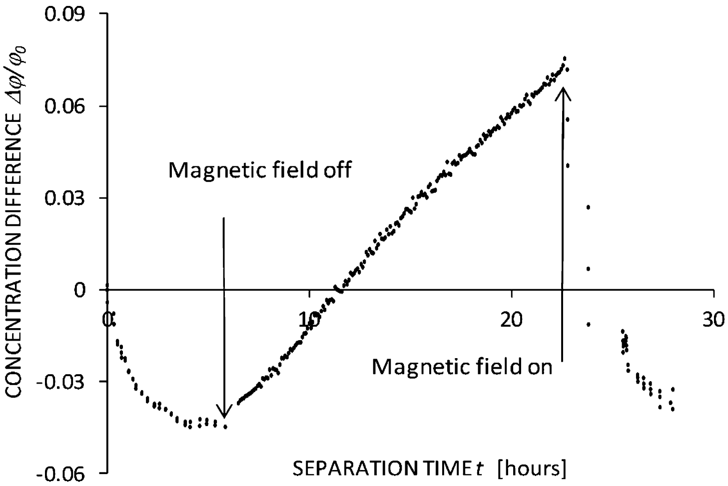

Fig. 7. The reversal of particle separation direction with on/off switched magnetic field $\mathbf{B} = 80$ mT. The saturated value $\Delta\phi/\phi_0 = -0.04$ agrees well with the dependence (13).

$$-\frac{\Delta\varphi_{\text{sat}}}{\varphi_0} = -\alpha_T \Delta T \tag{13}$$

the influence of the magnetic field on the separation process should saturate. The corresponding negative separation curves in Fig. 5 agree well with this prediction.

The performed experiments allow us to propose that a reason of the observed strong influence of the magnetic field on transient particle transfer may be a microconvection induced by nonmagnetic grains of the wall sheets [9] discussed above (see Fig. 2). Additional argument that supports the existence of such microconvective transport mechanism is the unexpectedly fast saturation of particle separation curves and the corresponding strong increase in D_{eff} in the presence of magnetic field (compare the data presented in Fig. 4 and in Fig. 6). Nevertheless, there should not be excluded also another mechanism of fluid mixing inside the layer, which is due to magnetoconvective instabilities. Numerical estimates say that the onset of thermomagnetic instability at low fluid layer thickness is not awaited, whereas appearance of a Soret-driven magnetic convection is very feasible because the solute-magnetic Rayleigh number $Rm_c = \mu_0(M\delta\Delta\varphi/\varphi_0)^2/(\eta D)$ exceeds the critical value, even under weak magnetic fields (for more about the solute-magnetic convection and the critical value Rm_c, see Refs. [1] and [10]). Both, the wall grain-induced microconvection and the solute-magnetic convection accelerates fluid mixing inside the layer. Still, even in presence of internal convection, the translation mass transfer is preserved because it is determined by difference in particle concentrations in both separation chambers and, correspondingly, inside the layer permeable walls. To support this conclusion, we have performed some separation measurements periodically switching on and off the magnetic field. Even at higher fields, when the effective Soret coefficient becomes negative, stable and reversible changes in the direction of the particle separation are observed (see Fig. 7). Relatively slow development of the concentration difference whilst the field is switched off agrees well with the characteristic relaxation time of the particles' thermodiffusive separation, whereas the very fast reversal of the particle transfer direction after the field's switching on, obviously, reflects grid-induced convection.

It is interesting to note that the measured separation curves of present experiments differ from those of similar experiments reported in [8]. Obviously, the reason of that is the significantly lower temperature difference used in those experiments and the attempt to exclude convection in separation chambers by cooling and heating only the ends of cylinder volumes. Under such conditions, the particle transfer through the layer does not evoke a concentration gradient related with that originating from the solute-magnetic driving force. Therefore, now the limit of particle separation level (13) does not exist. By the way, it is surprising that, even at the high particle separation level of 40%, when the solute-magnetic Rayleigh number Rm_c reaches extremely high values, no influence of convective instability was detected. Nevertheless, the dependences of the magnetic Soret coefficient on the longitudinal field $\mathbf{B}//\nabla T$ in the low-field region in both experiments coincide relatively well. For example, the field values of particle transfer direction's reversal in both present experiments and those reported in [8] are almost the same.

4. Conclusion

Experiments on non-isothermal nanoparticle transfer through a thin ferrofluid layer of permeable nonmagnetic walls are performed. A magnetic field that is aligned parallel to the temperature gradient across the layer causes a reduction of the thermophoretic separation significantly stronger than that of the known magnetic Soret effect. Increasing the field, even a reversal of particle transfer direction is observed. The obtained results are interpreted as manifestations of a specific microconvective mass transfer induced by nonmagnetic grid elements of permeable walls.

Acknowledgements

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