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Drift of ferrocolloids through a cylindrical grid by magnetic force

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Abstract
The subject of the paper is to investigate the coupling phenomena of magnetic and non-uniform temperature fields in ferrofluids. The coupling creates a special kind of mass transfer and an inhomogeneous concentration of ferrofluid arises especially near bodies, where higher field gradients are present. Particular attention is paid to the oriented mass transfer, i.e. the magnitude and direction of ferrofluid flux with respect to the temperature gradient and magnetic field. Quantitatively, oriented phoretic transport can be characterized by the magnetic Soret coefficient and osmotic pressure difference. The problem is solved using two-dimensional (2D) numerical simulations for the periodic structure of the bodies. Special attention is paid to the magnetic bulk force as the driving force.

1. Introduction

Experimental observations show that the presence of a temperature gradient and magnetic field creates a notable separation of ferrofluid in a system divided by a grid [1]. Separation of ferrofluid occurs also without the presence of magnetic field due to other (thermal expansion, surface tension) effects of colloidal particles, leading to conventional thermodiffusion or the so-called Soret effect [6], where the ferrofluid particles predominantly move to lower temperatures. Our aim here is to discuss in more detail the ferrofluid convection and subsequent separation by the action of the magnetic field force, resulting in the magnetic Soret effect, whereas other mechanisms, such as gravitational sedimentation, are neglected. Various types of directed mass transfer around bodies due to thermal, electric, magnetic perturbations near them are discussed in [4]. The presence of phoretic transport there is explained by inhomogeneous fields near the bodies creating interfacial forces. The phoretic transport in magnetic fluid due to magnetic field can be explained in a similar manner, as will be described later. Knowledge about directed mass transfer in ferrofluid due to magnetic force is still insufficient and different opinions about the magnitude and even the direction of the flow exist. Therefore, detailed descriptions of heat and mass transfer processes are required [2]. Analytical estimations are possible only in the simplest cases, e.g. ferrofluid convection around a sphere [5].

The heat and mass transfer processes here are analysed numerically by a two-dimensional model (2D) in order to estimate the role of convection around micron size bodies. As a particular example, a grid consisting of a large number of equidistantly spaced non-magnetic or magnetic cylinders is considered. Such a system is shown in figure 1. The temperature gradient is applied in the x-direction while cylinders of the grid are placed along the y-axis. The magnetic field can be oriented arbitrarily but we are particularly interested in magnetic field oriented either in x or y directions. The orientation of magnetic field is very important as it can even invert the direction of phoretic transport. Due to significant convective exchange far from the grid the distributions of temperature and concentration of the ferrofluid are approximately homogeneous far from the grid but significant gradients of concentration and temperature are possible close to the grid. As the characteristic Lewis number is much higher than 1 the concentration boundary layer in reality could be smaller than the temperature boundary layer. The possible break up of the periodical symmetry is neglected. Non-uniform magnetization of ferrofluid surrounding each cylinder creates a body force in the ferrofluid and subsequent microconvection. Therefore, distributions of temperature, magnetic field, concentration, and velocity have to be obtained for a correct description of the microconvection.

2. Equations describing microconvection

The convective heat transfer near the grid is neglected as the characteristic Peclet numbers are very small: typically

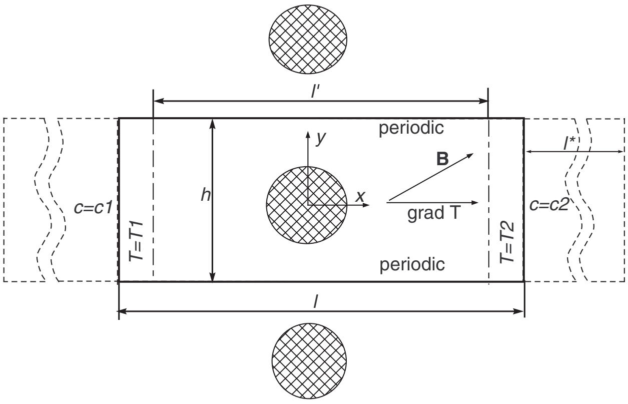

Figure 1. Schematic shape of the grid in the system surrounded by ferrofluid.

10^{-4}–10^{-2}. Therefore, stationary temperature distribution is only required $\nabla \cdot (\lambda \nabla T) = 0$. The analytical solution for one cylinder in ferrofluid with homogeneous heat conductivity of ferrofluid is:

$$T = T_0 + \left(\frac{\partial T}{\partial x}\right)_0 x \left(1 + \frac{r_0^2}{r^2} K_\lambda\right), \quad (1)$$

where $(\frac{\partial T}{\partial x})_0$ is the temperature gradient far from a cylinder with radius r_0, $K_\lambda = \frac{\lambda_e - \lambda_i}{\lambda_e + \lambda_i}$. Because the stationary flow and magnetodiffusion does not necessary converge in the direction of temperature gradient, as tested by analysis and calculations, the area with inhomogeneous temperature is chosen to be finite: $-l'/2 \leqslant x \leqslant l'/2$ and $l' < l$. Therefore, it is assumed that the temperature is T_1 at $x < -l'/2$ and $T = T_2$ at $x > l'/2$.

Magnetic field intensity $\mathbf{H}$ is calculated by the gradient of the scalar magnetic potential Ψ: $\mathbf{H} = \nabla \Psi$. Requiring divergence free magnetic induction one obtains $\nabla \cdot (\mu \nabla \Psi) = 0$. The particular solutions are analogous to those of the temperature field if the relative magnetic permeability μ in the ferrofluid is homogeneous. However, the magnetic permeability depends on magnetic field, ferrofluid concentration, and temperature. The last one can give the oriented convection along the temperature gradient or in the opposite direction. We use the Langevin equation to estimate the relative magnetic susceptibility χ:

$$\chi = c\frac{M_s}{H} L(\xi), \qquad L(\xi) = \frac{1}{\tanh \xi} - \frac{1}{\xi}, \qquad \xi = \frac{V_p \mu_0 M_s H}{k_B T}, \quad (2)$$

where M_s is saturated magnetization, V_p the volume of the ferroparticle, k_B the Boltzmann constant, c the volume fraction of ferrofluid. The dependence on temperature is not straightforward because the size of the particle and saturated magnetization are also temperature dependent. Therefore, we will use the experimentally obtained dependence of magnetization on temperature:

$$\mu = 1 + c\frac{M_s}{H_0} L(\xi_0) + \frac{1}{H_0}\frac{\mathrm{d}M}{\mathrm{d}T}(T - T_0), \qquad \frac{\mathrm{d}M}{\mathrm{d}T} = -Ac. \quad (3)$$

Thus, the magnetic susceptibility is calculated by the Langevin equation with experimentally observed slope of susceptibility on temperature characterized by parameter A, which works well in the required range of temperatures ≈15–30 °C. Magnetic force density usually is approximated by the Kelvin formula $\mathbf{f} = \mu_0 \mathbf{M} \nabla \mathbf{H} = \frac{1}{2}\mu_0 \chi \nabla H^2$. If magnetic susceptibility varies linearly with temperature as in (3) then the curl of magnetic force around the cylinder is

$$\nabla \times \mathbf{f} = 2\mu_0 H_0^2 \frac{r_0^2}{r^6} K_\mu \left(\frac{\partial \chi}{\partial x}\right)_0 \times \left[r^3 \sin(3\varphi - 2\varphi_0) - K_\mu r_0^2 r \sin\varphi - K_\lambda r_0^2 \left(K_\mu \frac{r_0^2}{r}\sin\varphi + r\sin(\varphi - 2\varphi_0)\right)\right], \quad (4)$$

where φ_0 is the angle between the magnetic field and temperature gradient, $K_\mu = \frac{\mu_e - \mu_i}{\mu_e + \mu_i}$. There exists also a Helmholtz [3] representation of magnetic force instead of the Kelvin one which could change the description of the ferrofluid at high concentrations. Recently, there has been some controversy [7] about the role of the Helmholtz force. A later article by Engel [8] showed that experiments do not necessarily invalidate the Kelvin force. Consequently, the Kelvin formulation of magnetic force will be used in the current studies. Gravitational force is usually lower than the magnetic force for particles with diameters lower than 10 nm and is neglected.

Magnetic force acting on ferrofluid particles drives the fluid and causes a redistribution of ferrofluid concentration. Incompressible fluid flow is calculated in potential formulation with velocity potential ψ and vorticity ω in the 2D case

$$\partial_t \omega + (\mathbf{v} \cdot \nabla)\omega = \frac{1}{\rho}\left(-\nabla \times (\nabla \times (\eta\omega)) + (\nabla \times \mathbf{f})\right), \quad (5)$$

$$\omega = \nabla \times \mathbf{v}, \qquad \mathbf{v} = \nabla \times \psi. \quad (6)$$

Typically, the resulting Reynolds numbers for the given system with respect to grid elements are small ($Re \ll 10$) resulting in laminar behaviour of the fluid flow. The analytical solution of velocity potential in the asymptotic limit for one cylinder and infinite ferrofluid according to (4) is

$$\psi = \frac{\mu_0 H_0^2}{32\eta} K_\mu \left(\frac{\partial \chi}{\partial x}\right)_0 \times \left[r\left(1 - \frac{r_0^2}{r^2}\right)^2 \times \left(\sin(3\varphi - 2\varphi_0) - \frac{\sin\varphi}{3} K_\mu K_\lambda\right) - \left(\frac{r^2}{r_0^2} - 1 - \ln\frac{r^2}{r_0^2}\right) \times \frac{2r_0^2}{r}\left(K_\mu \sin\varphi + K_\lambda \sin(\varphi - 2\varphi_0)\right)\right] \quad (7)$$

with six vortexes around the cylinder. The velocity does not decay with the distance from the cylinder, i.e. flow of ferrofluid occurs in all the area with inhomogeneous temperature. Therefore, the area with a temperature gradient in the real case should be bounded as before. As the velocity is periodic along the grid in the system represented in figure 1, a constant difference of velocity potential exists between both sides: $\psi_z|_{y=h_2} - \psi_z|_{y=-h_2} = const$. The constant is obtained either by setting the total flow of fluid through the system to zero,

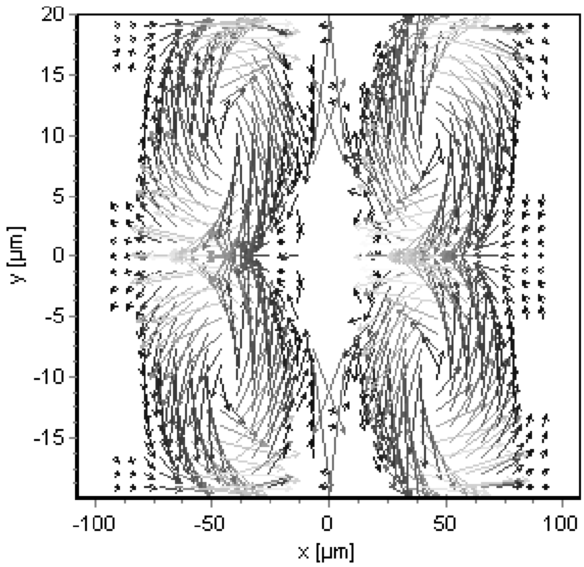

Figure 2. Stationary flow in ferrofluid with $c_0 = 0.05$ around the cylinder. Magnetic field $B = 0.0375$ T is oriented along the temperature gradient.

which yields $const = 0$, or by setting the pressure difference between boundaries $x = \pm l/2$. For the boundary conditions at $x = \pm l/2$ it is assumed that the tangential velocity component $\partial_x \psi$ and the normal vorticity gradient $\partial_x \omega$ are zero at these boundaries.

Because the magnetic force acts on nanoscale ferroparticles of the fluid, the concentration of them is no longer homogeneous [9]. Moreover, the distribution of ferrofluid concentration c (volume fraction of ferroparticles) is important to describe the oriented drift of ferroparticles, i.e. the Soret effect. The distribution of ferroparticles satisfies the mass conservation law $\frac{\partial c}{\partial t} + \nabla \cdot \mathbf{j} = 0$. If the ferroparticles are spherical and with equal radius r_p then the ferroparticle flux in Stokes' formulation is

$$\mathbf{j} = \mathbf{v}c - D\nabla c + D\frac{V_\text{p}}{kT}\mathbf{f}, \qquad D = \frac{kT}{6\pi r_\text{p}\eta}, \qquad (8)$$

where flux $\mathbf{j}$ contains convection, diffusion, and drift terms, respectively. The boundary conditions at $x = \pm l/2$ are chosen in such a way that reservoirs with intense mixing are present on both sides. Absence of deposition is assumed near the bodies, i.e. $j_\text{n} = 0$. Experiments, however, show that deposition is possible near the bodies and it should be discussed in future studies. The Soret coefficient is proportional to the ferroparticle flux through a vertical cross-section of the system [2]:

$$S_t(x, t) = -\frac{1}{D(T_0)Sc_0\left(\frac{\partial T}{\partial x}\right)_0}\int_x j_x \,\mathrm{d}S, \qquad (9)$$

where c_0 is the average concentration of ferrofluid.

The non-magnetic Soret effect can be included by adding a term to the drift part $S_0 D c \nabla T$ of the flux (8), where S_0 is the usual Soret coefficient. However, this term is omitted in numerical studies in order to see the effect of inhomogeneous magnetization.

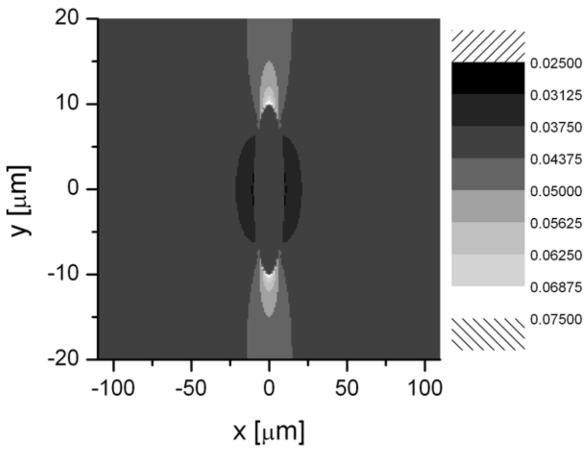

Figure 3. Stationary distribution of concentration corresponding to figure 2.

3. Ferrofluid flow around cylindrical bodies

Let us consider the flow of ferrofluid through the grid in figure 1. At first, magnetic field oriented parallel to the temperature gradient is considered. The parameters are assumed as in the previous subsection with the exception that magnetic field strength is $B = 0.05$ T and initial concentration $c = 0.04$. In addition, we set the radius of the cylinder to be $r_0 = 10$ μm; vertical spacing $h = 40$ μm; temperature difference $\Delta T = 10$ K is applied on the spatial distance $l' = 0.4$ mm; heat conductivities are $\lambda_e = 0.2$ W m^{-1} K^{-1} and $\lambda_i = 10$ W m^{-1} K^{-1} for ferrofluid and body, respectively; density of ferrofluid $\rho = 1000$ kg m^{-3}. Reservoirs with homogeneous concentrations are attached at both sides $|x| > l/2$ of the system as shown in figure 1. The length of each reservoir l^* is set equal to l.

As can be seen from figure 2, the flow of ferrofluid converges well in the horizontal direction, i.e. the intensity of flow drops considerably in the dimensions of the system. Nevertheless, the concentration and pressure varies in all the area with a temperature gradient. Figure 3 shows that the area with higher concentration is located in the grid plane along the y-axis, similarly to that in [2]. If the magnetic field is oriented along the y-axis then the areas with higher and lower concentrations would be opposite.

Figure 4 shows the development of the concentration difference between both sides of the grid. One notes that the ferroparticles are transferred towards higher temperatures in the case of magnetic field orientation along the x-axis, i.e. concentration in the right reservoir c_2 becomes higher than in the left c_1 and the resulting magnetic Soret coefficient is negative. That can also be seen in figure 5, where one can see that the resulting force far from the cylinder inside the area with a temperature gradient is directed towards increasing temperatures (decreasing μ). Various factors that can influence the relaxation time and stationary difference of concentrations with respect to the default case are examined in figure 4: a grid of double the thickness gives lower magnetic field, lower temperature and lower concentration. Curve 1 corresponds

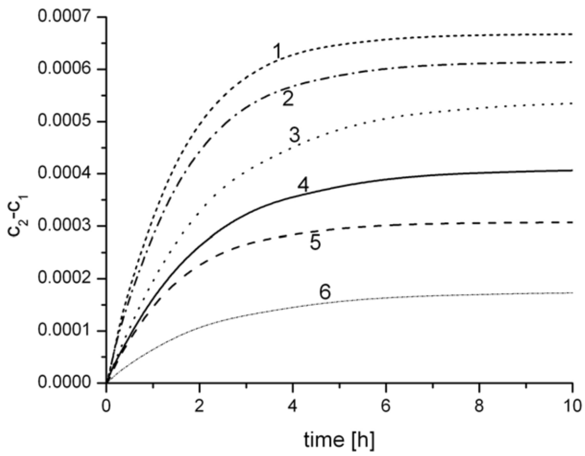

Figure 4. Development of concentration difference on both sides of the grid $x = \pm l/2$ with elliptic elements at $B = 0.05$ T. (1) A grid twice as thick, (2) default parameters, (3) $B = 0.025$ T, (4) infinitesimally thin grid elements, (5) $\Delta T = 5$ K, (6) $c_0 = 0.02$.

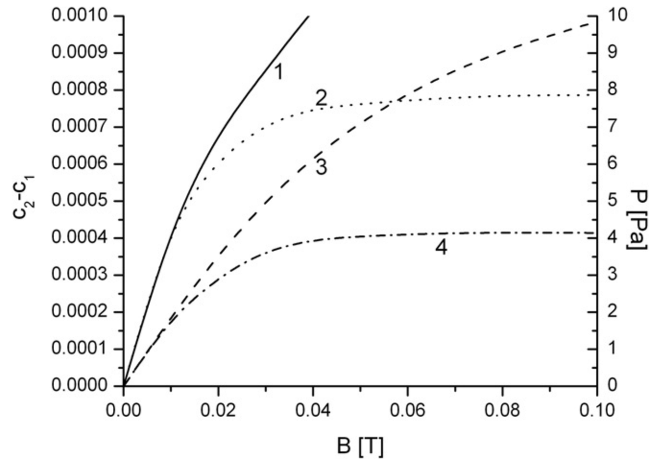

Figure 6. (1), (3) Stationary difference of concentration and pressure without change of magnetic field; (2), (4) with change of magnetic field.

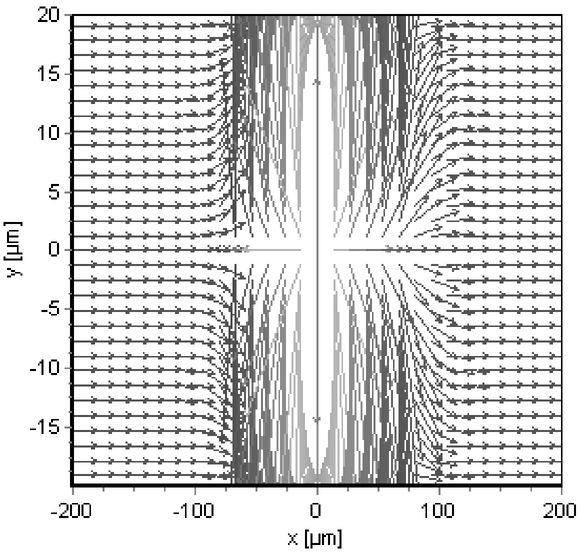

Figure 5. Stationary distribution of magnetic force density **f** (scaled).

to the case where the cylinder is twice as thick in the x-direction. As can be seen, it only slightly increases the effect of separation. This suggests that the influence of the body in total transfer of ferrofluid is quite small. More influence comes from the inhomogeneous magnetic intensity H far from the cylinder, as will be shown later. Curve 3 corresponds to twice as small magnetic field $B = 0.25$ T. Then the stationary difference of concentrations becomes smaller and the characteristic relaxation time longer. The case with reduced temperature difference $\Delta T = 5$ K is plotted by curve 5, where Δc_∞ is approximately halved while the characteristic relaxation time remains the same. Curve 6 corresponds to a halved concentration $c_0 = 0.025$, where Δc_∞ is more than halved and the relaxation time is only slightly longer. Curve 4 represents the case where the grid is infinitesimally thin but the transfer of ferroparticles towards higher temperatures is still present. This is because magnetic induction **B** is fixed at both sides of the system and equal whereas magnetic intensity **H** is changing due to change of magnetic permeability μ. Then the Kelvin force equals

$$\mathbf{f} \approx \frac{\chi B^2}{\mu\mu_0}\nabla\frac{1}{\mu} \tag{10}$$

far from the cylinder, where B^2 is nearly constant according to the given boundary conditions ($\nabla \cdot \mathbf{B} = 0$). We can conclude that effects of the microconvection around the grid and the oriented magnetic force in the inhomogeneously magnetized magnetic fluid account for the transfer of ferroparticles towards higher temperatures in the present formulation of boundary conditions and force. The time development of concentration difference on both sides of the system from the initial state in figure 4 can be described fairly well by an exponential law $\Delta c(t) \approx \Delta c_\infty(1 - \mathrm{e}^{-\alpha t})$, where Δc_∞ is the stationary difference of concentrations. The Soret coefficient is equal to

$$S_t = \frac{1}{2}\frac{\partial \Delta c}{\partial t}\frac{l^* l'}{D c_0 \Delta T}. \tag{11}$$

As the Soret coefficient characterizes the initial stage of the process, the derivative is $\partial\Delta c/\partial t|_{t=0} \approx \Delta c_\infty \alpha$. The coefficients Δc_∞ and α can be estimated from (8) and (10):

$$\Delta c_\infty \approx \frac{\frac{A}{M_s} c_0 \Delta T}{\frac{1+\chi_0}{\xi_0\chi_0} + L(\xi_0)},$$
$$\left.\frac{\partial \Delta c}{\partial t}\right|_{t=0} \approx \frac{2\pi r_p^2 M_s \mu_0}{9\eta(1+\chi_0)} \frac{c_0^2 L(\xi_0) A \frac{\Delta T}{l'}}{l^* + \frac{l-l'}{2} + \frac{l'}{8}}. \tag{12}$$

The resulting osmotic pressure differences are given in figure 6. The stationary state is achieved with an order of days, depending on the size of the system around 1 cm as the directed ferroparticle flow is relatively weak through the layer of $l = 0.46$ mm. The process agrees quite well with experiments carried out in our Institute of Physics, giving the right value

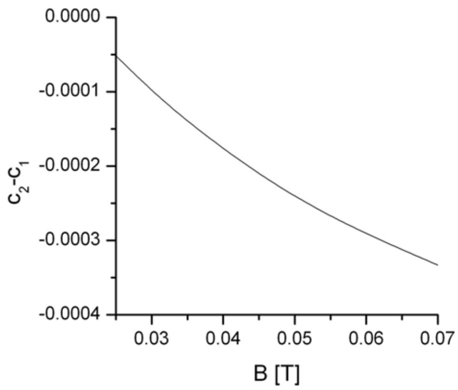

Figure 7. Stationary difference of concentrations on both sides of the grid at a perpendicular orientation of the magnetic field to the temperature gradient.

of pressure difference. That of course depends on how we set the size parameters l and l' of the concentration and temperature boundary layers, respectively. Figure 6 shows that equilibrium concentration and pressure difference between both sides becomes saturated at $B \approx 0.05$ T. The change of magnetic field distribution is very important considering non-magnetic bodies, as can be seen in figure 6 especially at magnetic field induction over 0.03 T when the difference of concentrations starts to saturate. The curve 4 in figure 6 agrees quite well with (12) where the direct absence of a grid is neglected.

Figure 7 shows the development of the concentration difference in the case of magnetic field $B = 0.05$ T oriented perpendicularly with respect to the temperature gradient. In this case the ferroparticles are transferred towards lower temperatures but the quantitative effect is approximately half that in the parallel case. The problem of magnetic force far from the grid is absent since inhomogeneous magnetization does not create bulk magnetic force in this orientation of magnetic field.

4. Conclusions

Interesting coupling phenomena of magnetic, concentration, and flow fields take place in the case of a ferrofluid around a non-magnetic body in the presence of both a magnetic field and temperature gradient. Particular attention should be paid in considering the Soret effect and osmotic pressure difference in order to strictly maintain the mass conservation laws, e.g. solving the system by finite volume methods. The problem related to the convergence of flow far from the grid is solved by setting the area with a temperature gradient to be finite. The calculations show that, from the viewpoint of the model, the directed transfer of ferroparticles is caused not only by the separating grid but also by the magnetic bulk force. If the magnetic field is oriented parallel to the temperature gradient then both these effects add to the transfer of ferroparticles towards higher temperatures. The effect takes the opposite sign in a perpendicular orientation of the magnetic field with respect to temperature gradient. The main problems in the model that should be solved are a correct description of the temperature and concentration boundary layers, having different scales, and an unambiguous description of the resulting magnetic force far from the grid.

Acknowledgments

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