THERMODIFFUSION IN MAGNETIC FLUIDS

T. Völker, S. Odenbach

ZARM, Universität Bremen, Am Fallturm, 28359 Bremen, Germany
(voelker@zarm.uni-bremen.de)

Investigations were made to determine the Soret coefficient of magnetic particles in a ferrofluid under the influence of a magnetic field. This so-called magnetic Soret effect was theoretically determined to be two to three orders of magnitude smaller than the conventional Soret effect. In contrast. former experiments have shown that the magnetic Soret effect is much higher than the theoretical predictions. However, in the experiments the influence of buoyancy and magnetic driven convection cannot be excluded. So the question how large the magnetic Soret effect can be is still open. Thus an experimental setup was developed to minimize parasitic effects and thus to simplify the analysis of the experimental results. These results show that the magnetic Soret effect can even be higher as the conventional one and confirm therewith former experimental results.

Introduction. The Soret effect also called thermodiffusion is a composite effect in which an external temperature gradient leads to a concentration gradient of one component in a two-component fluid [1, 2]. The strength of the effect is characterized by the Soret coefficient which takes values of about 10^{-4} K^{-1} for molecular mixtures. In suspensions it can be about three orders of magnitude higher, making the effect a phenomenon of technical importance. For ferrofluids it was shown that the Soret coefficient can be additionally influenced by magnetic fields [3]. To determine the Soret coefficient and the influence of a magnetic field thermodiffusion columns have been employed [4, 5]. In such kind of experiments the interplay between thermodiffusion and thermal convection is used to amplify the separation process. A temperature gradient in a vertical flat gap ($d \approx 0.5$ mm) induces, on the one hand, the thermodiffusion of particles towards the cold wall producing a concentration along the distance between the walls. On the other hand, it drives thermal convection that leads to a change of concentration of magnetic particles in two chambers connected to the upper and lower end of the channel. This concentration change in the chambers is used to determine the Soret coefficient of the nanoparticles indirectly. The advantage of this technique is the possibility to measure the particle separation dynamics during the separation process with a high accuracy. With this technique the Soret coefficient of magnetic particles in a ferrofluid was determined [6]. A backdraw of this method is obviously the combination of convective effects with diffusive ones. The determination of the Soret coefficient can only be undertaken under an assumption that the convective flow profile is well known and not disturbed by any parasitic effects. The restrictions of the thermodiffusion column as a tool for determination of the Soret coefficient gain even more serious importance if experiments are performed to determine the Soret effect in a magnetic fluid in the presence of magnetic fields. In such a situation the appearance of thermomagnetic convection in a fluid layer which is subjected to a magnetic field can strongly influence the flow in the diffusion column and can thus lead to intractable problems in the interpretation of the data and, in particular, in the determination of magnetically driven changes of the Soret coefficient. To overcome the problems resulting from the coupling of the magnetic influences on the convective flow as well as on the thermodiffusive transport, we developed an experimental setup which allows the investigation of thermophoretic changes in the concentration distribution of magnetic particles in a ferrofluid in an arrangement suppressing convective effects.

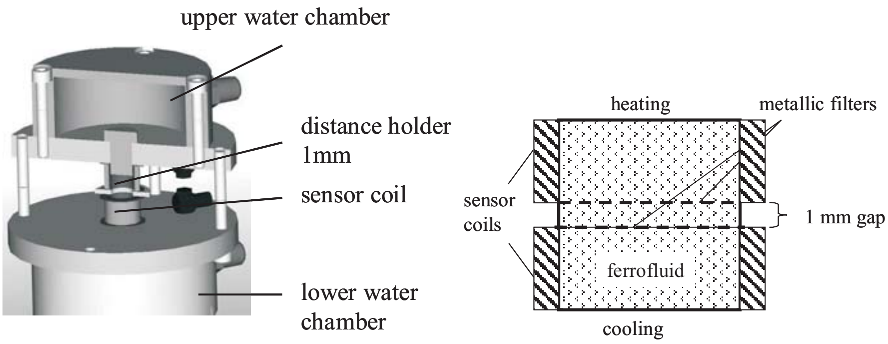

Fig. 1. The convectionless free column.

1. Experimental setup. This setup consists of a cylindrical fluid layer with a height and a diameter of 10 mm. To ensure that the fluid is at rest and that thus no parasitic effects affect the determination of the Soret coefficient, any kind of convective motion has to be suppressed.

Thermal convection can in principle be suppressed by heating the fluid layer from the top and cooling it from the bottom. For the investigation of ferrofluids under the influence of magnetic fields it has to be observed that a magnetic field aligned with the temperature gradient can drive thermomagnetic convection even if the fluid layer is heated from above [7].

To suppress this so-called thermomagnetic convection over a wide range of magnetic field strengths, two nonmagnetic metallic filters have been placed in the center of the fluid cylinder in a way that they form a fluid gap with an effective height of 1 mm (see Fig. 1). Within this layer the magnetic Rayleigh number as well as the conventional one is of the order of 1 ($\mathrm{Ra} \sim d^3$) and thus no convection will appear in this fluid region even if strong magnetic fields are applied.

To measure the Soret coefficient of the nanoparticles under the influence of a magnetic field, the cell can be orientated parallel or perpendicular to the direction of a magnetic field produced by an electromagnet. The determination of particle concentration in the ferrofluid is carried out by means of inductance changes in sensor coils as seen in Fig. 1 [8].

Experiments have been performed with a kerosene based ferrofluid containing magnetite particles with a volume concentration of $c_0 = 0.02$ for all experimental results shown here. The mean particle diameter $d = 9$ nm with a small size distribution has been calculated by magnetogranulometry from magnetization curves [9]. The temperature difference over the whole fluid cylinder of 10 mm was 10 K ($T_{\mathrm{bottom}} = 20°\mathrm{C}$, $T_{\mathrm{top}} = 30°\mathrm{C}$), providing a temperature gradient of 1 K/mm over the fluid layer between the metallic filters. The viscosity $\nu = 2.4$ mm^2/s was measured by means of a capillary viscosimeter. The Brownian diffusion coefficient $D_0 = 2 \cdot 10^{-11}$ m^2/s was calculated from the experimentally determined fluid data.

2. Experimental results. Experiments were carried out for 8 different magnetic field strengths between $H = 0$ kA/m and $H = 320$ kA/m. The magnetic field direction was either orientated parallel or perpendicular to the temperature gradient.

Fig. 2 shows the separation dynamics, i.e., the temporal development of the changes of the normalized concentration difference for different magnetic field strengths and orientations, respectively.

Starting with the situation $H = 0$, the linear fit of the experimental data gives a Soret coefficient for the magnetic nanoparticles of $\mathrm{S_T} = +0.16\,\mathrm{K}^{-1} \pm 0.02\,\mathrm{K}^{-1}$ [9]

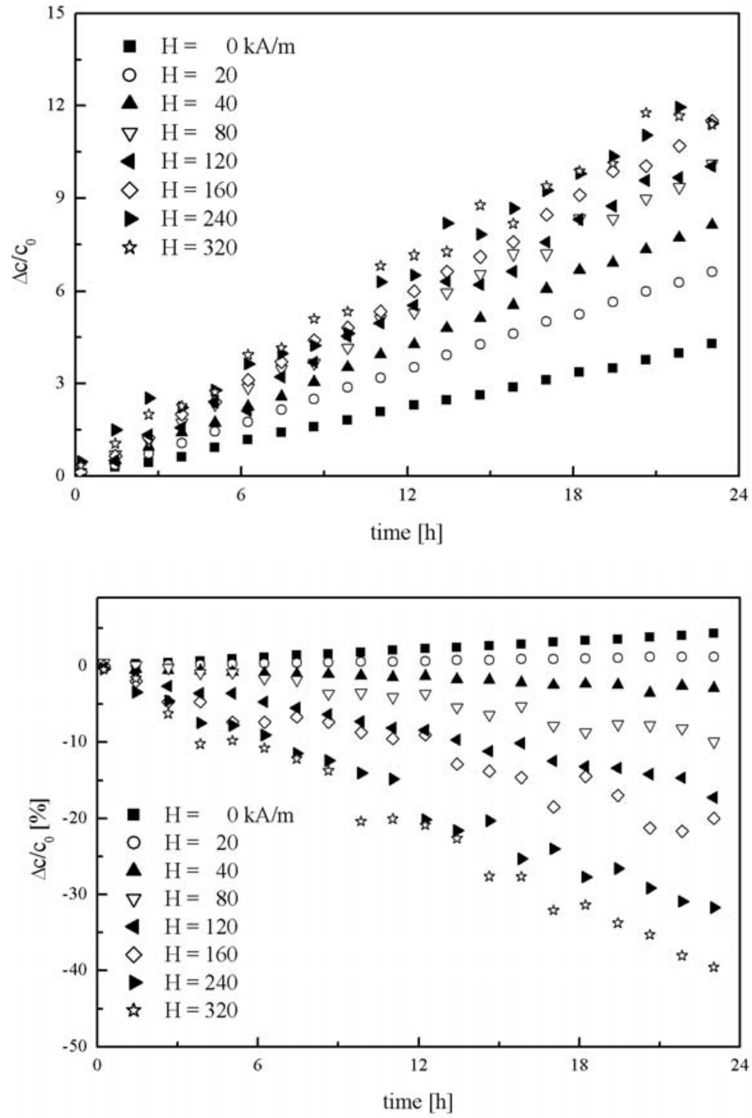

Fig. 2. The separation dynamics for (*a*) $H \perp \nabla T$ (top) and (*b*) $H \parallel \nabla T$ (bottom).

which is practically the same value ($S_T = +0.15\,K^{-1}$) as it has been found using the thermodiffusion column [4, 5].

As seen in Fig. 2*a*, the separation increases with the increasing magnetic field strength for the perpendicular orientation of magnetic field direction and temperature gradient, while a decrease is observed for the parallel orientation (see Fig. 2*b*). This agrees qualitatively well with the standard theories on thermodiffusion in magnetic fluids under influence of magnetic fields. But it is also seen that the field dependent change of the strength of the separation is of the same order of magnitude as the ordinary Soret effect in the absence of a magnetic field. Thus it is about three orders of magnitude stronger than predicted theoretically. The effective Soret coefficient calculated from these measurements is plotted in Fig. 3 as a function of the magnetic field strength for both relative directions of the magnetic field. If it is assumed that the effective Soret coefficient $S_{T,\mathrm{eff}}$ can be separated into the ordinary Soret coefficient and a so-called magnetic Soret

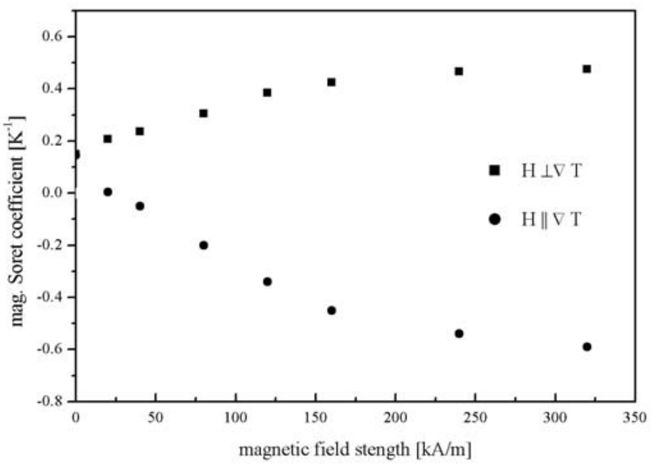

Fig. 3. The field dependence of the Soret coefficient for $H \perp \nabla T$ and $H \parallel \nabla T$.

coefficient accounting for all magnetic field induced changes, one can evaluate the magnetic part by subtracting $\mathrm{S_{T,0}}$ from $\mathrm{S_{T,eff}}$.

As seen, the effect saturates for magnetic fields of about 300 kA/m leading to a maximal magnetic Soret coefficient for the perpendicular arrangement of $\mathrm{S_T^m} = +0.35\,\mathrm{K}^{-1} \pm 0.02\,\mathrm{K}^{-1}$ which is more than two times larger than the conventional Soret coefficient. In the parallel arrangement the effective Soret coefficient becomes even negative for a magnetic field strength larger than 20 kA/m and the saturation value of the magnetic Soret coefficient equals $\mathrm{S_T^m} = -0.75\,\mathrm{K}^{-1} \pm 0.02\,\mathrm{K}^{-1}$ [10].

A theoretical approach for an explanation of the strength of the magnetic Soret effect found in the experiments discussed above is given in [11]. This approach bases, in contrast to the microscopic theory in [3], on the macroscopic description of ferrofluid dynamics given by Müller and Liu [12].

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