Thermodiffusion in magnetic fluids

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Abstract

Separation experiments in a ferrofluid are performed in a thermodiffusion column, which consists of a vertical flat ferrofluid layer between two walls held at different temperatures. By using a modified separation theory which takes into account the particle mass flux due to thermodiffusion and a convective transport flow, one can calculate the Soret coefficient from the unsteady part of experimentally found separation curves. In addition we performed experiments in which we reach the steady state of the process. A new model allows to calculate the Soret coefficient also from this part of the separation curve. The value found here is in very good agreement with that from the unsteady part.
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1. Motivation and theoretical background

Transport properties in magnetic colloids play an important role concerning the problem of long-term stability of ferrofluids. An attempt to measure particle mobility in the fluid was made by Blums in 1983 [1]. The investigations were based on non-stationary particle separation measurements using a thermodiffusion column. It consists of a vertical flat channel—a small gap between two plates held at different temperatures T_1 and T_2—and two connected separation chambers. A concentration profile in the gap develops, caused by thermal diffusive particle transfer along the temperature gradient. In combination with convective transport in the gap this leads to an increase of particle concentration in the lower and an analog decrease in the upper chamber.

At the moment two analytical calculations for the Soret coefficient based on a description of the separation problem exist: One is valid for the initial part of separation and the other for the steady concentration difference. Up to now the corresponding Soret coefficient is commonly evaluated from the unsteady part of separation curves by using an empirical analysis [2]

because of limited experimental time and experimental problems at the initial part of separation. To get a closer insight into these problems, we have designed two independent experimental setups with identical thermodiffusion columns and measuring devices, one for long-time experiments and the other for short-time experiments.

In the initial regime ($t \approx 200\,\mathrm{s}$) of the separation process, until the concentration boundary layers reach the center of the channel, an exact analytical solution for the concentration in the reservoirs is valid:

$$\frac{\Delta c}{c_0} \propto t^{2.5}. \tag{1}$$

The present paper deals also with experimental results of separation dynamics in the thermodiffusion column in which we reach the regime of steady concentration difference. In this state of separation the concentration difference follows [3]:

$$\frac{\Delta c}{c_0} \approx \frac{\sigma L D_0}{\delta D_c}. \tag{2}$$

Here $\Delta c/c_0$ is the normalized concentration difference between the upper and lower chambers, σ is the non-dimensional particle separation parameter, L is the height of the separation channel, D_0 is the Brownian diffusion parameter, δ is the width of the gap and D_c is

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the convective diffusion coefficient. For small values of S ($S = k\,Gr_c/Gr_T$, $Gr_c = g\beta_c c d^3/v^2$ is the concentration Grasshof number, $Gr_T = g\beta_T\,\Delta T\,d^3/v^2$ the thermal Grasshof number and $k = S_T\Delta T$ is a non-dimensional thermodiffusion parameter, which is proportional to the Soret coefficient S_T) the concentration difference in the column chambers is given in the asymptotic regime by [4]

$$\frac{\Delta c}{c_0} = 504\,\frac{kL}{Gr_T\,Sc\,\delta},\tag{3}$$

where $Sc = v/D_0$ is the Schmidt number.

The time to reach a steady concentration difference is in the order of weeks. If concentration increases ($S \gg 1$), a higher separation difference is reachable but the experimental time increases rapidly as well.

In the present paper we compare the Soret coefficient for magnetite particles found from the steady and unsteady part of the separation process.

2. Numerical calculations

A one-dimensional numerical integration program was developed to calculate the time dependence of the concentration profile in the gap. Calculations were made by assuming a hydrodynamic particle diameter of 13 nm, viscosity of $2.3\,\text{mm}^2/\text{s}$, $S_T = 0.16\,\text{K}^{-1}$ and $\nabla T = 20\,\text{K}/\text{mm}$. From the equation of motion of the particles it follows, that in the initial regime of the separation process a non-linear dependence is awaited. This dependence is valid until the thickness of the concentration boundary layers reaches the center of the channel. For the calculated Brownian diffusion coefficient and for the given gap width ($d = 0.5\,\text{mm}$) this time is determined from the numerical calculation as 180 s. This value agrees well with the analytically calculated times predicted in Ref. [3].

The time development of the concentration distribution in the gap is shown in Fig. 1a–d. After a transition time ($t \geqslant 9\,\text{h}$) the concentration profile follows an exponential law (see Fig. 1d).

3. Experimental procedure and setup

The separation measurements are performed using a vertical flat column (Fig. 2) of width $\delta = 0.5\,\text{mm}$ and height $L = 90\,\text{mm}$. The heated and cooled walls are held at constant temperatures $T_1 = 20°\text{C}$ and $T_2 = 30°\text{C}$. Particle concentration in both chambers is determined by measuring the resonance frequency of two independent LC-oscillators. Therefore the coils inside the two chambers are connected with two independent oscillators. The inductance of the coils increases linearly with volume concentration of magnetic particles, leading to a
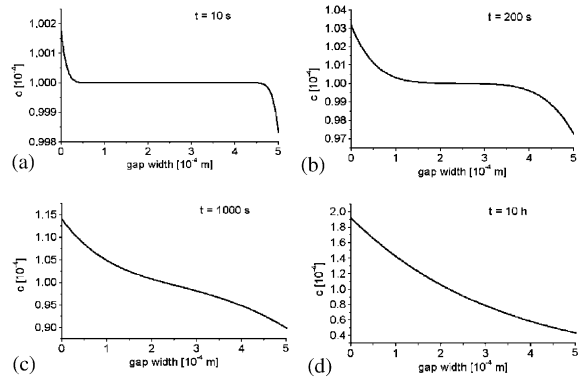

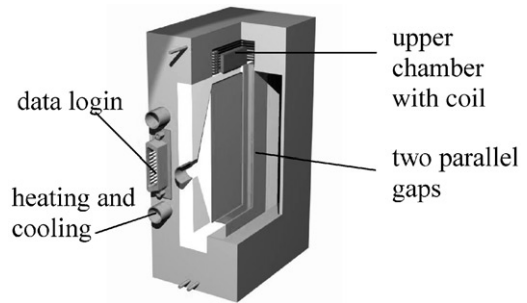Fig. 1. (a–d) Concentration profile in the gap for the various times.

Fig. 2. The thermodiffusion column.

decrease of the resonance frequency of the connected oscillator.

4. Results and discussion

Fig. 3 shows the separation curve in the initial part measured in zero field with $\Delta T = 10\,\text{K}$ ($T_1 = 20°\text{C}$, $T_2 = 30°\text{C}$, $T_0 = 25°\text{C}$). The plotted thin lines are the curves from the analytical model for different values of the Soret coefficient. The measured separation curve shows the $\Delta c/c_0 \sim t^{2.5}$ behaviour for times $t \leqslant 200\,\text{s}$. The small difference to the calculated curves reflects not only the measurement error but also the uncertainty of D_0. Expressions used for the calculation are based on the assumption of monodispersity of particles and negligibility of particle interaction. The comparison between calculated and experimentally found curves gives a value for the Soret coefficient of $S_T = +0.15K^{-1} \pm 0.02K^{-1}$.

Fig. 4 shows the long-term development of the separation process up to saturation, which is reached after approximately 51 days [5]. This investigation has been performed for $H = 0$, using a temperature difference of 8 K ($T_1 = 29°\text{C}$, $T_2 = 37°\text{C}$).
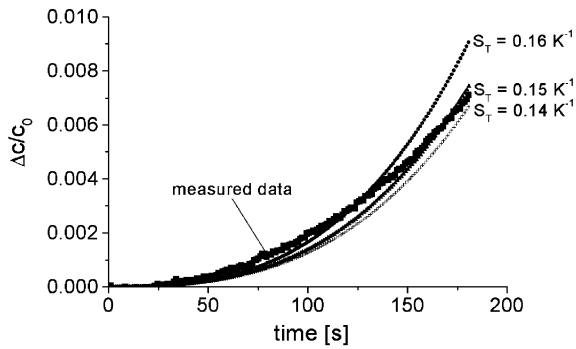
Fig. 3. Initial part of the thermodiffusion process.

Fig. 4. Steady state of the separation process.

The separation level $\Delta c/c_0 \approx 1.72$ is extremely high; from the initial concentration $c_0 = 0.017$ the concentration in the lower chamber is $c_l = 0.0326$ and $c_u = 0.0012$ in the upper one. Between the initial and saturation part we found time ranges with linear $(250 \leqslant t \leqslant 2000\,\mathrm{s})$ and square root time dependence of Δc $(2100\,\mathrm{s} \leqslant t \leqslant 10\,\mathrm{h})$, the latter one being predicted theoretically. The Soret coefficient determined from the steady part of the separation curve is $S_\mathrm{T} = 0.13\,\mathrm{K}^{-1}$. This is in very good agreement with the value found from the unsteady part of the separation process.
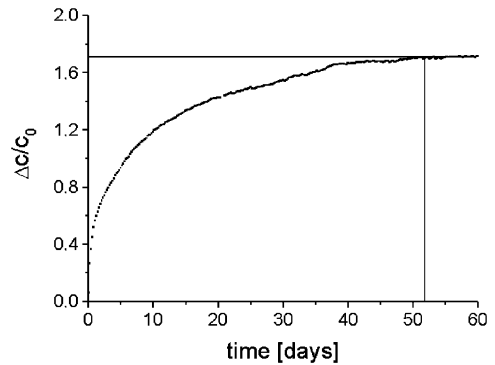
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