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# Thermophoretic separation of a ferrofluid–water emulsion: preliminary experiments

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#### Abstract

The Soret effect in an emulsion (microdroplets of hydrocarbon based ferrofluid in water) is evaluated from the separation measurements in vertical thermodiffusion column. The particle concentration in column ends is detected from the resonance frequency of LC oscillators with inductance coils mounted inside the column ends. The Soret coefficient  $S_{\rm T}$  of droplets is calculated from the initial part of dynamic separation curves. The measured value  $S_{\rm T} = -480 \, {\rm K}^{-1}$  agrees qualitatively well with the calculated one found assuming the Maragoni-type transfer of liquid particles.

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#### 1. Introduction

Magnetic fluids in many applications are subjected to strong temperature gradients. Recent experiments refer to a high thermodiffusive mobility of nanoparticles [1– 3]. Therefore, the investigation of Soret effect in colloids is of a great basic and practical interest.

Usually, the volume fraction of particles in ferrofluids  $\varphi$  is relatively small, therefore the mass flux  $j_{\varphi}$  can be written in a simplified form

$$j_{\varphi} = -D\rho(\nabla\varphi + S_{\mathrm{T}}\varphi\nabla T) \tag{1}$$

(*D* and  $S_{\rm T}$  are the mass diffusion coefficient and the Soret coefficient of a particle). The surfacted ferrite nanoparticles have positive Soret coefficient [1], whereas the charged particles in electrically stabilized ferrocolloids usually have negative values of  $S_{\rm T}$  [2,3]. The known experiments have examined only magnetic particles of a density  $\rho$  significantly higher than that of a carrier liquid  $\rho_0$ . In this paper we describe the first

results of experiments on dispersions consisting the light particles (microdroplets of a hydrocarbon-based ferrofluid in water) with density less than that of the carrier liquid ( $\rho < \rho_0$ ). One of the aim of the experiments is to clarify whether the direction of particle thermodiffusive transfer does not depend on the density difference  $\rho - \rho_0$ . Of course, we are aware of the non-possibility to make the final conclusions, because the mechanisms of particle transfer in suspensions and in emulsions might be different. The motion of liquid droplets can also be affected by the surface tension force.

### 2. Experimental

The emulsion is prepared in Mediport Kardiotechnik GmbH by the known Bibette method [4]. The mean diameter of ferrofluid droplets d=480 nm. The ferrofluid consists of lyophilized magnetite nanoparticles stabilized by sodium dodecyl-sulfate in *n*-heptadecane. The particle mean concentration calculated from the colloid density ( $\rho = 0.82 \times 10^3$  kg/m<sup>3</sup>), is equal to  $\varphi_0 \approx 0.02$ . Taking into account that both the particle concentration

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in ferrofluid and the droplet concentration in emulsion are very low, we assume that the coefficients, demanded in the analysis of the separation measurements, are equal to those for pure liquids. The surface tension  $\sigma$ and its dependence on temperature at the interface water-ferrofluid are extra measured. We found these coefficients being less than those for the *n*-heptadecane:  $\sigma = 9.8 \times 10^{-3} \text{ N/m}, \ \sigma^{-1} \partial \sigma / \partial T = -3.1 \times 10^{-3} \text{ K}^{-1}.$  Experiments are performed employing a thermodiffusion column consisting of a vertical flat channel (a gap  $\delta = 2a = 0.5 \,\mathrm{mm}$  between two plates of different temperatures  $T_1$  and  $T_2$ ) of a length L/a = 173. Both channel ends are connected with two equivalent separation chambers of an effective length  $L_{\rm c} = 0.059 \,{\rm m}$ . ( $L_{\rm c}$ represents the chamber volume divided by the crosssectional area of the channel). Inside each chamber an induction coil is mounted, playing a part of the LC oscillator, the resonance frequency of which determines the mean concentration of magnetic phase in the corresponding chamber.

#### 3. Results and discussions

Fig. 1 represents the dynamics of droplet separation in the column. The particle concentration in the upper chamber increases. It means, the droplets are transferred across the channel towards increasing temperature. The character of the separation curve is similar to that of the thermodiffusive separation [1] for negative Soret coeffi-



Fig. 1. Relative concentration of droplets in the upper chamber versus the non-dimensional time  $\tau = Dt/a^2$ .

cients. In our emulsion the gravitational sedimentation of droplets could be an additional reason for the separation. From the Adamar–Ribcinsky formula we have calculated that the raising velocity of droplets is equal to 28 nm/s. Thus, the effect of gravitational sedimentation can be neglected. According to Ref. [5], in initial stage of separation the development of particle concentration in the upper chamber follows the law:

$$\frac{\varphi_{\rm u} - \varphi_0}{\varphi_0} = -\frac{2}{45\sqrt{\pi}} k \; Gr_{\rm T} \; Sc \frac{a}{L_{\rm c}} \tau^{2.5} \tag{2}$$

with Sc = v/D being the Schmidt number,  $Gr_T = \beta_T g \Delta T a^3/v^2$  the Grashof number,  $\Delta T = T_2 - T_1$ , v the viscosity of the water,  $k = S_T \Delta T$  the non-dimensional Soret parameter and  $\tau = Dt/a^2$  the non-dimensional time. From the results represented in Fig. 1 it follows that the separation measurements follow to the law  $\Delta \varphi \sim \tau^{2.5}$  in a short regime of time until  $\tau < 0.001$ . The best fit between the experimental points and formula (2) is achieved with k = -4800. Experiments are performed at  $\nabla T = 10$  K. Thus, calculations return the value of the Soret coefficient  $S_T = -480$  K<sup>-1</sup>.

Such unexpectedly intensive thermodiffusion, obviously, is a reason why the initial part of separation curve with  $\Delta \phi \sim \tau^{2.5}$  is significantly shorter than that for colloids of smaller Soret coefficients. Conventional theories of thermodiffusion columns are developed under the assumption that the development of concentration boundary layers across the channel does not depend on the parameter k. In accordance with this model the law  $\Delta \phi \sim \tau^{2.5}$  takes place till  $\tau \approx 0.1$  [6]. Such simplified approach is valid only for small k values [7]. At k > 2 the development of concentration boundary layers starts to depend on the Soret coefficient, the unsteady regime of separation becomes shorter and the steady separation flux decreases [7]. Fig. 2 represents the dependence of the time limit  $\tau_0$  for regime  $\Delta \phi \sim \tau^{2.5}$  on parameter k. At  $k = 10^3$  the  $\tau_0 \approx 10^{-3}$ . This result agrees qualitatively well with the separation curve represented in Fig. 1.

We suspect that the observed high thermodiffusive mobility of droplets is as a result of Marangoni-type transfer. According to Ref. [8], the droplet velocity in a non-isothermal fluid follows the expression

$$u_{\sigma} = \frac{d}{2(3\eta + 2\eta_0)} \left( -\frac{\partial \sigma}{\partial T} \right) \nabla T \tag{3}$$

with  $\eta$  and  $\eta_0$  being the dynamic viscosity's of ferrofluid and of water. Regarding to this formula, we obtain the Soret coefficient  $S_{\rm T} = -550 \,{\rm K}^{-1}$ . This result agrees qualitatively well with the value of  $S_{\rm T}$  found from the separation measurements.



Fig. 2. Limit of unsteadiness  $(\tau_0)$  of the vertical mass flux versus the separation parameter *k*. Numbers near curves denote the ratio between the solute and the thermal buoyancy forces  $Gr_{\varphi}/Gr_{\rm T} = \beta_{\varphi} \varphi/\beta_{\rm T} \Delta T$ .

## 4. Conclusions

Performed experiments indicate high negative values of the Soret coefficient with ferrofluid droplets dispersed in water. The results can be interpreted as a Marangonitype transfer of liquid particles. The emulsion is relatively unstable in time. Additional experiments are needed to obtain quantitatively precise results.

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