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Experimental research of surfaced nanoparticle thermal transport in a porous medium



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ABSTRACT

Experimental research on thermally induced nanocolloid transport in a porous environment is presented. Addition of excess surfactant to a colloid with dispersed phase made of surfaced nanoparticles results in a decrease of effective Soret coefficient in a porous environment. It is shown that with sufficient amounts of surfactant added, this effect extends to a reversal of nanoparticle thermophoretic transport direction, and that the effect shows a tendency of saturation. A mechanism of thermal transport that involves slip velocity of surfactant molecules appearing near pore walls is evaluated as a possible cause of the decrease of Soret coefficient. Effects of temperature on particle thermophoresis are analyzed, revealing a decrease of Soret coefficient with the increase of temperature. In order to better understand surfactant motion in the porous environment, a series of experiments characterizing thermoosmotic motion of a mixture of surfactant diluted in pure carrier fluid through a porous layer is performed.

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1. Introduction

Experimental research of surfaced nanoparticle thermal transport in a porous environment has revealed a decrease of Soret coefficient of magnetic nanocolloids, or ferrofluids, when compared to that in a free fluid [1]. This is in contradiction to other research findings, both theoretical [2] and experimental [3,4], that have shown that, while a porous environment has an effect on the thermodiffusion process, it should not affect the Soret coefficient. Later research established that this unexpected behavior is characteristic to a wide range of surfaced ferrofluids, including several materials for colloidal particles and surfactant [5]. In continuation of that research, it was found that adding excess surfactant to the colloid resulted in this effect becoming more profound, suggesting a relation between the decrease of Soret coefficient and the presence of surfactant, and a significance of temperature dependence of the Soret coefficient [6]. An understanding of the mechanisms at work here would be beneficial not only for applications of surfaced colloids in a similar environment, but to experimental research of such colloids as well, where a porous environment gets used in experimental setups, such as those described in Refs. [3,7].

In this article, we present further experimental research regarding this phenomenon. Surfactant effect on nanoparticle thermal

* Corresponding author. E-mail address: viesturs.sints@lu.lv (V. Sints). transport is characterized in a series of thermophoresis experiments in a porous layer with a range of excess surfactant concentration values. It is demonstrated that a change of direction of particle transfer can be achieved. Such a change of sign of Soret coefficient is linked to temperature dependence of S_T [8], to which we turn our attention by conducting thermophoresis experiments at various temperatures. All of the aforementioned experimental work is done with ferrofluids: colloids made of magnetic nanoparticles and stabilized, in this case, by surfactant coating of the particles. To investigate thermal transport mechanisms of components of such a colloid in a porous environment, thermoosmosis experiments are performed. There, surfactant molecules are mixed not in a colloid of surfaced particles, but just the carrier fluid of that colloid.

Apart from experimental data to further map the dependence of particle transfer on the amount of excess surfactant, we turn to two mathematical models of thermophoretic particle transfer to characterize the observed phenomena. A challenge that interpretation of experimental observations and understanding of the underlying processes counters is lack of a definitive theoretical model for particle thermophoresis in liquids [9].

A model proposed by Morozov [10] describes the colloidal particle thermophoresis as driven by interaction between the particles and smaller species, like surfactant molecules. This transfer mechanism is similar to the slip velocity model described by Anderson [11]. Under a temperature gradient, the lyophilic interface of pore walls would drive a slip velocity transfer of molecules toward higher temperatures. At the time of publication of Ref. [1], an interpretation based on Morozov's work was proposed, suggesting that the effects of a porous environment on particle thermophoresis be explained by this slip velocity of surfactant molecules appearing near pore walls and contributing to motion of the colloid. This would cause a thermoosmotic transfer that would be directed opposite to the regular thermophoresis of surfaced nanoparticles, which is towards lower temperatures. It is then assumed that the flow of surfactant molecules by pore walls can induce thermophoretic bulk motion of the colloid that would be significant enough to account for the changes in Soret coefficient. The is one of the interpretations considered in this article.

Other theoretical approaches pay increased attention to interfacial effects on particle surface. Ruckenstein [12] linked thermophoresis, along with diffusiophoresis, to the Marangoni effect, suggesting gradient of interfacial tension as a driving mechanism of thermophoresis. Before him, Derjaugin and Sidorenkov [13] employed local enthalpy density for a model of thermophoresis, providing an expression for velocity of a fluid driven through a pore by the same mechanism. Parola and Piazza [14] arrived to an expression for the Soret coefficient that involved relation between surface tension and temperature. Iacopini et al. proposed an empirical model for Soret coefficient [15] with an emphasis on temperature dependence, later [16] citing the theoretical model described by Parola and Piazza as a firmer base for their phenomenological equation. The empirical Iacopini model is another approach employed for experimental data analysis in this article.

2. Experimental setup

Two experimental setups are used. One, to investigate thermophoretic nanoparticle transfer in a porous environment. It is referred throughout the article as *thermophoresis* setup. In experiments with this setup, concentration distribution of magnetic nanoparticles within a porous layer is determined at a single point in time, which also marks the end of experiment.

The other is used to research thermoosmotic processes concerning only the carrier fluid and surfactant. Referred to as *thermoosmosis* setup, it lets us investigate the flow of a fluid through a porous layer and allows real time measurements of the flow.

2.1. The porous environment

The porous layer used in both thermophoresis and thermoosmosis experiments is made of ten sheets of filter paper, tightly packed together to form a continuous porous environment.

The filter paper used is VWR Grade 410 with the relevant properties, as specified by the manufacturer, being: thickness $\delta_{filter} = 0.15$ mm, minimum pore size $d_{pore,min} = 9 \,\mu\text{m}$ and maximum pore size $d_{pore,max} = 20 \,\mu\text{m}$. The filters are found to be chemically inert to all the fluids used in experiments. The resulting layer then has thickness of $\delta = 1.5$ mm. Porosity is determined experimentally by comparing the weight of a filter paper saturated with a fluid of known density to that of an "empty" one, and it is found that $\varepsilon = 0.328$.

To estimate permeability of the porous medium, we use the Kozeny-Carman relation in the form given in [17]:

$$K = \frac{d^2 \varepsilon^3}{172.8(1 - \varepsilon^2)} \tag{1}$$

and get $K = 5.149 \cdot 10^{-14} \text{ m}^2$. It must be noted that this is only an initial estimation and the value of permeability used in calculations is obtained experimentally, as described in Section 5.1.

Concerns of convective stability for mass transfer in such a layer are addressed in [5], where it is concluded that no convective instabilities should arise in top-heated conditions.

2.2. Thermophoresis experiments

In thermophoresis experiments, illustrated in Fig. 1, the porous layer is fully enclosed by walls, allowing for no mass transfer to or from the layer. Side walls are thermally insulated, while top and bottom walls are kept at a constant temperature, with top wall being heated and bottom wall being cooled.

The porous layer is saturated with a colloid, which is a ferrofluid (fluids used in experimental work are expanded upon in Section 2.4). Diameter of the layer is 67 mm and the setup is positioned so that the axis of the flat cylindrical porous layer is aligned with the gravitational field. The temperature gradient is created at the start of the experiment and being kept constant throughout. Experiment is ended after 24 h, which is a well sufficient time for Fourier time $\tau = \frac{DI}{\sigma^2}$ to reach values of $\tau > 1$ and therefore the particle distribution to reach a steady state.

After the experiment is ended, the ten layers are split apart and samples are extracted from each layer. Concentration of magnetic nanoparticles in each layer is measured with the method of vibrational magnetometry, by using Lake Shore Cryotronics Co., model 7404 VSM vibrational sample magnetometer.

The accuracy of vibrational magnetometry measurements is given by the manufacturer as better than 2% of value reading. There are, however, other sources of error, possibly the most prominent being associated with splitting apart the layers post experiment and possible parasitic mass transfer during that process. To better estimate the measurement error, an experiment is performed with no temperature gradient present and, therefore, an expected uniform distribution of particles. The deviation of particle concentrations in this experiment gives us a 7% measurement error.

2.3. Thermoosmosis experiments

Thermoosmosis experiments are illustrated in Fig. 2. If colloid thermophoresis experiments investigate flow within a closed porous layer, in carrier fluid – surfactant thermoosmosis experiments that layer is "opened up", allowing for flow through it.

A porous layer with a diameter of $d_{layer} = 15$ mm is placed between two equal cylindrical volumes, forming an experimental cell that allows for both mass and heat transfer to and from the layer and the volumes. Axis of the cylinders are again aligned with gravitation field. Height of one volume is 15 mm. Both of the volumes have exits to a manometer, which is designed so that it's tubes run parallel and both at equal height from the ground at a point near the experimental cell, with the lower manometer tube being bent to meet the higher. From that point, the tubes can run either parallel to the ground (a setup that is referred to as "flow setup" in the article), or at an incline angle α (this setup is referred to as the "pressure setup"). Fluid levels in the manometer are monitored with a camera during experiments.



Fig. 1. Schematic layout of thermophoresis experiments.

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