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Translational thermophoresis and rotational movement of peanut-like colloids under temperature gradient

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Abstract Thermophoresis can be used for particle manipulation and separation in microfluidics. This work reports an experimental investigation on thermophoresis and the associated rotational patterns of dilute peanut-like colloids in DI water and SDS surfactant solutions. A microfluidic device is utilized for generating a linear temperature gradient and for directly visualizing the thermophoretic motion and rotation of peanut-like particles. Thermophilic behavior was observed for the peanut-like particles, and their thermophoretic mobilities were found smaller than those of the spherical particles of similar sizes. The peanut-like particles' rotation characterized by the rotational diffusion coefficient is found to be free diffusive dominated. A very small orientation order toward a preferred direction is distinguished along with the random Brownian rotation.

Keywords Thermophoresis · Thermal diffusion coefficient · Rotational diffusion · Orientation · Microfluidics

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1 Introduction

The development of techniques for manipulations of micro-/nanoparticles suspended in liquid media can significantly benefit the design of miniaturized sensors and probes in biotechnology (Miller and Jones 1993; Serey et al. 2012; Venu et al. 2013). For non-spherical particles, its diffusive translational and rotational movement due to Brownian motion can be both modulated into directed patterns under external force fields (Jiang et al. 2010; Filipovic et al. 2011; Kang et al. 2012; Dong and Cao 2014). The temperature gradient-induced thermophoresis (Piazza 2008; Piazza and Parola 2008; Würger 2010), which was discovered around 150 years ago, can be used as a lateral translational control means for particle separation (Sajeesh and Sen 2014). Extensive theoretical and experimental studies have been reported on colloidal thermophoresis under various effects of particle concentration (Zhao et al. 2013), particle size (Braibanti et al. 2008; Duhr and Braun 2006), solution temperature (Iacopini et al. 2006) and concentration (Putnam and Cahill 2005). Surfactants were sometimes added into colloidal solutions to standardize the particle–solvent interface (Braibanti et al. 2008) or to significantly alter the thermophoretic phenomena and even cause sign reversal (Jiang et al. 2010). It has been found that for colloid aqueous suspensions the electrokinetic effect due to the interaction of the electric double layer (EDL) of colloidal particles and electrolyte plays a major role in thermophoretic phenomena (Würger 2010).

The existing works on colloid thermophoresis mostly focus on spherical particles, while the discussion of non-spherical ones is scarce. Wang et al. (2013) studied the rod-like *fd* virus and extended the Dhont–Briels model (Dhont and Briels 2008) by representing the thermal diffusion coefficient of the rods with a superposition of that of the

same-diameter spherical beads such that good agreement was obtained with their experimental results. However, they did not explore the corresponding rotational movement of the rods under temperature gradient. This is actually a nontrivial issue since the coupling between translation and rotation can substantially complicate dynamic analyses (Han et al. 2006). Moreover, the non-spherical shape effect may influence electrical polarization and electrokinetic mobilities, even leading to anomalous orientations observed for charged colloids under external electric fields (Jiméneza and Bellini 2010). Similarly, a situation arising from the non-spherical shape in the presence of a temperature gradient should be addressed.

Thus, the aim of this paper is to experimentally investigate the thermophoresis and its associated rotation of highly dilute peanut-like polystyrene (PS) colloids in aqueous solutions. A recently developed microfluidic device (Zhao et al. 2013) is utilized for generating a linear temperature gradient and for directly visualizing the dynamic behavior of such peanut-like particles. The thermal diffusion coefficients of the non-spherical particles are compared with spherical ones in DI water and aqueous surfactant solutions. The rotational characteristics are studied through analysis of the rotational diffusion coefficient. It is found that the rotations are free diffusive dominated with a small orientation order observed.

2 Experimental section

2.1 Sample preparation

The non-spherical PS particles were purchased from Magsphere Inc., and their sizes are 1.75 μm × 3.3 μm, 3.5 μm × 5.1 μm and 4.85 μm × 7.7 μm. These particles are claimed to be peanut-like, with a scanning electron microscope (SEM) image for 1.75 μm × 3.3 μm particles shown in Fig. 1. For a comparison, spherical PS particles purchased from Thermo Fisher Scientific Inc., with their sizes of 2.0, 3.1 and 4.8 μm in diameter were also used. Our thermophoresis experiments consisted of both peanut-like PS particles and spherical PS particles in DI water and DI water–sodium dodecyl sulfate (SDS) surfactant solutions of various SDS concentrations including 0.001, 0.01, 0.025, 0.1, 1 and 5. The anionic SDS surfactant (with a purity of ≥99 %) purchased from Sigma-Aldrich was used for altering particle surface properties and thus the thermophoresis of particles in the following experiments.

The purchased particle suspensions were centrifuged three times with a centrifuge to wash off unknown surfactants and other impurities. Then, the washed particles were resuspended with DI water (Millipore grade I or SDS surfactant–DI water solutions) to form desirable
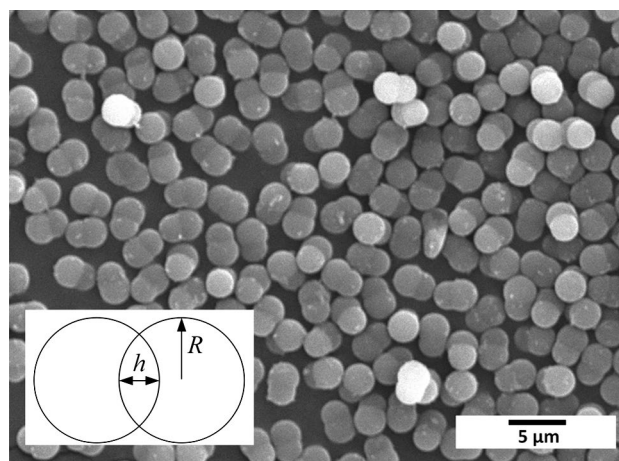

Fig. 1 A SEM image of 1.75 μm × 3.3 μm peanut-like colloids. The *inset* is a cartoon of the peanut-like particle treated as a dicolloid with the parameters R being the radius and h being the depth of the overlapped region of the two linked identical spheres

particle samples with a particle volumetric fraction of about 0.019 %. For such dilute particle suspensions, particle–particle interactions become insignificant (Zhao et al. 2013). Prior to thermophoresis experiments, all the prepared particle samples were sonicated for about 15 min in an ultrasonic cleaner so as to obtain better homogeneity of particle samples for testing.

2.2 Experimental setup

The literature studies have reported that microfluidic devices with two side channels at preset temperatures can generate adequate temperature gradient (Vigolo et al. 2010; Velve-Casquillas et al. 2011; He 2013), which is one of the important experimental conditions for thermophoresis studies. In the present work, the microfluidic device, shown in Fig. 2, is a stainless steel substrate used for loading samples and establishing temperature gradients and was developed in our previous work (Zhao et al. 2013). Three channels are fabricated on the substrate and sealed with an optical adhesive film to form an enclosed space; Among them, the middle channel of 400 μm wide, 35 μm deep and 20 mm long is for loading and testing the particle samples (the 35-μm channel depth ensures a negligible free convection and boundary effect); the two larger channels on each side of the middle channel is for running hot and cold water streams such that a transverse temperature gradient is established across the middle channel. The hot and cold water streams are sucked from two water baths with their temperatures set as 80 and 5 °C, respectively, by a dual-channel syringe pump. The counter flow heat transfer mode is adopted to ensure a nearly constant temperature difference along the longitudinal direction of the middle test
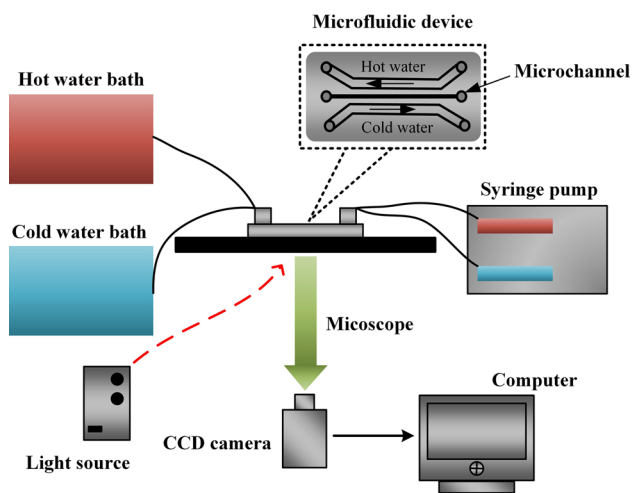
Fig. 2 Schematic diagram of the experimental system. The *inset* inside a *dashed rectangle* presents a magnified view of the microfluidic device

channel, and hence, along the transverse direction, a linear temperature distribution can be obtained. Since the test channel has 400 μm width and 35 μm depth, direct measurement of liquid temperature inside the channel is difficult. By using a Rhodamine B solution-based thermometry method (Ross et al. 2001; Zhao et al. 2013), the solution temperatures were measured to vary transversely from 48.3 to 42.1 °C across the test channel, thereby resulting in an average temperature of 45.2 °C and a temperature gradient of about 1.55×10^4 K/m. An inverted microscope was used to observe the thermophoretic behaviors of PS particles illuminated by a mercury lamp. Experimental images were captured with a CCD camera and then stored in a personal computer for analyses.

3 Results and discussion

3.1 Thermophoresis of both peanut-like and spherical particles

In the experiments, the single-particle tracking method (Dupont and Lamb 2011) was employed to observe the thermophoretic motion of both peanut-like and spherical particles. For each kind of particles, the thermal diffusion coefficient D_T is determined by using Eq. (1) expressed as.

$$u_T = -D_T \nabla T \tag{1}$$

Here, ∇T is the imposed transverse temperature gradient that was characterized in Sect. 2.2, and u_T is the thermophoretic velocity. For all particle samples, u_T was obtained by measuring the distance traveled along the transverse direction for a single particle and then dividing it by the

time elapsed. Over 400 measurements were averaged to eliminate the interference of translational Brownian motion. A positive D_T indicates particle motion from hot to cold regions (i.e., thermophobic behavior), while a negative D_T denotes from cold to hot regions (i.e., thermophilic behavior).

To effectively compare the thermophoretic characteristics of the peanut-like and spherical particles, the equivalent diameters of the non-spherical peanut-like particles need to be determined at first. The peanut-like particles, as shown in Fig. 1, can be treated as dicolloids, which are a combination of two overlapped spherical colloids
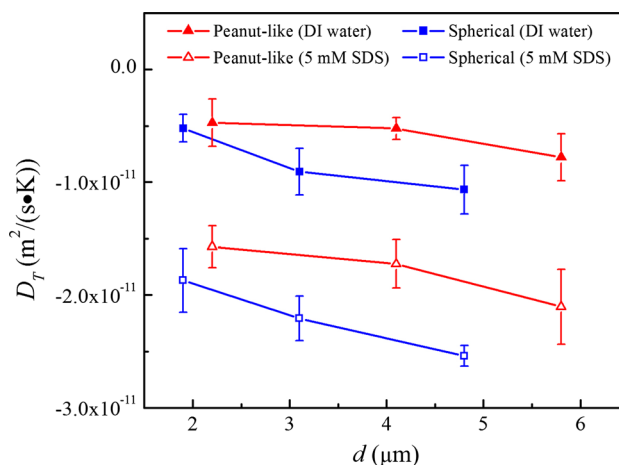

Fig. 3 Dependences of the thermal diffusion coefficients of the peanut-like colloids (1.75 μm × 3.3 μm, 3.5 μm × 5.1 μm and 4.85 μm × 7.7 μm) and spherical colloids (2.0, 3.1 and 4.8 μm) in DI water and 5 mM SDS solutions on particle (equivalent) diameter. The applied temperature gradient is about 1.55×10^4 K/m

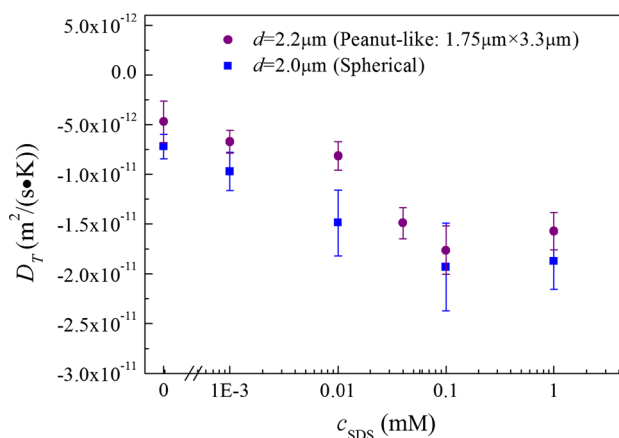**Fig. 4** Dependences of the thermal diffusion coefficient D_T on SDS concentration c_{SDS} for 1.75 μm × 3.3 μm (with the equivalent diameter of 2.2 μm) peanut-like colloids and 2.0 μm spherical colloids in the presence of a temperature gradient of about 1.55×10^4 K/m

with the diameter of the decomposed sphere set as the short-axis length of the peanut-like particle. Although the SEM image exhibits two spheres with slightly different sizes, for simplification, they are treated as two identical spheres. Then, the volume of the "peanut" is expressed as $V = 2\left(\frac{4}{3}\pi R^3 - \pi h^2\left(R - \frac{h}{3}\right)\right)$, with R being half of the short-axis length and h being the depth of the overlapped segment of the two linked spheres (shown in the inset of Fig. 1). Making such calculated volume equal to that of a sphere, one can obtain the equivalent diameters as 2.2, 4.1 and 5.8 µm for 1.75 µm × 3.3 µm, 3.5 µm × 5.1 µm and 4.85 µm × 7.7 µm particles, respectively. Figure 3 shows the thermal diffusion coefficients D_T of these peanut-like particles in both DI water and 5 mM SDS solution. As a comparison, the thermal diffusion coefficients of the spherical particles with their diameter of 2.0, 3.1 and 4.8 µm are also included. It is observed that both the peanut-like particles and the spherical particles in DI water and 5 mM SDS solution have negative values of D_T, showing their thermophoretic motions toward the hot region. Moreover, it is found that for similar sizes, the peanut-like particles all have lower D_T values than the spherical particles, indicating smaller thermophoretic velocities.

Figure 4 shows the dependence of the thermal diffusion coefficient on SDS concentration for 1.75 µm × 3.3 µm (with an equivalent diameter of 2.2 µm) peanut-like particles and 2 µm spherical particles. With increasing SDS concentration, for both types of particles, D_T becomes more negative and thus more thermophilic. Consistent with Fig. 3, the peanut-like particles have smaller thermophoretic mobilities than the spherical particles for all SDS concentrations studied. It is known that SDS belongs to an anionic surfactant with the same negatively charged sign as the surface of PS particles, and its critical micelle concentration (cmc) is around 8.7 mM at 45 °C (Zhao and Zhu 2003), which is close to our experimental temperature. The effect of surfactants on colloid thermophoresis can be quite complicated, while the biggest contribution may come from the adsorption of surfactant molecules onto the particle surface, leading to changes of the colloid–water interfacial properties such as electrostatic charge and surface free energy. Specifically in this work, since PS particles are hydrophobic with small amount of polar sulfate groups, the hydrophobic interaction results in the hydrophobic tails of the SDS molecules in contact with the PS surface and the hydrophilic heads of the SDS molecules pointing toward aqueous solution (Brown and Zhao 1993; Myers 1991). Consequently, with more SDS molecules added into the solution, the interface of PS particles will become less hydrophobic and even hydrophilic; this in turn will cause the PS particles to be more negatively charged, thereby giving rise to more thermophilic behaviors. This is also in line with our separated experiment which showed the more

hydrophilic the colloidal particles, the stronger the thermophilic behaviors.

Two points should be noted here: First, our observed thermophilic behavior and the corresponding negative values of the thermal diffusion coefficient cannot be predicted by existing theories. In the literature, Duhr and Braun (2006) related thermophoresis to temperature-dependent Gibbs free energy of a single particle–solvent system and introduced a hydration entropy term (s_{hyd}) in their model which always leads to positive D_T. If the model for rods by Wang et al. (2013) is used, it also predicts positive D_T when the Debye length changes from ~10^2 nm in DI water to several nanometers as SDS molecules are added (no more than 5 mM) in our case. The inclusion of the interfacial tension contribution in Würger's model (Würger 2007) can yield a negative D_T, but the absolute value is in the order of 10^{-10}–10^{-8} m²/(s K), much larger than our results of 10^{-12}–10^{-11} m²/(s K). We argue that this discrepancy between our experimental results and theoretical predictions may come from the surfactant–particle surface interactions that are not properly treated in the existing models. Second, an interesting phenomenon is noted that spherical particles exhibit a larger thermophilic tendency than the non-spherical ones at the same equivalent diameters. Since the ζ-potential measurements via Malvern Nano Zetasizer (ZEN3600) given in Fig. 5 do not show a noticeable difference between these two kinds of particle samples, this indicates that though the peanut-like PS particles and the spherical PS particles are made by two different companies, their surface properties in terms of surface charges are indistinguishable. Therefore, the difference in thermophoretic behaviors of these two kinds of PS particles may be attributed to the non-spherical shape effect.
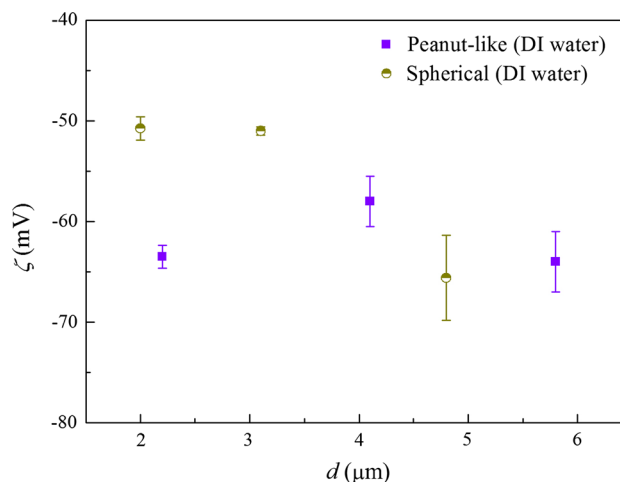

Fig. 5 Variation of ζ-potentials with particle size for the spherical and peanut-like colloids in DI water

3.2 Rotation and orientation of the peanut-like particles

At equilibrium, non-spherical particles in liquid will rotate indefinitely due to rotary Brownian motion. However, it is hard to tell what kind of intriguing patterns the peanut-like particles may fall into when a temperature gradient is present and when the particles are experiencing thermophoretic movement. The rotational diffusion coefficient serves as an indicator of a non-spherical particle Brownian rotation and a measure for the rotational mobility. The typical Einstein relation for calculating such diffusivity D_r can be expressed as (Cao and Dong 2014; Dong and Cao 2015).

$$D_r = \frac{\langle (\Delta\varphi)^2 \rangle}{2t} \qquad (2)$$

Here, $\langle \ldots \rangle$ denotes the ensemble average, $\Delta\varphi$ is the angular displacement and t is time. For convenience, two simplifications are made: First, the peanut-like particle is treated as a cylinder with its long axis being the cylinder length and its short axis being the cylinder diameter. Then, only the rotation around the axis that is perpendicular to the symmetric axis, i.e., the long axis of the particle, is studied here. Second, only the particle rotation on the focus plane is analyzed, which renders our analysis to be a two-dimensional (2D) rotational study (Cao and Dong 2014). In particular, for our rotation analysis, the rotation of a single particle is tracked, and then, the ensemble average is taken over both time and 20 different particles under the same experimental conditions.
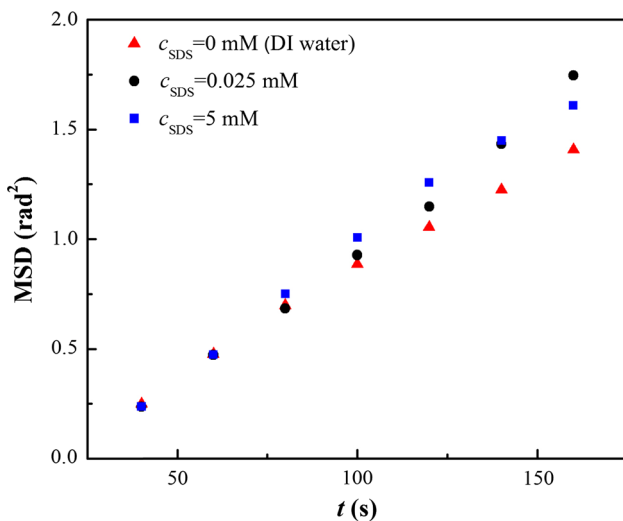
Fig. 6 Angular mean-squared displacements (MSD) for 4.85 μm × 7.7 μm peanut-like colloids in 0 mM (DI water), 0.025 and 5 mM SDS solutions in the presence of a temperature gradient of about 1.55×10^4 K/m

Fig. 7 Angle probability density distributions over angle φ for 4.85 μm × 7.7 μm peanut-like colloids in 0 mM (DI water), 0.025 and 5 mM SDS solutions in the presence of an applied temperature gradient of about 1.55×10^4 K/m. The *inset* is a schematic viewing of the angle definition
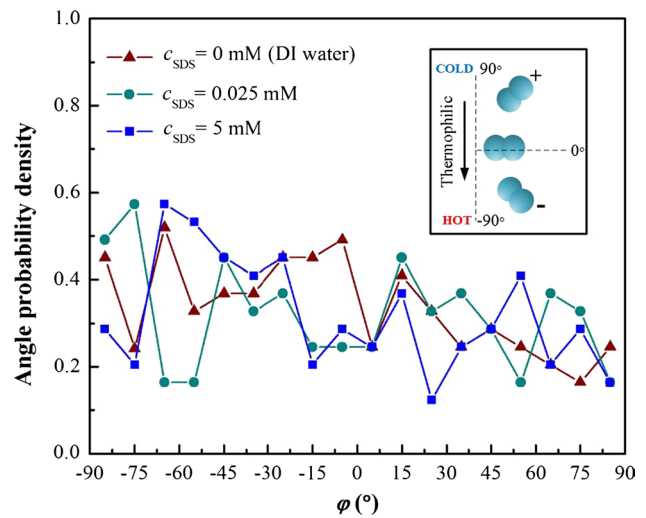
Figure 6 depicts the time-dependent mean-squared angular displacement (MSD), $\langle (\Delta\varphi)^2 \rangle$ for selected 4.85 μm × 7.7 μm particles in three different solutions, namely DI water, $c_{SDS} = 0.025$ mM, and $c_{SDS} = 5$ mM when a temperature gradient of about 1.55×10^4 K/m is present. The thermal diffusion coefficients for the above three cases are $D_{T\_0} = -7.78 \pm 2.1 \times 10^{-12}$ m^2/(s K), $D_{T\_0.025} = -15.3 \pm 3.0 \times 10^{-12}$ m^2/(s K) and $D_{T\_5} = -21.3 \pm 3.3 \times 10^{-12}$ m^2/(s K), respectively. The curves can all be fitted well with linear functions, implying that the rotational motion is diffusive dominated and in essence random Brownian motion. Furthermore, from Eq. (2), the corresponding rotational diffusion coefficients can be obtained as $D_{r\_0} = (4.4 \pm 0.26) \times 10^{-3}$ rad^2/s, $D_{r\_0.025} = (6.1 \pm 0.36) \times 10^{-3}$ rad^2/s and $D_{r\_5} = (5.9 \pm 0.43) \times 10^{-3}$ rad^2/s, respectively. Here to note is that the theoretical prediction for a cylinder (Cao and Dong 2014; Tirado and de la Torre 1984) under the same conditions also yields $D_{r\text{-theory}} = 6.7 \times 10^{-3}$ rad^2/s, which is comparable to our experimental results.

To further examine the rotational characteristics of these peanut-like particles, we analyzed the angle probability density distributions during the rotation. Shown in the inset of Fig. 7 is the definition of the angle φ. When a particle's long axis is perpendicular to the imposed temperature gradient, $\varphi = 0°$. For other cases, the angles are within a range of $(-90°, 90°]$ with the negative and positive angles indicated in the figure inset. Figure 7 shows that the normalized angle probability density distributions for the above three cases do not possess clear isolated peaks around a specific direction. Instead, nearly random

distributions can be observed, which coincides with the above analyses that the process is indeed free diffusive dominated.

A more rigorous way to quantitatively determine the orientation preference of non-spherical particles is through introducing the order tensor expressed by (Dong and Cao 2014):

$$T_{ij} = \frac{3}{2} \left\langle l_i l_j - \frac{1}{3} \delta_{ij} \right\rangle, \tag{3}$$

where $\vec{l}$ is the unit axial direction vector of the particles and δ_{ij} is Kronecker delta. The subscripts i and j are selected between the temperature gradient direction and the direction perpendicular to it on the focus plane. Thus, this 2D analysis gives **T** to be a 2×2 symmetric matrix. The largest eigenvalue of **T**, denoted as λ, is actually the orientation order along the most preferred angle, i.e., orientation angle φ_{orient}, which is the corresponding eigenvector. If $\lambda = 0.25$, the angles exhibit a completely random distribution; If $\lambda = 1$, the particles are perfectly oriented at orientation angle φ_{orient}. For the above three cases shown in Figs. 6 and 7, one can obtain $\lambda_0 = 0.37 \pm 0.03$, $\lambda_{0.025} = 0.33 \pm 0.01$ and $\lambda_5 = 0.37 \pm 0.01$, which are slightly larger than 0.25, suggesting a weak preference to be oriented toward a specific direction. Correspondingly, the preferred angles are $\varphi_{\text{orient\_0}} = -(61 \pm 5)°$, $\varphi_{\text{orient\_0.025}} = -(72 \pm 8)°$ and $\varphi_{\text{orient\_5}} = -(48 \pm 3)°$, and around these particular angles, one can observe relatively large angle probability densities from Fig. 7 as well.

Through the analysis of the rotation and orientation of the peanut-like particles during thermophoretic motion, the following can be highlighted: (1) The above results illustrate that the rotation accompanying particle thermophoresis is more of a random Brownian motion-dominated process. Here we use "dominated" to emphasize that it seems not a free diffusive behavior, because during a single particle rotation, it shows a small orientation order toward a particular direction. (2) When a single particle shows a preferred orientation along with time, all the particles in the experiment are aligned toward the same direction at any instantaneous moments. This will be supported by the ergodic hypothesis if the observation time is long enough and the particle number is large enough. (3) The slight orientation or alignment found in this work may not be a trivial issue, because as the anisotropy of the studied non-spherical colloid is further increased or the temperature gradient is increased, a stronger orientation order may be observed. This motivates our further studies to clarify this issue, by decoupling the effects of hydrodynamic flow, temperature field and Brownian motion.

4 Conclusion

Using a microfluidic approach for generating a linear temperature gradient, we have experimentally investigated the temperature gradient-induced translational motion (thermophoresis) and rotational motion of dilute peanut-like PS colloidal particles in aqueous solutions. We have found that for the peanut-like particles with their sizes of 1.75 µm × 3.3 µm, 3.5 µm × 5.1 µm and 4.85 µm × 7.7 µm, their thermophoresis exhibits a thermophilic behavior in DI water, and their thermophilic mobilities become stronger with increasing the concentration of anionic SDS surfactant from 0.001 to 5 mM in DI water. Also we have found that under the same experimental conditions, the thermophilic mobilities of the peanut-like PS particles are lower than the spherical PS particles of similar sizes. Furthermore, we have observed that the non-spherical particles rotate in additional to their thermophoretic translational movement. Through the analysis of the rotational diffusion coefficient, we have identified that such rotation is rotary Brownian motion dominated with a weak preferred orientation toward a specific direction. To the best of our knowledge, this work is the first experimental investigation of the rotational patterns and the thermophoresis of dilute peanut-like colloidal particles. This study is beneficial for manipulating non-spherical particles, such as DNA and bacteria cells in bioscience and micro-/nanoparticles in smart materials.

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