Electronic Supplementary Material (ESI) for Soft Matter.

Kinetics of Colloidal Particle Deposition in Microfluidic Systems under Temperature Gradients: Experiment and Modelling

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1. Fabrication of the temperature-gradient microchip

The PDMS monomer and the curing agent are fully mixed in a mass ratio of 10:1, and are pumped vacuum for 45 minutes in order to evacuate air bubbles remaining in the mixture. Thereafter, the mixture is poured onto the master mould made by SU8 on a silicon wafer. The wafer together with PDMS is placed in an oven for curing about 1 hour at 80℃. A thin layer of polymerized PDMS (1 mm in thickness) is peeled off from the mould. Two cylindrical openings with diameter of 0.75 mm are punched at both ends of the microchannel as the inlet and outlet for sample fluids. The PDMS block with microchannel structure is irreversibly bonded with the PDMS side of the treated ITO glass slide to form a closed microchannel, by using oxygen plasma of 35 Watt radio frequency (RF) power under vacuum condition. At last, the bonded microchip is put in the oven for 1 hour at 80℃ to reinforce the bonding strength between the PDMS block and the tread ITO glass slide.

2. Numerical Simulation for temperature gradients generated in the microchannel

To estimate the temperature gradient inside the microchannel along the vertical direction, a three-dimensional numerical simulation of the steady temperature distribution in the microchannel is performed by using the commercial FEM software package *COMSOL Multiphysics*. The numerical model consists of a water flow in the microchannel, the PDMS block and the glass slide. Four major sets of inputs are given as boundary conditions for the simulation: the temperature of the sample solution at the microchannel inlet is set as room temperature (25 °C) with a fixed flow rate of 0.1 mL/h, the surface temperature of the top surface of the PDMS block covered by the TEC unit is set as a constant value measured by the thermocouple (T_TEC), a surface heat source is imposed on the region covered by the ITO film according to the applied power (P_ITO) in experiments, and a convection boundary condition is applied on the outer surfaces of the microchip for heat loss. The heat transfer coefficients are assumed at 30 $W/m^2 \cdot K$ for the bottom surface of the glass slide, 5 $W/m^2 \cdot K$ over the exposed area of the top of the PDMS block and the sides of the glass slide and the PDMS block after considering the working conditions in the lab [1]. With the given boundary conditions, the temperature distribution of the temperature-gradient microchip can be numerically solved, and the simulation results of the temperature gradient the microchannel centreline are shown in Fig. S1. It is found that the average temperature gradient in the observation region varies from 78.9 K/m to 6846.9 K/m along the vertical direction of the microchannel centreline. Meanwhile, the average bulk temperature in the microchannel is changed from 332.73 K to 316.84 K.

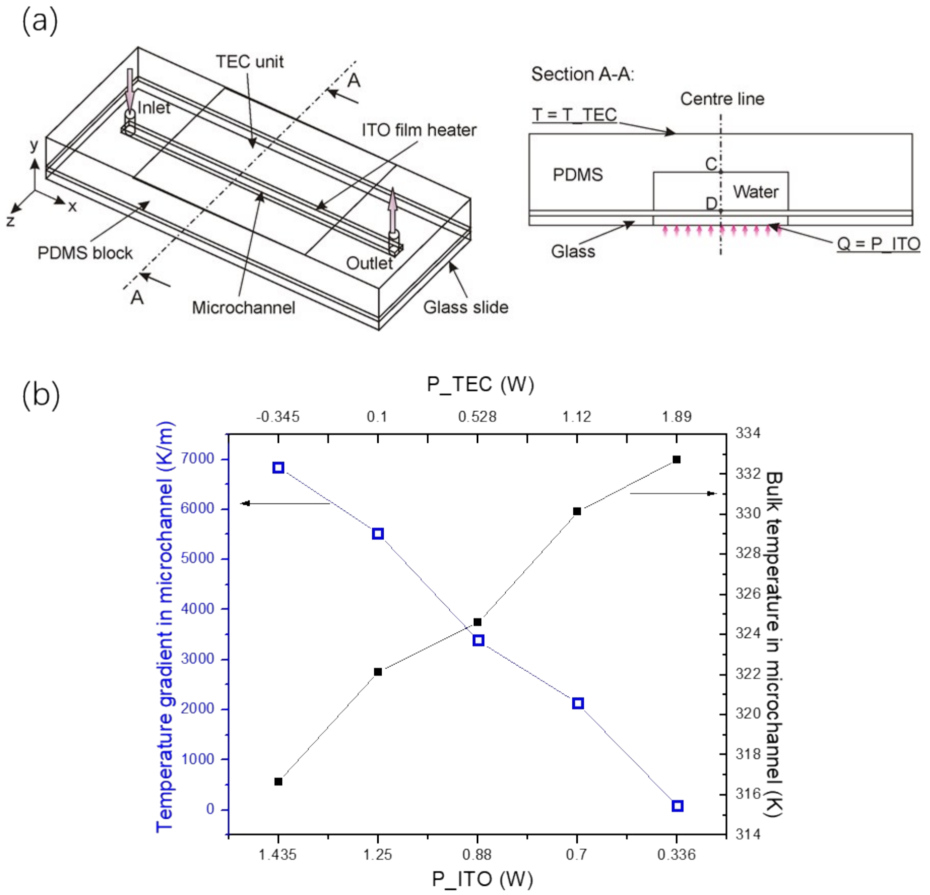

Fig. S1 (a) The numerical model consisting of the microchannel and the PDMS block and the glass slide. (b) Temperature gradient (-□-) along the centreline (CD shown in Section A-A) of the microchannel and bulk temperature (-■-) in the observation region versus the power inputs to the TEC cooling unit (P_TEC) and ITO film heater (P_ITO). The bulk temperature is estimated as the average temperature along the centreline, $(T_C+T_D)/2$. Data are obtained by a three-dimensional simulation with boundary conditions: The surface temperature covered by TEC unit is set as constant (T = T_TEC), Constant surface heat source is applied onto the ITO film heater (Q = P_ITO), and convective heat transfer (h = 30 W/m^2 K for bottom surface and 5 W/m^2 K for side and top surfaces of the PDMS block) is set between the chip and the ambient air to account for heat loss, a hydrodynamic flow with constant flow rate (0.1 mL/h) and inlet temperature (25 °C) is applied inside the microchannel.

2. Derivation of thermophoretic force caused by temperature gradient

Thermophoresis in colloidal suspensions has been both experimental and theoretically challenging subject. Akin to the forces underlying other driven transport processes including electrophoresis and diffusiophoresis, we derived the expression of thermophoretic force based on the available literature on thermophoresis in colloidal suspensions[2-4]. The derivation of thermophoretic force in Eq. (4) is shown in details as below:

By definition, thermophoresis is particle motion induced by thermal gradients [2, 3]. The steady-state thermophoretic velocity acquired by a particle can be simply given by

$$u_T = -D_T \nabla T, \tag{S1}$$

where D_T is the thermophoretic coefficient of the particle [4], also known as 'thermal diffusion coefficient' [3]; ∇T is the thermal gradient in the system.

Meanwhile, according to theories of colloidal transport phenomena [5], the particle moves under the effect of the thermophoretic force, one can write its velocity, (*i.e.* steady-state thermophoretic velocity) as

$$u_T = \omega F_T. \tag{S2}$$

Here, F_T is the thermophoretic force; ω is defined as the mobility of the particle (velocity per unit applied force) which is shown by Einstein as

$$\omega = \frac{1}{f} = \frac{D}{k_B T}, \tag{S3}$$

where *f* is the Stokes-Einstein friction factor; D is the Einstein coefficient, also known as Brown diffusion coefficient; k_B is the Boltzmann constant and *T* is the absolute temperature of the particle.

Substitute Eqs. S2 and S3 into Eq. S1, we can have the expression of the thermophoretic force as

$$F_T = -\frac{D_T}{D} \nabla T k_B T. \tag{S4}$$

While the Soret coefficient is defined as $S_T = D_T/D$ and its unit is 1/K [3], the thermophoretic force can be expressed as

$$F_T = -S_T \nabla T k_B T. \tag{S5}$$

The thermophoretic force is non-dimensionlized with ${k_B T}/{a_p}$, which has been widely implemented as an appropriate scaling factor for forces in studies of particle deposition and other colloidal transport phenomena [6-8]. Thus, we have the dimensionless thermophoretic force along vertical direction (y-axis in Fig. 1) shown in Eq. (4) of the manuscript as

$$\bar{F}_T = k_B T S_T \frac{dT\ a_p}{dy k T_0} = \frac{dT\ T}{dy T_0} S_T a_p. \tag{4}$$

Here, the positive sign of the dimensionless thermophoretic force means the repulsive effect on the particle deposition. Moreover, Yang and Ripoll [9] has proposed a similar form for the thermophoretic force exerting on a suspended particle by using the so-called thermodiffusion factor.

3. Dimensionless individual particle-microchannel interaction potential versus the dimensionless separation distance for different temperature gradients

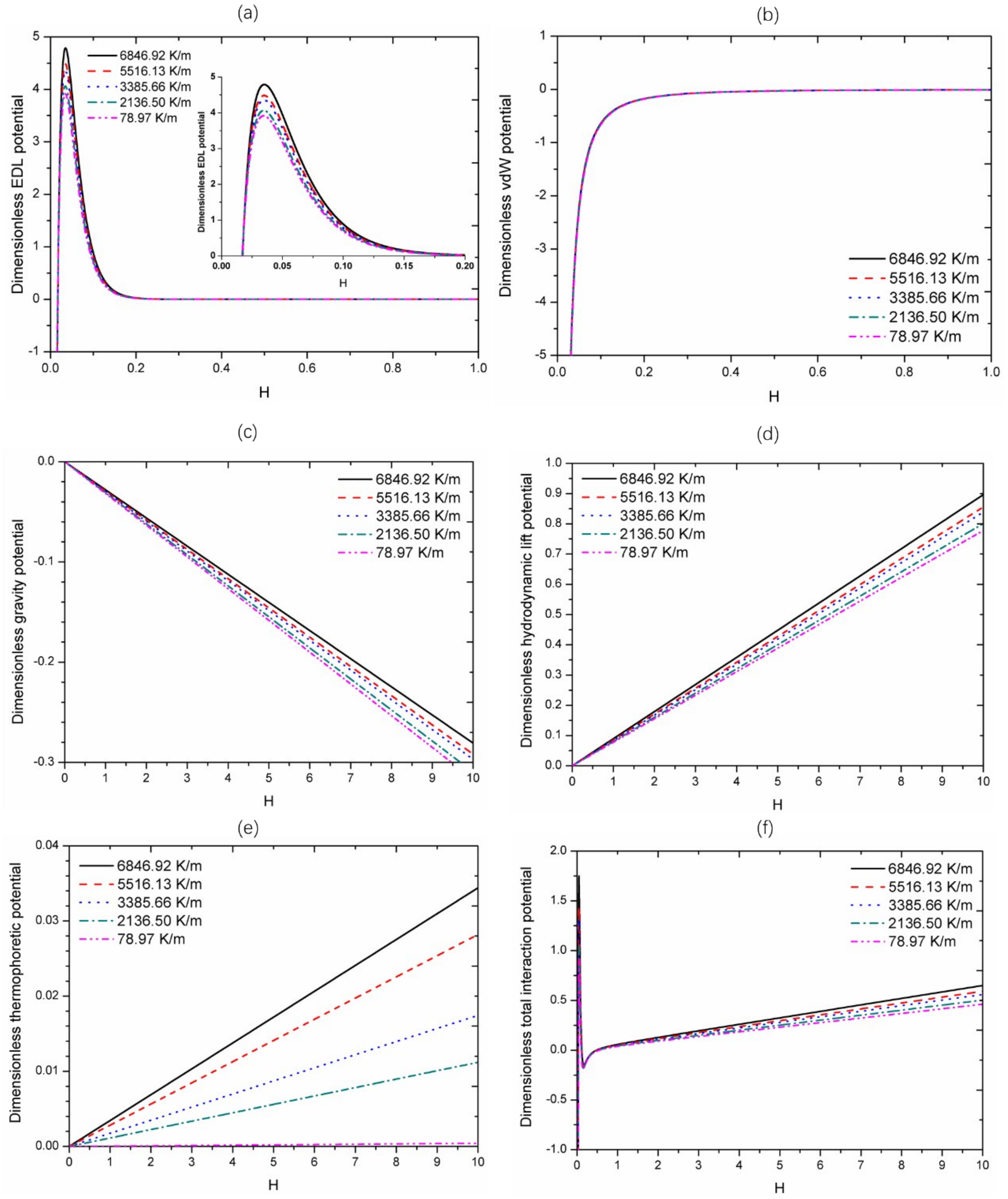

Fig. S2 Dimensionless particle-microchannel individual interaction potential versus the dimensionless separation distance H for different temperature gradients: (a) EDL interaction, (b) vdW interaction, (c) gravity interaction, (d) hydrodynamic lift interaction, (e) thermophoretic interaction, and (f) total interaction potential at H = 1~10.

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