Supporting Information

Contact Force Mediated Rapid Deposition of Colloidal Microspheres Flowing Over Microstructured Barriers

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[a)] Present affiliation: Department of Applied Mathematics and Theoretical Physics, University of Cambridge, Wilberforce Road, Cambridge, CB3 0WA, United Kingdom.
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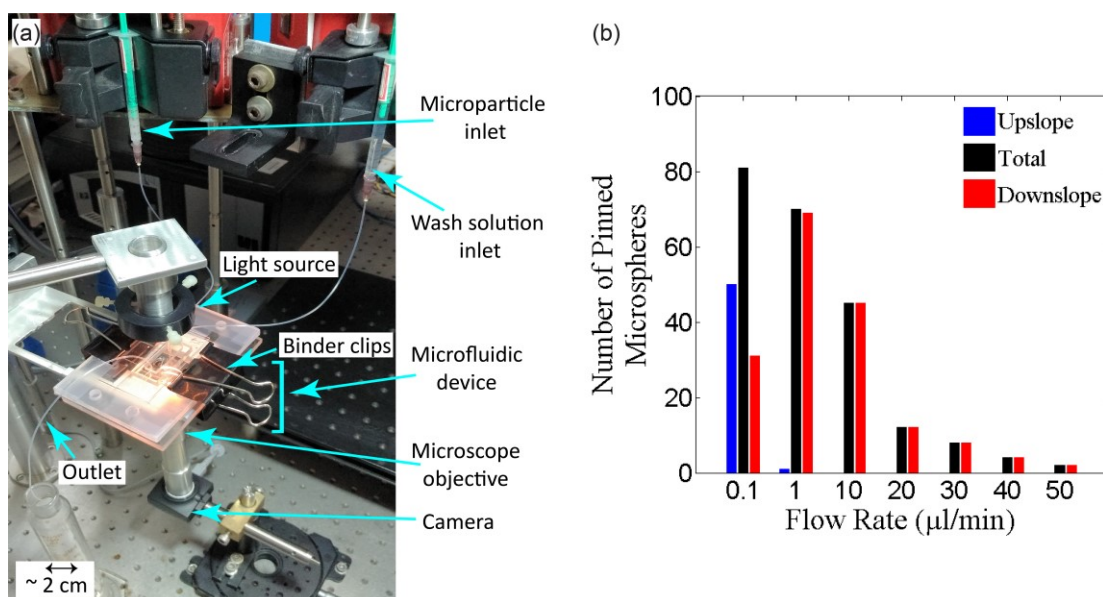## 1. Pinning of 10 μm microspheres in DI water medium

Figure S1. (a) Experimental setup showing PDMS microfluidic device and glass substrate sandwiched between acrylic plates using binder clips. (b) Total number of pinned microspheres in 2 min at different flow rates. Here the microspheres are flown in DI water and single 10 μm microspheres pin on the downslope similar to Figure 2(a) of the main text. At low flow rates (< 10 μl/min) microspheres pin both on the upslope and downslope[1]. 10 μl/min is the lowest flow rate at which there is no upslope pinning.

2. Hydrodynamic equations for COMSOL simulation

The force on microspheres is calculated by setting up hydrodynamics equations in accordance to the channel geometry and dimension. The Navier-Stokes equation (Eq. 1) together with continuity condition ($\nabla \cdot u = 0$) for an incompressible fluid ($d\rho/dt = 0$) determines the flow.

$$\rho\frac{\partial u(t)}{\partial t} + \rho(u \cdot \nabla u) = -\nabla p + \mu\nabla^2 u \qquad (1)$$

Where, u is the velocity and ρ, μ are density and dynamic viscosity of water respectively.
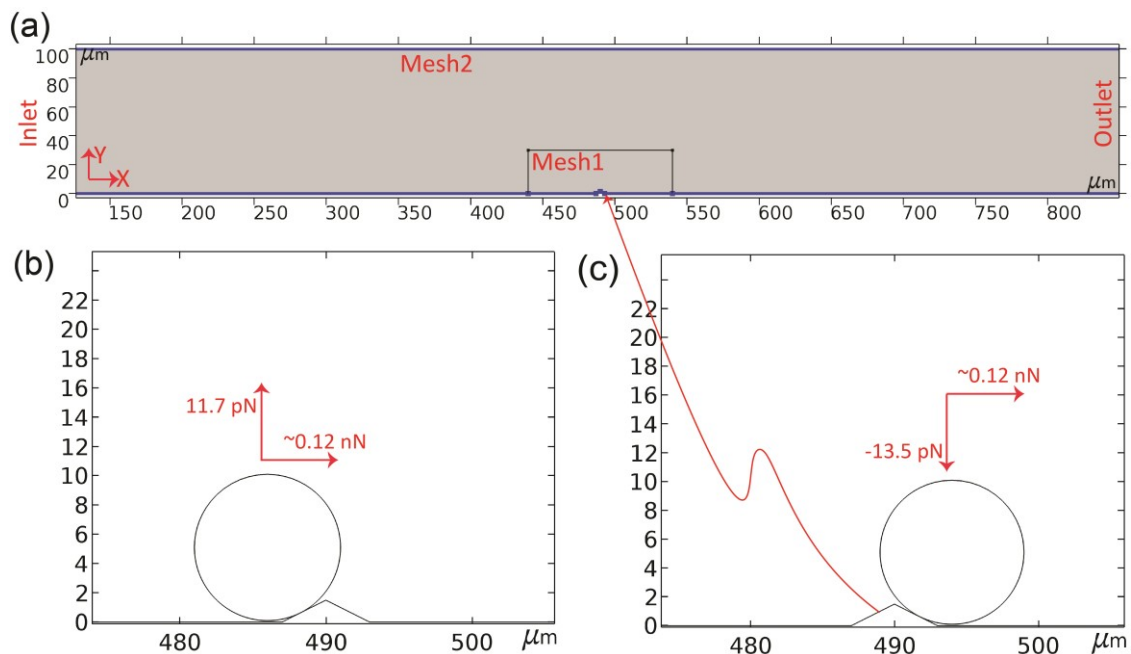

Figure S2. FEM simulation geometry. (a) Microfluidic channel having a height of 100 μm with wedge microstructures at the centre. (b) Force on 10 μm microsphere if it is assumed to be pinned on the upslope region. (c) Force on 10 μm microsphere in the downslope region.

We have used an inbuilt module 'Laminar Flow' in COMSOL to implement Navier-Stokes equation. No-slip boundary condition ($u = 0$) is applied at all the surfaces including microspheres (Figure S2(a)). The flow is initiated by assigning an average flow speed ($u_{avg} = 0.003$ ms^{-1}) to the microsphere solution at the 'Inlet' (Figure S2(a)). The average speed is calculated by taking ratio of the flow rate (10 μl/min) and cross-sectional area (100 μm × 500 μm) of the microfluidic channel. The 'Outlet' boundary condition (Figure S2(a)) is set as the pressure $P = 0$. We have split the microfluidic channel into two regions and applied inbuilt meshing option 'Extremely Fine' to Mesh1 and 'Extra Fine' to Mesh2. Using 'Extremely Fine' mesh settings for the entire channel is computationally very expensive. Hence, we have used a finer mesh setting near the microstructure and

comparatively coarser mesh setting for the remaining area. For simulating different situations, the locations of microspheres ia sppropriately modified in accordance to Figure 3(c), Figure 5(b) and Figure 5(c) of the main text.

3. DLVO force calculation

The electrostatic double layer repulsion (F_{EDL}) and attractive Van der Waals force (F_{VDW}) are together known as DLVO force ($F_{DLVO} = F_{EDL} + F_{VDW}$). The forces between Particle-Surface and Particle-Particle (same size) are defined as[2]:

Particle-Surface	$F_{VDW} = -AR/6D^2$	(2)
Particle-Surface	$F_{EDL} = KRZe^{-KD}$	(3)
Particle1-Particle1	$F_{VDW} = -AR/12D^2$	(4)
Particle1-Particle1	$F_{EDL} = KRZe^{-KD}/2$	(5)
Particle1-Particle2	$F_{VDW} = -A/6D^2(R_1R_2/R_1 + R_2)$	(6)
Particle1-Particle2	$F_{EDL} = KZe^{-KD}(R_1R_2/R_1 + R_2)$	(7)

Where A is the Hamaker constant, $R = 5$ μm is the radius of microsphere, and D is the gap between Particle-Surface and Particle-Particle. The Hamaker constant for polystyrene-water-glass system is $A \approx 1 \times 10^{-20}$ J, i.e. Particle-Surface interaction[2]. Similarly, the Hamaker constant for polystyrene-water-polystyrene system is $A \approx 1 \times 10^{-20}$ J i.e. Particle-Particle interaction[2]. $K^{-1} = 0.3/\sqrt{[C]}$ nm is the Debye length for a monovalent electrolyte. The DI water is a very weak monovalent electrolyte with $[H^+] = [OH^-] = 10^{-7}$M, accordingly the Debye length $K^{-1} = 0.3/\sqrt{[C]}$ nm = 948.7 nm. The interaction constant 'Z' for two surfaces having similar surface potential 'ψ_o' is defined as $Z = 9.22 \times 10^{-11}\tanh^2(\psi_o/103)$ Jm^{-1}. The electrostatic potential of polystyrene microspheres in DI water determined by zeta potential measurement is $\psi_o \approx -30$ mV. The streaming zeta potential measurements on glass is $\psi_o \approx -30$ mV as reported previously[3]. Reports on zeta potential and Hamaker constant measurements typically differ by $1 - 1.5$ times[4,5] and may accordingly require modifications in the expressions used. The expression for van der Waals and electrostatic double layer force for Particle-Particle interaction is exactly half of Particle-Surface interaction when interacting particles are identical. Also, constants A and Z are similar for both glass and polystyrene. Hence, the DLVO force between two polystyrene microspheres (Particle-Particle) is of the similar order as between a polystyrene microsphere and glass substrate (Particle-Surface).
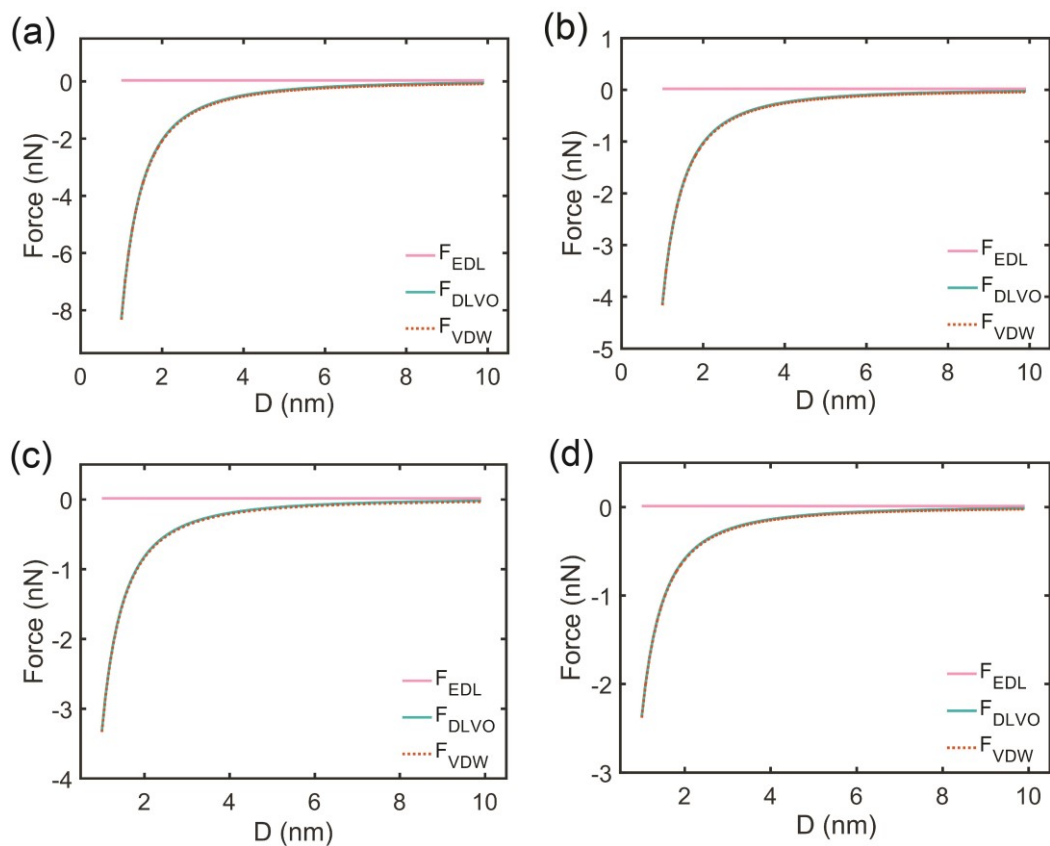
Figure S3. DLVO forces in DI water medium. **(a)** DLVO force as a function of gap between polystyrene microsphere of size 10 µm and glass substrate. (b) DLVO force as a function of gap between two polystyrene microspheres of size 10 µm. (c) DLVO force as a function of gap between polystyrene microsphere of size 4 µm and glass substrate. (d) DLVO force as a function of gap between two polystyrene microspheres of size 4 µm and 10 µm.

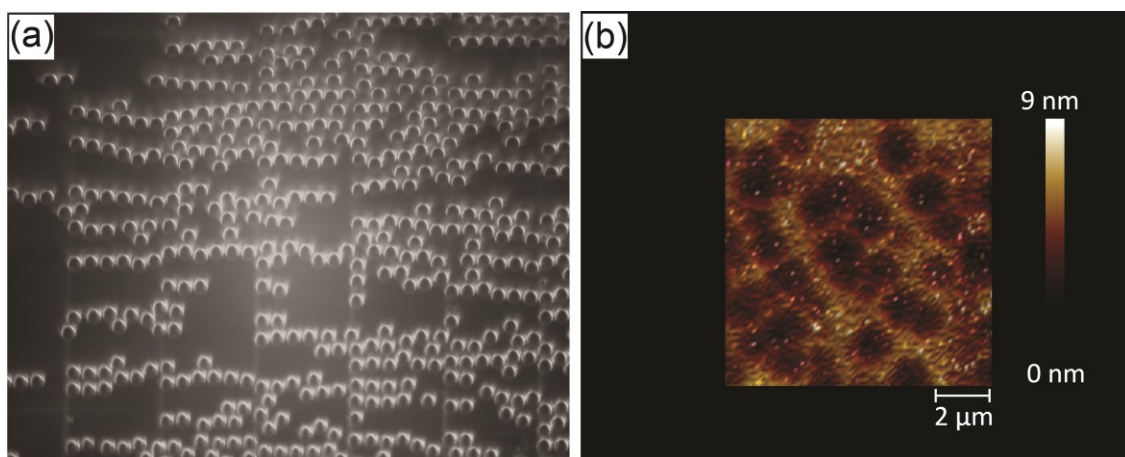**Figure S4.** (a) Linear chains of 10 µm microspheres eventually cover 70 % of the total area. (b) AFM of glass substrate reveals its roughness as ~8.0 nm.

4. Analytical expression of drag force near a surface

The drag force of a particle flowing near a surface can be analytically derived as $f_d \times F_{Stokes}$ where $F_{Stokes} = 6\pi\eta rV$ is the stokes drag on a sphere moving with speed 'V', where $\eta = 8.9 \times 10^{-4}$ Pa.s is the viscosity of water, and r = 5 μm is the radius of microspheres. f_d is expressed as[6–8]:

$$f_d = \left[1 - \frac{9}{16}\left(\frac{r}{r+z}\right) + \frac{1}{8}\left(\frac{r}{r+z}\right)^3 - \frac{45}{256}\left(\frac{r}{r+z}\right)^4 - \frac{1}{16}\left(\frac{r}{r+z}\right)^5 \right]^{-1}$$

Where 'r' is the radius of particle and 'z' is the distance of particle from wall. In the event when particle grazes a wall or is pinned, z = 0 and f_d = 1.7. To verify this expression, we can compare the calculated force on a particle pinned near a wall from COMSOL simulation. The drag force on a particle of size 10 μm pinned on the bottom of a channel of height '100 μm' and width '500 μm' at a flow rate of '10 μl/min' as calculated from simulation comes out to be 0.12 nN. The force using analytical expression is $1.7 \times 6\pi\eta rV = 0.128$ nN, assuming speed 'V= 0.0009 m/s' to be same as the flow speed experienced by the particle at its centre when pinned on the bottom. The flow speed 'V= 0.0009 m/s' at the height of 5 μm from bottom of the channel at a flow rate of 10 μl/min is estimated for a parabolic flow profile in a microfluidic channel.

5. Materials and methods

Photoresist – S1813, Camera – Thorlabs, DCC1545M; Light source - EO, 63-306; Microspheres of size 10 μm – Sigma, 72986-5ML-F; Smooth microspheres (R_a = 1.5 nm) of size 4 μm – Sigma, 81494-5ML-F; Rough microspheres (R_a = 4.7 nm) of size 4 μm – Spherotech, PP-40-10.

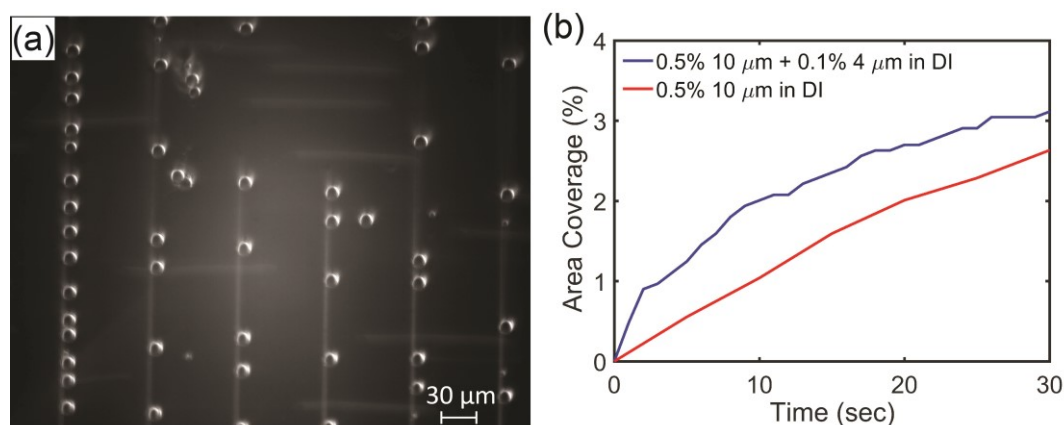## 6. Mixture of 0.1% w/v 4 μm microspheres of lower roughness ($R_a = 1.5$ nm) and 0.25% w/v 10 μm microspheres

Figure S5. (a) Selective deposition of 10 μm microspheres on downslope at a flow rate of 10 μl min^{-1} (b) Area of coverage with and without 4 μm microspheres in DI medium is nearly same.

7. Description of Supplementary Videos

a) SI Video 1: Microsphere of size 10 μm in DI water pins on the downslope when passed at a flow rate of 10 μl/min.

b) SI Video 2: Microsphere of size 10 μm in DI water pins on both the upslope and downslope when passed at a flow rate of 0.1 μl/min.

c) SI Video 3: Microsphere of Size 10 μm in PBS 0.1 M forms linear chain when passed at a flow rate of 10 μl/min. This is not observed when the microspheres are flown in DI water medium and is due to reduced Debye length in PBS solution.

d) SI Video 4: Longer version of SI Video 3 to show coupling in nearby chains.

e) SI Video 5: Microsphere of size 10 μm and 4 μm (mixed) in DI water deposits rapidly when passed at an initial flow rate of 10 μl/min and later increased to 70 μl/min. This is observed only when 4 μm microspheres of higher roughness (R_a = 4.7 nm) is used and is not observed when 4 μm microspheres of lower roughness (R_a = 1.5 nm) is used. Further, the initiation of rapid deposition happens only at a higher flow rate.

f) SI Video 6: Deposition of first few microspheres in the case of rapid deposition as shown in SI Video 5.

g) SI Video 7: Microsphere of size 10 μm and 4 μm (mixed) in PBS 0.1 M aggregate to form clumps. This is similar to the coagulation process.

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