**Supplementary Material**

**Colloid Retention and Mobilization Mechanisms under different Physicochemical Conditions in Porous Media: A micromodel Study**

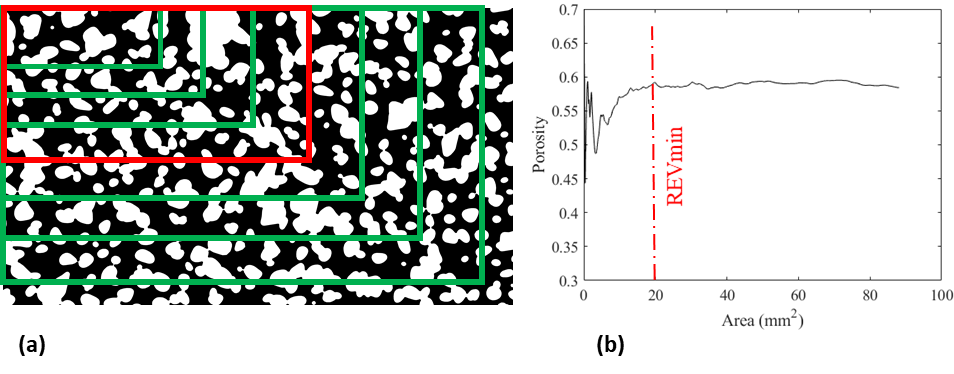
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## S1. Image processing

The quantity of colloids retained in the micromodel was computed from the captured images and represented as a percentage of the pore space. Images were captured at the Representative Elementary Area (REA) of the microchip. REA was determined by calculating the porosities of an expanded rectangular area, as shown in Fig. S1-a. The minimum REA represents an area of 20 mm2 with a porosity of 0.58, which matches that of the entire microchip (Fig. S1-b). The images were captured at 2.5x magnification with a 2x plan apochromatic objective (0.234 numerical aperture, 0.94 µm/pixel resolution, 2.4 x 1.8 mm2) to obtain images at a higher resolution. Nine adjacent images were combined using image registration to obtain a large image of an area of 29.6 mm2 (6.3x 4.7 mm2) at a resolution of 0.94 µm/pixel, which is REA. A moving stage was used to move the sample holder at a spatial movement in the x and y direction (2 mm and 1.5 mm, respectively), which in turn provides sufficient overlap for image registration to allow imaging at different locations to assemble a large image.



**Fig. S1:** REA calculations: (a) The REA was determined by increasing the sampling area of the base image by 3.5 µm width and 1 µm depth until the porosity values converged (b) the area of REA was found to be ~ 20 mm2.

Pore space in the micromodel was identified by segmentation of image captured with dye-mixed water using a threshold value determined by Otsu’s method [55]. Gas-phase and colloids in the pore space were segmented using a flood fill algorithm and Otsu’s method, respectively [55,56]. Each segmentation step included the application of an edge-preserving Gaussian bilateral filter followed by a median filter to enhance the contrast of the phases and remove any possible noise in the image. Pixel counting was adopted to obtain the percentage of particles retained after single and two-phase flow experiments. Colloids on GWI were quantified by counting the pixels in the colloid phase that has gas-phase in the neighboring pixel (Table 2).

# S2. Theoretical Considerations

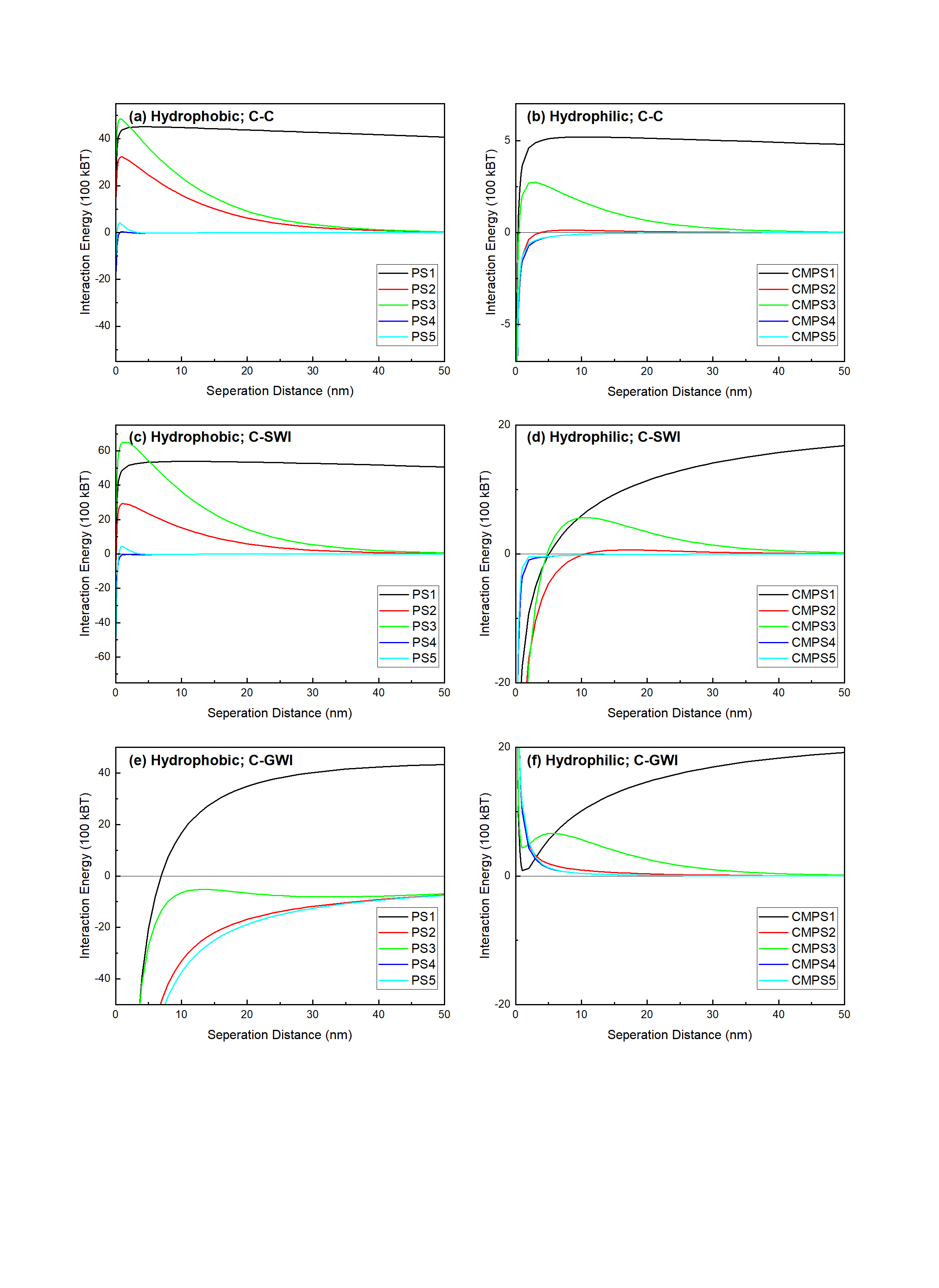
## S2.1. DLVO forces

The theoretical DLVO profiles of the colloids interacting with other colloids or interfaces were calculated as the sum of van der Waals (EvdW), electrostatic (Ee), and hydrophobic (Eh) energies [57–61]. Surface potentials replaced the measured zeta potential values in electrostatic energy calculations. All parameters and equations used to compute the DLVO energy profiles are given in Supporting Material. In the DLVO energy profiles, negative interaction energies indicate an attraction while the positive energy represents a repulsion for colloid-colloid, colloid-SWI or colloid-GWI interactions. The computed DLVO energy profiles for each experimental condition used in this work are presented in Fig. S2 for all possible interactions: colloid-colloid, colloid-SWI, and colloid-GWI interactions. The computed primary energy minimum (ϕmin1), the energy barrier (ϕmax1), and secondary energy minimum (ϕmin2) are given in Table S1.

## S2.2. Detachment forces

While fluid drag force is acting as the detachment force in single-phase flow, capillary forces are dominant in mobilizing attached colloids from SWI in two-phase flow. The drag forces generated on a partially submerged particle will be smaller in magnitude compared to the adhesive forces during two-phase flow [11,17,62]. The capillary forces acting on a colloid along the contact line between SWI and GWI (can be termed as GWSI, i.e., Gas-Water-Solid Interface) can be calculated as a function of surface tension, the contact angle of the colloid (θ) and the angle determining the position of GWI on the colloid surface (φ) [11]. Two magnitude maxima can be obtained for capillary force; one for φ < θ (capillary forces directing towards Gas-phase) and other for φ > θ (capillary forces directing away from Gas-phase, Supporting Material) [11]. The two components of capillary forces can be derived based on the contact angle of the solid surface (α). A friction force acts at the contact point that opposes the colloid movement on the solid surface.

Fig. 5 provides a conceptual schematic of forces acting on a colloid as GWI advances along the colloid surface. While GWI encounters a deposited particle, a small initial value of φ (φ < θ) can result in colloid lifting by GWI assuming the capillary force exceed the magnitude of adhesion force on SWI (Fig. 5-a). The capillary force acting on a colloid at GWSI was reported to be two orders of magnitude higher than DLVO forces [53,54]. Further wetting of colloid (φ > θ) can change the direction of the capillary force, as shown in Fig. 5-b. The frictional force can balance the horizontal component of the capillary force that pushes the colloid back into bulk fluid. The retained colloids remain attached to GWSI and later on thin films while GWI advances further on the solid surface, as shown in Fig. 5-c. Thick water films, as shown in Fig. 5-d prevent colloid pinning on GWSI; instead, the interaction is similar to the case in Fig. 5-a where the colloid remains attached to GWI. Thick water films formed around the solid surfaces due to the etched shape of the channel, as shown in Fig. 5-e.



**Fig. S2:** Computed DLVO interaction energies of colloids with other colloids (C-C), or SWI (C-SWI) or GWI (C-GWI) at different solution chemistry for hydrophobic and hydrophilic colloids.

**Table S1:** Estimated values of φmin1, φmax1 & φmin2 for hydrophobic and hydrophilic colloids at various interfaces

|  |  |  |  |  |  |  |  |
| --- | --- | --- | --- | --- | --- | --- | --- |
|  |  | Hydrophobic colloids (PS) | | | Hydrophilic colloids (CMPS) | | |
| ID | Interaction | φmin1 (100 kBT) | φmax1  (100 kBT) | φmin2 (100 kBT) | φmin1 (100 kBT) | φmax1  (100 kBT) | φmin2 (100 kBT) |
| 1 | Colloid-Colloid | na | 45.2 | na | -12.8 | 4.6 | na |
| 2 | Colloid-Colloid | na | 31.7 | na | -25.8 | 0.03 | na |
| 3 | Colloid-Colloid | na | 47.7 | na | -22.3 | 2.6 | na |
| 4 | Colloid-Colloid | -23.6 | 0.08 | na | -25.9 | na | na |
| 5 | Colloid-Colloid | -3.72 | 3 | -0.3 | -25.3 | na | na |
| 1 | Colloid-SWI | na | 53.8 | na | -70.3 | 8.5 | na |
| 2 | Colloid-SWI | -3.2 | 29.36 | na | -71 | 0.63 | na |
| 3 | Colloid-SWI | na | 65 | na | -55 | 5.6 | na |
| 4 | Colloid-SWI | -31 | 0.02 | -0.42 | -63.6 | na | na |
| 5 | Colloid-SWI | -4.8 | 10.4 | -0.25 | -58.2 | na | na |
| 1 | Colloid-GWI | -361 | 43.3 | na | na | 78.3 | na |
| 2 | Colloid-GWI | -922 | na | na | na | 109 | na |
| 3 | Colloid-GWI | -853 | na | na | na | 79.8 | na |
| 4 | Colloid-GWI | -917 | na | na | na | 110.9 | na |
| 5 | Colloid-GWI | -903 | na | na | na | 96.8 | na |

**Movie 1:** shows the mobile colloids interacting with GWI and attachment occur due to hydrophobic interaction. The effect of hydrodynamics near GWI on colloid movement near GWI is visible from the distracted colloids due to flow reversal near the solid phase.

**Movie 2:** shows the detachment of deposited colloids by the moving GWI and subsequently retained on GWI, GWSI, and thin films.