Supplementary Materials
Progressive colloidal clogging mechanism by dendritic build-up in porous media

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1 DLVO interaction potential approximations

We apply DLVO theory to approximate particle-particle and particle-surface interactions. It represents the sum of attractive potentials due to van der Waals forces and repulsion potentials due to the interaction of the electric double layers:

$$V^{DLVO} = V^{VdW} + V^{EDL}.$$

In Equation 1 we present the particle-particle interaction potential for particles of radius a_p separated by a distance h (where $h \ll a_p$), as proposed by Schenkel and Kitchener [7]. Additionally, Equation 2 presents the particle-surface interaction potential as described by Hogg et al. [4]:

$$V_{pp}^{DLVO} = -\frac{Aa_p}{12h} + \frac{\epsilon_0 \epsilon_r a_p \phi_p^2}{2} \left(\ln \left(1 + \exp(-\kappa h) \right) \right), \tag{1}$$

$$V_{ps}^{DLVO} = -\frac{Aa_p}{6h} \left(\frac{1}{1 + 14h/\lambda} \right) + \frac{\epsilon_0 \epsilon_r a_p}{4} (\phi_p^2 + \phi_s^2) \left(\frac{2\phi_p \phi_s}{\phi_p^2 + \phi_s^2} \ln \left(\frac{1 + e^{-\kappa h}}{1 - e^{-\kappa h}} \right) + \ln \left(1 - \exp(-2\kappa h) \right) \right), \tag{2}$$

where all the parameters with their corresponding values are defined and listed in Table 1. Additionally, we plot the DLVO potentials for particle-particle and particle-surface interactions as a function of the separation distance (Fig. 1).

2 Dendrite clogging at multiple locations within the porous domain

Fig. 2 presents averaged experimental images capturing the progressive formation of a dendritic clog in a specific region of the porous domain at a flow rate of 8 µL/min, as the number of

Table 1: Parameters and their corresponding values used in the DLVO interaction potential approximations. The annotations p, m, and s refer to the particle, medium, and PDMS surface.

| Fixed parameters	Value
Particle radius, a_p	2.25 μm
Vacuum permittivity, ε_0	8.85×10^{-12} C^2/Jm
Medium dielectric constant, ε_r	78.5 for water [1]
	40.2 for glycerol [1]
	70.1 (weighted average)
Boltzmann constant, k_B	1.38×10^{-23} J/K
Temperature, T	293 K
Avogadro's number, N_A	6.023×10^{23}
Electron charge, e	1.6×10^{-19} C
Hamaker constant, A	$A_{polystyrene} = 6.5 \times 10^{-20}$ J [3]
	$A_{PDMS-surface} = 4.4 \times 10^{-20}$ J [2]
	$A_{water} = 3.7 \times 10^{-20}$ J [3]
	$A_{glycerol} = 1.38 \times 10^{-20}$ J [5]
$A_{33} = A_{medium}$	3.19×10^{-20} J (weighted average)
$A_{131} = (\sqrt{A_{33}} - \sqrt{A_{11}})(\sqrt{A_{33}} - \sqrt{A_{11}})$ [5]	2.7×10^{-21} J for polystyrene-polystyrene interaction
$A_{132} = (\sqrt{A_{33}} - \sqrt{A_{11}})(\sqrt{A_{33}} - \sqrt{A_{22}})$ [5]	7.3×10^{-21} J for polystyrene-surface interaction
Retardation length, λ	100×10^{-9} m
Solution Ionic strength, I	100 mM
Measured particles zeta potential, ϕ_p	-21.3 mV
PDMS surface zeta potential, ϕ_s	-23 mV [6]
Reciprocal Debye length, $\kappa = \left(\frac{2 \times 10^3 N_A e^2 I}{\varepsilon_0 \varepsilon_r K_B T} \right)^{\frac{1}{2}}$	1.2×10^9 m
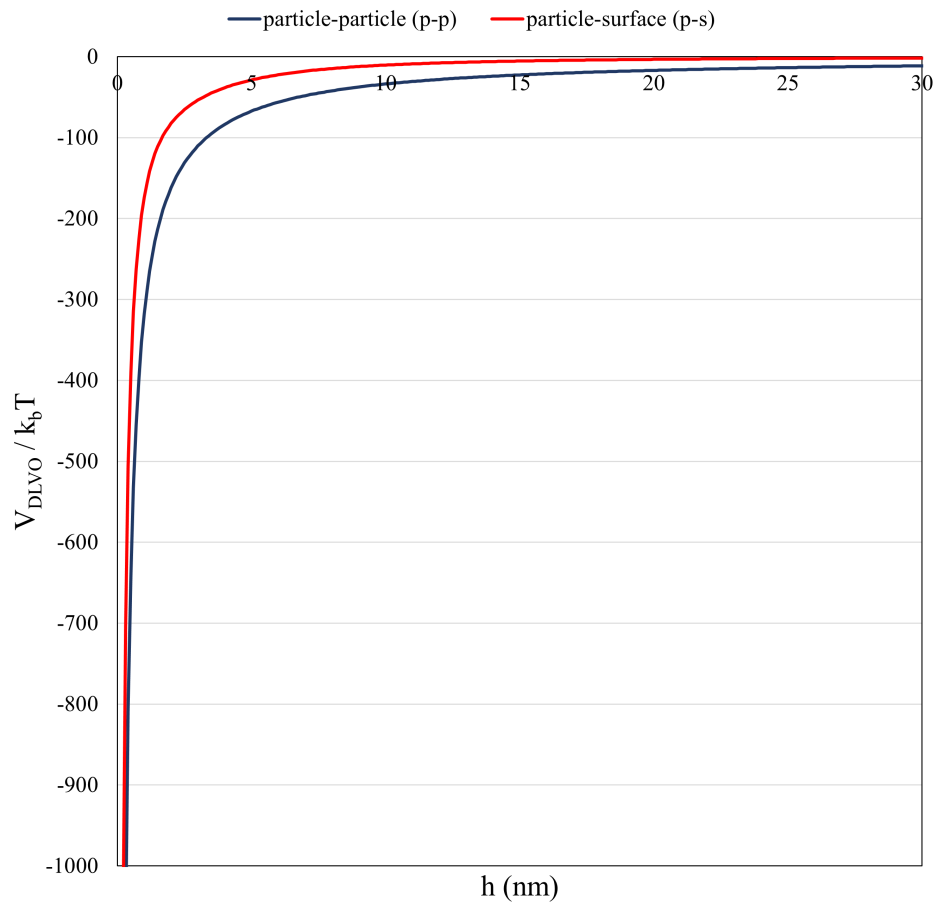
Figure 1: DLVO profiles for particle-particle and particle-surface interactions as a function of separation distance (h) in nanometers (nm).

injected pore volumes (PVs) increases, leading to the complete clogging of the local flow pathway. Fig. 3 further illustrates dendritic clogging occurring in two distinct regions: (a) after 2370 PVs and (b) after 2261 PVs, emphasizing the variability in dendrite-clogging dynamics within the porous domain.
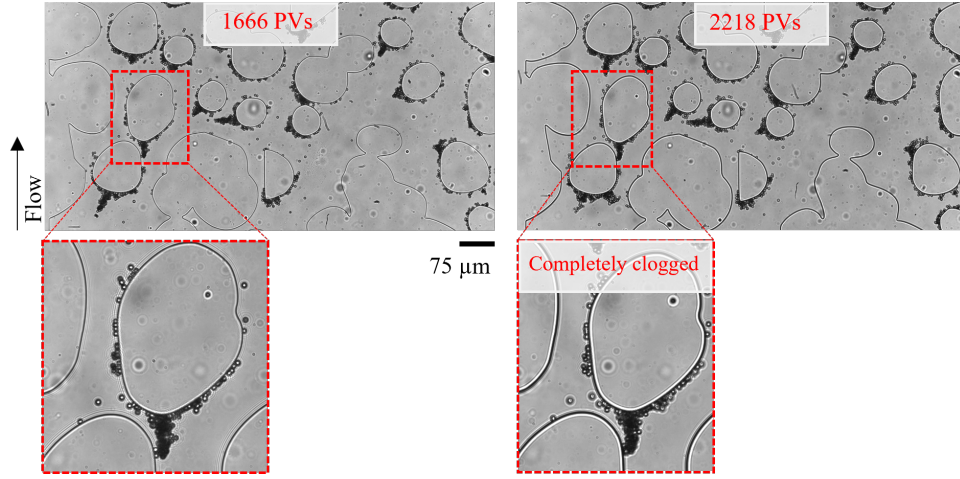

Figure 2: Averaged experimental images at a flow rate of 8 μL/min, showing the progressive formation of a dendritic clog in a specific region of the porous domain over successive pore volumes of particle injection (PVs).
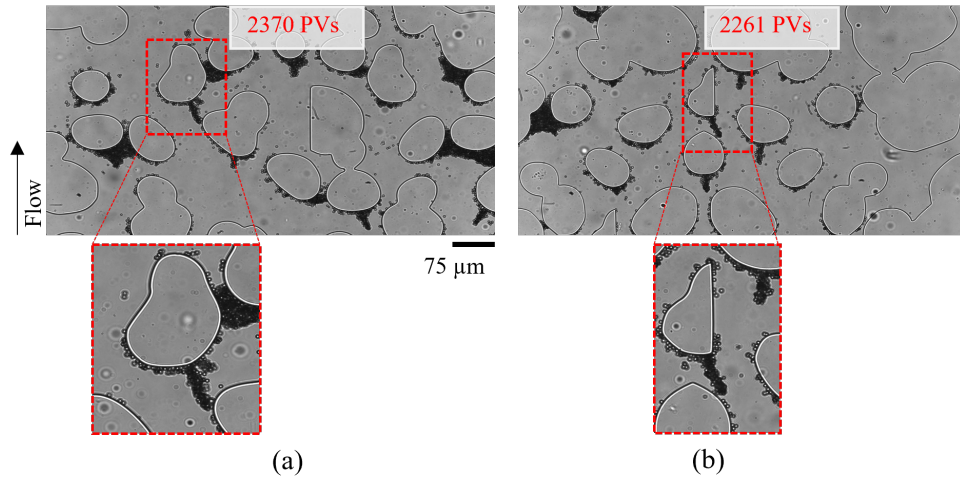

Figure 3: Averaged experimental images at a flow rate of 8 μL/min, illustrating dendritic clogging in two distinct regions of the porous domain: (a) after 2370 pore volumes (PVs) of particle injection and (b) after 2261 PVs.

3   Movies of the experiments

The movies were recorded using a FASTEC camera at 271 fps and are presented at a playback speed of 10 frames per second:

- Movie corresponding to Figure 3(a) in the main manuscript after 1664 PVs injected: `Movie_Figure_3a_8uL_min_1664_PVs`.

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- Movie corresponding to Figure 3(a) in the main manuscript after 2225 PVs injected: `Movie_Figure_3a_8uL_min_2225_PVs`.

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