Experimental results indicate that lowers the permeability of the porous medium more and submicron fluid, and J. C. Santamarina Re...
2. Experimental Study: Materials and Methods

2.1. Microfluidic Chips

We use soft lithography to fabricate microfluidic chips for convergent radial flow. The process includes (i) mask layout using computer-aided design software, (ii) mask printing, (iii) fabrication of the silicon wafer master with negative photoresist (SU-8 2050), (iv) polymerization of polydimethylsiloxane PDMS using the master as a mold, and (v) bonding of the PDMS slabs onto a glass substrate with the use of oxygen plasma (see Mazutis et al., 2013 for a detailed protocol). The microfluidic chip consists of 300-μm cylindrical columns separated by d_c = 40-μm-wide pore constrictions; all pore channels are 50 μm high. The inlet cavity of the porous network ensures a uniform flow field (Figure 1a).

2.2. Particle Suspensions

We use glass particles (440345 and 44054, Sigma-Aldrich, specific gravity Gs = 2.60) and polystyrene latex particles (polystyrene plain, PS010UM and PS005UM, Magsphere, Gs = 1.05) of two diameters d = 5 and 10 μm. These four particle types create different conditions among governing forces and geometric ratios (constriction-to-particle size ratios d_c/d = 4 and 8, Figure 1b). All particle suspensions are prepared with deionized water at a 0.2% mass concentration.

2.3. Test Protocol

Figure 1c presents the flow system. We saturate the radial flow microfluidic chip with deionized water (trapped air escapes through the gas permeable PDMS walls) and use a peristaltic pump to withdraw the suspensions from the “central port” at a constant flow rate (PeriWave microfluidic pump). A pressure sensor monitors the pressure at the central port (uPS0250, LabSmith). A magnetic stirrer prevents settlement or coagulation prior to injection. Digital video microscopy records particle movements within pores and captures emerging clogging patterns at the chip scale.

2.4. Image Analysis

We wrote an image processing algorithm to automatically detect pore clogging events at any of the 1,078 constrictions parallel to the flow direction in the full chip. The algorithm extracts the image of each constriction and assesses clogging based on (1) the gray value relative to the maximum, minimum, and threshold gray values and (2) the total number of pixels in the dark area (red box, Figure 2). Data gathered with the calibrated algorithm allow the interpretation of spatial and temporal correlation among clogging events.
3. Results and Analyses

3.1. Dimensionless Ratios

A migrating particle experiences its buoyant weight, the drag force, inertia against motion changes, and electrical attraction toward other particles and the pore walls. Dimensionless ratios capture the governing processes in terms of the particle size $d$, obstacle size $D$, particle mass density $\rho_p$, fluid mass density $\rho_f$, fluid viscosity $\mu$, fluid velocity $v$, gravity $g$, and attraction force $F_A$ to pore walls (the sum of van der Waals force, electrostatic force, and hydrophobic adhesion force). The three main dimensionless ratios are as follows:

\[ \text{Ratio} = \frac{F_A}{D g} \]

\[ \text{Ratio} = \frac{\rho_p - \rho_f}{\rho_f} \]

\[ \text{Ratio} = \frac{D}{d} \]

Figure 2. Image analysis. (a) The plugged constrictions shown within red boxes are identified by the image analysis algorithm. (b) Gray value distribution of a pore constriction.

Figure 3. Clogging: pore-scale observations. The solid columns are 300 $\mu$m in diameter in all cases (refer to Figure 1). Note: there is no clogging with 5- $\mu$m glass particles.
Adhesion number:

\[ N_{ad} = \frac{N_{at}}{\text{Drag}} = \frac{F_A}{3 \pi \mu d v} \]  

(1)

Archimedes number:

\[ Ar = \frac{\text{Terminal velocity}}{\text{Flow velocity}} = \frac{g d^2 (\rho_p - \rho_f)}{\mu v} \]  

(2)

Stokes number:

\[ Stk = \frac{\rho_p d^2 v}{18 \mu D} \text{ (inertial effect)} \]  

(3)

The Stokes number \( Stk \) is the ratio between the particle response time to the fluid field response time; a particle with a large Stokes number is dominated by inertia and tends to follow its trajectory, while a particle with a low Stokes number follows fluid streamlines. The ratio \( N_G \) between the constriction size \( d_c \) and the size of the migrating particle \( d \) is the main geometric descriptor:

\[ \text{Geometric ratio} : N_G = \frac{d_c}{d} \]  

(4)

In addition, the Reynolds number \( Re = \rho f D / \mu \) characterizes flow conditions around an obstacle.

### 3.2. Clogging Mechanisms

Figure 3 illustrates salient pore-scale observations gathered for the four particle suspensions. Distinct clogging mechanisms take place when glass particles and latex particles are involved.
3.2.1. Glass particles

Nonbuoyant glass particles (specific gravity $G_s = 2.60$) experience gravity and fall toward the bottom substrate (high $Ar$ value); on the other hand, their inertia causes collisions against obstructing pore walls (high $Stk$, Figure 4a), and particles suffer significant retardation relative to the fluid displacement along the flow paths. Inertial retardation develops where streamlines bend and the flow velocity changes at a high Stokes number $Stk$. Retardation results in a gradual increase in the local volume fraction of particles at the pore scale (Figure 3). High local concentrations near pore constrictions facilitate the formation of granular bridges. Figure 4b illustrates the retardation-accumulation bridging process.

Dimensionless ratios $Ar$ and $Stk$ scale with the square of the particle size $d^2$; hence, retardation becomes negligible when particles are small. Figure 3 shows no evidence of retardation and accumulation for the $d = 5$-μm-glass particle suspension.

Clogging for a geometric ratio $N_G = 4$ (Figure 3, top left) exceeds previously reported thresholds from single-pore constriction experiments with spherical particles that showed stable bridge formation when $N_G < 3$ (Marin et al., 2018; Valdes & Santamarina, 2006). This result hints to stabilizing electrostatic effects when small micron-scale particles are involved. Furthermore, a clogged pore throat in porous medium alters flow pathways instead of generating a large pressure drop across the clogged pore throat, which is the case for the single channel systems. Both effects combine to allow for bridging at $N_G$ values larger than 3 when small particles migrate in pore networks.

Figure 5. Time lapse photographs: clogging sequence for migrating 10-μm latex particles ($N_G = d_c/d = 4$).
3.2.2. Latex particles

Quasi-buoyant latex particles ($G_s = 1.05$) experience negligible retardation and follow along streamlines ($Ar \approx 0$ and small $St_k$). However, latex particles experience attraction toward PDMS walls and may be captured if in close proximity (high adhesion number $N_{ad}$). Figure 4c illustrates the process of direct particle interception. Indeed, latex particles experience a high adhesion number $N_{ad}$ toward the hydrophobic PDMS pore walls which ensures a strong particle-surface bonding. Captured particles reduce the size of pore throats, and particle-particle interaction favors aggregate formation near constrictions (Dersoir et al., 2015; Dersoir et al., 2017). Reduced constriction size and grain aggregation combine to form bridges even at large constriction-particle size ratios $N_G$ ($N_G = 4$ and 8; Figures 3 and 4). The sequence of time lapse photographs shown in Figure 5 highlights the capture-clogging process at pore constrictions.

3.3. The Effect of Flow Rate

Experimental results suggest that both particle retardation and capture are flow velocity dependent (Note: Velocity $v$ is involved in governing dimensionless ratios $N_{ad}$, $Ar$, $St_k$, and $Re$). We study the influence of the suspension injection rate on clogging at the pore scale using the convergent radial flow chips (Figure 1). The Reynolds number is $Re < 35$ in all experiments. Let us define the clogging ratio as the number of clogged pore constrictions with respect to the total number of constrictions. Figure 6 shows the effect of flow rate on the clogging ratio evolution for both glass and latex particles.

Clearly, a high flow rate leads to fewer clogged pores for glass particles (Figure 6a). The prevalent retention mechanism for glass particles is retardation-accumulation bridging. A high flow rate decreases the Archimedes number $Ar$ and minimizes gravity retardation (the maximum Archimedes number for the

![Image](https://www.agu.org/journals/j耕地固地/10.1029/2019JB017813/)

**Figure 6.** The effect of flow rate on the evolution of clogging. (a) Glass particles. (b) Quasi-buoyant latex particles. Flow rates $q = 20, 40$, and $60 \mu l/min$. In both cases: $d = 10 \mu m$.

![Image](https://www.agu.org/journals/j耕地固地/10.1029/2019JB017813/)

**Figure 7.** Evolution of clogging ratio distribution across radial flow microfluidic chips from 0 to 500 permeated pore volumes. Glass particles: (a) flow rate $q = 40 \mu l/min$ and (b) $q = 60 \mu l/min$. Latex particles: (c) flow rate $q = 40 \mu l/min$ and (d) $q = 60 \mu l/min$. The row sequence is defined in Figure 1a. In all cases, $d = 10 \mu m$, so that $N_G = d_c/d = 4$. Refer to (a) for color coding of pore volumes.
various flow rate is $Ar_{\text{max}} = 2$ when $q = 20 \mu l/min$, $Ar_{\text{max}} = 1$ when $q = 40 \mu l/min$, and $Ar_{\text{max}} = 0.7$ when $q = 20 \mu l/min$.

The flow rate exerts a reversed effect on the clogging behavior of latex particles compared to glass particles (Figure 4b): The clogging ratio increases with the increase in flow rate $q$ from $q = 20 \mu l/min (Re_{\text{max}} = 11)$ to $q = 60 \mu l/min (Re_{\text{max}} = 33)$. Streamlines compress at high Reynolds numbers and improve particle “capture efficiency by direct interception” $\eta_{DI}$ (Figure 4c). A detailed hydrodynamic formulation by Espinosa-Gayosso et al. (2012) shows the relationship between the capture efficiency $\eta_{DI}$, Reynolds number $Re$, and the relative size $d/D$ between the migrating particle size $d$ and the obstacle size $D$. Their results can be approximated as (valid for passive particles and $Re \leq 47$) follows:

$$\log \eta_{DI} = 0.4 \log Re + 6 (d/D) - 3$$

Hence, the particle capture efficiency by direct interception $\eta_{DI}$ increases with both Reynolds number $Re$ and the particle-to-obstacle size ratio $d/D$.

### 3.4. The Effect of Radial Flow

The spatially varying velocity field in convergent radial flow changes local dimensionless ratios $Ar$, $Stk$, $N_{adv}$, and $Re$ from the far field to the central producing well. Figure 7 shows the evolution of row clogging ratios in the radial flow microfluidic chip during the injection of 500 pore volumes at two different flow rates. The clogging ratio for glass particles decreases linearly from the far field to the central port as glass particles experience higher gravity retardation far from the central extraction port (Figures 7a and 7b).

On the other hand, streamline compression at high flow velocity increases the capture efficiency of latex particles (Figure 4c, equation (5)); yet the very high near well velocity drags particles away and hinders capture (low $N_{adv}$). These two competing mechanisms lead to a maximum clogging ratio at a characteristic radial distance away from the well wall (Note: Annular clogging was reported in Valdes & Santamarina, 2006.).
3.5. Dependent Clogging: Implications

Experimental observations show that a clogged pore constriction alters the flow patterns in nearby open paths, affects retardation, and may promote further clogging. We wrote a postprocessing algorithm to quantify the spatial dependence of clogging in convergent radial flow. Let us call a new clogging event "dependent" when one or more of the neighboring pore constrictions are already clogged; otherwise, the new clogging event is called “independent” (Figure 8a). Figures 8b and 8c show the evolving clogging ratios for dependent and independent pore clogging events in the case of glass and latex particles under the same flow rate \( q = 40 \, \mu \text{l/min} \). The increment of the clogging ratio \( \Delta N/N_{\text{ava}} \) is calculated within 5-min time intervals, given the fact that the number of open pore constrictions for dependent/independent clogging \( N_{\text{ava}} \) evolves with time. Dependent clogging is significantly more frequent than independent clogging, especially for glass particles due to the heightened inertial retardation in the surroundings. These results underscore the "cross talk" between pores observed in parallel microchannel experiments (Liot et al., 2018; Sauret et al., 2018; Van Zwieten et al., 2018).

The distribution of clogged pore throats profoundly influences the reduction in permeability of porous media (Dai & Seol, 2014; Sauret et al., 2018). We use a simple pore network model to explore the effect of dependent clogging on the evolution of permeability. The porous medium is a 2-D square network model made of 50 × 50 tubes of the same radius. Each clogging event is treated as a random event. Initially, all constrictions have the same clogging probability \( P_{\text{ind}} \). We assume Hagen-Poiseuille flow in each tube until it clogs; thereafter, the conductivity of the tube becomes zero. Once a tube clogs, neighboring tubes have a dependent probability of clogging \( P_{\text{dep}} \). Figure 9 shows the influence of \( P_{\text{dep}}/P_{\text{ind}} \) on the evolution of permeability. Clearly, spatially correlated dependent clogging lowers the permeability of the porous medium more effectively than independent clogging.

4. Conclusions

Migratory particles in porous media interact with fluids, pore walls, and other particles. Dimensionless ratios capture the effect of particle-level forces (\( N_{\text{ad}}, Ar, \) and \( St_k \)), flow conditions (\( Re \)), and geometric characteristics (\( N_G \)). These ratios define the domains for particle retardation, adhesion, and bridging. Micron-scale nonbuoyant glass particles and quasi-buoyant latex particles exhibit distinct clogging mechanisms. Glass particles experience retardation from gravity and inertial effects; the increase in the local volume fraction of particles promotes the formation of multigrain bridges at pore constrictions. Quasi-buoyant latex particles emphasize electrical interactions and may adhere to pore walls. Captured latex particles and aggregates may eventually plug constrictions, even at relatively high geometric ratios \( N_G \).

Flow rates exert opposite effects on the clogging behavior of glass and latex particles. A high flow rate diminishes gravity retardation and hinders the formation of stable multigrain bridges (case: nonbuoyant glass particles). On the other hand, high flow rates compress streamlines and facilitate adhesive capture to pore walls (case: quasi-buoyant latex particles).

Particles experience varying fluid velocity fields in convergent radial flow. Therefore, clogging distributions in radial flow reflect the local conditions (\( N_{\text{ad}}, Ar, St_k, \) and \( Re \)). Glass particles show a higher clogging ratio in the far field (gravity retardation under low flow velocity), while electrically affected latex particles tend to clog pores at a characteristic distance from the well wall and form an annular clogging pattern.

Clogging in porous media is not a random process. In particular, clogged pores alter flow conditions and promote further clogging nearby. Pore network model simulations suggest that dependent clogging lowers the permeability of the porous medium more effectively than independent clogging.

List of Notations

| Symbol | Description |
|--------|-------------|
| \( d \) | Migrating particle diameter |
| \( d_c \) | Constriction size |
| \( D \) | Obstacle size |
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\[ G \text{ Specific gravity} \]
\[ \rho_p \text{ Particle mass density} \]
\[ \rho_f \text{ Fluid mass density} \]
\[ \mu \text{ Fluid viscosity} \]
\[ v \text{ Fluid velocity} \]
\[ G \text{ Gravity} \]
\[ N_{ad} \text{ Adhesion number} \]
\[ Ar \text{ Archimedes number} \]
\[ St_k \text{ Stokes number} \]
\[ N_G \text{ Geometric ratio} \]
\[ Re \text{ Reynolds number} \]
\[ V \text{ Permeated volume} \]
\[ q \text{ Flow rate} \]
\[ \eta_{DF} \text{ Capture efficiency} \]
\[ p_{\text{dep}} \text{ Probability of dependent clogging} \]
\[ p_{\text{ind}} \text{ Probability of independent clogging} \]
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