Modeling microscale flow and colloid transport in saturated porous media

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Abstract

The microscale flow of water in natural soil porous media affects the transport of colloids and other contaminants contained in groundwater. In this study, two completely different computational approaches are applied to simulate pore-scale viscous flows in saturated porous media. The first is the lattice Boltzmann method based on the mesoscopic lattice Boltzmann equation. The second method, referred to as Physalis by its developers, is a hybrid representation in which a numerical solution based on discretized Navier-Stokes equation is coupled with analytical Stokes flow solutions valid locally near the surface of porous-medium grains. The porous medium is represented by a channel partially filled with circular (in 2D) or spherical (in 3D) particles. We demonstrate that the two methods can produce almost identical viscous flow at the pore scale, providing a rigorous cross-validation for each approach. A Lagrangian particle-tracking approach is then used to study the transport of colloids in these flows, considering hydrodynamic forces, Brownian force, and electro-chemical surface-interaction forces acting on each colloid. Due to the competing effects of hydrodynamic transport and electro-chemical interactions, it is shown that enhanced removal of colloids from the fluid by solid surfaces occurs when the residence time of colloids in a given flow passage is increased, in qualitative agreement with pore-scale visualization experiments using confocal microscopy.

Key words: Porous medium; saturated soil; lattice Boltzmann equation; colloid; deposition.

1 Introduction

Understanding the mechanisms of colloid retention and transport in soil porous media is of importance to the management of groundwater contamination by contaminants that could sorb to and migrate with mobile colloids or by pathogenic microorganisms. Even for the relatively simple case of saturated soil and aquifer, the transport of colloids and their attachment to solid surfaces are governed by a multitude of physical processes: transport by low-speed microscale water flows, Brownian motion due to random thermal fluctuation, and a variety of electro-chemical interactions between colloids and solid surfaces [8]. These physical processes together encompass a large range of length scales from millimeter scale to nanometer scale, with each possibly dominating the motion of a colloid depending on the colloid’s relative location within a pore-scale passage. A quantitative modeling tool

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requires both an accurate pore-scale flow simulation and a realistic representation of all important colloid-surface interaction forces.

This paper concerns mainly the accurate simulation of complex flows at the pore-scale. This will be addressed by employing two completely different computational approaches to a given flow problem. First, we will explore the use of lattice Boltzmann method (LBM) as a simulation tool for viscous flow through a porous medium. The LBM approach is based on a kinetic formulation and have certain advantages over the traditional Navier-Stokes based CFD [26,4]. While LBM models capable of addressing thermal flows, flows through porous media, multiphase flows, electro-osmotic flows, and contact line, etc., have been proposed in recent years, two general aspects remain to be studied before they can be applied to complex flow modeling. The first aspect concerns the accuracy and reliability of these LBM models for practical applications. Since these models have typically only been tested for idealized problems, their applications to complex flow problems need to be critically examined and different possible LB models be compared. The second aspect concerns a variety of LBM implementation issues when dealing with practical applications.

Since accurate local measurements of pore-scale microscale flows are not usually feasible, our strategy here is to introduce a second, Navier-Stokes based computational approach. The hybrid approach, referred to as Physalis by its original developers [28], integrates a numerical solution of discretized Navier-Stokes equation on a simple uniform grid with an analytical representation of local flow near the surface of a solid particle. Direct comparisons between LBM and Physalis offer an opportunity for cross-validating each approach as well as contrasting their pros and cons.

As the first step, we focus our attention on a two-dimensional model of porous media, namely, a two-dimensional channel partially filled with fixed circular cylinders. After the accuracy of the flow simulation is established, the trajectories of colloids are simulated under the influence of Stokes drag, Brownian force, and electro-chemical surface-interaction forces. The rate of deposition of colloids on solid surfaces at a given solution ionic strength is then analyzed for several flow speeds. A thorough analysis of colloid deposition under various conditions can be found in our companion paper [12].

2 Methodology

Consider a viscous flow in a two-dimensional channel with 7 fixed cylinders as shown in Fig. 1. This two-dimensional setting is used to mimic a slice of a 0.8mm×0.8mm channel packed with glass beads (0.20 mm in diameter) (i.e., see Fig. 10 below). Flow is driven by a constant pressure gradient or a body force in the y direction. Periodic boundary condition is assumed in flow direction with a periodicity length of \( L \), while no-slip velocity condition is applied on the two sidewalls at \( x = 0 \) and \( x = H \), and on the surface of the 7 glass cylinders. The channel width \( H \) is set to 200 and the cylinders have an identical diameter of 30. The computational domain size in terms of the grid-spacing (\( dx = dy = 1 \)) is 200 in the \( x \) direction and 93 in the \( y \) direction. The centers of the 7 cylinders are located at (50,25), (100,25), (150,25), (25,68), (75,68), (125,68), and (175,68), respectively.

At the initial time \( t = 0 \), the fluid is at rest. The body force per unit volume is set to \( F_B = 8 \rho \nu U_c / H^2 \), such that the centerline velocity of the channel would be \( U_c \) at long time when the body force is balanced by the viscous effects, if there were no glass cylinders in the channel. Here \( \nu \) and \( \rho \) are the fluid kinematic viscosity and density, respectively. The magnitude of \( U_c \) is adjusted to match the flow rate in our microchannel flow experiment. For the results discussed here, \( U_c \) is such that the Reynolds number based on \( U_c \) and \( H \) is \( U_c H / \nu = 0.20025 \).

2.1 The lattice Boltzmann approach

In the LBM approach, the lattice-Boltzmann equation for the distribution function \( f_i \) of the mesoscopic particle with velocity \( \mathbf{e}_i \)
Fig. 1. A sketch of the two dimensional channel with 7 fixed glass beads.

\[ f_i(x + e_i \delta_i, t + \delta_t) - f_i(x, t) = -\frac{1}{\tau} \left[ f_i(x, t) - f_i^{(eq)}(x, t) \right] + \psi_i(x, t) \]  

(1)

is solved with a prescribed forcing field \( \psi_i \) designed to model the driving pressure gradient or body force. In this work, \( \psi_i \) is specified as \( \psi_i(x, t) = W_i e_i \cdot F / c_s^2 \), where \( F \) is the macroscopic force per unit mass acting on the fluid. The standard D2Q9 lattice model in 2D and the D3Q19 model in 3D [26] are used with the following equilibrium distribution function

\[ f_i^{(eq)}(x, t) = W_i \left[ \rho + \frac{\rho_0 e_i \cdot u}{c_s^3} + \frac{\rho_0 uu : (e_i e_i - c_s^2 I)}{2 c_s^4} \right], \]  

(2)

where \( W_i \) is the weight, the sound speed \( c_s \) is \( 1/\sqrt{3} \), and \( I \equiv [\delta_{ij}] \) is the second-order identity tensor. The mean density \( \rho_0 \) is set to 1.0. The macroscopic hydrodynamic variables are computed as

\[ \rho = \sum_i f_i, \quad \rho_0 u = \sum_i f_i e_i, \quad p = \rho c_s^2, \]  

(3)

where \( \rho \), \( u \), and \( p \) are the fluid density fluctuation (the local fluid density is \( \rho_0 + \rho \)), velocity, and pressure, respectively. The above form of the equilibrium distribution was suggested by He and Luo [13] to best model the incompressible Navier-Stokes equation.

A uniform lattice is used to cover the computational domain. The straight channel walls are located in the middle of lattice links so a second-order accuracy is achieved with a simple bounce-back implementation. The inlet and outlet are also located half way on the lattice links to facilitate the implementation of the periodic boundary condition in the \( y \) direction.

The key implementation issue here is the treatment of solid particle surfaces. For each lattice node near a particle surface, we identify all links moving into the surface and their relative boundary-cutting location, namely, the percentage \( (\alpha) \) of a link located outside the surface. Since particles are fixed, this information is pre-processed before the flow evolution. Before the streaming step, the missing population is properly interpolated in terms of \( \alpha \) and two populations lying before and after the path of the missing population [32,20]. For results in this paper, we used the first-order interpolation based on two known populations, and found that the results are quite similar to the second-order interpolation based on three nodes [20]. All lattice nodes lying within the solid particles (including
the fluid-solid interface) are excluded from LBE evolution, their velocities are simply set to zero. As a validation check, the total mass for the fluid nodes (excluding the fluid-solid interface nodes) is computed and found to remain constant as time is advanced.

We also tested the generalized lattice Boltzmann equation or the multiple-relaxation-time (MRT) model as presented in [19,16]. The MRT has been shown to improve numerical stability so flows at higher Reynolds numbers can be simulated. Here our flow is at low flow Reynolds number, but we will demonstrate an interesting robustness of MRT which allows a wider range of relaxation parameter (or viscosity setting) to be used in the LBM approach, when compared to the usual BGK collision model shown in Eq. (1).

2.2 The Navier-Stokes approach: Physalis

To validate the lattice Boltzmann approach and compare it with traditional Navier-Stokes based computational approaches, we also developed a code using the hybrid method proposed by Prosperetti and co-workers [28,33,34]. The method was named *Physalis* [28]. Physalis combines a numerical discrete representation of the Navier-Stokes viscous flow around particles and an analytical representation imbedded near the surface of each particle.

![Fig. 2](image-url)

Fig. 2. The cage used to represent a glass bead surface in Physalis, for a glass bead with a radius of 15 grid spacings. Solid circles denote pressure cage nodes, open circles are vorticity cage nodes, filled triangles are u-velocity cage nodes, and open triangles are v-velocity cage nodes. The thick line denotes the glass bead surface.

The basic idea behind Physalis is as follows. Because of the no-slip boundary conditions on its surface, a solid particle induces a specific local flow structure that could be used to linearize the Navier-Stokes equations in the neighborhood of the particle surface. The fluid velocity, pressure, and vorticity near the particle surface can be expressed analytically using series solutions of Stokes flow equations. As a result, the geometric surface of the particle can be replaced by a Stokes flow solution valid in a narrow but finite region near the surface, known as the cage region as indicated by the two dash circles in Figure 2.
There are three main components in Physalis. The first component is an analytical representation of the flow within the cage region. This is obtained by the method of separation of variables applied to Stokes flow equations. The general form in 2D is given in [33] and in 3D is found in [34,10]. The second component is the numerical method for Navier-Stokes equations on a regular mesh (the flow solver). The second-order project method [3] is used. The intermediate velocity in the fractional step procedure is solved by a factorization method [17], while the Poisson equation for the projection step is solved by a combination of transformation and tridiagonal inversion. This mesh extends to the interior of the particle surface. The velocity cage essentially defines an internal boundary for the viscous flow where the Stokes solution is employed to specify the boundary conditions there.

The most essential component is the coupling or matching between the numerical solution on the regular mesh and the Stokes solution in the cage. This coupling is achieved by an iterative procedure in which (a) the numerical solution is used to refine the coefficients in the Stokes flow representation and in turn (b) the numerical solution is refined by an updated boundary conditions at the velocity cage from the refined Stokes flow. The first part is accomplished by a Singular Value Decomposition algorithm since an overspecified linear system (the number of cage nodes used for coupling is larger than the number of coefficients) is to be solved. The second part currently relies only on the specific method of defining the cage velocity nodes or the internal boundary, so the analytical nature of the Stokes solution may not be fully taken advantage of. There are more than one way to specify the cage region [28]. For accuracy of the Stokes flow representation, it is desirable to select the cage nodes as close to the surface of the particle as possible.

An important advantage of this hybrid method is that the force and torque acting on the particle can be calculated directly from the Stokes solution, avoiding often tedious numerical integration of local viscous force on the particle surface that is necessary for other non-hybrid numerical methods.

2.3 Dynamics of colloids

When the steady viscous flow is established in the microscale porous channel, colloids are randomly injected into the flow at the inlet with a velocity equal to the local fluid velocity, at a rate that corresponds to a concentration of 1 ppm. Since this concentration is very low, the fluid flow is assumed to be unaffected by the presence of colloids. Each colloid is treated as a discrete entity (point-like particle) and moves according to the following equation of motion

\[ m_c \frac{d\mathbf{v}(t)}{dt} = \mathbf{F}^{\text{drag}} + \mathbf{F}^{\text{b}} + \mathbf{F}^{\text{g}} + \mathbf{F}^{\text{B}} + \mathbf{F}^{\text{c}}, \]

where \( \mathbf{v}(t) \) is the instantaneous (Lagrangian) velocity of the colloid, \( m_c \equiv 4\pi \rho_c a_c^3/3 \) is the mass of the colloid, \( \rho_c \) is the material density of the colloid, and \( a_c \) is the radius of the colloid. All relevant physical parameters and their corresponding value in the numerical simulation are shown in Table 1. The hydrodynamic forces include the viscous drag \( \mathbf{F}^{\text{drag}} \) and the buoyancy force \( \mathbf{F}^{\text{b}} \). Other forces such as the pressure-gradient force, the added mass, Basset history term [22] are neglected here due to the very slow solvent (water) flow. \( \mathbf{F}^{\text{g}} \) is the gravitational body force. \( \mathbf{F}^{\text{B}} \) is a random force designed to simulate Brownian motion of the colloid due to local thermal fluctuations of solvent molecules. Finally, \( \mathbf{F}^{\text{c}} \) represents interaction forces of the colloid with the glass (grain and wall) surface or other deposited colloids. The colloid is assumed to have a radius of \( a_c = 0.5 \, \mu m \), this together with the slow Stokes flow of the solvent implies that a Stokes drag could be assumed, namely,

\[ \mathbf{F}^{\text{drag}} = \zeta (\mathbf{u}(\mathbf{Y}(t),t) - \mathbf{v}(t)), \quad \text{with} \quad \zeta \equiv 6\pi \mu a_c, \]

where \( \mathbf{u}(x,t) \) is the Eulerian solvent velocity field, \( \mathbf{Y}(t) \) is the instantaneous location of the colloid obtained from the kinematic equation \( d\mathbf{Y}(t)/dt = \mathbf{v}(t) \), and \( \mu \) is the solvent viscosity. As the first step, we neglected the effects of local fluid shear and any corrections of the viscous force due to glass cylinder surface or channel wall. Local shear flow may induce viscous force (i.e., lift force) normal to the flow [27,23]. Hydrodynamic interaction of the colloid
<table>
<thead>
<tr>
<th>Symbol</th>
<th>Physical value</th>
<th>Value in simulation</th>
</tr>
</thead>
<tbody>
<tr>
<td>water density</td>
<td>( \rho )</td>
<td>1000 kg/m^3</td>
</tr>
<tr>
<td>water viscosity</td>
<td>( \mu )</td>
<td>0.001 kg/(m.s)</td>
</tr>
<tr>
<td>grid spacing</td>
<td>( d_x, d_y )</td>
<td>2 ( \mu m )</td>
</tr>
<tr>
<td>channel width</td>
<td>( H )</td>
<td>800 ( \mu m )</td>
</tr>
<tr>
<td>periodicity length</td>
<td>( L )</td>
<td>372 ( \mu m )</td>
</tr>
<tr>
<td>glass cylinder radius</td>
<td>( R )</td>
<td>60 ( \mu m )</td>
</tr>
<tr>
<td>colloid radius</td>
<td>( a_c )</td>
<td>0.5 ( \mu m )</td>
</tr>
<tr>
<td>velocity used in setting ( F_B )</td>
<td>( U_c )</td>
<td>679.5 m/day</td>
</tr>
<tr>
<td>mean speed realized</td>
<td>( U_s )</td>
<td>2.887 m/day</td>
</tr>
<tr>
<td>porosity</td>
<td>( \varepsilon )</td>
<td>0.734</td>
</tr>
<tr>
<td>nominal flow speed</td>
<td>( U_s/\varepsilon )</td>
<td>( \sim 4.0 ) m/day</td>
</tr>
<tr>
<td>colloid density</td>
<td>( \rho_c )</td>
<td>1055 kg/m^3</td>
</tr>
<tr>
<td>time step</td>
<td>( dt )</td>
<td>( 6.4 \times 10^{-5} ) s</td>
</tr>
<tr>
<td>mass of colloid (actual)</td>
<td>( m_c )</td>
<td>( 5.524 \times 10^{-16} ) kg</td>
</tr>
<tr>
<td>mass of colloid (assumed)</td>
<td>( m_c^* )</td>
<td>( 6.033 \times 10^{-12} ) kg</td>
</tr>
<tr>
<td>response time (actual)</td>
<td>( \tau_c )</td>
<td>( 5.86 \times 10^{-8} ) s</td>
</tr>
<tr>
<td>response time (assumed)</td>
<td>( \tau_c^* )</td>
<td>( 6.4 \times 10^{-4} ) s</td>
</tr>
<tr>
<td>force</td>
<td>( F^{\text{drag}}, F^c, \text{etc.} )</td>
<td>( (1.563 \times 10^{-9})F_N )</td>
</tr>
</tbody>
</table>

with a surface can result in a modified drag, additional lift, and non-zero torque [11,25]. These modifications could be included in our Lagrangian colloid-tracking approach. Here we chose to keep the force formulation simple for the following reasons: (a) in a study conducted by Arcen et al. [2], it was shown that the results of particle statistical properties from numerical simulation based on the standard drag force only are not much different from these obtained using wall-corrected drag and lift forces; (b) computations of wall and shear corrections are computationally expensive; (c) there appear to be inconsistencies in the literature regarding the general expression of viscous force acting on a particle in a shear flow near a surface; and (d) we intend to develop a systematic understanding by gradually adding complexity to the description of hydrodynamic forces, one step at a time.

The Stokes inertial response time of the colloid \( \tau_c \equiv m_c/\zeta = 2\rho_c (a_c^2)/(9\mu) \) is about \( 5.86 \times 10^{-8} \) s, which is much smaller than the typical flow time scale. Therefore, the colloid would move along a streamline if no other forces were considered. In the numerical simulation, we assumed a value of \( \tau_c^* = 6.4 \times 10^{-4} \) s, which is several order of magnitude larger than the actual value, but is still very much smaller than the flow time scale, in order to reduce the stiffness of the system, Eq. (4), so a large integration time step can be used. Note that the typical flow time scale is on the order of \( a_c/U_s = 0.5 \) s. Numerical tests show that the results are not sensitive to the value of \( \tau_c \) as long as it is much smaller than the typical flow timescale.

The buoyancy force and body force together is given as
Fig. 3. Different colloidal interaction forces normalized by $(\zeta U_s)$ for colloid-surface interaction: (a) the region $h_0 < h < a_c$; (b) the zoom-in region $0.005a_c < h < a_c$. The thin vertical line in (a) marks the location where $h = h_0$. The three horizontal lines in (b) mark the value of 1, 0, and 1, respectively.

The Brownian force is specified as $F^B = (F^B_1, F^B_2)$, where each component $F^B_i$ is an independent Gaussian random variable of zero mean and the following standard deviation [9]

$$
\sigma_{F^B} = \sqrt{\frac{2\zeta k T}{dt}},
$$

where $dt$ is the time step size, $T$ is the temperature (assumed to be 293 K), $k = 1.38 \times 10^{-23} \text{ J/K}$ is the Boltzmann constant. When a simple explicit Euler scheme is applied, the Brownian force would generate the desired mean square value $(kT/m_c)$ of velocity fluctuation in each direction [8]. The ratio of the Brownian force to the drag force is estimated to be $\sqrt{2\zeta U_s^2/(m_c U_s^2 dt)} \approx 3.5$, implying that the Brownian effect is as important as the drag force in transporting the colloidal particles. The above treatment for the Brownian motion is based on a stochastic (Langevin equation) model. It should be noted that an alternative would be to directly introduce fluctuating hydrodynamics [21] within the lattice Boltzmann equation, as performed in [18].

While the drag force and the Brownian force are active in all regions of the flow domain, the colloidal interaction force is a short-range force that is only important when a colloid is very close to a glass surface or another deposited colloid. It consists of the electrostatic, Lifshitz-van der Waals, and Lewis acid/base interaction forces [30],

$$
F^c = F^{\text{EDL}} + F^{\text{LW}} + F^{\text{AB}},
$$
where all interaction forces are assumed to act in the direction normal to a surface, with a positive value indicating a repulsive force and negative an attractive force. The formulation of these forces is primarily based on the Derjaguin-Landau-Verwey-Overbeek (DLVO) interaction potential [7,31]. The electrostatic double layer (EDL) force results from the interaction of a charged particle with the ions in the liquid medium. For colloid-glass surface interaction, the EDL force may be written as [12,14]

\[ F_{\text{EDL,cg}} = \frac{a_c \kappa}{1 - \exp(-2\kappa h)} \left[ \alpha_1 \exp(-\kappa h) - \alpha_2 \exp(-2\kappa h) \right], \]

where \( h \) is the minimum gap between the colloid and a glass surface (the distance from the center of the colloid to the surface minus \( a_c \)), \( \kappa \) is the inverse Debye-Huckel screening length which depends on the solution ionic strength. Here we shall only consider an ionic strength at 100 mM in NaCl electrolyte solution, and in this case \( 1/\kappa = 0.963 \text{nm} \). The coefficients \( \alpha_1 \) and \( \alpha_2 \) are related to the surface potentials of the glass surface \((-69.74 \text{mV})\) and colloid \((-76.99 \text{mV})\) as well as the dielectric constant of the medium [12]. The surface potentials are computed based on the measured \( \zeta \)-potentials \((-41.31 \text{mV} \text{ and } -45.56 \text{mV}, \text{respectively})\) [12]. For the electrolyte solution used in this study [12], \( \alpha_1 = 4.648 \times 10^{-11} \text{ N} \text{ and } \alpha_2 = 4.671 \times 10^{-11} \text{ N} \). With the above parameters, the EDL force is repulsive and \( F_{\text{EDL}}/(\zeta U_s) > 1 \) when \( h/a_c < 0.02 \) (Fig. 3). For the case of colloid-colloid interaction at 100 mM ionic strength, the EDL force is computed by [12,8]

\[ F_{\text{EDL,cc}} = (1.847 \times 10^{-11} N)a_c \kappa \exp(-\kappa h), \]

where \( h \) is the minimum gap between the two approaching colloids.

The attractive Lifshitz-van der Waals (LW) interaction accounts for intermolecular interaction including London dispersion, Keesom dipole-dipole, and Debye induction. For the purpose of this paper, the LW force can be written as [12,30]

\[ F_{\text{LW,cg}} = -\beta a_c \left( \frac{h_0}{h} \right)^2, \]

where \( h_0 \) is an equilibrium distance used to model the occurrence of physical contact and is set to 0.157 nm [8]. The constant \( \beta \) has been determined to be 0.0434 J/m² for colloid-surface interaction and 0.0469 J/m² for colloid-colloid interaction, based on the thermodynamic parameters of colloids, glass, and the liquid solution.

The Lewis acid/base (AB) force originates from the bonding reaction of a Lewis acid and a Lewis base. It can be expressed as [12,30]

\[ F_{\text{AB}} = -\gamma \frac{h_0 a_c}{\chi} \exp \left( \frac{h_0 - h}{\chi} \right), \]

where \( \chi = 0.6 \text{nm} \) is the water decay length [30], the constant \( \gamma \) is determined, using the relevant electron-acceptor and electron-donor parameters, to be \(-0.0322 \text{ J/m}^2\) and \(-0.170 \text{ J/m}^2\) for colloid-surface and colloid-colloid interactions, respectively.

In Fig. 3, we compare the three different colloidal interactions forces and also their magnitudes relative to the drag force, for the case of colloid-glass surface interaction. Several important observations can be made. First, as a colloid approaches a surface, the Lifshitz-van der Waals attractive force is the first active force and it plays a role starting at \( h \approx 0.2a_c \). This attractive LW force dominates the colloidal interaction until \( h \approx 0.02a_c \), with a peak magnitude at least 10 times the drag force. Then the EDL force quickly takes over to turn the net interaction force a repulsive force. An important parameter here is \( \kappa a_c = 519.2 \), implying that the EDL force will play a role when the gap is on the order of \( a_c/519.2 = 0.00193a_c \). This scale indeed falls in the range when EDL force is the dominant force. Eventually at \( h/a_c \approx 0.0004 \), the LW force wins over to change the net force back to an attractive force (i.e., towards the primary energy minimum). This overall picture implies that the LW force is the dominant force acting over most of the small gap distances. The two locations where the net force is zero, namely, \( h/a_c = 0.000383 \) and \( h/a_c = 0.0152 \), correspond to the secondary minimum and the energy barrier in the net energy potential [8].
Fig. 4. Different colloidal interaction forces normalized by $(\zeta U_s)$ for colloid-colloid interaction: (a) the region $h_0 < h < a_c$; (b) the zoom-in region $0.005a_c < h < a_c$. The thin vertical line in (a) marks the location where $h = h_0$. The three horizontal lines in (b) mark the value of $-1, 0, 1$, respectively.

Similar plots are shown in Fig. 4 for the interaction of a colloid with another deposited colloid. In this case, the net interaction force at $h = h_0$ is repulsive, implying the surface location where a colloid has been deposited will not attract a second colloid. The net force variation near the secondary minimum is similar.

In our simulation, both the grain and wall surface are treated as a flat glass surface. For a given colloid particle, all possible colloid-surface and colloid-colloid binary pair interactions are summed to obtain the final $F^c$. Such pairwise summation of all the binary interactions is a reasonable approach since here $\kappa a_c >> 1$ [6]. The equation of motion was solved numerically by first integrating the drag force and colloidal force using a mixed fourth-order Adam-Bashforth and Adman-Moulton scheme. The Brownian force was then added using the explicit Euler scheme.

3 Results

3.1 Viscous flow simulation

Before discussing the results for the flow problem shown in Fig. 1, we shall validate both our LBM and Physalis codes by a unit cell flow problem in which a fixed cylinder of radius $a$ is located at the center of a two-dimensional channel with walls at $x = 0$ and $x = H$. Periodic flow condition is assumed in the $y$ direction with periodicity length of $L$. Both channel walls are moving at a constant velocity of $U$. The force $F_y$ acting on the cylinder in the flow direction is of interest. This simulates a problem of viscous flow over a line of cylinders with spacing equal to $L$, driven by two moving walls both parallel to the line and located $H/2$ away from the line. Clearly, the normalized force $F_y/(2\pi\rho U^2 a)$ is a function of three dimensionless parameters: the Reynolds number $Re = \rho U (2a)/\mu$ and the two geometric scale ratios $L/(2a)$ and $L/H$, where $\rho$ and $\mu$ are fluid density and viscosity. Here we set $Re = 1$ and
Table 2
Comparison of the normalized force acting on a fixed cylinder in a unit cell flow driven by two moving walls.

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<thead>
<tr>
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<tbody>
<tr>
<td>A</td>
<td>11.8</td>
<td>1.033</td>
<td>1.033</td>
<td>1.034</td>
<td>1.053</td>
<td>0.966</td>
</tr>
<tr>
<td>B</td>
<td>6.1</td>
<td>1.241</td>
<td>1.243</td>
<td>1.224</td>
<td>1.251</td>
<td>1.158</td>
</tr>
<tr>
<td>C</td>
<td>2.09</td>
<td>2.098</td>
<td>2.083</td>
<td>2.079</td>
<td>2.093</td>
<td>2.067</td>
</tr>
</tbody>
</table>

Fig. 5. Comparison of results on the normalized volumetric flux as a function of time.

$L/H = 1$. The resulting normalized forces are shown in Table 2 for three different $L/(2a)$ ratios. In our simulations, the unit cell is discretized at a mesh resolution of 144 × 144, 64 × 64, and 24 × 24 for cases A, B, C, respectively, which corresponds to a very moderate resolution of the cylinder radius at $a/\delta x = 6.102, 5.246, 5.742$, for the three cases, respectively. The density and the mesh spacing are assumed to be one for all runs. For the LBM simulations, the relaxation time is set to $\tau = 1$ so the kinematic viscosity is $\mu/\rho = 1/6$. For Physalis, the wall velocity $U$ is set to 0.1. Only the force at the steady state is presented here. Table 2 shows that our LBM and Physalis results are always within 1% to each other. Our results are also in very good agreement with the previous results of Inamuro et al. [15] and Zhang & Prosperetti [33].

We now consider the flow problem shown in Fig. 1. Both LBM and Physalis codes were developed to simulate this flow. The steady-state mean flow speed $U_c$, defined as the volumetric flow rate per unit depth normalized by $H$, is much smaller than $U_c$, and is found to be 0.00425$U_c$. Therefore, the flow Reynolds number based on the mean speed is $U_cH/\nu = 8.51e - 4$ or the flow in the microchannel is essentially a Stokes flow. It is then expected that the flux is linearly related to the applied forcing.

Fig. 5 compares the resulting flow volumetric flux normalized by 0.01$HU_c$. The parameters used in Physalis are $U_c = 8.01e - 4, \rho = 1, \nu = 0.8$, and the time step size $dt = 10$, while in the LBM simulation, $U_c = 4.171875e - 5, \rho = 1, \nu = 0.0416667$ (or $\tau = 5/8$), and $dt = 1$. These settings imply that one Physalis time step corresponds to roughly 192 LBM time steps, although we can adjust the LBM parameters to improve the efficiency of the LBM.
simulation (see below). During the transient development, the flux increases monotonically with time, reaching to its steady-state value at a nondimensional time of about $tU_c/H = 0.001$. The steady state value for LBM is only 1% different from the Physalis result. This comparison between LBM and Physalis is obtained when the truncation order for the local Stokes flow representation in Physalis is set to 6 (or a total of 26 expansion coefficients are considered, see [33] for detail). It was also found that even with a low truncation order of 2 in Physalis (i.e., 10 expansion coefficients), the steady-state flux is only 2% different from the IBM result. This shows that, on the one hand, the local flow structure near the cylinder surface is relatively smooth, but at the same time, the interaction between the cylinders and with the channel walls induces secondary higher-order corrections. The Stokes flow truncation order at 6 will be assumed for all the comparisons below.

In Table 3, we compare forces and torques acting on each cylinder as labeled according to Fig. 1. The forces are normalized by $F_0 = F_{BLHL}$ and torques by $F_0a$. Due to the symmetry of the geometric arrangement, only results for cylinders 1, 2, 4, and 5 are shown. For most of the cases, the relative differences between LBM and Physalis results are less than 1%. The results show that cylinder 1 experiences clockwise torque but cylinders 4 and 5 counterclockwise torques. The transverse (lift) force on cylinder 4 is the largest due to its close proximity to the wall, it also has the largest $x-$component force (drag) due to the combination of the flow blocking and the wall effect. We further confirm that the net force acting on fluid at the steady state, namely, the sum of reacting forces from the walls and the cylinders, and the applied body force is identically zero. The normalized total tangential force acting on each wall is 0.0370 and 0.0372 for the LBM and Physalis runs, respectively.

Figs. 6 and 7 display velocity distributions at three line cuts marked in Fig. 1. For the streamwise velocity component, the velocity profiles are essentially identical at all three locations inside the fluid regions. It is noted that Physalis produces flows within the cylindrical particle surfaces, while in LBM no flow is computed within the cylinders. The transverse velocity also matches precisely at the AA cut where its magnitude is comparable to the streamwise component. For the other two cuts, the transverse velocity is only about one fiftieth the magnitude of the streamwise velocity. Some small differences are visible in the fluid regions, but perhaps are not important due to its very small magnitude. Note that both LBM and Physalis have roughly a second-order accuracy in spatial representation.

In the above comparisons, the actual number of time steps used in LBM is 16,000 while only 80 time steps are used in Physalis. For a given flow geometry and grid resolution, the flow Reynolds number is the only remaining governing parameter. One could therefore increase both $U_c$ and $v$ proportionally in LBM to effectively reduce the number of time steps needed to produce the same macroscopic behavior (governed by the dimensionless time $tU_c/H$). Taking the base case $U_c$ value, we performed four additional runs at the same flow Reynolds number by increasing $U_c$ by a factor of 2 (run2, $\omega = 1.333$), 4 (run3, $\omega = 1.0$), 16 (run5, $\omega = 0.40$), and 32 (run6, $\omega = 0.222$). The dimensionless flux curves are shown in Fig. 8(a) for the single-relaxation time BGK collision model and in Fig. 8(b) for the multiple relaxation time model of Lallemand and Luo [19]. Here $\omega$ is the relaxation frequency and

<table>
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<th>Physalis $\tilde{F}_x$</th>
<th>LBM $\tilde{F}_y$</th>
<th>Physalis $\tilde{F}_y$</th>
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Fig. 6. Comparison of steady-state, streamwise velocity profiles from LBM and Physalis at three locations shown in Fig. 1. The horizontal lines mark the glass-bead boundaries when the $y$ location cuts through glass beads. (a) AA cut; (b) BB cut; (c) CC cut.

Fig. 7. Comparison of steady-state, transverse velocity profiles from LBM and Physalis at same three locations shown in Fig. 4: (a) AA cut; (b) BB cut; (c) CC cut.

is equal to $1/\tau$ in the BGK model or equal to the relaxation parameter for the stress tensor in the MRT model [19]. The relaxation parameters for other moments in the MRT collision model follow the suggested values in [19]. Several interesting observations can be made. For the steady-state flow, the BGK model produces the correct result when $\omega > 1$ or there is a sufficient number of time steps (about 1000) for the flow to evolve to steady state. On the other hand, the MRT moment-space collision model can always produce the correct steady-state even when the transient behavior is not correct, showing an desirable advantage of MRT over BGK. During the transient time period, care must be taken to properly set the relaxation parameter in LBM, as otherwise non-physical oscillatory
Fig. 8. The normalized flux as a function of time from LBM runs at different parameter settings but the same flow Reynolds number \((U_c H/\nu = 0.2)\). Run2: \(\nu = 0.08333, \ U_c = 8.34e-5, \ \text{and} \ T = 8,000\); Run3: \(\nu = 0.1667, \ U_c = 1.67e-4, \ \text{and} \ T = 4,000\); Run5: \(\nu = 0.6667, \ U_c = 6.675e-4, \ \text{and} \ T = 1,000\); Run6: \(\nu = 1.3333, \ U_c = 1.335 - 3, \ \text{and} \ T = 500\). Here \(T\) is the total number of time steps used. (a) BGK collision model; (b) the MRT model of Lallemand and Luo [19].

Fig. 9. Two snapshots of vorticity visualization for a test run at Reynolds number of 1,429 based on the steady-state average streamwise velocity \((U_s = 0.014295)\). The parameters are \(U_c = 15.0\) and \(\nu = 0.004\).

behaviors can occur, due to the contamination of hydrodynamic modes by inherent acoustic waves in the LBM approach [19,24].

While our current application here only concerns low Reynolds number viscous flows, our MRT LBM method can simulate a variety of flow Reynolds numbers. Fig. 9 demonstrates this capability with the same lattice
Fig. 10. A 3D geometric configuration with 25 nearly packed spherical glass beads in a 3D channel with a square cross section: (a) 3D view, (b) xy view; (c) xz view.

Fig. 11. Simulated velocity field on two plane sections shown Fig. 10, with LBM approach. Similar results are obtained with Physalis. (a) AA plane cut; (b) BB plane cut.

resolution, for a flow at a Reynolds number of about $U_sH/\nu = 1.429$. Interestingly, at this Reynolds number, the long-time flow is actually unsteady due to quasi-periodic vortex sheddings and the complex interactions of vortices with cylinder surfaces and channel walls. Only a half of the flow domain is shown in Fig. 9. The two time snapshots in Fig. 9 are both taken at the quasi-steady stage, but locally they have quite different vorticity distributions.
We now briefly discuss some preliminary results in three dimensions for a porous channel with a square cross-section, filled with 25 spherical glass beads almost packed in the channel, as illustrated in Fig. 10. A moderate lattice resolution of 80 by 35 by 80 is used. Fifteen beads are located at \( y = 10 \), with \( x, z = 10, 30, 50, 70 \). Another nine beads are located at \( y = 25 \), with \( x, z = 20, 40, 60 \). The diameter of glass beads is set to 20. The flow is driven by a body force of magnitude equal to \( 8\rho \nu U_c/H^2 \) where \( H = 80 \). The magnitude of \( U_c \) is set to give \( U_c H/\nu = 288 \).

Fig. 11 shows the velocity distributions in two plane cuts. One can see a complex three-dimensional flow with a large variation of local velocity magnitudes and directions, implying strong streamline curvatures in the flow. The effect of glass bead surfaces is well represented. The steady-state mean flow speed is found to be roughly 0.0005\( U_c \). A quantitative analysis and comparison between LBM and Physalis for this case is being undertaken and will be reported separately.

### 3.2 Preliminary results on colloid transport and deposition

In this section, results on the transport of colloids in the 2D porous channel with 7 fixed cylinders are presented. The flow may be extended in the \( y \) direction indefinitely using the periodicity condition to allow a colloid to travel any length in the \( y \) direction. We used bilinear interpolation to obtain the fluid velocity at the location of a colloid. To ensure the accuracy of simulated colloid trajectories, we purposely doubled the grid resolution to 400 \( \times \) 186 (see Table 1), although this is not necessary for the flow simulation. Fig. 12 displays the locations of colloids within a same region from the inlet at a time when a total of 1,898 colloids have been released into the channel, for four different mean flow speeds ranging from 1 to 8 m/day. The solution ionic strength is set to 0.1 M in these simulations. Suspended colloids are marked as open squares while deposited colloids are shown as filled squares. Since the colloid concentration is fixed at 1 ppm, a fixed total number of injected colloids corresponds to a same total solvent volume passing through the inlet. The numbers of deposited colloids at the times shown in Fig. 12 are 508, 358, 231, and 105 for 1 m/day, 2 m/day, 4 m/day, 8 m/day, respectively. These results show that the overall colloidal deposition decreases with increasing flow speed.

The results were further analyzed quantitatively by calculating and comparing the average surface coverage, defined as the fraction of the solid surface covered by deposited colloids. Fig. 13 shows the surface coverage as a function of the total number of injected colloids. The total number \( N(t) \) of injected colloids is proportional to the time \( t \) as \( N(t) = \theta t \) (s), where the coefficient \( \theta \) represents the number of colloids released per unit time and is 0.737, 1.474, 2.949, and 5.899 s\(^{-1}\) for the 4 flow speeds. The surface coverage has a quadratic dependence on the total number of injected colloids since the net rate of deposition increases roughly linearly with the injection time or the total number of injected colloids. The observed dependence on the mean flow speed may be qualitatively explained as follows. While fluid flow transports colloids along curved streamlines, Brownian motion can cause colloids to cross streamlines. The lower the mean flow speed, the longer it takes for a colloid to move through a given distance in the \( y \) direction. This then increases the possibility for Brownian force to shift the colloid across streamlines and bring the colloid to regions very close to a solid surface where the surface interaction forces can act to remove the colloid from the flow.

Another analysis is that we counted the number of colloids deposited on each cylinder including its periodic images. It appears that more colloids are deposited on the cylinders near the center of the channel (Cylinders 2, 5, and 6; see Fig. 1) for high flow speeds (4 m/day and 8 m/day). But at low flow speeds (1 m/day and 2 m/day), there is no detectable bias. This shows that the channel walls can have an effect on the deposition at high flow speeds. It should be noted that there are 170, 122, 74, 25 colloids deposited on the channel walls for the 4 flow speeds, corresponding to 33.5\%, 34.1\%, 32.0\%, and 23.8\% of total deposited colloids, respectively. This shows that the percentage of deposition on channel walls also decreases with flow speed when the flow speed is high. There are large statistical uncertainties on the data shown in Table 4, as judged by the non-symmetric distribution of the deposited colloids. Therefore, the conclusion here needs to be checked with longer simulations.
We have also considered the effect of the solution ionic strength on the surface coverage and found a very nonlinear dependence of surface coverage on the ionic strength [12]. A thorough analysis of surface coverage at different flow speed and solution ionic strength has been presented in our companion paper [12], where we also show that our own experimental observations using confocal microscopy confirm the simulated dependence of colloid deposition on flow speed and ionic strength.
4 Conclusions

In this paper, we have simulated viscous flows in a model porous medium, using two different computational approaches. The fact that the mesoscopic lattice Boltzmann approach and a macroscopic CFD approach produce almost identical velocity profiles shows that both approaches are capable of handling this model geometry. Our next step will be to establish a quantitative agreement between LBM and Physalis for a realistic porous medium in three dimensions. It is believed that cross validations of two completely different computational approaches will partially eliminate the need to perform pore-scale flow measurements.

The mesoscopic lattice Boltzmann approach is perhaps a better choice due to its simplicity in treating the no-slip boundary condition on a solid surface and its potential to extend to arbitrary flow geometry, provided that the parameters in LBM are set properly such that the results are not contaminated by acoustic waves or other numerical instabilities. The generalized LBE or multiple-relaxation-time collision model is shown to be more robust in simulating steady-state viscous flows. Further research is needed to gain a theoretical understanding of acoustic waves in LBM for complex geometries.

A Lagrangian modeling approach for colloid transport has been developed to study the deposition of colloids
on solid surface. The key finding is that the rate of deposition depends on the flow speed and solution ionic strength, and the nature of the dependence agrees qualitatively with previous observations and our visualizations using confocal microscopy [12]. This demonstrates the feasibility of our computational model as a quantitative research tool and its potential for revealing transport mechanisms at the pore scale. We intend to develop this model further to include other hydrodynamic interaction forces and three-dimensional flow effects, so a quantitative comparison with pore-scale experimental observations can be made possible. It is important to note that, because typically $\kappa a >> 1$, the hydrodynamic interaction forces become active far before the electrostatic double layer interaction force.

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References