Investigation of gravity effect on biocolloid and colloid transport through water-saturated porous media

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Abstract
The role of gravitational force on biocolloid and colloid transport in water-saturated columns packed with glass beads was investigated. Transport experiments were performed with biocolloids (bacteriophages: ΦX174, MS2) and colloids (clays: kaolinite KGa-1b, montmorillonite STx-1b). The packed columns were placed in various orientations (horizontal, vertical, and diagonal) and a steady flow rate of Q=1.5 mL/min was applied in both up-flow and down-flow modes. All experiments were conducted under electrostatically unfavorable conditions. The experimental data were fitted with a newly developed, analytical, one dimensional, colloid transport model, accounting for gravity effects. The results revealed that flow direction has a significant influence on particle deposition. The rate of particle deposition was shown to be greater for up-flow than for down-flow direction, suggesting that gravity was a significant driving force for biocolloid and colloid deposition.

Keywords: gravity effects, ΦX174, MS2, clay minerals, KGa-1b, STx-1b, biocolloid transport

1. INTRODUCTION

The transport of colloids and biocolloids (e.g. viruses, bacteria) in porous and fractured media has long been recognized to be of considerable importance to a number of environmental practical applications, including groundwater pollution by microbial pathogens, in situ bioremediation of contaminated aquifers, and granular filtration of water and wastewater. Numerous investigators have examined theoretically and experimentally the various factors that affect colloid and biocolloid transport in porous and fractured media, especially the effects of interstitial velocity [1], colloid particle size [2], collector size [1,3], solid matrix porosity [4], collector roughness [5], ionic strength [6], water chemistry [7], gravitational settling [2], and presence of suspended clays [8].
The effect of flow direction on colloid or biocolloid fate and transport in porous media has received relatively minor attention. Laboratory bench scale experiments are traditionally conducted with flow direction orthogonal to gravity (horizontal flow) [9], whereas packed column experiments are carried out with flow orthogonal to gravity [1], against gravity (up-flow) [3, 7]), or in the direction of gravity (down-flow) ([10]). However, the majority of the published studies do not report the flow direction used, and either neglect or regard insignificant the influence of gravitational settling, which is potentially a significant retention mechanism. The Happel sphere-in-cell [11] and hemispheres-in-cell [4] are widely employed models for colloid retention during transport in porous media. Both models behave similarly for favorable deposition conditions. However, the hemispheres-in-cell model incorporates a grain-to-grain contact, which allows for colloid retention in the presence of energy barriers (unfavorable conditions). This is the main advantage of the hemispheres-in-cell model [4]. Note that based on the Happel sphere-in-cell model, zero retention is expected in the presence of energy barriers; however, experimental evidence indicates that some colloids are retained despite the unfavorable conditions.

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There are numerous mathematical models available that describe colloid and biocolloid transport in fractured and porous media. These models rely on either continuum or statistical approaches. Continuum approaches are based on macroscopically derived conservation equations and do not consider the morphology of the pore space within the solid matrix. In this study, the frequently employed continuum approach was adopted, and the phenomenological colloid transport model developed by Sim and Chrysikopoulos [12] was extended to account for colloid sedimentation. The present study aims to improve our understanding of how the flow direction relative to gravity influences colloid and biocolloid fate and transport in porous media.

2. EXPERIMENTAL APPROACH

2.1 Biocolloid and Clay colloid Selection and Preparation

The bacteriophage MS2 (F-specific, single-stranded RNA phage with 31% nucleic acid content, whose host bacterium is *E. coli* ATTC 15597-B1) and ΦX174 (icosahedral, single-stranded DNA phage with 26% nucleic acid content, whose host bacterium is *E. coli* ATTC 13706-B1) were used in this study as surrogates for human viruses. Both bacteriophage were assayed by the double-layer overlay method.

The clays used in this study were kaolinite (KGa-1b, is a well-crystallized kaolin from Washington County, Georgia) and montmorillonite (STx-1b, a Ca-rich montmorillonite, white, from Gonzales County, Texas), purchased from the Clay Minerals Society, Columbia, USA. The optical density of the clay colloids was analyzed at a wavelength of 280 nm by a UV-vis spectrophotometer, and the corresponding clay concentrations were determined twice as outlined by Chrysikopoulos and Syngouna [13].

2.2. Column Experiments

Flow through experiments were carried out with ΦX174 and MS2 bacteriophages as model biocolloids, and KGa-1b and STx-1b as clays as model colloids in a 2.5 cm diameter and 30 cm long Chromaflex glass column packed with 2 mm in diameter glass beads (Fisher Scientific, New Jersey). Glass beads were chosen as model porous media because they are chemically non-reactive with the solutions used in this study. Prior to the experiments, the beads were cleaned and dried in an oven at 105°C and then stored in screw cap sterile beakers until use in the column experiments. The column was packed with glass beads under standing ddH_2O to minimize air entrapment. The estimated dry bulk density was 1.61 g/cm3, and the porosity was 0.42. Constant flow of sterile ddH_2O at flow rate of Q=1.5 mL/min, corresponding to pore water velocity of U=0.74 cm/min, was maintained through the packed column with a peristaltic pump. For each experiment, three pore volumes of the clay colloid or the bacteriophage suspension was injected into the packed column, followed by three pore volumes of ddH_2O. One set of flow through experiments was performed with virus and clay colloid individual transport in horizontal (β=0°), a second set in vertical (β=±90°), and a third set in inclined (β=±45°) columns. For all sets, three pore volumes of solution was injected into the packed column, followed by three pore volumes of ddH_2O.

3. MATHEMATICAL DEVELOPMENT

3.1 Transport of Dense Colloids

Based on the continuum approach, the transport of dense biocolloids in one-dimensional, homogeneous, water saturated porous media with first-order attachment (or filtration) and inactivation, assuming that an effective velocity term accounts for both the interstitial as well as the particle settling velosity is governed by the following partial differential equation:

$$\frac{\partial C(t,x)}{\partial t}+\frac{\rho_b}{\theta}\frac{\partial C^*(t,x)}{\partial t}=D\frac{\partial^2 C(t,x)}{\partial x^2}-U_{tot}\frac{\partial C(t,x)}{\partial x}-\lambda C(t,x)-\lambda^*\frac{\rho_b}{\theta}C^*(t,x) \quad (1)$$

where C is the concentration of biocolloids in suspension; C* is the concentration of biocolloids attached on the solid matrix; t is time; ρb is the bulk density of the solid matrix; θ is the porosity of the porous medium; λ is the transformation rate constant of biocolloids in solution (e.g., inactivation of suspended viruses); λ* is the transformation rate constant of attached biocolloids; and Utot is the total (or effective) particle velocity, which for biocolloids subject to gravitational forces accounts for gravitational settling:

$$U_{tot}=U+U_s \quad (2)$$

where U is the interstitial velocity, and U_s is a modified version of the traditional "free particle" settling velocity in static water columns to "restricted particle" settling in granular porous media under directional flow conditions:

$$U_s=-f_s\frac{(\rho_p-\rho_w)d_p^2}{18\mu_w}g_{(i)} \quad (3)$$

where f_s is the correction factor accounting for particle settling in the presence of the solid matrix of granular porous media, ρ_w and ρ_p are the densities of the suspending fluid (water) and the colloid particle, respectively; μ_w is the dynamic viscosity of water, and g(i) accounts for the directional interstitial flow as follows:

$$g_{(i)}=g_{(-z)}\sin\beta\ \mathbf{i} \quad (4)$$

where g(-z) is the acceleration due to gravity in the negative z-direction (indicated by the subscript in parentheses), β is the angle of the main flow direction with respect to the horizontal x-direction, and i is the unit vector parallel to the flow (see definition sketch in Figure 1, also note that for up-flow 0°<β<90° whereas for down-flow -90°<β<0°); and D is the hydrodynamic dispersion coefficient. The correction factor f_s converts the average free particle sedimentation velocity to the average sedimentation velocity through water saturated porous media [14].

It should be noted that the governing biocolloid transport equation (1) is essentially the biocolloid transport model provided by Sim and Chrysikopoulos [12] with U replaced by U_{tot}. Also, (3) is the balance among gravity, buoyancy, and viscous forces, implicitly assumes that the colloids are small, uniform spheres, and there is a distinct density difference between the biocolloids and the suspending fluid. The rate of biocolloid attachment onto the solid matrix is described by a first-order equation [15, 16]. The various model parameters can be estimated by fitting the analytical solution to the experimental data with the nonlinear least squares regression package "Colloidfit" [12], which can be obtained for free from the authors upon request.

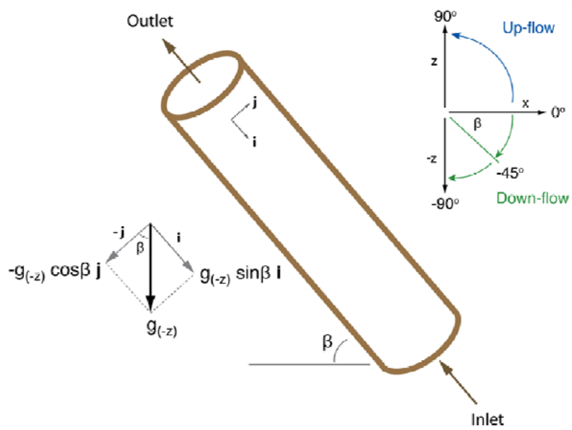

Figure 1. Schematic illustration of a packed column with up-flow velocity having orientation (-**i**) with respect to gravity. The gravity vector components are: $g_{(\mathbf{i})}= g_{(-z)}\sin\beta\ \mathbf{i}$, and $g_{(-\mathbf{j})}= -g_{(-z)}\cos\beta\ \mathbf{j}$.

4. RESULTS AND DISCUSSION

The normalized ΦX174 and MS2 flowthrough experimental data are presented in Figure 2, together with the fitted model predictions. The parameters Utot, D and kc were estimated by fitting the analytical solution to the experimental ΦX174 and MS2 breakthrough concentrations. From the fitted U_{tot} values, the fitted restricted particle setling velocity was easily obtained as Us=Utot-0.74 cm/min, because for all experiments of this study U was fixed at 0.74 cm/min. In a similar fashion, the KGa-1b and STx-1b flowthrough experimental data were fitted with the nonlinear least squares regression package "Colloidfit". The experimental data together with the fitted concentration history are presented in Figure 3.

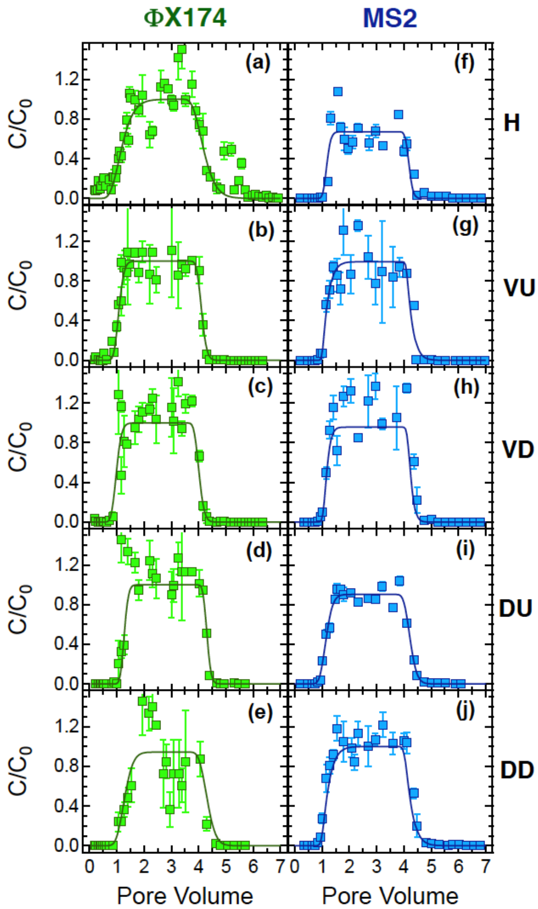

Figure 2. Experimental data (symbols) and fitted model simulations (curves) of (a-e) ΦX174 and (f-j) MS2 breakthrough in columns packed with glass beads with (a,f) horizontal, (b,g) vertical up-flow, (c,h) vertical down-flow, (d,i) diagonal up-flow, and (e,j) diagonal down-flow directional flow conditions.

With the exception of the case for ΦX174 with horizontal flow conditions, all fitted U_s values followed the theoretical trend suggested by (3) (U_s=0 for horizontal flow, Us>0 for down-flow, and U_s<0 for up-flow). Note that the absolute Us values for the clays were much higher than those for the bacteriophages owing to their lager size and density. Based on the fitted k_c values, particle attachment is generally higher for up-flow than down-flow experiments. The fitted D values are smaller for the smaller (bacteriophage) than the larger (clay) particles, suggesting that the dispersivity is increasing with particle size.

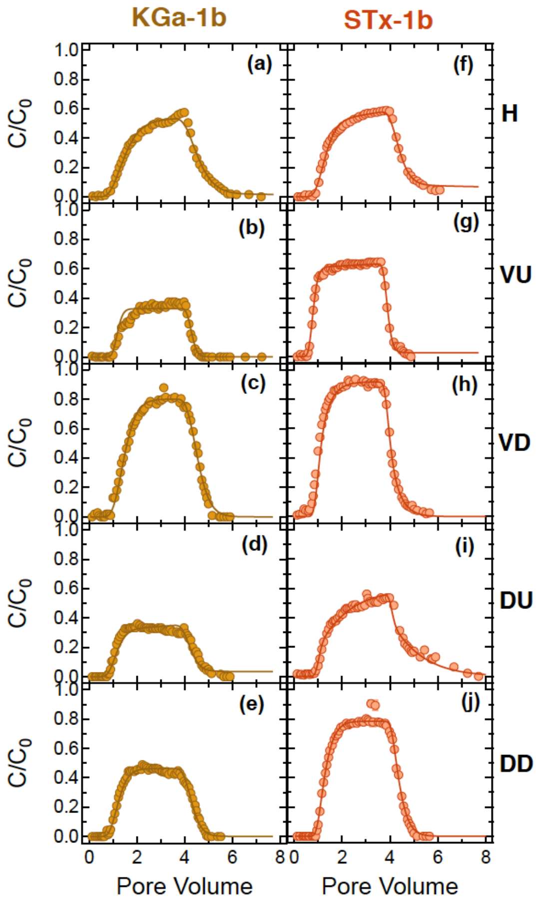

Figure 3. Experimental data (symbols) and fitted model simulations (curves) of (a-e) KGa-1b and (f-j) STx-1b breakthrough in columns packed with glass beads with (a,f) horizontal, (b,g) vertical up-flow, (c,h) vertical down-flow, (d,i) diagonal up-flow, and (e,j) diagonal down-flow directional flow conditions.

The calculated mass recovery values indicated that, with the exception of the case for MS2 with horizontal flow conditions, there was very little virus retention in the packed columns. The Mr values suggested that, with the exception of the case for ΦX174 with diagonal column orientation, there was more mass retained in the columns under upward than downward flows. However, significant amount of both clays were retained in the packed columns. For most cases examined, higher mass recoveries were observed for both clays for the vertical than diagonal column orientation, and for downward than upward flows. Velocity enhancement was observed for both biocolloids, with slightly higher values for down-flow than up-flow experiments. Moreover, it was shown velocity enhancement for both clay colloids and most of the flow conditions considered in this study.

5. SUMMARY

A mathematical model to describe colloid transport in homogeneous, water saturated porous media, accounting for the gravitational force was presented. The colloid transport model successfully matched the bacteriophage as well as the clay particles breakthrough data. The fitted Us values were positive for up-flow and negative for down-flow experiments, as suggested by theory. The experimental results verified the theoretical prediction that gravity is a significant force for colloid transport in water saturated porous media under conditions unfavorable to deposition, especially for the larger and denser particles. It was shown that particle attachment is generally higher for up-flow than down-flow experiments, and that more mass is retained in porous media under upward than downward flows. Finally, it was shown that biocolloid and colloid particle velocities were enhanced compared to the tracer for most of the cases considered in this study.

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