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Experimental investigation of virus and clay particles cotransport in partially saturated columns packed with glass beads

Vasiliki I. Syngouna [a], Constantinos V. Chrysikopoulos [b,*]

[a] Environmental Engineering Laboratory, Civil Engineering Department, University of Patras, Patras 26500, Greece
[b] School of Environmental Engineering, Technical University of Crete, Chania 73100, Greece

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ABSTRACT

Suspended clay particles in groundwater can play a significant role as carriers of viruses, because, depending on the physicochemical conditions, clay particles may facilitate or hinder the mobility of viruses. This experimental study examines the effects of clay colloids on the transport of viruses in variably saturated porous media. All cotransport experiments were conducted in both saturated and partially saturated columns packed with glass beads, using bacteriophages MS2 and ΦX174 as model viruses, and kaolinite (KGa-1b) and montmorillonite (STx-1b) as model clay colloids. The various experimental collision efficiencies were determined using the classical colloid filtration theory. The experimental data indicated that the mass recovery of viruses and clay colloids decreased as the water saturation decreased. Temporal moments of the various breakthrough concentrations collected, suggested that the presence of clays significantly influenced virus transport and irreversible deposition onto glass beads. The mass recovery of both viruses, based on total effluent virus concentrations, was shown to reduce in the presence of suspended clay particles. Furthermore, the transport of suspended virus and clay-virus particles was retarded, compared to the conservative tracer. Under unsaturated conditions both clay particles facilitated the transport of ΦX174, while hindered the transport of MS2. Moreover, the surface properties of viruses, clays and glass beads were employed for the construction of classical DLVO and capillary potential energy profiles, and the results suggested that capillary forces play a significant role on colloid retention. It was estimated that the capillary potential energy of MS2 is lower than that of ΦX174, and the capillary potential energy of KGa-1b is lower than that of STx-1b, assuming that the protrusion distance through the water film is the same for each pair of particles. Moreover, the capillary potential energy is several orders of magnitude greater than the DLVO potential energy.

1. Introduction

Several theoretical and experimental investigations have shown that suspended mobile colloids can either facilitate or hinder the mobility of various contaminants in water saturated porous and fractured media [1–11]. However, colloid facilitated virus transport in unsaturated porous media is substantially different and more complex than that in saturated porous media. In addition to the retention mechanisms governing colloid and virus transport in saturated porous media (e.g., pore straining and attachment onto solid-water interfaces (SWI)), colloids in unsaturated porous media can be retained at air–water interfaces (AWI), in thin water films (film straining), and air–water–solid (AWS) interfaces [12–26]. Furthermore, in unsaturated porous media, colloids and viruses can also be retained in air–water meniscus–solid (AW_mS) interfaces [27,28]. Note that AW_mS interfaces are essentially areas where significant colloid attachment occurs and water meniscuses diminish to thin water films.

Capillary forces are known to be important for colloid attachment at the AWI as well as for film straining [12,14,25,29,30]. Derjaguin–Landau–Verwey–Overbeek (DLVO) interactions cannot always explain the observed colloid deposition in unsaturated porous media [17,30], especially at the contact line of the AW_mS interface. Colloid retention of the AW_mS interface is explained more convincingly by capillary force interactions [23,24,31]. Despite of these and other related research efforts, the role of capillary forces on colloid retention in unsaturated porous media is not fully understood and deserves more attention.

The objective of this paper was to explore further the specific interactions of simultaneously transported colloids and viruses with the various interfaces (SWI, AWI, and AWS) present in

* Corresponding author.
E-mail address: cvc@enveng.tuc.gr (C.V. Chrysikopoulos).

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unsaturated porous media. Also, the synergistic effects of suspended clay colloids and water saturation level on the attenuation and transport of viruses in unsaturated porous media is examined. Furthermore, the surface properties of viruses, clays, and glass beads are used to construct classical DLVO and capillary potential energy profiles, which are evaluated.

2. Materials and experimental procedures

2.1. Bacteriophages and assay

The bacteriophage MS2 (F-specific single-stranded RNA phage with effective particle diameter ranging from 24 to 26 nm), and ΦX174 (somatic single-stranded DNA phage with effective particle diameter ranging from 25 to 27 nm) were used as model viruses. Both bacteriophages are infecting *E. coli,* and were assayed by the double-layer overlay method [32], as outlined by Syngouna and Chrysikopoulos [33]. Viruses attached onto clay particles were separated from viruses suspended in the liquid phase by centrifugation at 2000*g* for 30 min [10]. The suspension of unattached viruses in the supernatant was pipetted out, and the concentration of the suspended viruses was determined. The absence of clay colloids in the supernatant was verified by a UV–vis spectrophotometer (UV-1100, Hitachi) at a wavelength of 280 nm. The concentration of attached viruses was determined by subtracting the mass of viruses that remained in suspension from the initial virus concentration in each sample.

2.2. Clays

The clays used in this study were kaolinite (KGa-1b, 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 < 2 μm colloidal fraction of each clay mineral suspension in sterile ddH_2O was separated by sedimentation and was purified following the procedure described by Rong et al. [34]. 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 as outlined by Chrysikopoulos and Syngouna [35]. The hydrodynamic diameter of the clay colloids was measured by a zetasizer (Nano ZS90, Malvern Instruments) and was found to be equal to $d_p = 843 \pm 126$ nm for KGa-1b, and $d_p = 1187 \pm 381$ nm for STx-1b [35].

2.3. Porous media

Glass beads were used as the packing material of the columns in order to eliminate possible experimental difficulties associated with real soil, which may provide numerous uncertainties that can complicate considerably the analysis of the experimental data. Following the procedure previously described by Syngouna and Chrysikopoulos [10], the glass beads were purified until the water conductivity, as determined by a conductivity meter, was negligible. The glass beads were dried in an oven at 105 °C, and then stored in screw cap sterile beakers until use in the column experiments.

2.4. Electrokinetic measurements

The Zeta potentials, ζ [V], of the bacteriophages and clays, measured at pH 7 in sterile ddH_2O by the zetasizer (Nano ZS90, Malvern Instruments, Southborough, MA), were $\zeta = -40.4 \pm 3.7$ mV for MS2, $\zeta = -31.8 \pm 1.3$ mV for ΦX174, $\zeta = -26.0 \pm 2.8$ mV for KGa-1b, and $\zeta = -20.5 \pm 0.8$ mV for STx-1b [35]. Furthermore, the zeta potential of glass beads stored in ddH_2O at pH 7 was determined to be $\zeta = -54.6 \pm 2.4$ mV [10]. The zeta potential for the AWIs present in the unsaturated packed columns was obtained from the literature ($\zeta = -65$ mV in ddH_2O solution) [15,36].

2.5. Column experiments

All flow-through experiments were conducted using Plexiglas columns with length 15.2 cm and internal diameter 2.6 cm. The experimental setup is similar to that described in detail by Mitropoulou et al. [37]. Briefly, the column was uniformly wet-packed with glass beads. Several pore volumes of the de-aired sterile ddH_2O were passed through the column from the bottom to avoid the capture of air bubbles. The packed column was vertically attached to a vacuum chamber (Soil Measurement Systems, Tucson, AZ) with a fraction collector inside, which allowed for various levels of water saturation. Due to the relatively small size of the viruses used in this study, gravity effects were assumed to be negligible [38]. The water potential and the uniformity of water in the column were verified with tensiometer readings, which were collected continuously using a CR800 datalogger (Campbell Scientific, Inc., Logan, UT). Liquid samples were collected at regular time intervals from the column effluent in small fractions with an automatic fraction collector.

A fresh column was packed for each experiment. The entire packed column and glassware used for the experiments were sterilized in an autoclave at 121 °C for 20 min. Constant flow of ddH_2O at flow rate of $Q = 1.5$ mL/min in the vertical-down direction was used. The mean pH of the column influent remained constant at 7.0 ± 0.2 for the duration of each experiment. One set of experiments was performed with viruses and clay particles alone in order to determine their individual transport characteristics. Another set of cotransport experiments was performed to investigate the effect of the presence of clay colloids on virus transport. The clay colloidal suspension and the viral suspension were injected simultaneously into the packed column, at the same flow rate, for 3 pore volumes, followed by 3 pore volumes of ddH_2O. All experiments were carried out at room temperature (~25 °C). Chloride, in the form of potassium chloride (KCl), was chosen as the nonreactive tracer. All effluent chloride concentrations were measured using ion chromatography (ICS-1500, Dionex Corp., Sunnyvale, CA).

3. Theoretical considerations

3.1. Moment analysis

The colloid concentration breakthrough data obtained at the end of the packed column ($x = L$) were analyzed by the first normalized temporal moment, M_1 [t] [39]. In this study, four different $M_{1(i)}/M_{1(t)}$ ratios of the first normalized temporal moment, indicating the degree of velocity enhancement of colloid (i) relative to the conservative tracer (t), were calculated based on the effluent concentrations for $C_{Total-v}$, C_c, C_v, and C_{vc}. Furthermore, the mass recovery, M_r [–], of the tracer and the suspended $C_{Total-v}$, C_c, C_v particles, as well as the produced mass of C_{vc}, M_p [–], were quantified.

3.2. Filtration theory

The classical colloid filtration theory (CFT) was used to quantitatively compare the attachment of viruses and clay particles onto SWIs. The dimensionless collision efficiency, α [-], which represents the ratio of the collisions resulting in attachment to the total number of collisions between particles and collector grains [40], was calculated for the unsaturated transport experiments by the following expression [3,41]:

$$\alpha_{exp} = -\frac{2}{3}\frac{d_c}{L(1-\theta_m)\eta_0}\ln\left[\frac{C_{iss}}{C_{i0}}\right] \quad (1)$$