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Colloid transport in unsaturated porous media: The role of water content and ionic strength on particle straining

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Abstract

Packed column and mathematical modeling studies were conducted to explore the influence of water saturation, pore-water ionic strength, and grain size on the transport of latex microspheres (1.1 μm) in porous media. Experiments were carried out under chemically unfavorable conditions for colloid attachment to both solid–water interfaces (SWI) and air–water interfaces (AWI) using negatively charged and hydrophilic colloids and modifying the solution chemistry with a bicarbonate buffer to pH 10. Interaction energy calculations and complementary batch experiments were conducted and demonstrated that partitioning of colloids to the SWI and AWI was insignificant across the range of the ionic strengths considered. The breakthrough curve and final deposition profile were measured in each experiment indicating colloid retention was highly dependent on the suspension ionic strength, water content, and sand grain size. In contrast to conventional filtration theory, most colloids were found deposited close to the column inlet, and hyper-exponential deposition profiles were observed. A mathematical model, accounting for time- and depth-dependent straining, produced a reasonably good fit for both the breakthrough curves and final deposition profiles. Experimental and modeling results suggest that straining — the retention of colloids in low velocity regions of porous media such as grain junctions — was the primary mechanism of colloid retention under both saturated and unsaturated conditions. The extent of stagnant regions of flow within the pore structure is enhanced with decreasing water content, leading to a greater amount of retention. Ionic strength also contributes to straining, because the number of colloids that are held in the secondary energy minimum increases with ionic strength. These weakly associated colloids are prone to be translated to stagnation regions formed at grain–grain junctions, the solid–water–air triple point, and dead-end pores and then becoming trapped.
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Keywords: Colloid transport; Deposition; Straining; Ionic strength; Hydrodynamics

1. Introduction

Colloid movement through porous media is of great significance in a number of environmental fields including contaminant transport, soil profile development, and subsurface migration of pathogenic microorganisms. Pathogenic microorganisms such as bacteria and viruses

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(often referred to biocolloids) pose a high risk to water resources through land application of raw and treated wastewater, septic systems, leaking sewage pipes, and animal manure (Hurst, 1980; Powelson et al., 1993; Redman et al., 2001). Colloid transport in porous media is also of concern due to colloid-facilitated transport of a wide variety of inorganic and organic contaminants that adsorb onto these particles and travel significant distances (de Jonge et al., 1998; Ryan et al., 1998; McGechan and Lewis, 2002). Hence, comprehensive knowledge of the transport of colloidal particles in subsurface environments is essential for predicting biological and chemical contaminant fate. Considerable research has been devoted to the fate and transport of colloids in saturated porous media (reviews are given by Schijven and Hassanizadeh, 2000; Harvey and Harms, 2002; de Jonge et al., 2004); however, colloid transport in the unsaturated (vadose) zone, which often act as a first natural layer against the pollution of groundwater, has received little systematic research and the governing mechanisms are poorly understood.

Mechanisms of colloid retention in unsaturated porous media include adsorption (attachment/detachment processes) to solid–water interfaces (SWI) (reviews are given by Ryan and Elimelech, 1996, Schijven and Hassanizadeh, 2000), air–water interfaces (AWI) (e.g. Wan and Wilson, 1994a), straining (McDowell-Boyer et al., 1986; Bradford et al., 2002, 2003), and film straining (Wan and Tokunaga, 1997; Saiers and Lenhart, 2003a). Attachment to the SWI involves colloid collision with and attachment to grain surfaces, and consequently depends on the chemical and physical characteristics of the colloids and soil surfaces as well as the solution chemistry (Ryan and Elimelech, 1996, Walker et al., 2004). The AWI present within unsaturated porous media is also believed to serve as a collector for colloid particles, reportedly in an irreversible manner, retained by either capillary or electrostatic forces (Wan and Wilson, 1994b; Schafer et al., 1998). Colloid attachment to the AWI, therefore, depends on pH, ionic strength, and colloid surface properties (DeNovio et al., 2004; Torkzaban et al., 2006b). Additionally, Wan and Tokunaga (2002) demonstrated that only positively charged colloidal particles attached at the AWI, and Crist et al. (2004, 2005) observed in three-dimensional porous media that negatively charged-hydrophilic colloids did not attach to the AWI.

Straining involves the retention of colloids in the smallest regions of the pore space (McDowell-Boyer et al., 1986; Cushing and Lawler 1998; Bradford et al., 2002, 2003, 2006a) such as those formed near grain-to-grain contact points. It should be noted that retention of colloids near to grain-to-grain contact points has also been referred to in the literature as wedging (Herzig et al., 1970; Johnson et al., 2007). In the smallest regions of the pore space the water velocity is very low and these locations can be considered as zones of relative flow stagnation. Straining may also occur in pore throats that are too small to allow the passage of multiple colloids (Herzig et al., 1970; Bradford et al., 2002), a process often referred to as bridging (Ramachandran and Fogler, 1999). Criteria for colloid straining in porous media has traditionally been assumed to be simply a function of the ratio of colloid to collector diameters (d_p/d_g) and the pore size distribution of the medium (Herzig et al., 1970; McDowell-Boyer et al., 1986; Bradford et al., 2002, 2003). Herzig et al. (1970) computed that when d_p/d_g exceeds 0.05, straining significantly contributes to retention of colloids in porous media. Recently, a few researchers have suggested that the theoretical criteria of Herzig et al. (1970) underestimates the extent of straining in colloid retention and that straining can be occurring at d_p/d_g as low as 0.002 (Bradford et al., 2002, 2003; Li et al., 2004).

In unsaturated media, straining has received relatively less attention (Gargiulo et al., 2007). In fact, straining may be more pronounced in unsaturated versus saturated porous media because capillary forces constrain water flow within regions having smaller pore spaces. Furthermore, unsaturated systems also contain solid–water–air triple points at the intersection of solid–water and air–water interfaces. Triple points and grain–grain contact points share many similarities in that these locations are both low velocity zones. In this work, we consider retention at the triple point as an additional form of straining. Moreover, the extent to which saturated and especially unsaturated colloid straining is sensitive to changes in the chemical properties of the system (i.e. aqueous phase, colloids, and collectors) remains poorly understood (Bradford et al., 2006a, 2007). In addition to straining, film straining (Wan and Tokunaga, 1997) is another potential retention mechanism needing further investigation. Film straining is the removal of colloids in partially saturated porous media occurring as a result of the physical restrictions to colloid transport through water films with thicknesses smaller than the diameter of the colloids.

Previous studies on unsaturated colloid transport have mainly focused on the determination of the colloid concentration in the effluent (e.g. Saiers and Lenhart, 2003a; Torkzaban et al., 2006a,b). Breakthrough concentrations are typically simulated by using a variety of parameters to best fit the data that consider SWI and/or AWI adsorption rate coefficients, and/or film straining coefficients. Few studies have reported the shape of the