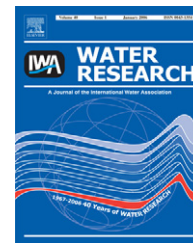


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Coupling of physical and chemical mechanisms of colloid straining in saturated porous media

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ABSTRACT

Filtration theory does not include the potential influence of pore structure on colloid removal by straining. Conversely, previous research on straining has not considered the possible influence of chemical interactions. Experimental and theoretical studies were therefore undertaken to explore the coupling of physical and chemical mechanisms of colloid straining under unfavorable attachment conditions (pH = 10). Negatively charged latex microspheres (1.1 and 3 μm) and quartz sands (360, 240, and 150 μm) were used in packed column studies that encompassed a range in suspension ionic strengths (6–106 mM) and Darcy water velocities (0.1–0.45 cm min^{-1}). Derjaguin–Landau–Verwey–Overbeek (DLVO) calculations and torque analysis suggests that attachment of colloids to the solid–water interface was not a significant mechanism of deposition for the selected experimental conditions. Effluent concentration curves and hyperexponential deposition profiles were strongly dependent on the solution chemistry, the system hydrodynamics, and the colloid and collector grain size, with greater deposition occurring for increasing ionic strength, lower flow rates, and larger ratios of the colloid to the median grain diameter. Increasing the solution ionic strength is believed to increase the force and number of colloids in the secondary minimum of the DLVO interaction energy profile. These weakly associated colloids can be funneled to small regions of the pore space formed adjacent to grain–grain junctions. For select systems, the ionic strength of the eluant solution was decreased to 6 mM following the recovery of the effluent concentration curve. In this case, only a small portion of the deposited colloids was recovered in the effluent and the majority was still retained in the sand. These observations suggest that the extent of colloid removal by straining is strongly coupled to solution chemistry.

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1. Introduction

Considerable research has been devoted to the fate and transport of microbes (biocolloids) and colloids in porous media (reviews are given by Schijven and Hassanizadeh, 2000; Harvey and Harms, 2002; Jin and Flury, 2002; Ginn et al., 2002). The bulk of existing colloid literature considers deposition to be controlled by attachment of colloids on grain surfaces.

Mass transfer of colloids to the solid–water interface occurs via diffusion, interception, and sedimentation (Yao et al., 1971). Attachment involves collision with and subsequent retention of colloids at the solid–water interface. Once a colloid approaches the solid–water interface, attachment depends on a combination of forces and/or torques that act on the colloid (Cushing and Lawler, 1998; Bergendahl and Grasso, 1998; Li et al., 2005) at this location. These forces

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include gravity and buoyancy, hydrodynamic drag and lift, electrical double-layer repulsion (or attraction), and London–van der Waals interaction.

In the presence of an energy barrier (unfavorable attachment conditions) the transport and deposition behavior of colloids is frequently found to be inconsistent with clean bed filtration theory (CFT) predictions (Albinger et al., 1994; Camesano and Logan, 1998; Bolster et al., 1999; Redman et al., 2001; Bradford et al., 2002; Tufenkji et al., 2003; Li et al., 2004; Tong et al., 2005; Li and Johnson, 2005; Tufenkji and Elimelech, 2005a,b). Specifically, the deposition coefficient has commonly been observed to exhibit a depth dependence and the concentration of retained colloids do not decrease exponentially with distance as CFT would predict. Deviation between CFT predictions and experimental data has been reported to be increasingly significant for larger colloids and finer textured porous media (Bradford et al., 2002; Tufenkji and Elimelech, 2005a). Bradford et al. (2003) reported that filtration theory predictions no longer adequately describe the measured deposition profiles when the ratio of the colloid diameter to the median grain diameter is greater than around 0.005.

Filtration theory assumes a model collector (such as Happel's sphere-in-cell model which is an isolated collector in an assemblage of collectors) that neglects the actual pore structure and pores formed by grain–grain contacts which occur in porous media. However, recent experimental and theoretical research has indicated that the pore structure can play an important role in colloid deposition under unfavorable attachment conditions (Cushing and Lawler, 1998; Bradford et al., 2002, 2003, 2004, 2005, 2006a–d; Li et al., 2004, 2006a,b; Tufenkji et al., 2004; Bradford and Bettahar, 2005, 2006; Foppen et al., 2005). This removal likely occurs by straining, which is the retention of colloids in the smallest regions of the soil pore space formed adjacent to points of grain–grain contact (Cushing and Lawler, 1998). Pore spaces occurring at grain contacts provide optimum locations for colloids that are weakly associated with the solid phase (secondary energy minima) to be retained because of reduced hydrodynamic forces, pore size limitations, and enhanced Derjaguin–Landau–Verwey–Overbeek (DLVO) interactions (Hoek and Agarwal, 2006).

Most published research on straining has focused on the role of physical factors such as the relative size of the colloid and porous medium, and little attention has been given to the potential interrelated influence of solution chemistry and hydrodynamics on straining. Attachment under unfavorable conditions is known to be highly dependent on solution chemistry (e.g., Li et al., 2004; Tufenkji and Elimelech, 2004, 2005a), and system hydrodynamics (Wang et al., 1981; Tan et al., 1994; Kretzschmar et al., 1997; Compere et al., 2001; Li et al., 2005). Under such attachment conditions, hydrodynamic forces may be sufficient to overcome the weak association of colloids in the secondary energy minimum of the DLVO interaction energy distribution (Kuznar and Elimelech, 2007). In this case, attached colloids may be lifted from the solid surface and detach, or they may roll or slide down gradient on the grain surface to locations where the hydrodynamic shear is less significant. Bergendahl and Grasso (1998) demonstrated that rolling is often the primary mechanisms for hydrodynamic

motion of detached colloids. It is logical to anticipate that some of the colloids rolling on the grain surface may be retained in straining locations (i.e., smallest regions of the pore space formed at grain–grain contact points). One can therefore expect that solution chemistry and hydrodynamic forces will play an important interrelated role in straining.

The objective of this research is to investigate the role of solution chemistry and system hydrodynamics on colloid transport and straining. Negatively charged latex microspheres and quartz sands were used in packed column studies that encompassed a range of solution IS (6–106 mM) and Darcy water velocity (0.1–0.45 cm min⁻¹). All experiments were conducted using electrolyte solution buffered to a pH of 10 to ensure highly unfavorable attachment conditions. Data analysis and interpretation was aided through DLVO calculations of the total interaction energy between the colloids and quartz surfaces, analysis of torques, mass balance computation, and experimental effluent concentration curves and deposition profiles.

2. Material and methods

2.1. Colloids

Yellow–green fluorescent latex microspheres (Molecular Probes, Eugene, OR) were used as model colloid particles in the experimental studies (excitation at 505 nm, and emission at 515 nm). Two sizes of microspheres were used in the transport experiments, 1.1 and 3.0 μm. The uniformity of the colloid size was verified using a Horiba LA 930 (Horiba Instruments Inc., Irvine, CA) laser scattering particle size and distribution analyzer and by inspections of suspensions under an epi-fluorescent microscope. The microspheres had carboxyl surface functional groups, a density of 1.055 g cm⁻³, and are reported as hydrophilic by the manufacturer. The initial influent concentration (C_i) for the 1.1 and 3.0 μm colloids for the experiments was 2.7×10^{10} and $1.3 \times 10^9 N_c L^{-1}$ (where N_c denotes number of colloids), respectively. Several experiments with the 1.1 μm colloids were also conducted at a lower initial concentration of $C_i = 6.8 \times 10^8 N_c L^{-1}$.

2.2. Sand

Aquifer material used for the column experiments consisted of various sieve sizes of Ottawa sand (US Silica, Ottawa, IL). The porous media were selected to encompass a range in grain sizes, and are designated by their median grain size (d_{50}) as: 360, 240, and 150 μm. Specific properties of the 360, 240, and 150 μm sands include: the coefficient of uniformity (d_{60}/d_{10} ; here 10% and 60% of the sand mass is finer than d_{10} and d_{60} , respectively) of 1.88, 3.06, and 2.25; and intrinsic permeabilities of 6.37×10^{-11} , 1.12×10^{-11} , and $4.68 \times 10^{-12} m^2$, respectively. Ottawa sands typically consist of 99.8% SiO₂ (quartz) and trace amounts of metal oxides, have spheroidal shapes, and contain relatively rough surfaces. An estimate of the pore-size distribution for these sands can be obtained by using Laplace's equation of capillarity and measured capillary pressure–saturation curves presented by Bradford and Abriola (2001).

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