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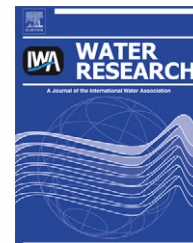
Colloid retention at the meniscus-wall contact line in an open microchannel

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ABSTRACT

Colloid retention mechanisms in partially saturated porous media are currently being researched with an array of visualization techniques. These visualization techniques have refined our understanding of colloid movement and retention at the pore scale beyond what can be obtained from breakthrough experiments. One of the remaining questions is what mechanisms are responsible for colloid immobilization at the triple point where air, water, and soil grain meet. The objective of this study was to investigate how colloids are transported to the air-water-solid (AWS) contact line in an open triangular microchannel, and then retained as a function of meniscus contact angle with the wall and solution ionic strength. Colloid flow path, meniscus shape and meniscus-wall contact angle, and colloid retention at the AWS contact line were visualized and quantified with a confocal microscope. Experimental results demonstrated that colloid retention at the AWS contact line was significant when the meniscus-wall contact angle was less than 16°, but was minimal for the meniscus-wall contact angles exceeding 20°. Tracking of individual colloids and computational hydrodynamic simulation both revealed that for small contact angles (e.g., 12.5°), counter flow and flow vortices formed near the AWS contact line, but not for large contact angles (e.g., 28°). This counter flow helped deliver the colloids to the wall surface just below the contact line. In accordance with DLVO and hydrodynamic torque calculations, colloid movement may be stopped when the colloid reached the secondary minimum at the wall near the contact line. However, contradictory to the prediction of the torque analysis, colloid retention at the AWS contact line decreased with increasing ionic strength for contact angles of 10–20°, indicating that the air-water interface was involved through both counter flow and capillary force. We hypothesized that capillary force pushed the colloid through the primary energy barrier to the primary minimum to become immobilized, when small fluctuations in water level stretched the meniscus over the colloid. For large meniscus-wall contact angles counter flow was not observed, resulting in less colloid retention, because a smaller number of colloids were transported to the contact line.

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1. Introduction

Colloids facilitate the transport of a wide range of contaminants through the vadose zone and into groundwater. Some typical contaminants that experience colloid-facilitated transport and are of great concern for human and ecological health include: radionuclides, pesticides and trace metals (Saiers and Hornberger, 1999; Williams et al., 2006). While most colloid transport studies have been carried out under saturated conditions, fewer studies have characterized colloid retention processes in unsaturated soils. These studies found that, in addition to retention mechanisms occurring in saturated soils, e.g., pore straining, and attachment to grain surfaces by the overall Derjaguin-Landau-Verwey-Overbeek (DLVO) energy minima and/or surface chemical heterogeneity or roughness that produce locally attractive sites on an overall repulsive surface (Johnson et al., 2010), the presence of a gaseous phase in unsaturated soils forms other potential retention sites such as the air-water interface (AWI) and the air-water-solid (AWS) contact line (Bradford and Torkzaban, 2008). Early work in unsaturated processes reported colloid retention at the air-water interface (Wan and Wilson, 1994) and in thin water films enveloping grains (Wan and Tokunaga, 1997; Veerapaneni et al., 2000). Research in the last decade has utilized micros-copy advances and identified two new retention sites: immobile water zones (Gao et al., 2006) and the AWS contact line, which is defined as the triple point where air, water and grain approach each other. Colloid attachment at the AWS contact line, which is the focus of this study, has been observed in soil media (Crist et al., 2004, 2005; Zevi et al., 2005, 2009) and in surrogate of soil pores (e.g., microchannels) (Lazouskaya et al., 2006; Lazouskaya and Jin, 2008). Capillary and DLVO forces have both been considered to explain colloid retention at this interface by Crist et al. (2005), Gao et al. (2008), and Shang et al. (2008).

This study sets out to explain why colloids adhere at the AWS contact line. To date several studies have indicated that a variety of colloids attach at the AWS contact line, especially when the DLVO forces do not favor colloid attachment at the solid-water interface (SWI) (Schäfer et al., 1998a; Sirivithayapakorn and Keller, 2003; Auset and Keller, 2006; Lazouskaya et al., 2006; Crist et al., 2004, 2005; Zevi et al., 2005, 2009). The studies by Crist et al. (2004, 2005) were likely the first time that the colloids were visualized to attach at the AWS contact line. Other earlier studies might have also observed this phenomenon, but attributed it to the attachment at the air-water interface (Wan and Wilson, 1994; Sirivithayapakorn and Keller, 2003). Although a controversy exists regarding the role of evaporation at the AWS contact line (Steenhuis et al., 2005; Wan and Tokunaga, 2005), since then the colloid attachment at the AWS contact line has been observed by Lazouskaya et al. (2006), Lazouskaya and Jin (2008), Gao et al. (2008), Morales et al. (2009), and Zevi et al. (2009). Despite these observations there has not been a satisfactory explanation on how colloids can approach the often energetically repulsive solid-water and/or air-water interfaces.

Hydrodynamic forces have been speculated to be responsible for supplying the necessary external energy to overcome the repulsive energy exerted at the AWS contact line (Lazouskaya et al., 2006). However, according to Shang et al. (2008), drag forces at realistic groundwater flow rates are too weak to thrust suspended colloids through the energy barrier at the AWS contact line so that capillary force can more permanently immobilize the colloids. Although the source of energy that colloids use to approach this energetically unfavorable retention site remains uncertain, experimental observations indicate that colloid immobilization at the AWS contact line ubiquitously occurs. For now, its mechanistic understanding remains elusive.

The studies of Gao et al. (2008) and Shang et al. (2008) indicate that capillary forces are generally at least two orders of magnitude greater than the system's DLVO forces. A detailed theoretical description of DLVO and capillary forces is presented in the auxiliary material. O'Brien and van den Brule (1991) suggested that colloid attachment on a solid substrate is controlled by capillary force of the film covering the solid, and can be determined by the liquid-colloid and liquid-substrate contact angles. Despite this critical observation for colloid retention in unsaturated systems, many studies have explained colloid attachment by employing the DLVO theory exclusively (e.g., Auset and Keller, 2006; Crist et al., 2004, 2005; Lazouskaya et al., 2006; Schäfer et al., 1998a,b; Sirivithayapakorn and Keller, 2003).

Numerical models for colloid retention in unsaturated porous media mainly consider the air-water interface as an additional retention site as compared to saturated media (Corapcioglu and Choi, 1996; Massoudieh and Ginn, 2007; Chen, 2008). Specific model for colloid attachment at the AWS contact line are unavailable; therefore, additional work in this area is needed. Shi et al. (2010) simulated a moving air-water interface in a microchannel. Other studies have used the finite element analysis software COMSOL in colloid transport studies (Cakmak et al., 2008; Kemps and Bhattacharjee, 2009; Torkzaban et al., 2007, 2008; Bradford and Torkzaban, 2008).

The objective of this study was to experimentally visualize and quantify how colloids flowing through a simplified "soil pore" (i.e., microchannel) are retained at the AWS contact line under a wide range of DLVO forces (varied by solution ionic strengths) and capillary force (varied by the air-water interfacial shapes exhibiting different meniscus-wall contact angles).

2. Materials and methods

2.1. Colloid attachment experiment in a microchannel

The colloids used were hydrophilic (water contact angle = 12°), negatively charged, 1-μm yellow fluorescent synthetic polystyrene microspheres (Polysciences, Inc., Warrington, PA). Colloid suspensions of 9.1×10^6 microspheres/mL were prepared in the solution of 0.001% Rhodamine B dye at three ionic strengths (i.e., 1 mM, 50 mM, and 100 mM NaCl), respectively. The very low concentration of Rhodamine B dye was added to allow for imaging the water phase and does not influence colloid behavior (Zevi et al., 2006, 2009). The colloid-free solutions of identical solution chemistry with that of the colloid suspensions were used as background solutions. Prior to each experiment the colloid suspensions were sonicated in