Journal of Colloid and Interface Science 406 (2013) 44–50

Contents lists available at SciVerse ScienceDirect

Journal of Colloid and Interface Science

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Colloid mobilization by fluid displacement fronts in channels

Volha Lazouskaya [a], Lian-Ping Wang [b], Dani Or [c], Gang Wang [c], Jeffrey L. Caplan [d], Yan Jin [a,*]

[a] Department of Plant and Soil Sciences, University of Delaware, 152 Townsend Hall, Newark, DE 19716, USA
[b] Department of Mechanical Engineering, University of Delaware, 126 Spencer Lab, Newark, DE 19716, USA
[c] Department of Environmental Systems Science, ETH Zurich, Universitaetstrasse 16, 8092 Zurich, Switzerland
[d] Delaware Biotechnology Institute, University of Delaware, 15 Innovation Way, Newark, DE 19711, USA

ARTICLE INFO

Article history:
Received 24 January 2013
Accepted 31 May 2013
Available online 7 June 2013

Keywords:
Colloid transport
Contact line
Drainage
Imbibition
Contact angle
Thin films

ABSTRACT

Understanding colloid mobilization during transient flow in soil is important for addressing colloid and contaminant transport issues. While theoretical descriptions of colloid detachment exist for saturated systems, corresponding mechanisms of colloid mobilization during drainage and imbibition have not been considered in detail. In this work, theoretical force and torque analyses were performed to examine the interactive effects of adhesion, drag, friction, and surface tension forces on colloid mobilization and to outline conditions corresponding to the mobilization mechanisms such as lifting, sliding, and rolling. Colloid and substrate contact angles were used as variables to determine theoretical criteria for colloid mobilization mechanisms during drainage and imbibition. Experimental mobilization of hydrophilic and hydrophobic microspheres with drainage and imbibition fronts was investigated in hydrophilic and hydrophobic channels using a confocal microscope. Colloid mobilization differed between drainage and imbibition due to different dynamic contact angles and interfacial geometries on the contact line. Experimental results did not fully follow the theoretical criteria in all cases, which was explained with additional factors not included in the theory such as presence of aggregates and trailing films. Theoretical force and torque analyses resulted in similar mobilization predictions and suggested that all mobilization mechanisms contributed to the observed colloid mobilization.

1. Introduction

Understanding mobilization of previously deposited colloids or in situ formed colloids in soil is important for assessing concentration of mobile colloids and colloid-associated transport of contaminants (e.g., [1,2]). Several reviews of colloid mobilization and transport in soil and model systems have been published (e.g., [3–5]). However, understanding of the mechanisms controlling colloid mobilization in unsaturated systems is still limited compared to saturated systems [4–6].

El-Farhan et al. [7] and Saiers et al. [8] were among the first to emphasize the role of air–water interface (AWI) in mobilization of soil colloids during transient events, i.e., drainage and imbibition, in unsaturated porous media. Zhuang et al. [9], Cheng and Saiers [1,10], and Chen et al. [6,11] investigated colloid mobilization in column experiments during both drainage and imbibition, but reported inconsistent results on which event was more efficient in colloid mobilization. Visualization experiments to date involved mostly indirect visualization methods such as enumeration of colloids after the passage of air bubbles or successive drainage and imbibition fronts [12–18], which did not distinguish effects of drainage and imbibition. Recently, Aramrak et al. [19] and Lazouskaya et al. [20] observed colloid mobilization during drainage and imbibition directly with a confocal microscope, but worked with a limited range of colloid and substrate properties.

Several researchers indicated that colloids were mobilized on the contact line (where AWI contacts the solid) [14,20,21] and then were transported with the AWI. Lazouskaya et al. [20] used the term front, which encompassed both AWI and the contact line, and therefore emphasized the importance of both in colloid mobilization.

Theoretical descriptions of particle detachment from substrates in either air or water phases are available in the literature (e.g., [22–25]); "detachment" in these studies is equivalent to "mobilization". An attached particle will detach from the substrate when the external forces exceed the adhesion force (between the particle and the substrate) or applied torques on the particle offset the corresponding resisting torque [24,26,27]. Depending on the directions and magnitudes of the forces and torques acting on the

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Abbreviations: AWI, air–water interface; DLVO, Derjaguin–Landau–Verwey–Overbeek theory.

* Corresponding author. Fax: +1 302 831 0605.

E-mail addresses: volha@udel.edu (V. Lazouskaya), lwang@udel.edu (L.-P. Wang), dani.or@env.ethz.ch (D. Or), gang.wang@env.ethz.ch (G. Wang), jcaplan@udel.edu (J.L. Caplan), yjin@udel.edu (Y. Jin).

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http://dx.doi.org/10.1016/j.jcis.2013.05.078

particle, it can be mobilized via lifting off from the substrate, sliding, or rolling along the substrate. It is important to recognize that mobilization by sliding or rolling may not necessarily lead to colloid separation from the substrate, which corresponds to the different modes (i.e., mobile and immobile) of adhesion as studied in Boks et al. [28]. Previously, rolling has been declared as the most likely mobilization mechanism for particles fully submerged in air or water phases (e.g., [24,25]).

Theoretical conceptualization of colloid mobilization with receding and advancing fronts (corresponding to drainage and imbibition, respectively) was originally developed by Leenaars [29] and Leenaars and O'Brien [30] and has been adopted in later studies (e.g., [12,19,20]). The conceptualization is based on the analysis of major forces acting on a colloid on the contact line including surface tension force, adhesion force, and hydrodynamic drag force. Among the forces, the surface tension force (also known as capillary force) was identified as the dominant force responsible for colloid mobilization with displacement fronts [16,29,31]. While these studies provided thorough investigation of the role of the surface tension force in colloid mobilization, the mechanisms of colloid mobilization in two-phase system (i.e., lifting, sliding, and rolling) and corresponding mobilization conditions have not been considered in detail. Shang et al. [31] considered torque balance and rolling possibility for a partially submerged colloid, but to the best of our knowledge, the role of rolling in colloid mobilization on the contact line during imbibition and drainage has not been previously addressed. Extensive analysis of the surface tension force was performed by Gao et al. [32] and Zevi et al. [33], but was applied to colloid retention on the contact line.

In addition to the forces acting on colloids, colloid mobilization and retention have been previously linked to water films that are ubiquitously present in unsaturated porous media (e.g., [34–36]). The ratio of colloid size to film thickness is the major parameter determining potential retention and mobilization (e.g., [34,36]). In addition to the "static" films, e.g., due to adsorbed water or higher water content [37], there are films associated with dynamic processes such as precursor and trailing films observed for advancing and receding fronts [38], which may play an exclusive role in colloid mobilization.

In this work, we investigated colloid mobilization during drainage and imbibition in rectangular capillary channels. The specific objectives were to (1) develop theoretical criteria of colloid mobilization via lifting, sliding and rolling mechanisms during both drainage and imbibition, and (2) examine these criteria experimentally in channels by employing hydrophilic (contact angle generally <90°) and hydrophobic (contact angle generally >90°) colloids and substrates. Special attention was given to thin film configurations and their potential effects on colloid mobilization.

2. Theoretical considerations

2.1. Forces acting on colloids on the contact line

Fig. 1 provides a conceptual schematic of a colloid attached to hydrophilic and hydrophobic substrates and interacting with imbibition and drainage fronts. Fig. 1 also shows two front positions relative to a colloid in each configuration, corresponding to the two surface tension force maxima (as discussed further). Also shown are the adhesion force (directed toward the substrate), hydrodynamic drag force (directed with the flow), and friction force.

Adhesion force (F_A), the physicochemical interaction force between a colloid and the substrate, is calculated with the extended Derjaguin–Landau–Verwey–Overbeek (DLVO) theory [3,39–41] as a sum of van der Waals (F_{vdW}), electrostatic (F_{el}), hydrophobic (F_H), and Born repulsion (F_B) forces. Details on each force can be found in the literature [41–44]. Inclusion of Born repulsion in DLVO calculations results in a finite depth of the primary minimum and a more accurate prediction of the force required for colloid mobilization [3,45]. The maximum adhesion force can be found as the maximum attractive (negative) force in the DLVO force profile.

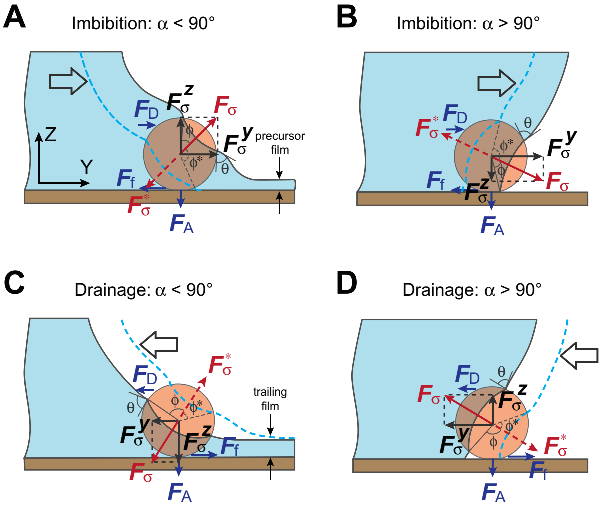

Fig. 1. Colloids interacting with imbibition (A and B) and drainage (C and D) fronts on a hydrophilic and a hydrophobic substrate. Two interface positions on the particle (for the two surface tension force directions, i.e., $\phi > \theta$ and $\phi < \theta$) are shown. The earlier interface position is represented with the dashed line; only surface tension force F_σ^* and position angle ϕ^* are shown for this interface. For the later interface position, the direction and components of surface tension force and other forces are shown. Force arrows do not represent force magnitudes. The large arrows indicate flow direction.

Drag force (F_D) exerted on the attached spherical particle in an imposed shear flow can be calculated as (e.g., [46,47])

$$F_D = \pm 1.701(6\pi\mu G r^2) \tag{1}$$

where the sign depends on whether drainage (−) or imbibition (+) is considered in Fig. 1, μ is fluid viscosity, r is colloid radius, and G is shear rate. The fluid velocity parallel to the substrate is modeled as $v_y = Gz$ where z is the distance from substrate. Because Eq. (1) is derived for a fully submerged stationary particle in a linear shear flow, the drag force on a partially submerged particle will be smaller (e.g., [19]).

Surface tension force (F_σ) acts along the contact between a colloid and AWI and is expressed as

$$F_\sigma = 2\pi r\sigma \sin\phi \sin(\theta - \phi) \tag{2}$$

where σ is the liquid surface tension, θ is the dynamic contact angle on the colloid surface, and ϕ is the angle determining the AWI position on the colloid surface ([29]; Fig. 1). As liquid advances (or recedes) along the particle surface, ϕ changes from 180° to 0° (or 0° to 180°), and surface tension force assumes two magnitude maxima, $F_\sigma = 2\pi r\sigma \sin^2(\theta/2)$ (surface tension force is directed away from the liquid for $\phi < \theta$ and this maximum occurs at $\phi = \theta/2$) and $F_\sigma = -2\pi r\sigma \sin^2(90^\circ + \theta/2)$ (surface tension force is directed toward the liquid for $\phi > \theta$ and this maximum occurs at $\phi = 90^\circ + \theta/2$). For a particle on the contact line, the components of maximum surface tension force (Fig. 1) also depend on the substrate dynamic contact angle, α, and are determined as

$\phi < \theta$:

$$F_\sigma^z = 2\pi r\sigma \sin^2(\theta/2)\cos\alpha \tag{3}$$