



Transport and retention of TiO₂ and polystyrene nanoparticles during drainage from tall heterogeneous layered columns



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ABSTRACT

Recent developments in nanotechnology have seen an increase in the use of manufactured nanomaterials. Although their unique physicochemical properties are desirable for many products and applications, concern continues to exist about their environmental fate and potential to cause risk to human and ecological health. The purpose of this work was to examine one aspect of nanomaterial environmental fate: transport and retention in the unsaturated zone during drainage. The work made use of tall segmented columns packed with layers of two different porous media, one medium sand and one fine sand. The use of tall columns allowed drainage experiments to be conducted where the water table remained within the height of the column, permitting control of final saturation profiles without the need for capillary barrier membranes which can potentially complicate analyses. Experiments were conducted with titanium dioxide (TiO₂) and polystyrene nanomaterials. For the strongly negatively-charged polystyrene nanomaterials, little retention was observed under the conditions studied. For the TiO₂ nanomaterials, results of the work suggest that while saturated fine sand layers may retain more nanomaterials than saturated coarse sand layers, significantly greater retention is possible in unsaturated media. Furthermore, unsaturated medium sand layers exhibited significantly greater retention than adjacent saturated fine sand layers when present at low saturations high above the water table. Retention by unsaturated media were found to correlate strongly with elevation. Free drainage experiments including both primary and secondary drainages in homogeneous columns showed evidence of redistribution during imbibition and secondary drainage, but still showed substantial unsaturated retention of TiO₂ nanoparticles high in the column, despite re-saturation with- and drainage of nanoparticle-free water.

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1. Introduction

Manufactured nanomaterials are engineered for use in a variety of products and applications. Nanomaterials may be found in consumer goods such as sunscreens, tires, and sports equipment, environmental remediation products, electronics, and medicine (Rao and Cheetham, 2001). It has been projected that as manufacturing costs decrease and more applications are found and become economically feasible, the quantity of nanomaterials produced each year will continue to increase substantially (Delany, 2006). As nanomaterials are produced and used in larger quantities, releases into the environment become increasingly likely. Recent studies have shown that environmental contamination with nanomaterials may be a cause for alarm because of the potential health implications of some nanomaterials (Chen and von Mikecz, 2005; Derfus et al., 2004; Dreher, 2004; Oberdorster, 2004).

It is important to note that these health and environmental concerns may be mitigated if the risk of exposure is low enough. One area of

significant concern is the fate and transport of nanomaterials in the subsurface. If mobility is significant, movement in the subsurface could potentially lead to dispersal of nanoparticles to environmentally sensitive areas or drinking water sources. Alternately, if mobility is low in a particular compartment of the environment, nanomaterials may accumulate, creating the potential risk of a substantial release if water conditions change to enhance mobility (for example, a change in pH or ionic strength).

The unsaturated zone is an environmental compartment of particular interest in understanding the fate and transport of contaminants, including nanomaterials. Contaminants entering the subsurface often need to pass through the unsaturated zone to achieve high mobility in groundwater. Furthermore, the air-water interface plays a significant role in the movement of contaminants in the unsaturated zone, both by providing a sink for adsorption, and, in the case of nanomaterials and colloids, by facilitating retention by receding films (Chen et al., 2008; Chen et al., 2010). Finally, because the conditions in the unsaturated zone are dynamically changing, as a result of changing water table levels, evaporation, and rainfall infiltration, mobility of nanomaterials in unsaturated media might be expected to vary substantially over time. As such, it is important to understand the potential impact of changing conditions on transport.

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While a substantial quantity of work has been conducted examining the movement of colloids and nanomaterials in saturated media (e.g., (Bradford et al., 2006; Elimelech and Omelia, 1990; Kocur et al., 2013; Raychoudhury et al., 2010; Seymour et al., 2013; Torkzaban et al., 2010)), much less is known about their movement in unsaturated media, in part because of the significant challenges associated with transport experiments in unsaturated media. Previous research in unsaturated media has found evidence of adsorption to air-water interfaces (Chen et al., 2008; Wan and Wilson, 1994a; Wan and Wilson, 1994b), straining by stationary and receding water films (Chen and Kibbey, 2008; Chen et al., 2008; Chen et al., 2010; Chen et al., 2012; Uyusur et al., 2010; Wan and Tokunaga, 1997), as well as retention at three-phase (air, water, solid) contact lines (Crist et al., 2004; Zevi et al., 2006; Zevi et al., 2012).

This work focused on studying the transport of two types of nanomaterials (TiO_2 and polystyrene) during gravity drainage from tall unsaturated columns packed with natural sand media. While previous studies of dynamic unsaturated transport in our laboratories used coarse packed glass beads for the porous medium to allow the use of a capillary barrier membrane coarse enough to allow nanoparticles to pass (e.g., (Chen and Kibbey, 2008; Chen et al., 2008; Chen et al., 2010; Chen et al., 2012)), unsaturated transport in finer grained natural materials might be expected to exhibit some differences from transport in packed glass beads. For example, unsaturated fine grained sands can potentially have significantly greater surface areas and air-water interfacial areas than unsaturated glass beads at high capillary pressures (e.g., (Kibbey and Chen, 2012)), so they might be expected to exhibit enhanced nanomaterial attachment to both interfaces. Interestingly, however, because sand grains are generally significantly rougher than glass beads, water films on sand grains would also be expected to be much thicker well above the water table (Kibbey, 2013), so straining film effects might be reduced, potentially increasing mobility under similar conditions.

A significant additional focus of the work described here was on exploring effects related to heterogeneity, by studying transport in columns consisting of layered medium and fine sands.

2. Materials and methods

2.1. Nanomaterials

Two nanomaterials were selected for this work: titanium dioxide (TiO_2) nanopowder and polystyrene latex nanospheres. Titanium dioxide was selected because it is a widely used nanomaterial, while the polystyrene nanospheres were selected as a model synthetic material. Both were used at pH 10 with an ionic strength of 0.4 mM. These conditions were selected to favor moderate-to-high mobility in saturated media so experiments could focus on differences between saturated and unsaturated transport that result from the presence of an air-water interface (Chen et al., 2008).

Titanium dioxide nanopowder (P-25 Aeroxide) was obtained from Degussa Corporation (Düsseldorf, Germany). The manufacturer reports an anatase/rutile composition of 80/20 and a primary particle size of 21 nm. As is the case with many nanomaterials, TiO_2 nanoparticles do not exist as individual primary particles in aqueous solutions, but rather as stable aggregates (Chen et al., 2008; Chen et al., 2010; Wiesner et al., 2006). Chen et al. (2008) reported the mean size of TiO_2 aggregates to be 123.2 ± 7.6 nm with a zeta potential of -26.4 ± 1.2 mV at pH 10 and 0.2 mM ionic strength, conditions similar to those used for this work.

Polystyrene microspheres were obtained from Duke Scientific (Palo Alto, CA). The manufacturer reports a nominal size of 240 ± 6 nm. A zeta potential of -36.5 mV was measured with a particle size/zeta potential analyzer (NICOMP 380 ZLS, Particle Sizing Systems, Santa Barbara, CA). The microsphere suspension was shipped with trace quantities of added surfactant to increase the long-term stability of

the suspension. Polystyrene latex microspheres are produced with high homogeneity in both their physical and chemical characteristics and are highly stable in suspension. The observed stability of the microspheres in suspension exceeded one week for this work.

All nanomaterial suspensions were prepared as concentrations of 25 mg/L in NanoPure water (Barnstead International, Dubuque, IA), with pH adjusted to 10 by addition of sodium hydroxide, and ionic strength adjusted to 0.4 mM with addition of sodium chloride.

Prior to experiments, all nanomaterial suspensions were sonicated using a Cole Parmer (Vernon Hills, IL) Ultrasonic Processor for 10 min per liter of solution, in order to break apart large aggregates and increase the stability of the suspension. The sonicated TiO_2 nanoparticle suspensions were found to be stable for more than 24 h at pH 10, longer than the typical run time for an experiment.

2.2. Sands

Two size fractions of foundry sands (quartz silica) were obtained from the U. S. Silica (Berkeley Springs, WV) Ottawa, Illinois mine: one medium grain sand, F45, and one fine grade sand, F95. F45 medium and F95 fine sands have mean grain sizes (d_{50}) of 323 μm and 140 μm , respectively, determined from the sieve data provided by the manufacturer. As packed for this work, the F45 medium sand had a measured porosity of 0.36, while the F95 fine sand had a measured porosity of 0.38. Prior to use, all sands were washed multiple times with pH 10 NanoPure water in order to reduce the dispersal of sand fines during drainage experiments. Fig. 1 shows measured primary drainage capillary pressure-saturation (P_c - S) relationships for the two sands used for the work, measured by weight using the tall columns described in the next section. Van Genuchten parameters for the two sands are: F45 medium sand ($\alpha = 0.04 \text{ cm}^{-1}$, $n = 14$), F95 fine sand ($\alpha = 0.0165 \text{ cm}^{-1}$, $n = 17$).

2.3. Tall-column drainage experiments

Experiments conducted for this work consisted of draining packed columns initially saturated with nanomaterial suspension, and then disassembling and analyzing the columns to determine the spatial distribution of nanomaterials retained during drainage. The columns used for the work were comprised of multiple segments of clear polyvinyl chloride plastic with lengths ranging from 2.5 cm to 25 cm, and an inner diameter of 2.5 cm. The sections of the column were assembled in different configurations to create columns up to 120 cm in height. The shortest column sections (2.5 cm) were glued together using silicone sealant, while longer sections were attached with threaded pipe adaptor unions that were sealed with integrated o-rings. Columns were packed in layers of the two sands, with interfaces between sand

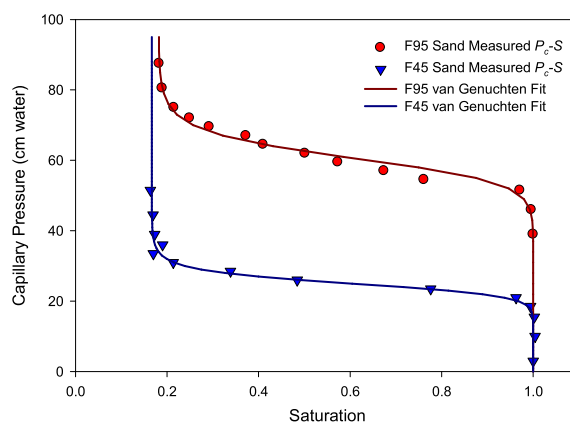


Fig. 1. Measured primary drainage P_c - S relationships for the two sands used for this work, F45 medium sand, and F95 fine sand.

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