

Effect of particle size and natural organic matter on the migration of nano- and microscale latex particles in saturated porous media

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Abstract

In the interest of fully assessing the potential environmental risks linked to “nanolitter,” we need to be able to predict the persistence, toxicity, and mobility of engineered nanomaterials in the natural subsurface environment. To examine the effects of particle size and natural organic matter on nanoparticle mobility, laboratory-scale filtration experiments were performed using different sized model nanomaterials (i.e., latex colloids having diameters of 50, 110, and 1500 nm) in the presence and absence of 5.0 mg/L Suwannee River humic acid (SRHA). At low ionic strengths (1–10 mM KCl), an increase in attachment efficiency (α) with increasing particle size was observed. This result contrasts with predictions of particle filtration based on attachment in the primary energy minimum of the particle–grain interaction energy profile evaluated using Derjaguin–Landau–Verwey–Overbeek (DLVO) theory. The presence of SRHA generally resulted in a decrease in α over the range of experimental conditions investigated. Results of particle transport experiments combined with particle characterization measurements suggest that the decrease in colloid attachment in the presence of SRHA is related to the combined influence of the mechanisms of charge stabilization and steric stabilization.

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1. Introduction

Global activities involving nanotechnologies are estimated at US\$45 billion [1] and in Quebec alone, more than \$400 million has already been invested in nanotechnology in an effort to promote research in this area. Nanomaterials are used in the fabrication of paints, coatings, cosmetics, and many other consumer goods. It is anticipated that by 2015, the global market for “nano” products will exceed US\$1 trillion, thereby affecting several economic sectors [2,3]. Although nanotechnologies promise many social and economic benefits, recent reports published in North America and Europe underline the potential environmental and public health risks linked to this “nano” revolution [1]. A portion of manufactured nanomaterials will end up in the natural environment (e.g., as a result of accidental spills or released in industrial wastewaters). A wide range of

manufactured nanoparticles have been shown to exhibit properties of increased reactivity and toxicity [1,4–6].

The impact of manufactured nanomaterials on the environment and human health will depend on their fate in natural systems. Despite the risks associated with the release of nanomaterials in natural soils and groundwaters, few studies have addressed their behavior in these systems. The few reported studies on the transport and fate of engineered nanomaterials in granular matrices have been conducted using laboratory-scale experimental systems [7–11]. In most cases, chromatography columns packed with glass beads have been used to study the filtration of various nanomaterials suspended in simple electrolytes. However, the natural subsurface environment exhibits significant physical, biological, and geochemical heterogeneities, including the presence of biofilms, natural organic matter (NOM), and changing groundwater and sediment chemistry that can influence the degree of nanoparticle migration in granular matrices. For instance, some researchers have demonstrated that adsorption of NOM onto suspended colloids and/or sediment grain surfaces generally facilitates colloid transport in the subsurface [12–14]; yet, little is known about its impact on

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the stability and migration potential of different-sized nanoparticles in groundwater environments.

Initial investigations into the mobility of nanomaterials in systems that model groundwater aquifers have shown that varying the surface functionalization of nanoparticles can lead to significant differences in their migration potential [7,11]. For example, nanoparticulate fullerene aggregates (nC_{60}) have very limited mobility in porous media while fullerol has been found to be highly mobile [11]. Findings such as these underscore the need to avoid using generalizations in the implementation of a regulatory environmental framework for protection of groundwater supplies from contamination by engineered nanomaterials. Rather, it is necessary to obtain a better mechanistic understanding of the key factors controlling nanomaterial migration over a broad range of environmentally relevant physicochemical and geochemical conditions.

Particles suspended in groundwater will contact the surface of a sediment grain (collector) as a result of three governing mechanisms: (i) interception, (ii) gravitational sedimentation, and (iii) Brownian diffusion [15,16]. The latter is of particular importance in the retention of nanoparticles as their high diffusivity leads to a higher incidence of collisions with the surface of soil grains. Classical colloid filtration theory takes into account the contribution of these three transport mechanisms to particle retention in a parameter referred to as the single-collector contact efficiency (η_0) [15,17]. However, simply making contact with a collector surface does not ensure that a particle will attach to the grain surface as various colloidal interactions and forces (e.g., electrical double layer repulsion) may prevent particle removal from the pore fluid. Hence, the actual single-collector *removal* efficiency (η) is expressed as the product of an empirical attachment efficiency (α) and the single-collector contact efficiency (η_0). The attachment efficiency represents the fraction of collisions between suspended particles and collectors that result in attachment. Currently, there are no satisfactory theoretical models to predict the attachment efficiency under conditions deemed *unfavorable* for particle attachment; namely in the presence of repulsive Derjaguin–Landau–Verwey–Overbeek (DLVO) interactions [18,19]. Attempts have been made to incorporate the classic DLVO theory into the convective-diffusion equation in an effort to predict particle deposition rates [16,20,21]. In general, though, experimentally determined particle deposition rates are orders of magnitude greater than theoretical values. Furthermore, results of particle transport and deposition experiments are often not in agreement with qualitative predictions based on DLVO theory [22–24]. For instance, some studies indicate that particle retention in granular porous media increases with particle size [23–25], whereas others have shown that particle attachment efficiency may be independent of particle size [22,26]. In contrast, consideration of *irreversible* particle deposition in the primary energy minimum of the DLVO interaction energy profile predicts a decrease in particle deposition rates with increasing particle size (under *unfavorable* conditions for deposition) [16]. Various explanations have been proposed to rationalize these discrepancies such as the surface roughness of the particles or collectors, hydrodynamic interactions, the non-homogeneity of

surface charge distribution, the dynamics of colloidal interactions, physical straining or trapping of particles, and deposition in the secondary energy minimum of the DLVO interaction energy profile [22–25]. While each of these factors likely merit attention, the current understanding of these factors precludes their inclusion in any rigorous quantitative assessment.

The general objective of this paper is to determine the effect of particle size in conjunction with that of the presence of NOM on the transport of nanomaterials in a model groundwater system. Laboratory-scale packed column experiments were used to examine the deposition behavior of model latex colloids over a broad range of solution ionic strengths (IS). Sulfate latex particles ranging in size from 50 to 1500 nm were selected as model anthropogenic nanomaterials for the transport studies revealing an increase in colloid retention with increasing particle size. In a second series of packed column experiments, the effect of NOM on the transport and retention of micro- and nano-sized colloids was investigated by including Suwannee River humic acid (SRHA; 5 mg/L) in the artificial groundwater solution. The use of different-sized colloids of similar surface chemistry allows us to examine whether the presence of NOM similarly impacts the transport and fate of nanoparticles as it does that of microscale colloids. Physicochemical properties of the particles characterized over the range of solution conditions investigated are used in the interpretation of data obtained from the packed column experiments.

2. Materials and methods

2.1. Model colloids, natural organic matter and granular media

Three different-sized (50, 110, and 1500 nm) surfactant-free polystyrene latex microspheres with sulfate functional groups were used as model colloids (Interfacial Dynamics Corporation, Portland, OR). These particles were selected for investigation because they range in size from nano- to microscale while having comparable surface charge densities ($0.9 \mu\text{C}/\text{cm}^2$). Colloid suspensions used in colloid filtration experiments were prepared by diluting stock colloidal samples in electrolyte of varying ionic strength (1–100 mM KCl in deionized MilliQ water) at a pH of 5.7 ± 0.2 .

Suwannee River humic acid (SRHA) was used as a representative organic matter (International Humic Substances Society). Humic substances represent a significant fraction of organic carbon present in aquatic systems [27] and are found in natural waters at concentrations of 0.1–200 mg/L dissolved organic carbon (DOC) [28]. In colloid deposition experiments conducted with NOM, SRHA was applied at a concentration of 5 mg NOM/L. Electrolyte solutions were adjusted to pH 5.7 ± 0.2 using 0.1 M NaOH.

High purity quartz sand (Sigma-Aldrich) was selected as a model granular material to allow careful control of the experimental system. Although not perfectly representative of natural soils, the pure quartz sand allows us to interpret the experimental results within the context of the DLVO theory of colloidal stability. Prior to use, the sand was sieved with Nylon sieves

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