



Co-transport of gold nanospheres with single-walled carbon nanotubes in saturated porous media



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ABSTRACT

Porous media transport of engineered nanomaterials (ENMs) is typically assessed in a controlled single-particulate environment. Presence of a secondary particle (either natural or engineered) in the natural environment though likely, is rarely taken into consideration in assessing ENMs' transport behavior. This study systematically assesses the effect of a secondary ENM (i.e., pluronic acid modified single-walled carbon nanotubes, PA-SWNTs) on a primary particle (i.e., gold nanospheres, AuNSs) transport through saturated porous media under a wide range of aquatic conditions (1–100 mM NaCl). AuNS hetero-dispersions (i.e., with PA-SWNTs) are transported through saturated sand columns, and the transport behavior is compared to AuNS-only homo-dispersion cases, which display classical ionic strength-dependent behavior. AuNS hetero-dispersion, however, is highly mobile with little to no ionic strength-dependent effects. This study also assesses the role of pre-coating of the collectors with PA-SWNTs on AuNSs' mobility, thereby elucidating the role played by the order of introduction of the secondary particles. Pre-existence of the secondary particles in the porous media shows enhanced filtration of primary AuNSs. However, the presence of natural organic matter (NOM) slightly increases AuNS mobility through PA-SWNT coated sand at 10 mM ionic strength. The study results demonstrate that the presence and order of addition of the secondary particles strongly influence primary particles' mobility. Thus ENMs can demonstrate facilitated transport or enhanced removal, depending on the presence of the secondary particulate matter and background solution chemistry.

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1. Introduction

Increased use of engineered nanomaterials (ENMs) makes their environmental release highly likely whether the release is intentional as remedial agents, accidental from manufacturing processes, or inevitable at the end of life of ENM-containing products (Köhler et al., 2008; Nowack et al., 2012), highly likely. Regardless of routes of release and exposure (Khin et al., 2012; Köhler et al., 2008) sediments and porous subsurface have been identified as likely environmental sinks for ENMs (Gottschalk and Nowack, 2011), in which the ENMs undergo subsequent transport and transformation (Lowry et al., 2012). Fate and transport, the primary process controlling ENM exposure and risk, is at the center of the nano-

environmental health and safety (nano-EHS) research (Petosa et al., 2010), which devotes most of its efforts to determining appropriate fate descriptors (deposition rate and efficiency or transport distances) with single-particle breakthrough studies (Petosa et al., 2010; Xin et al., 2016) or by studying deposition onto polished crystal surfaces (Chang and Bouchard, 2013; Quevedo and Tufenkji, 2009). Although a reliable prediction of ENM transport is the ultimate goal of nano-EHS studies, most, if not all, are performed in clean and controlled environmental platforms.

State-of-the-art ENM transport studies have systematically evaluated the effects of various material properties and background chemistries of a wide set of materials (Petosa et al., 2010), e.g., fullerenes (nC₆₀), carbon nanotubes (CNTs), nano-scale zero-valent iron (nZVI), titanium dioxide (TiO₂), zinc oxide (ZnO), and silver nanoparticles (AgNPs). The influence of solution chemistry [pH and Ionic Strength (Godinez and Darnault, 2011), organic matter (Thio et al., 2011)], ENM characteristics [size and size distribution

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(Phenrat et al., 2009), concentration (Phenrat et al., 2009), shape (Afrooz et al., 2013b), surface charge (El Badawy et al., 2013), synthesis method (Chowdhury et al., 2012a), surface coating (Tripathi et al., 2011), and magnetic properties (Hong et al., 2009), collector properties [grain size distribution (Mattison et al., 2011) and flow conditions (Cai et al., 2015)] on ENMs' mobility have also been reported. In addition to the aforementioned experimental investigations, efforts have been made to predict and validate ENM transport theoretically by employing clean-bed filtration theory (CFT) (He et al., 2009). However, experimental and theoretical studies of ENMs in homogeneous and single particulate systems have limited transferability to the natural environment in the presence of natural particles and other ENMs (Hotze et al., 2010).

Appreciating the need for assessment of environmental complexities, numerous studies have been conducted to delineate the effects of physical [i.e., surface roughness (Song and Elimelech, 1994) and shape (Saiers and Ryan, 2005)] and geochemical [i.e., surface charge (Song et al., 1994), surface coating (Joo et al., 2009), and mineralogical compositions (Kim et al., 2012)] heterogeneities of the collectors on ENM transport. Furthermore, the effect of pre-coating (Joo et al., 2009; Tong et al., 2010) the collector surfaces with natural organic matter (NOM), clay, or biofilm has also been evaluated, where ENM mobility is reported to decrease due to the competition between the ENMs and the NOMs for available attachment sites or to increase due to the availability of increased surface sites of the secondary particle coated collectors for NM deposition (Chowdhury et al., 2012a; Tong et al., 2010). The altered nanomaterials transport behavior in the presence of secondary entities necessitates further studies to assess the effects of environmental complexities on porous media transport of ENMs.

Altered interfacial interaction of the ENMs may also result from the co-existence of multiple particulate entities in the column influent suspension (Afrooz et al., 2013a; Cerbelaud et al., 2008; Huynh et al., 2012). These studies were performed with aquatic suspensions to understand their aggregation behavior under certain ionic strength conditions, but do not provide any information regarding the sub-surface transport processes of ENMs. ENM co-transport studies are needed to better understand of the influences of secondary entities; i.e., ENMs, bio-particles, or natural nanoparticles (co-dispersed with the primary ENM or pre-deposited on collector surfaces), on the transport of the primary particles through porous media. Thus far, ENM co-transport literature primarily assessed the effects of bacterial presence on ENM transport behavior; e.g., TiO_2 transport was observed to have enhanced in a co-transport scenario compared to that of TiO_2 -only case (Chowdhury et al., 2012a; Wang et al., 2012). Reduced bacterial mobility in the presence of CNT has also been reported (Yang et al., 2013a) at an elevated ionic strength (25–100 mM). Additionally, the effect of natural nanomaterials (NNM) on ENM transport has been reported (Wang et al., 2015): goethite nanoparticle showed an enhanced mobility of nano-hydroxyapatite at circumneutral pH due to heteroaggregation between the two particles. Such evidences suggest that ENM transport through porous media is strongly influenced by the presence of a secondary particulate entity, the likely mechanism being higher collision and facilitated filtration.

However, transport studies with multiple ENMs are scarce in ENM fate and transport literature. To-date, only two such studies have been reported: TiO_2 transport with multiwalled carbon nanotubes (Wang et al., 2014) and co-transport of TiO_2 and nC_{60} (Cai et al., 2013). These studies reported significant differences in the mobility of the primary TiO_2 in singular and binary matrices (in the presence of secondary nC_{60} s or CNTs) with strong pH and ionic strength dependence. Similarly, when TiO_2 was used as a secondary particle, transport of CNTs decreased under all examined

conditions. However, both of these studies used matrices in which primary and secondary particles would undergo simultaneous deposition at higher ionic strength (i.e., representing brackish or saline condition); thus they preclude enumeration of underlying transport mechanisms over a wide range of ionic strength. Therefore, contribution of the reported studies is restricted to low ionic strength (0.1–10 mM) conditions with limited environmental relevance. To summarize, mechanistic co-transport studies appear to be a persistent data gap in the ENM fate and transport literature, and the role of a wide range of environmentally relevant ionic strength conditions, presence of NOM, and the order of addition of the secondary particles need to be systematically assessed.

The objective of this study is to investigate transport of a model nanoparticle, i.e., gold nanospheres (AuNSs), in the presence of a well-characterized secondary ENM, i.e., single-walled carbon nanotubes (SWNTs). This study contributes to addressing the existing data gaps by providing a mechanistic understanding of ENM co-transport behavior over a wide range (1–100 mM) of ionic strength for a new ENM duo (AuNS-SWNT). In order to mechanistically assess the filtration mechanisms, especially at elevated ionic strengths, inherent SWNT-SWNT interaction is eliminated via nonionic pluronic acid (PA) functionalization (Afrooz et al., 2013a). A range of PA concentrations is utilized to generate PA-SWNTs with desirable stability, i.e., highly stable suspensions are used for co-transport studies, whereas, SWNTs with lower stability are used to pre-coat the collector surfaces. Co-transport under a wide range of ionic strength (1–100 mM NaCl) is studied and compared with primary AuNS transport. The effect of NOM on AuNS transport is also monitored for typical groundwater conditions (i.e., 10 mM total ionic strength). This is a first-of-its-kind study to mechanistically assess co-transport of AuNSs (in the presence of another ENM) over a wide range of ionic strengths. The results from this study can be considered the initial step to attain a more reliable assessment of ENM co-transport through porous media in cases of multiple ENMs' presence in an aquatic environment.

2. Materials and methods

2.1. Preparation of AuNS suspensions

Poly (acrylic acid) or PAA-coated AuNS suspension at a concentration of 50 mg/L was procured from Nanopartz Inc. (Loveland, CO). The supplier reported 10 nm nominal diameter for the AuNSs. Procured AuNSs were diluted 4 times to achieve a concentration of 12.5 mg/L for all transport experiments.

2.2. Preparation of PA-SWNTs

Dry SWNTs, produced by high-pressure carbon monoxide disproportionation or HipCO method (Bronikowski et al., 2001) was obtained from Tubes at Rice (Houston, TX). The supplier reported that average diameter and length of the SWNTs were 1.03 nm (a range of 0.7–1.45 nm) and 0.93 μm (a range of 0.1–2.6 μm), respectively. Commercially available pluronic acid (PA) F-127 was obtained from Sigma Aldrich (St. Louise, MO); and 0.02, 0.1, 1, and 5% (w/v) PA solution was prepared using filtered (0.45 μm inorganic membrane filters, VWR international LLC, Radnor, PA) deionized (DI) water (Millipore, Darmstadt, Germany). The PA solutions were prepared by magnetic stirring for 1 h at room temperature.

10 mg of procured SWNTs was added to 100 mL (0.02, 0.1, 1, and 5%) of PA solution and sonicated in an ice bath for 1 h employing an S-4000 ultrasonicator (Misonix, Inc. Farmingdale, NY) at a controlled input energy of 100–105 kJ. The sonicated samples were quiescently left aside for 24 h and the supernatant was decanted and centrifuged at 12,000 \times g for 1 h with a Beckman Coulter (Brea,

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