



Effects of *Escherichia coli* and phosphate on the transport of titanium dioxide nanoparticles in heterogeneous porous media

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ABSTRACT

Transport behaviors of titanium dioxide nanoparticles (nTiO₂) were examined in the individual- and copresence *Escherichia* (*E.*) *coli* and phosphate in heterogeneous sand (uncoated and iron oxyhydroxide-coated sand) columns. The results showed that for the individual presence of phosphate, the degree of nTiO₂ deposition was less in uncoated than in iron oxide-coated sands. In contrast, an opposite trend that greater deposition of nTiO₂ in uncoated than in coated sands occurred in the individual presence of *E. coli*. These observations are due to the phosphate adsorption changing the charge of NPs and iron oxyhydroxide-coated sand, or the preferential adhesion of bacterial to coated sand. In the copresence of *E. coli* and phosphate, interestingly, the phosphate level plays an important role in influencing nTiO₂ transport. At a high phosphate concentration (>1.0 mM), the deposition of nTiO₂ with the individual presence of *E. coli* was stronger than nTiO₂ in the copresence of both *E. coli* and phosphate, regardless of sand type. The potential mechanism was that phosphate adsorption led to the formation of more negatively charged NPs-bacteria complexes that have higher mobility in sand columns. At a low phosphate level (≤0.1 mM), a similar observation occurred in uncoated sand. Nevertheless, the deposition of nTiO₂ with copresence of *E. coli* and phosphate was greater than nTiO₂ with *E. coli* in oxyhydroxide-coated sand. It was attributed to the formation of large NPs-bacteria-phosphate clusters (less mobile) and the preferential adhesion of *E. coli* cells to iron oxyhydroxide coating simultaneously. Taken together, our findings provide crucial knowledge for better understanding the fate, transport, and potential risks of engineered nanoparticles in complicated environmental settings where bacteria and phosphate are ubiquitous.

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1. Introduction

Upon extensive application of nano-titanium dioxides (nTiO₂)-incorporated commercial products (e.g., photocatalysts, coatings, cosmetics, sunscreens, and ceramic membranes) (Chen et al., 2011; Thio et al., 2011), nTiO₂ are (un)intentionally entering the subsurface environment increasingly from various sources in diverse pathways (Robichaud et al., 2009). It is estimated that nTiO₂ may reach the highest concentration in surface waters (e.g., 21 ng L⁻¹) compared to other environmental settings (Gottschalk et al., 2009). Numerous studies demonstrate that nTiO₂ are toxic to

phytoplankton, microbes, and fish, thereby posing a potential threat to the aquatic ecosystems and human health (Farré et al., 2009; Scown et al., 2010; Miller et al., 2012; Lin et al., 2014). To reliably evaluate the potential risks of nTiO₂ to the environment, it is essential to understand the fate and transport including colloidal stability of nTiO₂ in natural ecosystems, and thus evaluate their persistence and bioavailability in aquatic environments.

In aquatic systems, nTiO₂ generally exist in the form of aggregates, and the aggregation affects the stability and mobility of nTiO₂ (Wu and Cheng, 2016). Aggregation and transport of nTiO₂ are documented to be influenced by physical (e.g., flow velocity, and grain size) (Chowdhury et al., 2011; Chen et al., 2015), chemical (e.g., pH, ionic strength (IS), and natural organic matter) (French et al., 2009; Chen et al., 2012; Fang et al., 2013), and biological factors (such as bacteria and extracellular polymeric substance)

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(Chowdhury et al., 2012). Porous media type and composition also determine the transport potential of nTiO₂ (French et al., 2009; Godinez and Darnault, 2011; Wang et al., 2011a). For example, surface charge heterogeneity on natural granular media could inhibit nanoparticles (NPs) transport (Johnson et al., 1996). The most common surface charge heterogeneities encountered in groundwater and soils are patches of iron and aluminum oxides that are normally positively charged at ambient pH conditions (Lin et al., 2011; Wang et al., 2012a; Han et al., 2014), due to their high points of zero charges (pH_{PZC}) (e.g., the pH_{PZC} values of iron and aluminum oxyhydroxides are 7.5 and 9.0, respectively) (Parks, 1965). Thus, collector grains covered with more positive charged iron and aluminum oxides affect the transport and retention of engineered NPs, which is sensitive to solution pH (Wang et al., 2012a).

Bacteria, which occur ubiquitously in soils and groundwater environments, can alter the surface properties and surface interactions of colloidal NPs and collectors via the adsorption processes (Vasiliadou and Chrysikopoulos, 2010; Chowdhury et al., 2012; Yang et al., 2013; Aal et al., 2015), thus influencing the fate and transport of NPs (Chowdhury et al., 2012). For example, *Pseudomonas aeruginosa* was found to stabilize nTiO₂ agglomerates by sorption of NPs onto cell surfaces (Horst et al., 2010). The presence of natural organic matter (NOM), nutrient conditions, and sulfate enhanced the cell transport in quartz sand, which may due to the competition of retention sites (e.g., humic acid or nutrient conditions) (Yang et al., 2012; Han et al., 2013; Shen et al., 2013). Comparably, the presence of extracellular polymeric substances (EPS) on cell surfaces enhanced cell deposition in porous media (Tong et al., 2010). Overall, interaction of nTiO₂ with bacterial cells can alter NPs fate and transport (increased or decreased mobility), depending on NPs properties (e.g., size, shape, surface chemistry, and coating), bacterial type, and solution chemistry (e.g., pH, ionic strength, and presence of natural organic matter). In addition to bacteria, phosphorus (P) is another major component in surface and groundwater due to the widespread application of phosphate-containing fertilizers or sewage sludge (Li and Schuster, 2014). Our previous study showed that phosphate favors the stability and transport of NPs (Chen et al., 2015; Liu et al., 2017), due to phosphate adsorption onto both NPs and collector surfaces (Li and Schuster, 2014; Chen et al., 2015). Phosphate can also attach to the surface of bacteria (Wang et al., 2011b), and may play different roles in bacterial interaction with porous media having different degrees of surface charge heterogeneity (e.g., cleaned and iron-coated sands), depending on its concentration (Park et al., 2009). Therefore, their interlaced reactions and adsorption mechanisms might trigger complicated transport behaviors of NPs when *E. coli* and phosphate co-present with the NPs. However, till now, very limited information is available regarding how and to what extent bacteria and phosphate influence the colloidal stability and transport of nTiO₂ in the presence of surface charge heterogeneity on collectors.

The overall objective of this study was therefore to systematically investigate the effects of individual- and co-presence of bacterial and phosphate on the colloidal stability and transport behavior of nTiO₂ in water-saturated sands with different degrees of surface charge heterogeneity (uncoated and iron oxyhydroxide-coated sand). Numerical modeling based upon the two-site kinetic attachment model (TSKAM) was employed to simulate the transport and unravel the mechanisms behind nTiO₂ transport in porous media.

2. Materials and methods

Anatase nTiO₂ (20 nm) were purchased from Gaoquan Chemical,

Inc. (Shanghai, China) with >99% purity. Physicochemical properties of the nTiO₂ were characterized in our previous study. Briefly, nTiO₂ exists as large agglomerates with some fine particles, and have a specific surface area of 103.47 m² g⁻¹ (Chen et al., 2015). A Gram-negative strain *Escherichia coli* (*E. coli*) was used as a representative bacterial in this study. *E. coli* (CICC 23,675) was obtained from the China Center of Industrial Culture Collection (CICC). The characteristics and procedures for culturing the *E. coli* were given in Supporting Information (SI). The phosphate solution was diluted from 1.0 MP stock solution which was prepared by dissolving NaH₂PO₄ (analytical grade) in deionized (DI) water. Quartz sands (~600 μm in average size) were obtained from Sinopharm Chemical Reagent Co., Ltd. (China).

2.1. Preparation of nTiO₂ influent suspensions

Stable aqueous nTiO₂ suspension at environmentally relevant NaNO₃ concentrations (0.1–10 mM) was mixed with 0.1–10 mM phosphate solutions or *E. coli* in single and binary systems of phosphate and *E. coli*, yielding final concentrations of nTiO₂ and *E. coli* of 1.0 g L⁻¹ and 2 × 10⁸ cfu mL⁻¹, respectively. Since the predominant species of phosphate are H₂PO₄⁻ and HPO₄²⁻ at pH 6.0 and 9.0, respectively, the pH of nTiO₂ suspension was adjusted to 6.0 and 9.0 using HCl or NaOH solution. The nTiO₂ suspension was stirred on a magnetic stir for 1 min, and then sonicated in a water bath for 30 min (KQ2200B, Kunshan Ultrasonic Instrument Co. Ltd., China) at room temperature to ensure a homogeneous suspension. The detailed physicochemical conditions for column transport experiments were summarized in SI (Table S1). Please note that, the used nTiO₂ concentration (1.0 g L⁻¹, i.e., 0.1%) is higher than those commonly encountered in the subsurface environments, but it can occur in scenarios such as accidental spills or in-situ nanoremediation of contaminated sites. Also, the method for determining nTiO₂ concentration in column experiments—diantipyryl methane photometric method requires an optimal concentration (~0.1%) for accurate quantification (described below).

2.2. Adsorption isotherms of phosphate on *E. coli*

Adsorption isotherms were determined to further understand the adsorption of phosphate on *E. coli* by adding 2 × 10⁸ cfu mL⁻¹ *E. coli* cells to series of phosphate solutions (0–600 μM) in 50 mL vials at pH 6.0. The 0–600 μM phosphate solutions were obtained by diluting the phosphate stock solution. The suspension pH was adjusted using HCl or NaOH solution. The vials were then placed on a rolling incubator (shaking rpm of 30). When adsorption equilibrium is achieved (2 h), the suspensions were centrifuged at 17,800 × g for 90 min, and then passed through 0.22 μm pore size filters. Soluble P concentration in the filtrates was analyzed using the molybdenum blue method via colorimetry (Xu et al., 2014). The amount of phosphate adsorbed onto *E. coli* cells was then determined by the difference between the initial and equilibrium phosphate concentration in filtrates. Triplicate experiments were performed for phosphate adsorption onto *E. coli*.

2.3. Column transport experiments

Quartz sands were chosen as granular media in column studies. Prior to use, the sand was cleaned thoroughly using a procedure described elsewhere (Chen et al., 2015). Iron oxide-coated sand was prepared using a procedure described in Stahl and James (1991), and the details are described in SI.

Transport experiments were conducted in 20-cm long and 2.6-cm inner diameter glass chromatography columns. After dry-packing with uncoated or 100% iron oxide coated quartz sands,

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