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Individual, co-transport and deposition of TiO_2 and ZnO nanoparticles over quartz sand coated with consortium biofilm

Jyoti Kumari[a], Natarajan Chandrasekaran[a], R. Nagarajan[b], Amitava Mukherjee[a,*]

[a] Centre for Nanobiotechnology, VIT University, Vellore, Tamil Nadu, India
[b] Department of Chemical Engineering, IIT Madras, Chennai, India

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ABSTRACT

The transport of TiO_2 and ZnO nanoparticles was assessed using column experiments in a porous media containing biofilm. In order to understand the influence of bacteria encountered in the natural aquatic environment when nanoparticles are introduced in the surroundings, nanoparticles transport has been studied through quartz coated with a biofilm composing of a consortium of *Pseudomonas aeruginosa* (Gram-negative), *Bacillus alitudinis* and *Bacillus subtilis* (Gram-positive). The influence on individual and co-transport mobility and deposition kinetics of TiO_2 (10, 15 mg/L) and ZnO (5, 10 mg/L) nanoparticles based on variations in ionic strength was also determined. Various environmentally related ionic strengths were studied for the experiment using NaCl (0.1–10 mM), by keeping the pH fixed to pH 7.0. At pH 7.0, TiO_2 NPs transport declined, when the ZnO nanoparticles were present, whereas, enhanced ZnO nanoparticles transport was observed in the presence of TiO_2 nanoparticles. The solution chemistries of the nanoparticles can be inferred from the breakthrough curves (BTCs), wherein an increase in ionic strength results in a decline in the BTC plateau. The inverse of BTC plateaus provided the retention profiles, which could also be anticipated from the mass balance contemplation. Altogether, the upshot of this work explains the transport behavior of both the nanoparticles in biofilm coated sand depends on suspension chemistries (pH and ionic strength) are likely the primary factors that control. Further research could thus be focused on enhancing our knowledge of the basic mechanisms governing nanoparticles transport and fate in typical biofilm containing aquatic environment.

1. Introduction

Diverse reports have indicated the noxious nature of TiO_2 NPs to organisms like microbes, invertebrates, algae, fish, and rat. The manufacture of metal oxide nanoparticles like TiO_2 NPs has increased substantially, being used in several consumer products and industrial applications, such as water treatment, cosmetics, and medicine [1]. Hence, these nanoparticles may inevitably reach the aquatic ecosystem, causing undesired consequences. Moreover, reports suggest the presence of 10–100 mg/L of Ti in waste water effluents [2]. Therefore, by comprehending the transport and distribution of TiO_2NPs in the natural environment, the potential ecological risk posed by them can be quantitatively assessed.

The substantial exploitation of ZnO NPs in various industries and commercialized outcomes like chemicals and cosmetics make them promising [3,4]. The direct and indirect discharge of ZnO NPs in the environment are an outcome of these applications. The ZnO NP production yearly has fattened to about 30,000 t/year globally (http://www.researchandmarkets.com) mostly due to their manifold applications [5,6] several contemporary works suggest that ZnO NPs can trigger acute toxic effect [7] and apoptosis in mammalian cells [8,9], phytotoxicity to herbage roots [10,11], etc. Thus, the nanoparticles fate and behavior in the natural ecosystem dictates their probable health and environmental risks [12]. Upon discharge, these nanoparticles release into soil and groundwater [13]. Thus, it is utmost exigent to perceive the transport and distribution profile of ZnO NPs in order to evaluate the probable ecological hazard in a natural porous media system. A considerable amount of studies has been done before on nanoparticle transport and retention in porous media [14]. These reports mostly focus on the transport and retention of one type of nanomaterials. However, there are very high chances for the conjoint entry of various nanoparticles types in the environment due to their broad applications. Co-transport of entities like virus, bacteria, and clay

* Corresponding author at: Centre for Nanobiotechnology, VIT University, Vellore 632014, India.
E-mail addresses: amit.mookerjea@gmail.com, amitav@vit.ac.in (A. Mukherjee).

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have demonstrated the complicated nature of co-transport in comparison to individual entity transport behavior, thus, necessitating the evaluation of nano-material co-transport [15,16].

Mineral surfaces in the environment, generally, tend to be coated by biofilm layers, comprising of microorganisms surrounded by their self-secreted matrix [17]. Bacteria deposition in porous media has been found to enhance in presence of biofilm [4,18], and the presence of biofilm in turn resulted in higher retention profiles for C60 nanoparticles [19]. In porous media, the transport of zero valent FeO NPs coated with acrylic acid were found to be dependent on the biofilm formation [20]. Another study suggested the influence of culture medium, as well as the biofilm and their EPS on the behavior of 4 nanoparticles, and the increased nanoparticles deposition in presence of biofilm could be observed irrespective of the nanoparticles size [21].

Hence, the current study aims to investigate the transport behavior of TiO_2 and ZnO NPs (both individual and co-transportation) at pH 7.0 at several environmentally relevant ionic strengths (NaCl 0.1–10 mM) using biofilm-packed column experiments. NaCl was used in solution as electrolyte for the current study. The resulting breakthrough curves (BTCs) and retention form have been compared for individual and co-transport of nanoparticles. This is the first-ever report on the co-transport of two important engineered nanoparticles, namely TiO_2 and ZnO NPs, in a consortium-coated sand column, and the study will be useful to assess the potential hazards posed by these nano-materials.

2. Materials and methods

2.1. TiO_2 and ZnO NPs suspension preparation

Titanium dioxide (anatase) NP powder (diameter: <25 nm; purity: 99.9%) and ZnO NPs (<100 nm) were procured from Sigma. For preparation of a stock suspension (100 mg/L) of TiO_2 nanoparticles, the TiO_2 nanopowders were suspended in deionised water and sonicated with the help of an ultrasonicator (Sonics., USA) for 10 min, at 130 W. For ZnO NPs aqueous suspensions (100 mg/L), ZnO nanopowders were suspended in deionised water and sonicated at 530 W for 20 min using a bath sonicator (Crest Ultrasonics).

NaCl solution was used to vary the ionic strengths of NPs suspensions from 0.1 to 10 mM. Using 0.1 M HCl or 0.1 M NaOH, the suspension pH was adjusted to pH 7.0 [22]. Under the above-mentioned conditions, the zeta potential measurements of nanoparticles and biofilm were studied at room temperature (25–30 °C) using Brookhaven Instruments Corporations, USA, and the values were checked repeatedly.

2.2. Porous media (quartz)

Quartz sand for the nanoparticles transport experiment has been procured from Sigma-Aldrich. Cleaning of the quartz sand for column study was performed as described by Cai et al. [17]. Under the experimental conditions, the zeta potential measurements of the consortium biofilm were done in NaCl.

2.3. Column studies

Biofilm-coated porous media were wet-packed in glass columns of 10-cm length and 2-cm inner diameter. Briefly, before packing, the rehydration of cleaned quartz sand was done for at least 1 h in boiling deionized water. Throughout the experiment, the temperature was 27 ± 5° C. To release the nanoparticles from quartz sand, approximately 5–10 mL of 0.1 M NaOH/$CaCl_2$ solution was added to each sediment segment, and the mixture was continuously shaken at 300 rpm overnight, and then manually shaken vigorously for a few seconds. The concentration of effluent samples and supernatant samples from the recovery of retained TiO_2 NPs and ZnO NPs were evaluated by UV spectrophotometer (U2910, Hitachi, Japan) at 330 and 370 nm. The schematic diagram of individual and co-transport study of TiO_2 NPs and ZnO NPs in consortium coated sand is explained in Fig. 1.

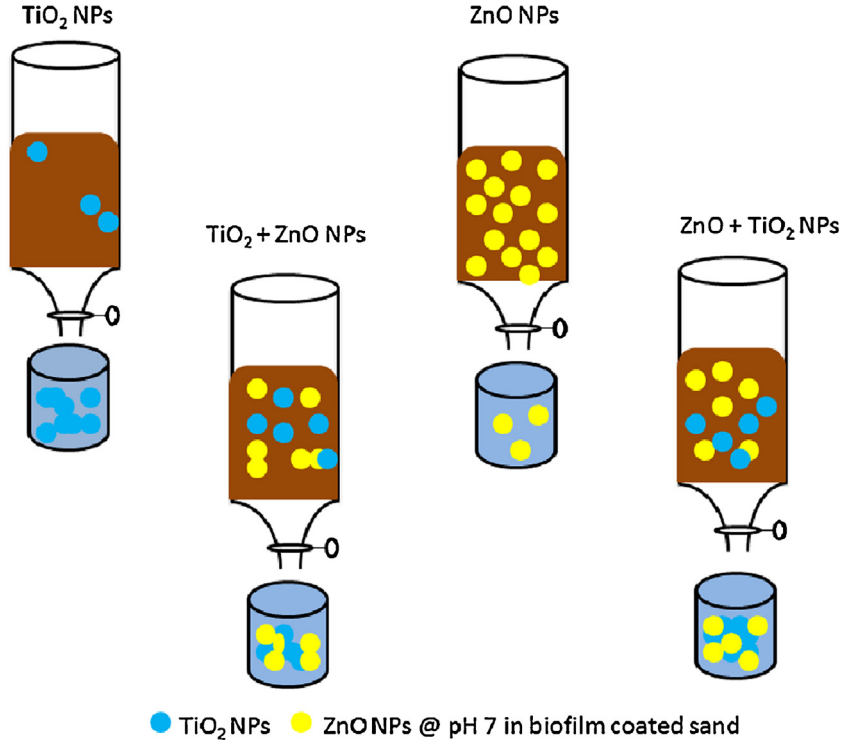

Fig 1. Schematic diagram of individual and co-transport study of TiO_2 and ZnO NPs consortium coated sand column.