



Transport and retention of surfactant- and polymer-stabilized engineered silver nanoparticles in silicate-dominated aquifer material[☆]

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ABSTRACT

Packed column experiments were conducted to investigate the transport and blocking behavior of surfactant- and polymer-stabilized engineered silver nanoparticles (Ag-ENPs) in saturated natural aquifer media with varying content of material < 0.063 mm in diameter (silt and clay fraction), background solution chemistry, and flow velocity. Breakthrough curves for Ag-ENPs exhibited blocking behavior that frequently produced a delay in arrival time in comparison to a conservative tracer that was dependent on the physicochemical conditions, and then a rapid increase in the effluent concentration of Ag-ENPs. This breakthrough behavior was accurately described using one or two irreversible retention sites that accounted for Langmuirian blocking on one site. Simulated values for the total retention rate coefficient and the maximum solid phase concentration of Ag-ENPs increased with increasing solution ionic strength, cation valence, clay and silt content, decreasing flow velocity, and for polymer-instead of surfactant-stabilized Ag-ENPs. Increased Ag-ENP retention with ionic strength occurred because of compression of the double layer and lower magnitudes in the zeta potential, whereas lower velocities increased the residence time and decreased the hydrodynamics forces. Enhanced Ag-ENP interactions with cation valence and clay were attributed to the creation of cation bridging in the presence of Ca²⁺. The delay in breakthrough was always more pronounced for polymer-than surfactant-stabilized Ag-ENPs, because of differences in the properties of the stabilizing agents and the magnitude of their zeta-potential was lower. Our results clearly indicate that the long-term transport behavior of Ag-ENPs in natural, silicate dominated aquifer material will be strongly dependent on blocking behavior that changes with the physicochemical conditions and enhanced Ag-ENP transport may occur when retention sites are filled.

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1. Introduction

Engineered silver nanoparticles (Ag-ENPs) are among the most widely used nanoparticles in consumer products due to their antimicrobial properties (Maillard and Hartemann, 2013; Zhang et al., 2016). Their use has among others been documented for textiles, paints, cleaning agents, electrical appliances, medical

technology and cosmetics (Goswami et al., 2017 for review). Ag-ENPs can be released into the environment during manufacturing processes, product distribution, product use, and disposal (Farkas et al., 2011; Kaegi et al., 2010; McGillicuddy et al., 2017). Once in the environment Ag-ENPs can become bioavailable (Navarro et al., 2008) and are potentially toxic or detrimental to a wide variety of organisms (Andrei et al., 2016; Juganson et al., 2017; Katsumiti et al., 2015; Makama et al., 2016; Malleve et al., 2014; Siller et al., 2013; Zuykov et al., 2011).

One major route for environmental release is through wastewater (Nowack and Bucheli, 2007; Nowack et al., 2012; Troester

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et al., 2016). Ag-ENPs can be retained in activated sewage sludge in wastewater treatment plants (Kaegi et al., 2011, 2013; Maier et al., 2012), which is then used as agricultural fertilizer or disposed of at landfills (Blaser et al., 2008). Ag-ENPs can subsequently be leached into the vadose zone and transported toward groundwater. Groundwater can exhibit variations in solution ionic strength (IS) because of changes in aquifer material composition, temporal variations in recharge, groundwater-surface water interactions, increased salinization, and mixing of groundwater of different age and compositions. In many countries, groundwater is a major source of drinking water serving millions of people (Danish Ministry of the Environment, 2013; Federal Statistical Office of Germany, 2013; United States Environmental Protection Agency, 2008). Thus, understanding Ag-ENP transport processes in the saturated zone is essential to the protection and future use of groundwater resources.

Colloid and nanoparticle transport studies have typically been conducted under clean bed conditions using highly idealized porous media and solution chemistries (Flory et al., 2013; Kanel et al., 2007; Lin et al., 2011; Liu et al., 2009; Mattison et al., 2011). For example, glass beads and/or coarse textured sand that have been rigorously cleaned and monovalent electrolyte solutions are commonly employed in these studies (Bradford et al., 2002; Lin et al., 2011; Liu et al., 2009). Clean bed conditions imply a constant rate of retention and an infinite retention capacity (Yao et al., 1971). Colloid filtration theory (CFT) is commonly employed to characterize colloid and nanoparticle retention under clean bed conditions (Bayat et al., 2015; Park et al., 2016). Filtration theory describes retention as a first-order process that depends on the rate of mass transfer (diffusion, interception and sedimentation) from the bulk aqueous phase to the solid surface, and a sticking or collision efficiency that is controlled by adhesive interactions between the nanoparticle and solid (Tufenkji and Elimelech, 2004a; Yao et al., 1971). Nanoparticle transport studies that apply CFT typically assume that lower effluent concentrations imply less risk of contamination (Tufenkji and Elimelech, 2004b). It is demonstrated in this work that this assumption is inaccurate in many instances for Ag-ENPs.

In contrast to the CFT assumption, soil and aquifer material always have a finite retention capacity (Johnson and Elimelech, 1995; Leij et al., 2015; Sasidharan et al., 2014; Torkzaban et al., 2012), and the retention rate coefficient, therefore, decreases over time as available retention sites fill (also referred to as blocking). Retention is completely eliminated when all of the available retention sites are filled, and the retained solid phase concentration reaches a maximum, thus enhancing the long-term potential for Ag-ENP transport (Leij et al., 2015). Langmuirian (Adamczyk et al., 2013) and random sequential adsorption blocking (Johnson and Elimelech, 1995) models assume that the retention rate coefficient decreases in a linear and nonlinear manner with increasing retention, respectively. This has been observed not only for Ag-ENPs but also for other colloids (e.g. Kuhn et al., 2000). An alternative approach to account for nonlinear blocking dynamics is to employ Langmuirian blocking on two retention sites, with separate retention and release rate coefficients and maximum retention capacities on each site (Sasidharan et al., 2014).

The rate of filling of retention sites increases with the input concentration, input pulse duration, the clean bed retention rate and decreasing values of the maximum solid phase concentration (Leij et al., 2015). In addition, the retention rate and the maximum solid phase concentration are strong functions of many physico-chemical variables (Sasidharan et al., 2014). Consequently, the selection of experimental conditions can have a large impact on whether or not blocking is apparent in transport experiments. Previous research on Ag-ENP transport has focused on the

determination of retention rate coefficients for varying physico-chemical conditions, including grain size (Liang et al., 2013b), water velocity (Braun et al., 2015), solution IS (Liang et al., 2013b; Wang et al., 2014) and pH (Flory et al., 2013), ionic composition (Liang et al., 2013a), organic matter (Hou et al., 2017; Mitzel and Tufenkji, 2014; Park et al., 2016), or stabilizing agent for the Ag-ENPs (El Badawy et al., 2013). These studies typically were not designed to investigate blocking dynamics that control the long-term potential of Ag-ENP transport. A limited number of studies have examined the transport and retention of Ag-ENPs in natural soils (Cornelis et al., 2013; Liang et al., 2013a; Sagee et al., 2012; Wang et al., 2014, 2015; Mäkelä et al., 2017). Some of these studies neglected the influence of blocking altogether (Cornelis et al., 2013; Sagee et al., 2012), whereas those that did consider blocking (Liang et al., 2013a; Wang et al., 2014; Wang et al., 2015) did not systematically investigate the role of stabilizing agent type, small amounts of silt and clay in silicate-dominated aquifer material, and the coupling of these factors with monovalent and divalent cations, IS, and water velocity. These gaps in knowledge are addressed in this research.

The objective of our work was to study the transport and blocking behavior of surfactant- and polymer-stabilized Ag-ENPs in silicate-dominated aquifer material, a material that can be found around the world in many unconsolidated aquifers essential to drinking water supply. To achieve this objective, we conducted laboratory column experiments and employed analytical methods (inductively coupled plasma mass spectrometry, dynamic light scattering) as well as numerical modeling techniques. During the transport experiments, we specifically focused on situations that are more representative of natural conditions than previous studies (Hou et al., 2017; Li et al., 2008; Mattison et al., 2011; Mitzel and Tufenkji, 2014), including (i) IS and major cation type (Ca^{2+} or Na^{+}); (ii) stabilizing agent (polymer or surfactant) for Ag-ENPs in the absence and presence of fine particle size fractions (<0.063 mm); and (iii) changing flow velocities in the column.

2. Materials and methods

2.1. Electrolyte solutions and aquifer material

All electrolyte solutions were prepared using ultrapure water (Merck Millipore Milli-Q 18.2 M Ω -cm, TOC: 1–2 ppb, 0.22 μm membrane filter) that was unbuffered with a pH of 5.8–6.2. Solutions of selected IS and composition were prepared by adding NaNO_3 (Sigma Aldrich, St. Louis, Missouri, USA) or $\text{Ca}(\text{NO}_3)_2 \cdot 4\text{H}_2\text{O}$ (Merck KGaA, Darmstadt, Germany) to the ultrapure water.

The silicate-dominated aquifer material was collected in Dormagen-Gohr, NRW, Germany from a depth between 16 and 19.5 m below surface during the drilling of a groundwater monitoring well. The shallow unconfined aquifer at that site consists mainly of unconsolidated fine to coarse Quaternary sands and gravels deposited during the Saale glaciation (Pleistocene). A sample of the aquifer material (0–2 mm grain size) was oven-dried at 40 °C, and the mineral and clay composition were determined by means of X-ray powder diffraction (XRD) using a Siemens D5000 (Table S1). The oven-dried aquifer material was also sieved into grain size fractions of 1–2, 0.5–1, 0.025–0.5, 0.125–0.25, 0.063–0.125, and <0.063 mm to obtain a representative grain size distribution (Fig. S1) with a median grain diameter of 0.7 mm. Hydrometer analysis was carried out on the fraction <0.063 mm to determine silt (fraction of 0.002–0.063 mm) and clay (<0.002 mm) percentages, and to recover them for studies examining their influence on Ag-ENP transport. The pH of the soil solution was measured according

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