



# The attachment of colloidal particles to environmentally relevant surfaces and the effect of particle shape



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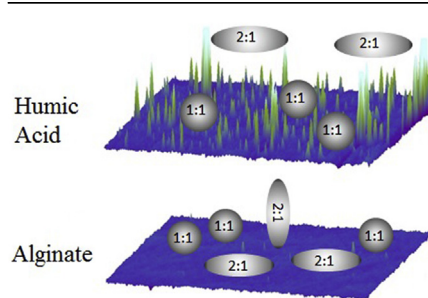
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## HIGHLIGHTS

- Colloid attachment onto NOM surfaces explored using QCM-D.
- NOM surfaces characterized with AFM.
- Particle attachment onto rough NOM dependent upon particle shape.
- Interaction energy of stretched particles dependent upon particle-surface orientation.

## GRAPHICAL ABSTRACT



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## ABSTRACT

Despite the prevalence of nonspherical colloidal particles, the role of particle shape in the transport of colloids is largely understudied. This study investigates the attachment of colloidal particles onto environmentally relevant surfaces while varying particle shape and ionic strength. Using quartz crystal microbalance and atomic force microscopy measurements, the role of particle shape was elucidated and possible mechanisms discussed. The attachment of both spherical and stretched polystyrene colloidal particles onto a smooth alginate-coated silica surface showed qualitative agreement with DLVO theory. Attachment onto a Harpeth humic acid (HHA) surface, however, significantly deviated from DLVO theory due to its high surface heterogeneity and extended confirmation from the silica surface. This extended confirmation provided increased potential for spherical particle entanglement, while the enlarged major axis of the stretched particles hindered their ability to attach. As ionic strength increased, the HHA layer condensed and provided less potential for spherical particle entanglement and therefore the selectivity for spherical particle attachment vanished. The findings presented in this study suggest that colloidal particle shape may play a complex and important role in predicting the transport of colloidal particles, especially in the presence of natural organic matter-coated surfaces.

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## 1. Introduction

Colloidal transport through the subsurface environment is a topic of great importance and intense study to the field of environmental engineering. Seemingly immobile toxins can attach to the surface of mobile colloids and be readily transported through

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the subsurface, greatly enhancing their risk to ecosystems and human health, or in some cases, the colloid itself is the toxin of concern (Ward et al., 1985). Many natural colloids are nonspherical in shape, including plate and rod shaped clay (Degueldre et al., 1996), various bacterial shapes (e.g. ellipsoid, ovoid, and rod shaped) (Hirsch and Rades-Rohkohl, 1990; Balkwill et al., 1989), and some engineered nanomaterials (e.g. carbon nanotubes) which exhibit an aquatic toxic potential (Nel et al., 2006). Despite the ubiquity of nonspherical colloids, only a limited number of studies have focused on the role of shape with regard to colloidal transport (Seymour et al., 2013; Baltus et al., 2008; Wang et al., 2008; Salerno et al., 2006; Weiss et al., 1995).

Weiss et al. (1995) demonstrated the systematic effect of bacterial cell shape on the transport of bacterial cells in packed columns and Salerno et al. (2006) observed higher retention of polystyrene latex microspheres with increasing aspect ratio, illustrating the direct impact of particle shape on retention in porous media. Furthermore, two additional studies Baltus et al. (2008); Wang et al. (2008) examined the microfiltration of various types of bacterial cells, also demonstrating a sensitivity to cell shape, with Wang et al. (2008) suggesting that cell shape may be the determining factor in the filterability of bacteria. Aside from advection, dispersion, and diffusion, colloidal transport also depends on the kinetic rate at which particles attach to soil and grain surfaces with which they collide. To date, only one study has attempted to isolate the role of shape on particle deposition. Seymour et al. (2013) reported a much higher deposition rate of spherical carboxylate-modified polystyrene particles as compared to 2:1 and 4:1 stretched versions of the same particles. These experiments, however, were conducted under favorable conditions (i.e. a positively charged, PLL-coated silica surface), lacking an energy barrier to deposition and limiting the environmental relevance of the findings. No study to date has investigated the role of shape on colloidal particle attachment to environmentally relevant surfaces.

When dissolved in solution, NOM adsorbs to the surface of colloidal particles and typically limits their deposition through steric hindrance. When the depositional surfaces are pre-coated with NOM, however, the results tend to be much more complex. Pre-coating silica surfaces with various NOM have been shown to hinder, enhance, or have no effect on attachment, depending on the type of NOM (McNew and LeBoeuf, 2015; Chen and Elimelech, 2008), ionic strength of solution (Chang and Bouchard, 2013; Qu et al., 2012; Chen and Elimelech, 2008), and temperature (McNew and LeBoeuf, 2015). Chen and Elimelech (2008) reported that an alginate layer enhanced  $n_{C60}$  attachment in the presence of 10 mM NaCl, while hindering it in the presence of 30 mM NaCl. The variable nature was attributed to the alginate layer's compaction at higher NaCl concentrations, resulting in a smoother, more rigid layer, with less room for  $n_{C60}$  particle entanglement. In our recent work (McNew and LeBoeuf, 2015), we reported that increasing the temperature of a humic acid layer while holding all other variables constant, raised the attachment efficiency of  $n_{C60}$  from low attachment ( $\alpha = 0.3$ ) to complete attachment ( $\alpha = 1$ ). We attributed this to the temperature-dependent hydration of the layer, opening up previously unavailable sorption sites.

Derjaguin-Landau-Verwey-Overbeek (DLVO) theory (Derjaguin, 1941; Verwey, 1947) describes the attractive and repulsive forces between a colloidal particle and a surface. Several studies have investigated the attachment of colloidal particles to bare mineral surfaces in simple electrolyte solutions (Chowdhury et al., 2014a; Chang and Bouchard, 2013; Shen et al., 2011; Yi and Chen, 2011; Jiang et al., 2010; Chen and Elimelech, 2008, 2006) and the results appear to be in good qualitative agreement with DLVO theory. The attachment deviates significantly from DLVO theory, however, in the presence of natural organic matter (NOM), whether dissolved

in solution (Akaighe et al., 2013; Qu et al., 2012; Thio et al., 2011; Jiang et al., 2010; Chen and Elimelech, 2008) or coated onto the depositional surface (McNew and LeBoeuf, 2015; Chowdhury et al., 2014b; Chang and Bouchard, 2013; Thio et al., 2011; Qu et al., 2012; Jiang et al., 2010; Tong et al., 2010; Chen and Elimelech, 2008).

The purpose of this study is to isolate the role of shape on the attachment of colloidal particles to environmentally relevant surfaces by combining quartz crystal microbalance with dissipation monitoring (QCM-D) and atomic force microscopy (AFM) experiments. For the first time, we present attachment efficiency curves for colloidal particles onto NOM coated silica surfaces, as a function of particle shape and ionic strength. By comparing the attachment results with DLS surface measurements and AFM topography images, we elucidate and discuss the mechanisms driving the complex attachment behavior observed. The results presented here suggest that colloidal particle shape will play an important role in predicting transport, especially in the presence of NOM. A better understanding of the role of shape in the attachment of colloidal particles to environmentally relevant surfaces will lead to a more accurate prediction of colloidal particle transport, aiding in risk assessment of potential exposure pathways.

## 2. Materials and methods

### 2.1. Materials

200 nm carboxylate-modified polystyrene (PS) spheres were purchased from Phosphorex (107, Hopkinton, MA). Dry Harpeth humic acid (HHA) was isolated and characterized as described in our previous publication (DeLapp et al., 2005). Alginate sodium salt (alginate, A2158), poly-L-lysine hydrobromide (PLL, P1274), and 4-(2-Hydroxyethyl)-1-piperazineethanesulfonic acid (HEPES) were purchased from Sigma-Aldrich (St. Louis, MO). Reagent grade NaCl (S671) was purchased from Fisher Scientific (Pittsburgh, PA). Silica-coated QCM-D sensors (Q5X-303) were purchased from Q-Sense (Biolin Scientific, Stockholm, Sweden). 900 nm silica microspheres were purchased from Polysciences Inc (Warrington, PA) as a surrogate for estimating the electrophoretic mobility of the silica sensor surface. All solutions were prepared using ultra pure water purified by the Milli-Q Water Purification System from Millipore (Billerica, MA).

### 2.2. Preparation of stretched PS particles

The particle stretching procedure used in this study is based on a method previously published by Champion et al. (2007) and the details can be found in Appendix A.1. In order to determine the concentration of the stretched particles, a calibration curve was constructed using known concentrations of stock PS particles and UV-vis spectrophotometry (NanoDrop 2000 Thermo Scientific, Wilmington, DE). The resulting calibration curve can be seen in Appendix A.1. The stock stretched PS suspension was determined to have a concentration of 129 mg L<sup>-1</sup>.

### 2.3. Preparation of HHA solution

In order to prepare the HHA solution, 50 mg dry HHA was introduced into 400 mL deionized water and stirred for more than 3 h. During this time, the pH was adjusted drop wise with 0.1 N NaOH to a pH of 10.0 to allow the HHA to dissolve. After nearly complete dissolution of the HHA, the pH was adjusted drop wise with HCl down to 7.0. The alginate stock solution was prepared by stirring 50 mg dry alginate in 400 mL deionized water overnight, after which time the alginate was adjusted drop wise with 0.1 N NaOH to a pH of 7.0. Both HHA and alginate stock solutions were

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