Geophysical Research Abstracts Vol. 21, EGU2019-11995-1, 2019 EGU General Assembly 2019 © Author(s) 2019. CC Attribution 4.0 license.



How Nanoscale Surface Heterogeneity Impacts Transport of Nano- to Micro-Particles on Surfaces under Unfavorable Attachment Conditions

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The impact of nanoscale surface heterogeneity on retention of nano-to-micro-scale particles (colloids) on surfaces under unfavorable (repulsion present) relative to favorable (repulsion absent) conditions is dependent on colloid size, yet this has not previously been recognized or explained despite unfavorable conditions being considered prevalent in the environment, where applications ranging from water resource protection to contaminant remediation require its understanding. We report experiments on soda lime glass (silica) performed for carboxylatemodified polystyrene latex particles across broad set of sizes $(0.1, 0.25, 1.1, 2.0, 4.4, 6.8 \mu m)$ under varied ionic strengths (0.006 and 0.02 M) and pH (6.7 and 8.0) in an impinging jet system representing upstream sides of porous media grains. These experiments demonstrate dramatically reduced attachment efficiencies (α) for ca. 1 μ m colloids relative to smaller (e.g., < 200 nm) or larger (e.g., $2 \mu m$) colloids with equivalent surface properties. We demonstrate via mechanistic trajectory simulations incorporating discrete representative nanoscale heterogeneity (DRNH) that for ca. 1 μ m colloids, their least combined diffusion and fluid drag in the near-surface fluid domain increased their residence times prior to encountering nanoscale heterogeneity on which to attach, both phenomena thereby reducing the likelihood of colloid attachment under unfavorable relative to favorable conditions. We explore the generality of this phenomenon by examining silica colloids of selected sizes, and by compiling values reported in literature for nano- and micro-particle transport in porous media. We speculate on how this new understanding may guide strategies for targeted delivery of nano- and micro-particles in porous media.