

# Comments

## Comment on Breakdown of Colloid Filtration Theory: Role of the Secondary Energy Minimum and Surface Charge Heterogeneities

Tufenkji and Elimelech<sup>1</sup> provide experimental evidence that deviation of retained colloid profiles from filtration theory is reduced as a result of treatments designed to reduce or mask heterogeneity in surface characteristics in the system of colloids and porous media. This finding is consistent with the expectations of many previous researchers regarding the possibility that the source of deviation from filtration theory is heterogeneity in surface characteristics among the colloid population, as reviewed in ref 2. However, the authors attributed the deviation from filtration theory to both the presence of secondary minima and heterogeneity in surface characteristics either among the colloid population or on the porous media grains. The nonspecific nature of this attribution is laudably open-minded. However, we would like to clarify that this general attribution can be refined on the basis of important distinctions between the effects of heterogeneity among the colloid population versus the effects of heterogeneity on the porous media grains, as explained below.

The observed deviation of retained colloid profiles from filtration theory signals that the colloid population experienced multiple deposition rates within the porous media and that these multiple deposition rates were distributed independently of one another in the spatial dimension. As was well stated by the authors,<sup>3</sup> the shapes of the deviant retained colloid profiles demonstrate that “a fraction of the particle population exhibits a fast deposition rate (implied by the steep slope at the top [influent] end of the column), whereas the remaining particles experience a slow deposition rate (denoted by the shallow portion of the retained profile)”. In the authors’ terminology, the fast deposition mechanisms are located up-gradient of the slow deposition mechanisms. It is this aspect of deviation from filtration theory that distinguishes the effects of heterogeneity in surface characteristics on porous media grains from the effects of heterogeneity in surface characteristics among a colloid population.

Secondary energy minima and heterogeneity in surface characteristics on porous media grains are characteristics

that operate on the nanoscale. These characteristics would be distributed evenly across a packed porous media column, resulting in an even distribution of fast and slow deposition across a packed column, assuming a homogeneous colloid population. As a result, the multiple deposition rates experienced by the colloid population at any one location would yield a combined deposition rate that would be constant with distance across the packed porous media. In other words, the fast depositing colloids would not be deposited up-gradient of the slow depositing colloids. This is demonstrated via simple mathematical relationships in the Supporting Information.

Deviation from filtration theory arises when the fast depositing colloids deposit up-gradient of the slow depositing colloids or, more generally, when the proportions of the multiple deposition rates vary across the length of the porous media, as demonstrated via simple mathematical relationships in the Supporting Information.

Changes with distance of transport in the proportions of the multiple deposition rates experienced by a colloid population can occur as a result of either heterogeneity in surface characteristics among the colloid population or macroscopic variations in the surface characteristics of the porous media with distance. In the absence of proposed mechanisms to produce macroscopic spatial variations in the surface characteristics of the porous media with distance, it is prudent to conclude that heterogeneity in the surface characteristics of the colloid population produced the observed deviations from filtration theory. The treatments used by Tufenkji and Elimelech<sup>1</sup> were shown to alter the surface characteristics of the colloids. Hence, the conclusion that deviation from filtration theory was driven by heterogeneity in surface characteristics among the colloid population is consistent with their experimental observations.

**Supporting Information Available:** Mathematical expressions demonstrating that deviation from filtration theory requires that the proportions of multiple deposition rates vary with distance across a packed column. This material is available free of charge via the Internet at <http://pubs.acs.org>.

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