Reactive transport in porous media: Visualization and modeling

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We present reactive transport flow cell experiments in a refraction index-matched, 3D, water-saturated porous medium of polyacrylamide beads. Two types of experiments were investigated, involving: a pH indicator as the reactant, and citrate-capped, negatively-charged gold and silver nanoparticles (AuNPs, AgNPs) and polyethylenimine-capped, positively-charged AgNPs to determine the effect of surface charge on NP transport behavior. The magnitude of reaction during transport through the porous medium can be related to the color change of the pH indicator or intensity of NP color, via detailed image analysis and sequential imaging of the flow cell. The setup yields measurements of the temporally evolving spatial concentration field. Experiments focused on point injection of (i) a pH-sensitive reactive tracer into a macroscopically uniform flow field containing water at a pH different from that of the injected tracer, and (ii) AuNPs and AgNPs into a uniform flow field. The measurements were matched with continuous time random walk particle tracking (CTRW-PT) models, to account for non-Fickian transport, and require a minimal set of fitting parameters. For the pH-indicator system, a term accounting for chemical reaction was established from analysis of the reactant concentrations; no other fitting parameters were required. The results emphasize the localized nature of transport and reaction, caused by small-scale concentration fluctuations. For the NP system, the negatively-charged NPs behaved similarly to the conservative tracer. However, the positive AgNPs displayed a decreasing tendency over distance to attach to the oppositely-charged porous medium. This transport behavior is understood by DLS and ζ potential measurements, which showed that aggregation processes and inversion in particle surface charge occurred during transport of the positive NPs. CTRW-PT modeling results indicated the transport to be more non-Fickian for the positive AgNPs, highlighting the connection between attachment and transport processes. Further analysis showed that decreasing tendency of these NPs to attach to the porous medium may be correlated to the particle residence time. We conclude that interactions between migrating particles and the porous medium may alter NP chemical properties and lead to significant changes in their mobility.