Colloid Transport and Retention in Fractured Media

Project ID: 55036

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Research Objectives

The goal of the project entitled "Colloid Transport and Retention in Fractured Media" was to identify the chemical and physical factors that control the transport of colloids in water-saturated fractured formations, and develop a generalized capability to predict colloid attachment and detachment based on hydraulic factors, physical structure, and chemical properties. The research targeted multiple scales, including (a) mechanistic description and experiments colloid dynamics in fractures; (b) colloid transport experiments in undisturbed geological monoliths; (c) field-scale colloid transport experiments; and (d) modeling of colloid transport in complex fracture networks.

Research Progress and Implications

Fundamental Description Of Particle Transport In Fractures – Using first principles, models were developed for particle deposition rate on the surface of fractures and results experimentally tested. The model is similar to those presented by Friedlander, Cookson, and Yao et al. for describing aerosol and aqueous filtration by stationary collectors, but is unique in accounting for the geometry of the collector surface; in this case, infinite planes of two separated by a finite aperture width. Using a particle tracking method, an algorithm has been developed to describe the colloid dynamics in fractures, including consideration of particle diffusion, sedimentation under gravity field, and electrostatic interactions of colloids with fracture walls under infinite plane geometry. A steady state solution is attained. The particle diffusion and collection by walls are most significant, compared to gravitational sedimentation. We have also derived an analytical solution for large Peclet numbers under steady state (expressed in terms of parabolic cylinder functions), and confirmed agreement between this solution and the steady state solution given by the particle-tracking model. An experimental system of 1-D parallel-plate artificial fracture with SiO_2-coated fracture walls was used to test and validate the model predictions (Fig. 1). The advanced mechanistically rigorous descriptions of colloid migration under infinite plane geometry will be incorporated in models of colloid transport in water films within partially saturated media.

Colloid Transport In Intact Geological Columns – Colloid tracer experiments using undisturbed monoliths of fractured saprolite under water-saturated conditions investigated the influence of particle diameter and the ionic strength and cation valence of the groundwater solution. There was an optimum size of 0.5 to 1.0 μm for transport of the latex microsphere tracers. Loss of the smaller-than-optimum particles was attributed to the more rapid diffusion of the smaller particles, leading to more collisions with, and retention by, the fracture walls. This was confirmed by dismantling of the core and mapping of distribution of the microspheres using petrographic thin sections and epifluorescent microscopy to visualize the fluorescent colloids. In experiments using aqueous solutions containing different ionic strengths of monovalent (Na^+) and divalent (Ca^{2+}) cations, colloid retention increased as ionic strength increased, with far greater retention with the divalent cations (Fig. 2).

Field-Scale Colloid Tracer Migration — Two series of field tracer experiments were completed. In the first experiment was conducted at a radionuclide-contaminated DOE site at which groundwater contained near-saturated levels of $CaCO_3$. Colloid transport was very limited (Fig. 3), consistent

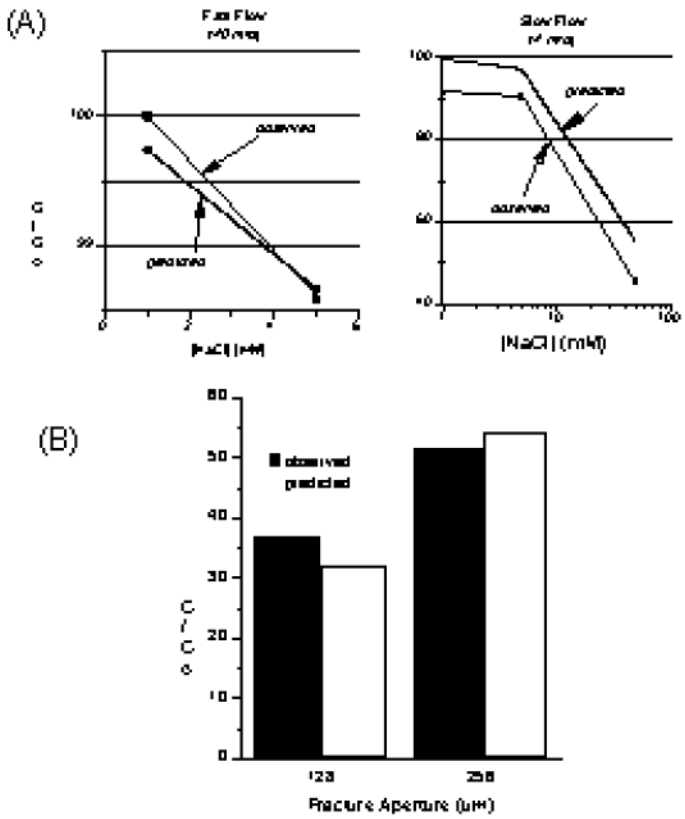


Figure 1. Comparison of analytical model predictions with experimental observations of colloid transport in artificial fracture apparatus. (A) Effect of flow rate and ionic strength on colloid transport through model fractures; (B) Effect of aperture width on colloid transport.

with the effects of high Ca levels observed in the column studies. However, colloids retained by the formation could be mobilized by experimental manipulation of ionic strength that increased the concentration of colloids by two orders of magnitude within a few days. Hypotheses concerning the effects of water chemistry and fracture network characteristics were tested at the second experimental site. The ionic strength of the groundwater at the second site was less than half that at the first site; furthermore, colloids were injected into wells screened in two contrasting lithologies: a highly weathered saprolite similar to the intact column (characterized by a dense network of small fractures), and an underlying rubbly, weathered bedrock (characterized by larger, more widely-spaced fractures). Data (Fig. 3) revealed 1) there is considerably greater mobility of colloids at the second site compared to the first site, consistent with the lower ionic strength, and 2) that there are very significant differences in the rate and extent of colloid transport in the two lithologies, with much greater retention of small colloids in the densely-fractured upper zone. These results are consistent with the ionic strength studies at the column scale (above), and with predictions from the fracture network model (below).

Fracture Network Models of Colloid Transport – Numerical simulations of colloid transport in discretely fractured porous media investigated the importance of diffusion into fine porosity as well as the retention and remobilization of colloids. The processes accounted for by the finite element code, COLDIFF, include advective-dispersive transport of colloids, filtration and mobilization of colloidal particles in both fractures and porous media, and diffusive interactions of colloids between the fractures and porous media. Results demonstrated the importance of the porosity of the matrix and colloid retention. Simulations successfully reproduced observed transport data in saprolite monoliths (Fig. 4), and highlighted the tailing caused by remobilization of colloids after the source was removed. Field-scale simulations indicated that diffusive transfer of colloids to fine porosity and colloid retention and remobilization on fracture walls were dominant processes affecting the long-

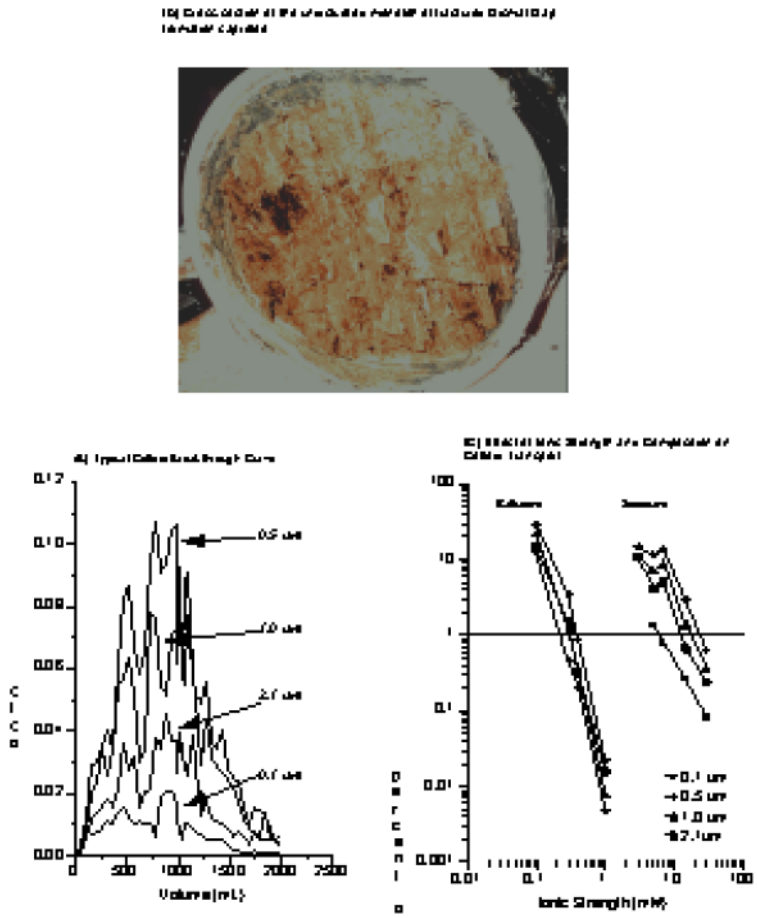


Figure 2. Colloid transport in undisturbed monoliths of saturated fractured saprolite from the Dismal Gap formation (Oak Ridge National Laboratory)

term colloid concentration and migration distance. In environments where porosity is relatively high and colloids are able to diffuse from the fractures, the properties of the porous matrix were more important than those of the fracture network. Because the properties of the fracture network are more difficult to know with certainty relative to those of the porous matrix, predictive uncertainties associated with colloid transport in discretely fractured porous media may be reduced.

Summary and Implications – Both aqueous chemistry and physical structure of geologic formations influence transport. Results of studies at all spatial scales reached consensus on the importance of several key controlling variables: 1) not unexpectedly, colloid retention is dominated by chemical conditions favoring colloid-wall interactions; 2) even in the presence of conditions favorable to colloid collection, deposited colloids are remobilized over long times and this process contributes substantially to the overall extent of transport; 3) diffusive exchange between water-conducting fractures and finer fractures and pores acts to "buffer" the effects of the major fracture network structure, and reduces predictive uncertainties. Predictive tools were developed that account for fundamental mechanisms of colloid dynamics in fracture geometry, and linked to larger-scale processes in networks of fractures. Although a great deal more remains to be learned about colloid transport in water-saturated media, the results of our study highlight the key role of physical and hydrologic factors, and processes of colloid remobilization that are potentially of even greater importance to colloid transport in the vadose zone than in saturated conditions. We therefore propose to focus this

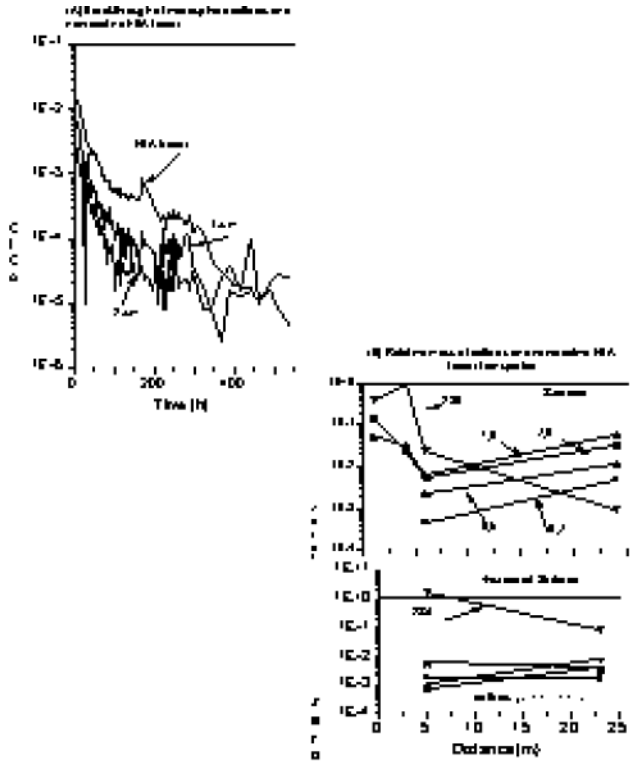
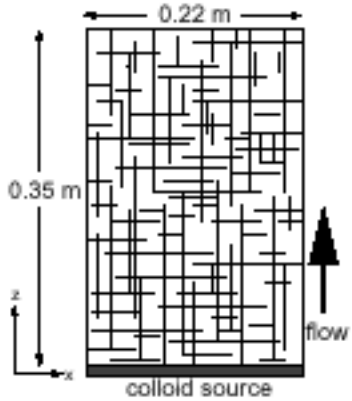


Fig. 3. Field-scale transport of different sizes of carboxylate-modified latex microspheres through water-saturated fractured formations. (A) Breakthrough of microspheres over a 3-m flow path of fractured aquifer. Note that colloids were injected for a 4-h period, yet deposited colloids continued to be released from the formation over extended times. (B) The relative mass of colloids (each marker) of colloids injected into two lithologies at the same location.



proposal on understanding vadose zone transport processes so that they can eventually be linked to the understanding and tools developed in our previous project on transport in saturated groundwater systems.

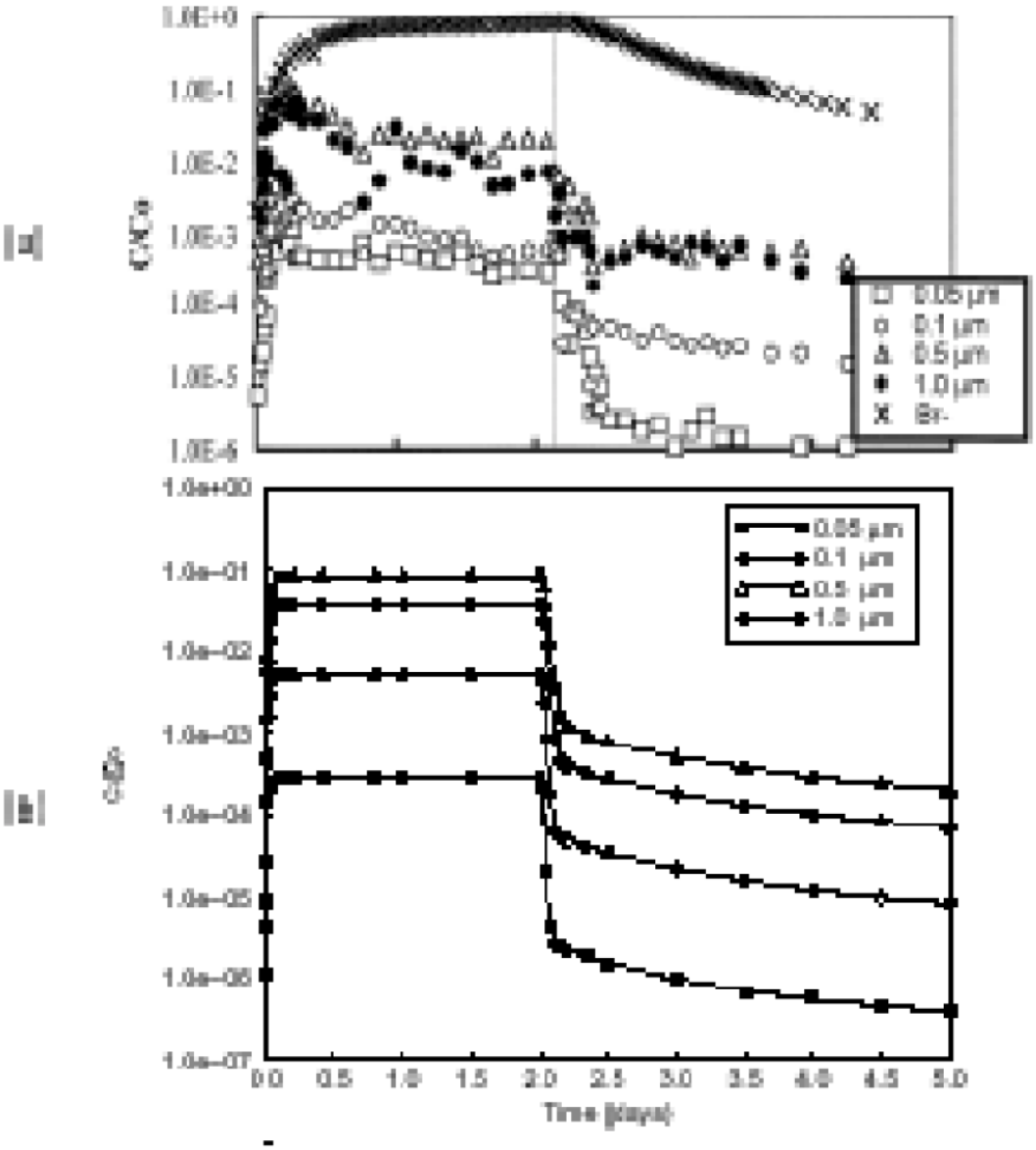


Fig. 4. Comparison of observed data and network model simulation of colloid transport through an undisturbed monolith of Dismal Gap saprolite. The top panel is the representation of the experimental core for the network model. The graphs show the relative concentration rations (C/Co) for the laboratory core-scale experiment and the results of the numerical simulation performed using COLDIFF.