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Colloid Transport and Retention in Fractured Deposits

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Research Objective

The goal of the DOE project is to identify the chemical and physical factors that control the transport of colloids in fractured formations, and develop a generalized capability to predict colloid attachment and detachment based on hydraulic factors (head, flow rate), physical structure (fracture aperture), and chemical properties (surface properties of colloids and fracture surfaces). The research approach targets multiple scales, including: (a) a theoretical description of colloid dynamics in fractures that extend concepts used for porous media to fracture geometry, with predictions experimentally tested in simplified laboratory "fractures"; (b) colloid transport experiments in intact geological columns, which provide natural complexity, but mass balance data and experimental control over flow, colloid size, ionic strength and composition; (c) field-scale transport experiments using colloidal tracers to examine realistic scales of fracture connectivity; and (d) modeling of colloid transport in complex fracture networks that include fractures with varying flow rates and permitting colloid diffusion into microfractures. Understanding the processes that control colloid behavior will increase confidence with which colloid-facilitated contaminant transport can be predicted and assessed at contaminated DOE sites. An added benefit is the expectation that this work will yield novel techniques to either immobilize colloid-bound contaminants in-situ, or mobilize colloids for enhancing remedial techniques such as pump-and-treat and bioremediation.

Research Progress and Implications

This report summarizes work after 20 months of a 3-year project. Progress in the four project elements (tasks) is summarized below:

A) Fundamental Description of Particle Transport in Fractures

This modeling effort represents the first theoretical description of the electrostatic and physical processes controlling colloid interactions with surfaces within a fracture geometry. 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 probability of particles sticking to fracture walls under infinite plane geometry. We found that 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). We next plan to check on the agreement between this solution and the steady state solution given by the particle-tracking model. An experimental system of 1-D parallel-plate "artificial fractures" with SiO_2-coated "fracture walls" is being used to test and validate the model predictions. The results of this task will advance fundamental descriptions of colloid migration in fractures, and will also provide a theoretical foundation for parameterizing filtration coefficients that are used in the geochemical transport model of colloid migration in fracture networks (below).

B) Colloid Transport in Intact Geological Columns

Two series of colloid tracer laboratory experiments have been completed using undisturbed column samples of weathered fractured saprolite. The first series of experiments investigated the influence of particle diameter on colloid transport in undisturbed columns of the fractured shale saprolite. The experiments showed that there was an optimum size of 0.5 to 1.0 mm for transport of the latex microsphere tracers. Loss of the smaller-than-optimum particles was attributed the more rapid diffusion of the smaller particles, leading to more collisions with, and retention by, the fracture walls. Dismantling of the soil column and mapping of distribution of the microspheres clearly showed evidence of size segregation of particles within the fractures, with the smaller microspheres penetrating into the fine-grained matrix between fractures.

A second series of experiments to examine the influence of ionic strength and cation valence in the carrier solution has also been completed. In these experiments 4 different sizes of latex microspheres (0.1-2.1mm) were passed through a saprolite column in carrier solutions containing different ionic strengths of monovalent (Na^+) and divalent (Ca^{2+}) cations. As expected, for both the monovalent and divalent cases, colloid retention increased as ionic strength increased. As well, retention was greater for the experiments with the divalent carrier solution, relative to experiments with the monovalent carrier solution. Further evaluation is underway to determine deposition rates and collector efficiencies for the different sizes of colloids.

This project is also being assisted through a research transfer element of DOE/OBER's Subsurface Science Program (SSP). A SSP project (G. Moline, PI) is developing improved techniques to characterize fracture networks, using, for example, x-ray tomography to visualize and analyze the statistical distribution of fracture networks. We hope to utilize these techniques in place of the expensive, time-consuming, and destructive core dissection so that transport models can accurately represent the column either as discrete fracture networks with representative characteristics, or in a Monte-Carlo fashion using actual statistical distributions for the characteristics.

C) Field-Scale Colloid Tracer Migration

Two series of field tracer experiments were completed. The first experiment at a radionuclide-contaminated DOE site was conducted in the same saprolite formation from which the intact columns were excavated. Groundwater at the site contained near-saturated levels of $CaCO_3$. Two sizes of colloids (0.8- and 2.0-μm) and a nonreactive tracer were injected. Results demonstrated that colloid transport was very limited (consistent with the effects of high Ca levels observed in the column studies). At a monitoring location only 1-m from the injection well, recovery of colloids was an order of magnitude below that of the nonreactive tracer (relative to the source concentrations of each tracer), and recovery decreased another order of magnitude at a distance of 3-m. However, the size discrimination observed in the column experiments was not observed in the field experiments, suggesting that interconnections between fractures occurring at larger spatial scales may permit greater transport of larger colloids than predicted from the column-scale studies. Colloids retained by the formation could be mobilized by experimental manipulation of ionic strength. When distilled water was introduced into the injection well three weeks after the colloid injection ceased, the concentration of colloids in the downgradient monitoring well increased by two order 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). Four sizes of colloid were injected into each lithology, with each colloid uniquely identified by a fluorescent dye. Preliminary data suggests: (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).

D) Modeling Colloid Transport in Complex Fracture Networks

The fracture network modeling has focused on determining the relative importance of the processes controlling the migration of colloids through fractured media, as well as further development of the FRACLD code to include the additional processes of remobilization of colloids and their diffusion into the matrix. Some of the processes considered for the sensitivity analysis were the filtration coefficient (reflecting the extent of colloid retention by the fracture walls), fracture network density, fracture lateral network extent, and dispersivity. Simulation results show that the size and, especially the density of these fracture networks were the single most important attribute determining the overall migration and eventual breakthrough of the colloidal particles. Increasing the density of fractures delays the arrival time for colloids. For example, a ten-fold increase in the number of fractures results in a five-fold increase in the time required for the colloid concentration to reach 80% of the source concentration. The importance of fracture density seen in these model simulations is consistent with the results of the field study in the contrasting lithologies (above). The results are also consistent with results from the collaborating SSP project, which examined solute and colloid transport in intact column experiments in which flow was oriented parallel-to-bedding or perpendicular-to-bedding. Less recovery of both tracers was observed for transport parallel-to-bedding, which is characterized by a high density of narrow fractures, than for transport perpendicular-to-bedding, where transport is through fewer, wider fractures, and a greater number of horizontal connecting fractures.

Significance

This work has yielded new insights into colloid transport and it's importance to contaminant mobility at DOE sites by quantifying the importance of both physical factors (particle diameter, flow velocity, and fracture density) and chemical factors (ionic strength, cation valence, and likely pH of the carrier solution) on the potential for colloid transport. The demonstrated effects of ionic composition in the column studies, and the observed release of colloids following ionic strength adjustment in the field experiment highlights the possibility that colloid transport behavior may change seasonally or over shorter time-frames due to variations in influx of meteoric water. For example, during seasonally wet periods there are typically large increases in flow rate and some decrease in ionic strength, which could result in mobilization and rapid transport of colloids during these periods. As a result, migration of colloidal contaminants might be highly intermittent, and this should be considered in designing monitoring strategies.

Planned Activities

The analytical model of colloid interactions in fractures will be compared with the particle-tracking model within the next few months. Single-fracture transport experiments to test the model predictions will be conducted this Summer and Fall. Another series of intact column experiments will be conducted over the next year to examine the effect of colloid surface charge on transport. A series of field experiments are planned in the Winter of 1999 to examine colloid transport in fractured bedrock, and to examine the effect of colloid surface charge on field-scale migration of colloids. The modifications of the FRACLD model to include the processes of diffusion into fine pores and colloid remobilization will be completed within the next six months. These modifications will also greatly increase the computational efficiency of the model which will allow for larger, more extensive, and field-scale simulations. Computer simulations of the column experiments are now underway and will be completed in the next year. Simulations of the field-scale experiments will be examined in the final phases of the project. The increases in the computational efficiency are expected to make the model available to users without access to high-end work stations. Thus, by the end of the project, a simulation tool will be available to predict the field-scale transport of colloids, based on improved fundamental understanding and tested on column and field experiments that incorporated the key variables controlling colloid transport in subsurface systems.