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Colloid Migration in Fractured Media
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Field studies at the Nevada Test Site by researchers at Lawrence Livermore National Laboratory have demonstrated that radionuclides are being transported by colloidal material suspended in groundwater. This observation is counter to most predictions from contaminant transport models because the models assume adsorbed species are immobile. The purpose of this research is to quantify the transport processes for colloidal materials and develop the mechanistic understanding necessary to predict radionuclide transport in fractured media. There were three areas of investigation during this year that have addressed these issues: chemical control of colloid deposition on clean mineral surfaces, colloid accumulation on fracture surfaces, and the influence of deposited colloids on colloid and tracer migration.

1. Chemical Control of Colloid Deposition and Release

Filtration theories are well-developed and physical transport processes can be accurately predicted for colloid transport in clean porous or fractured media (McDowell-Boyer et al., 1986). The major area of uncertainty is the sticking efficiency of colloids on surfaces when there are repulsive forces between colloids and surfaces at low ionic strengths. Under these conditions, limited experimental data differ by orders of magnitude from theoretical predictions. The purpose of McDowell-Boyer's (1989) thesis research was to investigate colloid deposition at low ionic strengths onto clean sand at flow velocities typical of the subsurface, 1 to 100 meters per day. All experiments were conducted with 1.46 micrometer diameter polystyrene latex microspheres that were manufactured without surfactants. The porous media was composed of cleaned, narrowly-sized Monterey sand composed of quartz and feldspar, and the sand was held in columns 5 to 60 centimeters long. The background electrolyte was NaCl, and it was buffered at pH 6.3 with sodium bicarbonate. Colloid concentrations were measured in the influent and effluent with an ELZONE particle counter. Erosion experiments were conducted by stopping the flow of influent particles while maintaining the flow rate and salt concentration. This was followed by a switch to distilled water as the influent. Since colloid concentrations were very low and experiments were of short duration, media surfaces were not altered by colloid attachment.

The experimental results showed that colloid attachment decreased as the salt concentration decreased, but still the results suggest limited mobility of colloids through clean porous media. At a flow velocity of 1 meter per day, the characteristic distance colloids would migrate for an NaCl concentration of 1.0 M was about 3 centimeters. The migration distance increased to approximately 300 centimeters at an NaCl concentration of 0.001 M (60 milligrams per liter). When colloids were removed from the influent fluid, colloids did not appear in the effluent indicating that erosion was not an important mechanism for fixed flow velocities and constant electrolyte concentration. When buffered distilled water flowed through the columns at the same water flow velocity, the previously deposited colloids were observed in the effluent. These results indicate that limited colloid migration is expected when fluid flows through clean porous media, but remobilization is likely by changes in aqueous solution composition that results in lowered colloid-surface binding forces.

2. Colloid Clogging of Fractured Media

Since porous and fractured media surfaces are rarely clean initially, and even if they were, long-term attachment of colloids would result in substantial alteration of the fluid-solid interface. Colloids are known to deposit at rates greater than expected from theories when the fluid ionic strengths are low, and filtration theories have not been able to predict long-term colloid deposition and the resulting decrease in media permeability. An experimental apparatus has been constructed to study long-term colloid deposition and permeability reduction. The ideal fracture consists of two glass plates separated by 100 micrometers and allows colloidal suspensions to flow through this channel. Colloids that deposit on the walls form a porous medium that alters further deposition and can substantially decrease the permeability. Colloid deposition studies have not been conducted as yet because of the need to assure pulse-free fluid flow and accurate pressure drop measurements. The intent is to quantify the mass of colloidal material retained in the fracture and the resulting reduction in permeability. These measurements will be compared with independent rheological measurements of colloid aggregates as one way of determining the strength of the deposited material. Cleaned clay minerals have been selected as initial experimental colloids.

3. Colloid and Tracer Migration

There are a number of field observations that demonstrate earlier arrival of colloidal tracers than conservative molecular tracers in porous and fractured media. Harvey et al. (1989) found that microspheres traveled more rapidly than bromide in a column that had a secondary pore structure giving selective flow through channels. Gerba and Bales (1988) have reported on more rapid bacteriophage migration than a non-sorbing tracer through fractured tuff in laboratory experiments. Toran et al. (1988) imposed an artificial fracture within a laboratory sand column and also observed early break through of colloids compared to a conservative tracer. Finally, Champ and Schroeter (1988) conducted field scale tracer tests with bacteria and non-sorbing, inorganic tracers in fractured crystalline rock. The peak of the bacteria concentration occurred at approximately one-fifth the time required for the inorganic tracers to peak. All these researchers have made qualitative arguments that colloids are excluded from part of the pore space, but they have not presented quantitative explanations for these very important observations. If colloidally transported contaminants are the first to arrive at some downstream location, this could require a complete reevaluation of monitoring strategies based on tritium detection.

In conjunction with previously described deposition experiments, Ph.D. student John Kessler has been evaluating various quantitative transport models that are available in the literature. He is specifically comparing colloid versus non-sorbing tracer migration in porous and fractured media. While molecular and hydrodynamic dispersion is recognized for flow through porous and fractured media, many researchers have not acknowledged that colloidal particles have diffusivities that are one-thousandth that of simple tracers. This results in very little diffusion but substantially greater Taylor dispersion for colloids than would be found for simple tracers. Model predictions for the elution of molecular and colloidal tracers from ideal fractured media and a fractured media containing a porous coating of previously deposited colloids are possible from an early analysis by Aris (1959). Preliminary results indicate that fractured media with a porous coating on the walls promotes the more rapid transport of colloids than molecular tracers. Given these model predictions, there may be experimentally feasible conditions for evaluating the predictions in the laboratory using cores collected from the Nevada Test Site or in the ideal fracture flow apparatus.

4. Summary

Laboratory and model investigations of colloid transport in porous and fractured media have supported ongoing field investigations at the Nevada Test Site. Aqueous chemistry has been shown to control colloid attachment and release from clean mineral surfaces. For colloidal deposits on fracture walls, the current experimental program will determine how this material responds to hydrodynamic forcing and if the porous colloidal deposit causes the more rapid transport of colloids than non-sorbing tracers.

5. References

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June 28, 1990

Ms. J. K. Carlson
Procurement Department
LLNL, L-650
P.O. Box 5012
Livermore, CA 94551

SUBJECT: Final Report, Intramural Order 2368303

Dear Ms. Carlson:

The research project on Colloid Migration in Fractured Media terminated on December 31, 1989, after being extended from September 30, 1989. I submitted an annual report to Dr. Kenneth Marsh in September, 1989, that provided a summary of the bulk of my activity. A copy of that report is attached.

The main emphasis during this final period was on modeling molecular and colloid transport through realistic fractured media. We have shown that there is a quantitative explanation for the earlier arrival of colloids compared to molecular tracers as has been observed in field and laboratory experiments. Preliminary modeling discussed in the earlier annual report has been refined and was presented at the May 1990 American Geophysical Union Meeting in Baltimore. The abstract for that invited presentation is also attached. The modeling results have shown that colloids, if excluded from a porous region on the fracture walls, will have a greater average velocity than molecular tracers. Also, colloids will be dispersed during fracture flow much differently from molecular tracers because of their lower molecular diffusivity. Laboratory experiments on fracture clogging and tracer migration are the subject of continued research supported by LLNL, Nuclear Chemistry.

I am sorry for the delay in reporting my activities.

Sincerely,

James R. Hunt

James R. Hunt
Associate Professor
Environmental Engineering

cc: Suzanne Kempton, Engineering ORS, UCB
Robert Edwards, SPO, UCB
Ken Marsh, Nuclear Chemistry, LLNL

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