Project ID: **55395**

Project Title: **Physics of DNAPL Migration and Remediation in the Presence of Heterogeneities**

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RESEARCH OBJECTIVE

The goal of our research is to develop a fundamental quantitative understanding of the role of physical heterogeneities on DNAPL migration and remediation in aquifers. Such understanding is critical to cost effectively identify the location of the subsurface zone of contamination and design remediation schemes focused on removing the source of the contamination, the DNAPL itself. To reach this goal, the following objectives for the proposed research are defined:

Objective 1: Develop fundamental understanding of the physics of DNAPL migration processes within heterogeneous porous media: a) Conduct a suite of two-dimensional physical experiments within controlled and systematically varied heterogeneous porous media at scales up to one meter. Vary system parameters to consider a range of capillary and bond numbers within these heterogeneous porous structures. b) Develop a new DNAPL migration model based on an up-scaling of invasion percolation (UIP) to model the migration process. Compare the model predictions to experimental results. Accomplishing objective 1 provides a series of experiments against which we will be able to evaluate the validity of existing multi-phase flow theory as formulated in both percolation codes and in continuum flow codes. These experimental results will also provide new insights into DNAPL migration behavior. Development of the UIP model will provide an exciting alternative to continuum multi-phase flow codes since UIP offers several advantages for modeling DNAPL migration. The UIP model is fast, allowing for: 1) modeling in three dimensions; 2) the incorporation of much more geologic detail; and, 3) its use in probabilistic modeling by way of Monte Carlo techniques.

Objective 2: Develop fundamental understanding of the physics of DNAPL remediation processes within heterogeneous porous media: Conduct a suite of physical experiments within controlled and systematically varied heterogeneous porous media at scales up to one meter that consider several remediation treatments. Accomplishing objective 2 will allow us to consider the efficacy of several promising DNAPL remediation techniques under realistic yet well-controlled conditions. We consider this work to be of the type of broad-based, initial studies needed to better understand the intricacies associated with various remedial processes. We expect that the results of this work will be used to focus subsequent research on those remedial approaches or combination of approaches that appear to offer the most promise.

RESEARCH PROGRESS AND IMPLICATIONS

This report summarizes work after 33 months of a 36-month project.

Objective 1: Develop fundamental understanding of the physics of DNAPL migration processes within heterogeneous porous media.

Our approach integrates experimentation using unique capabilities at Sandia's Flow Visualization and Process Lab with development of new approaches for modeling DNAPL migration. Our experimental system consists of a 2D, sand-filled, initially water-saturated chamber (60cm high, 30cm wide and 1cm thick) where we can fully visualize DNAPL migration using light transmission techniques. Image analysis tracks the differences in light intensity for each successive image thereby yielding a time series of the displacement process at exceptional spatial and temporal resolution. Such resolution allows us to perform detailed data analysis that is sufficient to increase our understanding of DNAPL migration processes and to test our upscaled percolation model. In order to make reproducible geologic structures for our experiments we use a unique computer-controlled sand-filling apparatus. With our filler we can make macro heterogeneities (such as discrete formation-scale features like facies) as well as smaller scale micro heterogeneities (such as micro layering and crossbedding found within lithologic units).

In macro-heterogeneous fields, we have conducted a suite of immiscible displacements through heterogeneous porous media in which we vary system parameters to consider DNAPL migration over a range of migration conditions. All our experiments clearly show the invasion field to evolve in a series of gravity stabilized and destabilized invasion periods resulting in a final configuration of fingers and pools. Behind the growing front, we found nonwetting phase saturation to pulsate in certain regions when viscous forces were low. We found also that pool height, finger diameter, and the degree of nonwetting phase redistribution to be affected by the presence of significant viscous forces. Through a scale analysis, we have derived and confirmed a series of length scales that completely describe these flow phenomena. However, with regard to DNAPL migration at the field scale, we expect that gravity forces and variable capillary forces imparted by lithology will play the primary roles in controlling migration path. A paper summarizing this work has been submitted to Water Resources Research.

We have also begun to explore the effects of micro heterogeneities (such as micro layering and crossbedding found within lithologic units). In an initial series of experiments, microstructures have been varied through their "intensity" in terms of property variation across individual laminae. Results obtained suggest that the degree of small-scale heterogeneities affect effective block saturations and may well affect accessibility rules for the upscaled percolations.

Additionally, in these laminated systems we found migration path to be more sensitive to the flow rate than in either homogeneous systems or in discrete/macro-scale systems that we have studied.

In our UIP model, DNAPL migration is dictated by capillary and gravity forces. Percolation models simply track growth order (instead of solving for a pressure field at each time step) resulting in very fast computations. The speed of the model allows much more detail about heterogeneities to be incorporated. The UIP model easily models gravity driven fingering, a process common to DNAPL migration. Data requirements for the UIP model are light and readily obtainable. All that is needed is DNAPL density and interfacial tension, and the boundaries of lithologic units and their textures.

Our UIP modeling shows an exceptional ability to predict migration path for the experiments we have run. Although testing in well-controlled field situations will be needed, the implication of these encouraging results is that the UIP model will likely produce meaningful simulations of migration behavior for the vast majority of DNAPL spill situations.

Over the past year, we have made a number of enhancements to the UIP model. The model now handles: 1) the simultaneous invasion of both wetting (water) and non-wetting (DNAPL) phases; 2) incorporation of viscous forces (multiple fingers and greater "fill in" of the migration field); and 3) an improved method for inputting the lithologic units into the model.

Objective 2: Develop fundamental understanding of the physics of DNAPL remediation processes within heterogeneous porous media

Our intent has been to work closely with developers of each remediation approach to attempt to optimize the remedial process and show each technique in its best possible light. It is hoped that our experimental results will be useful to the remediation developers in helping them better understand the physics and chemistry of their remediation processes as they observe performance under more realistic, less idealized, conditions while maintaining good experimental control. We set up the experiments so that they provided a realistic analog to field-scale remediation processes and we performed these experiments in such a way that we may directly visually observe the interaction between DNAPLs, the remedial fluids, and the heterogeneous aquifer material.

We began by running a series of micromodel experiments looking at surfactant mobilization and solubilization of DNAPLs. We collaborated in this work with Dr. Alex Meyer, a professor at Michigan Tech. Alex sent a doctoral student, Lirong Zhong, to work in our lab for several months to run these experiments. Results of this work was presented at 1998 AGU fall meeting. A paper describing this work is nearly ready for submittal.

Through our work on DNAPL migration, we have developed a unique experimental system where we can reproduce complicated heterogeneity structures. This system also allowed us to conduct experiments for the first time that can compare several competing approaches for DNAPL remediation on equal footing. Our work on DNAPL migration was critical to this work in that we were able to provide realistic initial DNAPL emplacement conditions for the remediation experiments. We emplaced the DNAPL by allowing it to migrate through the heterogeneous lithologic structure we constructed. Thus, each experiment for a remedial technique was conducted in a nearly identical geologic environment with the DNAPL in nearly identical locations. We designed and constructed a new chamber (dimensions: 60cm x 60cm x 1cm) that was equipped with the ports and plumbing necessary to run the remediation experiments. We purchased a gas chromatograph to measure effluent concentrations and trained a student operator.

Using this new experimental chamber, we conducted three remediation experiments. We looked at the performance of two surfactants and potassium permanganate, an oxidizer of TCE. We also performed a partitioning tracer test. The dramatic and unexpected results that we obtained in our experiments point to the necessity of performing well designed "transparent earth" experiments in the lab prior to deploying innovative remediation techniques in field tests. Throughout tests we observed a recurrent theme -- misconceptions about nature location in the subsurface can lead to flawed remedial design.

Partitioning tracer test - We worked with Dr. Varadarajan Dwarakanath of Duke Engineering to design the tracer test. It occurred to us that the certain conditions provided by our remediation experiments – subsequent to the emplacement if the DNAPL and prior to beginning the remediation – were ideal for performing a tracer test while requiring very little extra work. Partitioning tracer tests are designed to compare the breakthrough of partitioning and non-partitioning tracers. Retardation of the tracers that partition into the organic phase provides a means to calculate the mass of DNAPL contained in the region swept by the tracer test. We found that the test worked qualitatively, indicating the presence of DNAPL, but the calculations significantly underestimated the mass of DNAPL in the chamber. We believe that failure to account for the fact that the vast majority of the DNAPL mass existed in large pools resulted in under-prediction of DNAPL mass. When significant DNAPL mass exists in pools, typical tracer flow rates do not allow sufficient time for partitioning/diffusion of the tracers into and out of large pools.

MA surfactant experiment – For this experiment, we obtained surfactant advice from Alex Meyer and Lirong Zhong. The experiment used the surfactant MA and was designed to maximize solubilization while minimizing mobilization. Contrary to expectation, we observed dramatic mobilization. The DNAPL penetrated the aquitard and became inaccessible to the surfactant. Even though trapping number calculations predict some modest amount of mobilization, failure to account for DNAPL in pools resulted in significantly underestimating the potential for extensive downward mobilization. In observing the mobilization process, we discovered a previously unknown mobilization process that occurs when the surfactant front first encounters a pool. Very different interfacial tensions on either side of the surfactant front result in enhanced drainage of the DNAPL pool. For our particular experimental conditions, due to downward mobilization and penetration of the DNAPL into fine-grained units, introduction of the MA surfactant actually made the problem worse.

Tween surfactant experiment – For this experiment, we obtained surfactant advice from Dr. Kurt Pennell, a professor at Georgia Tech. We obtained much better results using the Tween surfactant. We observed only modest DNAPL mobilization because the Tween surfactant maintains a much higher water/organic interfacial tension. We also observed good solubilization. Complete cleanup was achieved after several pore volumes of flushing. Time-lapse animation of this experiment yielded important insights into remediation process.

Permanganate experiment – For this experiment, we worked with Dr. Jack Istok, a professor at Oregon State. Flushing with potassium permanganate has been investigated as an oxidizer that mineralizes TCE. Jack suspected that the manganese precipitate that forms as a mineralization product cause permeability reduction and thereby inhibit access between the TCE and the permanganate solution and this is precisely what we were able to visually observe in this experiment. The manganese precipitate formed a low permeability rind surrounding the DNAPL pools. Such results had not been seen previously, because for experiments run in uniform media, the DNAPL does not reside in pools. The permanganate oxidation process not likely to be as efficient as initially hoped in cases where DNAPL resides in pools. Perhaps intermittent flushes with a substance to dissolve away manganese precipitate might be possible.

We presented results of this experiment at the EMSP Workshop and at the 1999 AGU fall meeting.

PLANNED ACTIVITIES

Our experimental work is complete. We plan to apply for a no cost extension for the project, allowing us the time to publish 3 more papers -- percolation modeling of our migration experiments, results of our remediation experiments, and creating reproducible heterogeneous lithologic structures using our computerized sand filler.

INFORMATION ACCESS

Visit our web site at http://www.nwer.sandia.gov/flowlab

We have completed a paper that describes how our upscaled percolation model can be used within a Monte Carlo analysis to help determine DNAPL location by creating a probability map of where the DNAPLs might reside. Such a map could be used in conjunction with any DNAPL detection technology to suggest the most profitable sampling locations. Contact us to receive a copy. The citation is:

Borchers, B., S.H. Conrad, E.K. Webb, R.J. Glass Jr., R. Cox. 1997. A simulation and decision analysis approach to locating DNAPL in subsurface sediments. Sandia Report SAND97-2261.

We have submitted a paper describing the results of our migration experiments:

Glass, R.J., S.H. Conrad, and W.J. Peplinski. Gravity destabilized non-wetting phase invasion in macro-heterogeneous porous media: experimental observations of invasion dynamics and scale analysis. Water Resources Research, in review.

STUDENTS

Three undergraduate students have worked on this project. One doctoral student from Michigan Tech came to our lab to work collaboratively with us for several months.