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Investigation of Fluid Flow and Contaminant Transport Processes in Heterogeneous Multiphase Systems

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January 8, 1999

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Introduction

Goals

To develop improved methods for assessing and remediating subsurface environments that are contaminated with fluids that are immiscible with water, such as petroleum products and chlorinated solvents.

Objectives

to gain an improved fundamental understanding of flow and transport phenomena in heterogeneous multiphase systems

to utilize this improved understanding to advance more effective strategies for monitoring, modeling, remediating and making technically-based decisions for such systems

to disseminate/distribute this knowledge to the DoD groundwater community

Summary

Experimental Results
Due to our extensive laboratory capabilities, the Center for Multiphase Research (CMR) is in a position to capture unique and quantitative data sets, thus allowing us to examine processes at a much more fundamental level than most investigations. These data sets are also extremely beneficial for validating numerical simulations. This work will lead to improved evaluation of exposure and more efficient remediation methods. In particular, we have made progress in the following areas:

1. Improved understanding of multiphase flow phenomena, especially the residual-funicular relationship. One of the constitutive relations used in the modeling of multiphase flow is the pressure-saturation relation. The value of residual nonaqueous phase liquid (NAPL) saturation is a function of the funicular (connected) NAPL saturation reached during NAPL migration. Long-column experiments have been performed that will lead to a better understanding of the residual-funicular relationship, an important part of the pressure-saturation constitutive relation. This knowledge will lead to improved modeling of subsurface flow and transport.

2. Improved understanding of the mass transfer limiting processes during partitioning tracer tests. Partitioning tracer tests, used for estimating the mass of NAPL in a contaminated, subsurface system, have been validated only for systems where the NAPL saturation is at or below residual—our work is focusing on medium- and high-saturation NAPL pools. We believe that mass transfer limitations under these saturation conditions may lead to low estimates of the NAPL mass. In a one-dimensional column, a NAPL pool is created by injecting NAPL into a coarse layer bounded above and below by fine media, thus trapping a "pool" of NAPL. Quantitative visual, aqueous phase concentration, and saturation data is collected throughout the experiments. This data set allows us to develop mass transfer relations and to validate numerical models. This tracer work will also benefit our ongoing mesoscale investigation of NAPL pools.

3. Improved understanding of the mobilization processes during surfactant-enhanced remediation. It is very important during remediation techniques that the processes are well-understood and controlled in order to prevent the contaminant from migrating to formerly pristine regions of the aquifer. In a bench-scale rectangular cell, glass-bead media contaminated with NAPL were remediated using a mixture of two food-grade surfactants. Flow characteristics were found to depend on the relationship between the gravitational, viscous, and capillary forces. Quantitative visual, aqueous-phase concentration, and pressure data were collected during the experiments. This unique data set will allow quantitative validations of numerical models describing these processes and will allow us to provide guidelines and recommendations concerning flow and chemical conditions during surfactant-enhanced remediation schemes. In addition, pore-scale dynamics were observed that are not currently accounted for in standard multiphase numerical simulations—an observation that warrants further investigation.

4. Work is continuing on the meter-scale (mesoscale) experiments investigating the factors that control the formation and morphology of NAPL pools in heterogeneous subsurface systems. A second experiment is in progress and much-needed work was accomplished that will have a positive impact on the mesoscale experiments: improved X-ray techniques, optimization of X-ray methods, partitioning tracer column experiments, and the NAPL mobilization studies. This additional theoretical and experimental work will greatly enhance the quality of the mesoscale experiments.

Modeling Results

The CMR has access to a wide range of computing resources. In addition to the some of the latest and most powerful workstations, we have consistently received thousands of hours of supercomputer time from the North Carolina Supercomputing Center. Recently, we have been provided computer time and access to supercomputers at WES. These capabilities have allowed us to make many advancements in the modeling of groundwater flow and transport.

1. Improved understanding of the NAPL-aqueous phase mass transfer process, particularly in regard to dissolution fingering. Dissolution fingering refers to the pattern of a NAPL dissolution front that results from even minor heterogeneities in the initial distribution of NAPL. This fingering can cause the mass transfer zone to assume a complex shape that requires potentially long time and length scales to develop fully. Using a multiphase numerical simulator, we are continuing to conduct numerical experiments that simulate the development of dissolution fingers in a mildly heterogeneous porous medium. Current investigations include improvement of numerical methods for simulating these processes and the impact of field-scale heterogeneities on dissolution fingering.

2. Improved problem-solving environments (e.g., object-oriented programming) and numerical techniques (e.g., Richards equation). In the past year, we have made two significant improvements in the numerical modeling of Richards equation,; an equation for solving for water flow in partially saturated porous media. This problem is highly nonlinear and difficult to solve. Four existing and one new transformation method were compared for a wide range of media properties and shown to improve the efficiency and robustness compared to standard solution approaches. The other problem addressed was the numerical convergence difficulties associated with using typical capillary pressure-saturation-relative permeability relations. Here, an integral permeability approach was introduced and used. This technique has proved to be much more robust and economical for a wide range of test problems than standard solution techniques.

Stochastic Modeling Results

Part of the CMR Research Group is involved in the development and application of stochastic methods to solve groundwater flow and transport problems and environmental analysis.

1. Following up on the work of previous years, numerical aspects of the space-transformation approach for the solution of the groundwater flow equation have been

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further investigated. We have improved the numerical codes to increase the efficiency, and studied the effects of model parameters on performance. The method has also been compared with standard finite-difference solvers and proved superior in computational speed. We proposed a novel mathematical approach based on differential geometry for the solution of multiphase flow equations in stochastic porous media. The characteristic of this approach is that the flow equations are expressed along the flowpaths of the different phases, thus allowing a one-dimensional formulation of the problem along the pathlines. The flowpath method is strongly related to space transforms. An iterative numerical solver based on the flowpath method was developed and used successfully for determining the flowpaths in a two-dimensional random medium. The space transformation and diagrammatic techniques developed by our group were proven to be successful tools for modeling studies. The space transformation approach leads to more efficient numerical codes than other standard techniques. In addition, it has strong links to the flowpath method for multiphase flow, which also promises to be an efficient tool for solving flow models that include more than one contaminant phase.

2. We investigated the application of the diagrammatic technique to the calculation of effective parameters of three-dimensional, heterogeneous flow domains. In the past year, we further pursued this work in order to estimate the scaling of effective parameters in the pre-asymptotic, non-ergodic regime. We introduced a new variational method of stochastic averaging and thus obtained the equations which govern the pre-asymptotic behavior of effective parameters. The results of the variational approach are valid for larger heterogeneities than leading-order perturbation, and they agree with the latter in the case of weak heterogeneity. We tested our approach with laboratory measurements of permeabilities and found excellent agreement. Finally, we obtained a scaling function for the effective permeability as a function of the domain scale. Based on this result we predict that non-stationary random field models are required in order to capture the behavior of large-scale measurements of hydrologic parameters. The diagrammatic approach permits explicit calculations with better accuracy than currently used low-order perturbation approximations. We were the first to publish in the hydrologic literature detailed studies of contamination problems using these techniques. Since then several papers have appeared which base their analysis on our original results. A significant result of our work is the conclusion that the class of random models that is routinely used in subsurface hydrology is inadequate for characterizing large-scale behavior.

3. An extension of previous work on spatial indicators to space/time indicator parameters for nonhomogeneous/nonstationary processes is currently under way. Such indicator parameters provide useful tools for exposure and health risk assessment that incorporate quantitative measures of the uncertainty due to incomplete information and the space/time variability. Hazardous waste site characterization is extremely valuable in health management and environmental decision-making. The health effects of contaminants are closely related to the adequate characterization of the spatial/temporal heterogeneity and the information available. Stochastic indicator analysis can improve the accuracy of statistical estimates of contaminant concentrations leading to more accurate exposure

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assessment and consequently better evaluation of the health impacts of environmental contaminants.

4. We have built on previous progress in spatiotemporal analysis. New space/time covariance models have been constructed based on the theory of spatiotemporal random fields (S/TRF) that allow the rigorous characterization of space/time variabilities and heterogeneities. We have found an asymmetry in the time component of the generalized covariance that leads to more compact models than earlier work in the case of correlated residuals. In addition, the numerical codes for space/time estimation are being continuously updated and improved in order to optimize numerical operations. A new estimation approach based on the Bayesian Maximum Entropy (BME) concept has also been proposed and investigated. The BME estimator is non-linear, and it does not suffer from the restrictive assumptions of the linear kriging technique. Moreover, the BME can incorporate information from soft data, and it also allows for updating of the estimates in light of new information. Preliminary numerical studies have been conducted which provide strong evidence for the advantages of the BME method compared with the standard kriging methods. The new estimation techniques that we are developing based on the BME approach are powerful tools that allow far greater flexibility than the currently used kriging estimators.

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Inventions

None

Bibliography

All relevant references were listed in the "Publications and Technical Reports" section.

Appendixes

None