**ANNUAL REPORT FOR
ENVIRONMENTAL MANAGEMENT SCIENCE PROGRAM PROJECT NUMBER 86598
COUPLED FLOW AND REACTIVITY IN VARIABLY SATURATED POROUS MEDIA**

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RESEARCH OBJECTIVE

Improved models of contaminant migration in heterogeneous, variably saturated porous media are required to better define the long-term stewardship requirements for U.S. Department of Energy (DOE) lands and to assist in the design of effective vadose-zone barriers to contaminant migrations. The objective of our three-year project is to meet the DOE need by developing new experimental approaches to describe adsorption and transport of contaminants in heterogeneous, variably saturated media (i.e., the vadose zone). The research specifically addresses the behavior of strontium, a high priority DOE contaminant. However, the key benefit of this research is improved conceptual models of how all contaminants migrate through heterogeneous, variably-saturated, porous media. Research activities are driven by the hypothesis that the reactivity of variably saturated porous media is dependent on the moisture content of the medium and can be represented by a relatively simple function applicable over a range of scales, contaminants, and media. A key and novel aspect of our research is the use of the

2-meter radius geocentrifuge capabilities at the Idaho National Engineering and Environmental Laboratory (INEEL) to conduct unsaturated reactive transport experiments (Figure 1). The experimental approach using the geocentrifuge provides data in a much shorter time period than conventional methods allowing us to complete more experiments and explore a wider range of moisture contents. The vadose zone research being done in this project will demonstrate the utility of environmental geocentrifuge experimental approaches and their applicability to DOE's vadose research needs.

RESEARCH PROGRESS AND IMPLICATIONS

The objectives are being accomplished by conducting integrated conventional laboratory column experiments and geocentrifuge approaches to assess breakthrough curves for one- and two-dimensional systems with constructed heterogeneities under conditions of variable moisture content. In addition, we plan to conduct similar experiments using intact cores of natural soil formations. This report summarizes our progress as of June 15, 2004 and represents the first 1.5 years of a three-year project. Key activities to date are described in the following paragraphs.

Review of the theory of unsaturated flow in the geocentrifuge. Results suggest that a general theory of unsaturated fluid flow in a variably accelerated field such as found in a finite radius centrifuge is different from the scaling relationships that have previously been assumed. We presented these results at the National Meeting of the American Geophysical Union National in San Francisco, CA in Dec. 2003.

Development of numerical tools. We are collaborating with Dr. Jirka Simunek to modify the

Figure 1. One-dimensional column used to obtain EC breakthrough curves mounted on

Hydrus 1D and Hydrus 2D codes to account for variable acceleration encountered on the centrifuge. We are using these codes to design of our experiments as well to analyze and interpret our experiments. In addition, Dr. Simunek has provided a modified code that will allow us to include the dependence of the reactivity on the water saturation.

Design and construction of an in-flight fraction collector. While sample collection is well established for conventional laboratory experiments, there is little experience in identifying robust instrumentation that can be remotely controlled while in flight on the geocentrifuge. Much of our effort this year has been dedicated to resolving this issue. We have designed and constructed a vacuum operated fraction collection system that can be controlled in-flight. The current design will allow us to collect up to 26 samples of up to 10 ml each. The system can be easily cascaded to yield up to 104 samples.

Figure 2. Fraction collector for geocentrifuge experiments.

Design, construction, and calibration of soil moisture sensors. In-flight soil moisture measurements are required for studies of transport in unsaturated porous media. We have modified the design of the commercially available ECH₂O probes for measuring soil moisture content. The new design consists of a flexible electrode that is affixed to the inside wall of the column and wraps approximately $\frac{3}{4}$ of the circumference. Calibration of the electrodes suggests that water saturation can be measured to within $\pm 3\%$ over the range of 0 to 100% saturation.

Figure 3. Original design of ECH₂O probe (left) and new flexibel design (right).

Breakthrough curves. Experimental break through curves (BTCs) of a conservative tracer were obtained on a 30-cm long column to test fluid delivery systems and general design concepts. At the top of the column, a constant head of water was established over a porous ceramic plate to provide a constant flux at steady state. A water table established at the bottom of the column maintains a constant potential at the outlet. An electrical conductivity (EC) probe provided real-time monitoring of the tracer in the outflow at one-second intervals. Solute BTCs were obtained at 10 and 20 g acceleration. The BTC at 20 g was obtained in roughly half the time as it took to get breakthrough at 10 g. These curves were obtained in approximately $1/10^{\text{th}}$ and $1/20^{\text{th}}$ of the time it takes by conventional techniques.

Figure 4. EC breakthrough curves obtained from one-dimensional column on the geocentrifuge

PLANNED ACTIVITIES

We will begin our one-dimensional reactive transport experiments on ostensibly homogeneous systems to determine the properties of the materials that will be used in the two-dimensional experiments that will be conducted later. In addition, conventional column experiments using a vacuum chamber will provide comparison between techniques. Both types of experiments will utilize conservative (Cl^-) and reactive tracers (Sr^{2+} , Zn^{2+} , F^-) to obtain BTCs as functions of moisture content. In addition, moisture content-matric potential- hydraulic conductivity relationships will be experimentally obtained. Our experimental results will be used to determine reactivity-moisture content relationships.

Two-dimensional tracer experiments will be conducted using the geocentrifuge using obviously heterogeneous materials. The type of heterogeneities will include: 1) systems with continuous alternating horizontal layers of low and high permeability; 2) systems with continuous alternating vertical bands of low and high permeability that can simulate vertical fractures (INEEL) or clastic dikes (Hanford); and 3) systems with discontinuous high permeability layers in a low permeability matrix or discontinuous low permeability layers in a high permeability matrix that simulate clay lenses or the current conceptualization of the INEEL vadose zone. Although the experiments examine idealized-heterogeneity, the heterogeneity patterns capture the essential features and characteristics of common types of heterogeneities observed at DOE sites.

“Intact” sections of soil will be obtained for experiments in the geocentrifuge. We have initiated discussions with members of Phil Jardines’s research group (support through EMSP project number 86911) on collaborating in the collection of an intact core and the sharing of experimental results. We plan to conduct the intact section experiment in FY 2005. The intact section will be monitored in a manner similar to that used for the ideal heterogeneous packs described in the previous task. Cl^- , F^- , Sr^{2+} and Zn^{2+} will be run through the soil section, and breakthrough curves obtained for at least five different average water contents. The reactivity/moisture content obtained for this naturally heterogeneous soil will be compared to the relationships obtained from the two-dimensional experiments using idealized heterogeneities to determine if the same general function is obtained. To test for potential anisotropy, we will conduct another experiment where flow is perpendicular to the first experiments. The data will

be analyzed using the numerical tools previously described.

INFORMATION ACCESS

Discussions of our project were presented at the Geocentrifuge Workshop held at the Idaho National Engineering and Environmental Laboratory, Idaho Falls, ID, March 27-28, 2003. Information about the workshop and summaries of presentations can be found at www.inel.gov and under “geocentrifuge”.

Dr. E.D. Mattson gave a presentation the Southern Idaho Section ASCE on October 23 on Environmental and Geo-Engineering Research at the INEEL focused on our geocentrifuge studies.

A poster session of geocentrifuge techniques at the annual American Geophysical Union meeting in San Francisco was organized by Drs. E.D. Mattson (INEEL) and P. Culligan (Columbia University). We were associated with five posters at the session.

Project presentations and posters include:

Palmer, C.D., E.D. Mattson, and R.W. Smith. Coupled flow and reactivity in variably saturated porous media. EMSP Annual PI meeting, May 6-7, 2003, EMSL Facility, Richland, WA. (See www.pnl.gov/emsp/fy2003/presentations/palmer_carl_86598.pdf).

Mattson, E.D., K.E. Baker, C.D. Palmer, R.W. Smith, and J. Simunek, 2003. One-dimensional solute transport in variably saturated soil using a geocentrifuge apparatus. American Geophysical Union Meeting, San Francisco, CA, Dec. 8-12, 2003.

Simunek, J., E.D. Mattson, and C.D. Palmer, 2003. Modifications of the HYDRUS software packages for analyzing transient flow and solute transport experiments from the centrifuge. American Geophysical Union Meeting, San Francisco, CA, December 8-12, 2003.

C.D. Palmer, J. Crepeau, R.W. Smith, 2003. Fundamental equations of flow in the geocentrifuge. American Geophysical Union Meeting, San Francisco, CA, December 8-12, 2003.

Nakajima, H., E.D. Mattson, and A.T. Stadler. Unsaturated hydraulic properties determined from geocentrifuge tests. American Geophysical Union Meeting, San Francisco, CA, December 8-12, 2003.

R.M. Holt, R.J. Glass, J.M. Sigda, and E.D. Mattson. Phase structure in a dentrifugal field: Impact of capillary heterogeneity and angular velocity. American Geophysical Union Meeting, San Francisco, CA, December 8-12, 2003.