The Subsurface Flow and Transport Experimental Laboratory: A New Department of Energy User's Facility for Intermediate-Scale Experimentation

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Abstract. Intermediate-scale experiments for flow and transport are important for the physical simulation of subsurface features and conditions encountered in the field at government and private sites. To offer a capability where scientists and engineers are able to conduct such experiments, a new facility has been developed in the Environmental Molecular Sciences Laboratory at the Pacific Northwest National Laboratory. The new facility, called the Subsurface Flow and Transport Experimental Laboratory (SFTEL), offers a variety of columns and flow cells, a new state-of-the-art dual-energy gamma system, a fully automated saturation-pressure apparatus, and extensive analytical equipment for sample processing. The new facility, including qualified staff, is available free of cost for scientists interested in collaboration on conducting high-quality flow and transport experiments, including contaminant remediation. Close linkages exist between the SFTEL and numerical modelers to aid in experimental design and interpretation. In this paper, the new facility is discussed and some examples of recent intermediate-scale experiments are presented.

1. Introduction

Intermediate-scale experiments (ISE) offer the ability to study, under controlled laboratory conditions, complicated processes characteristic of mixed wastes and heterogeneous subsurface environments, in multiple dimensions and at different scales. ISEs may, therefore, result in major cost savings if employed prior to field studies. A distinct advantage of ISEs is that researchers can design physical and/or chemical heterogeneities in the porous media matrix that better approximate natural field conditions and therefore address research questions that contain the additional complexity of processes often encountered in the natural environment. An ISE is characterized by configurations that allow small-scale processes to manifest themselves at a larger scale so that their relative contributions to flow and transport

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phenomena can be studied and quantified (Lenhard et al., 1995). On the other hand, the size of the experiment has to be small enough for the environment to be controlled. In addition, the experimental column or container dimensions have to be compatible with measurement and sampling techniques.

To facilitate intermediate-scale flow and transport research, the Environmental Molecular Sciences Laboratory (EMSL) at Pacific Northwest National Laboratory (PNNL) has recently opened the SFTEL laboratory equipped with various columns, flow cells, equipment to determine fluid and porous medium properties, and analytic tools. The new laboratory is a U.S. Department of Energy User Facility. The main objective of this paper is to discuss the new facility and the procedures to obtain access to the equipment and PNNL support staff. In addition, a brief overview is presented of recently conducted ISEs at PNNL.

2. The Subsurface Flow and Transport Experimental Facility

The new SFTEL is part of the Environmental Spectroscopy and Biogeochemistry Facility of the EMSL. The EMSL is the cornerstone of the U.S. Department of Energy's (DOE) commitment to provide high-quality capabilities for enabling fundamental research on the physical, chemical, and biological processes. A more thorough understanding of these processes lays the foundation for new solutions to environmental problems. The EMSL is a national scientific user facility and research organization. It offers, at one location, a comprehensive array of leading-edge resources available on a peer-reviewed proposal base. The SFTEL has been recently added to the EMSL capabilities. Researchers from around the world who share their results from their selected proposals in the open literature are given access to the SFTEL – at no cost. Under normal operating procedures, the users are only responsible for travel and lodging expenses. Users of the facility have the option of staying at the User Housing Facility on the campus of PNNL.

The main focus of the SFTEL is on intermediate-scale experimentation. The laboratory offers several meter-scale flow cells and columns for research in saturated and unsaturated porous media. The potential research is certainly not limited to nonaqueous phase liquid (NAPL) investigations. Any flow, transport, or remediation research request will be considered. The laboratory is equipped with a newly constructed, fully automated, dual-energy gamma radiation system (Fig. 1). The apparatus can be used to simultaneously and nondestructively determine:

1 Saturation of two immiscible fluids (water/NAPL; water/air; NAPL/air)
2 Bulk density and water content
3 Water content and salt concentration

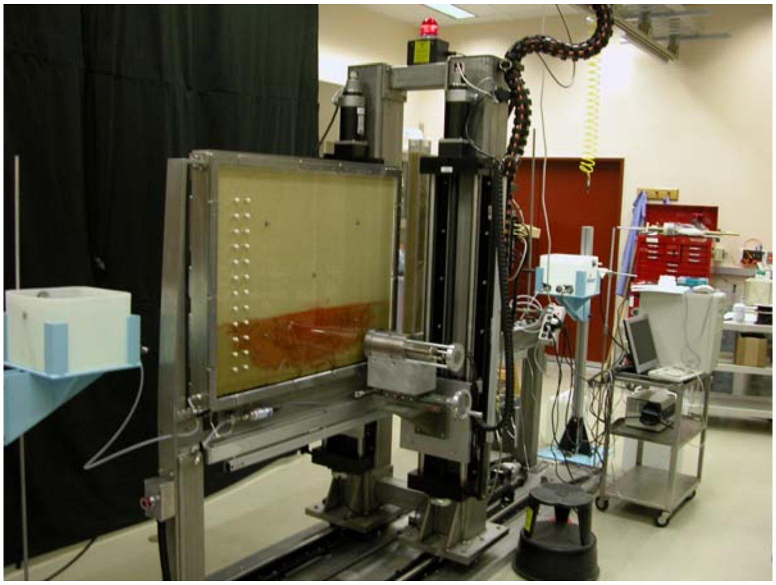

Figure 1. The new SFTEL dual-energy gamma radiation system

A fully automated saturation- fluid pressure apparatus is operational to determine constitutive relations of two- and three-phase fluid systems in porous media. The SFTEL has a well-equipped analytical laboratory to provide chemical analysis support. The instruments available for support of flow and transport research are Inductively Coupled Plasma Mass Spectrometer, Gas Chromatograph- Mass Spectrometer, Liquid chromatograph, Ion Chromatograph, Capillary Electrophoresis, and a Total Organic Carbon Analyzer. Staff, software, and computational facilities are available to design experiments and to compare experimental results with numerical predictions. An example of the available software is the STOMP (Subsurface Transport Over Multiple Phases) simulator (White and Oostrom, 2003).

The process for using the EMSL facilities begins with a User Proposal Form submitted via the EMSL User System. The web site for the User System is located at http://www.emsl.pnl.gov. The proposal will be screened for scientific content by an internal committee. Once the proposal has been approved, the requestor and PNNL scientists make further arrangements.

3. Overview of Intermediate-Scale Experimental Studies

Intermediate-scale flow and transport research has been conducted at PNNL for several years. The first detailed flow cell experiment was described by Lenhard et al. (1995). The experiment focused on investigation of density-dependent gas advection of trichloroethylene (TCE). The experimental results clearly indicated that vapor-density effects should be considered and that density-driven vapor advection may be an important mechanism for moving volatile organic components with high vapor pressures and molecular mass through the subsurface. The experiment was successfully modeled using the STOMP simulator.

Papers by Oostrom et al (1999a,b) discussed the movement and remediation of TCE in a saturated heterogeneous porous medium packing in an intermediate-scale flow cell. To remove a liquid TCE spill, pump-and-treat (P&T) as well as surfactant flushing (SF) techniques were used. Dissolved TCE concentrations were measured at 20 locations, while fluid saturations were obtained with a dual-energy gamma scanner. A total of six alternating P&T and SF periods were used to remediate the flow cell. During the first P&T period, most entrapped TCE was removed but TCE saturations in a substantial pool on top of a fine-grained sand layer were largely unaffected. During the first SF period, a dense plume was formed containing solubilized TCE which partially sank into the fine-grained sand. In addition, unstable fingers developed below the liquid TCE in the pool. In several samples, small TCE droplets were found, indicating mobilization of TCE. The SF considerably reduced the amount of TCE in the pool on top of the fine-grained sand. During the second P&T period, plume sinking and instabilities were not observed. After starting the second SF period, some unstable fingering and plume sinking resumed, starting at the upstream end of the TCE in the pool. The saturation distribution obtained after the second SF period was quite similar to the one obtained after the first SF period, indicating that additional removal of TCE through SF was difficult as a result of the limited accessibility of the TCE in the pool. A gamma scan, obtained after 3 weeks of pumping using the 3-well configuration, shows that all the liquid TCE had been removed from the coarse sand. Computations based on extraction rates and measured TCE concentrations show that only about 60% of the injected TCE were removed from the cell during the experiment. Part of the missing 40% might have moved downwards into the fine sand as a result of pure phase mobilization. The experimental results suggest that besides the positive effects of solubilization, possible detrimental processes such as pure phase mobilization and dense aqueous-phase plume behavior should be considered during SF.

Intermediate-scale experiments dealing with the partitioning interwell tracer test (PITT) to detect and quantify NAPLs were discussed in Brusseau et al (2002) and Nelson et al. (1999). The purpose of the PITT experimental work was to examine the effect of porous-media heterogeneity, nonuniform NAPL distribution, and sampling method for measuring saturation in saturated subsurface systems. The intermediate-scale flow cell contained two discrete zones of TCE and at residual saturation. One zone consisted of a 10% saturation in the same coarse sand as used for the flow-cell matrix. The other zone consisted of a 10% saturation in a finer sand emplaced within the coarse sand. Fluid saturation were obtained with a gamma system, while dissolved concentration were determined from horizontal and vertically-integrated samples. Results indicate that the presence of porous-media heterogeneity and a variable distribution of NAPL saturation can lead to reduced accuracy of the tracer test. The reduced performance can be improved, in part, by using depth-specific sampling.

Oostrom et al. (1997) and Oostrom and Dane (1997) conducted flow cell and column ISEs to test the ability of commonly used nonhysteretic and hysteretic constitutive relations between the relative permeability, fluid saturation, and capillary pressure to described multifluid flow in two dimensions. The results show a reasonable match between the experimental and numerical data, indicating that the constitutive relations are adequate to describe relatively slow NAPL infiltration and redistribution.

Schroth et al. (2001) used column ISE to demonstrate how in-situ oxidation of TCE by permanganate affects the porous medium hydraulic properties. The experiments demonstrate that successful application of this remediation technique requires consideration of detrimental processes such and MnO_2 precipitation and CO_2 gas formation. In addition, the results show that utilization of a buffered oxidant solution may improve the effectiveness of in-situ oxidation.

Williams and Oostrom (2002) used an ISE to demonstrate how oxygenation of anoxic water in a fluctuating water table system affects oxygen concentration levels. The hypothesis that water table fluctuations increase oxygen transfer from air to water through enhanced exchange from entrapped water was tested. The experimental results show that zones with entrapped air, formed during the imbibition portions of the experiment, were instrumental in re-oxygenation of the water. The fluctuating water table system also allowed oxygen to move deeper into the flow cell. The STOMP simulator was able to predict water and entrapped air saturations well.

The most current ISE work at PNNL is related to the formation of residual NAPL saturation in the vadose zone. Descriptions of mobile and entrapped NAPL behavior are now standard features of some multifluid flow simulators. However, theseDespite recent contributions describing NAPL residual saturation formation (Lenhard et al., 2003; Van Geel and Roy, 2002; Wipfler and van der Zee, 2001), these models do not generally incorporate retention of NAPL in the vadose zone following imbibition events (e.g., surface spills, tank leaks). Commonly used constitutive relations assume a nonzero NAPL relative permeability when the non-entrapped NAPL saturation is greater than zero. As a result, this NAPL is allowed to drain from the pore spaces. Neglecting residual NAPL saturation formation might cause an overestimation of the volume of NAPL that reaches the ground As NAPL retained in the vadose zone can serve as a long-term source for ground water contamination, via transport through the gas or aqueous phase, understanding and predicting the processes of residual NAPL formation and remobilization is critical when considering restoration or management of a contaminated subsurface site.

Laboratory experiments have shown that formation of residual NAPL might be an important process under certain conditions and needs to be considered in modeling NAPL migration, redistribution, dissolution and volatilization. Two recent intermediate-scale experimental studies (Oostrom and Lenhard, 2003; Oostrom et al, 2003) clearly demonstrate the formation of residual carbon tetrachloride saturation in heterogeneous porous media. Fig. 2 shows a picture of the final carbon tetrachloride distribution in a fine-grained sand layer in a coarse-grained sand matrix. Fig.3 shows a gamma scan after movement of the carbon tetrachloride has ceased.

Figure 2. Final carbon tetrachloride distribution in fine-grained sand layer.

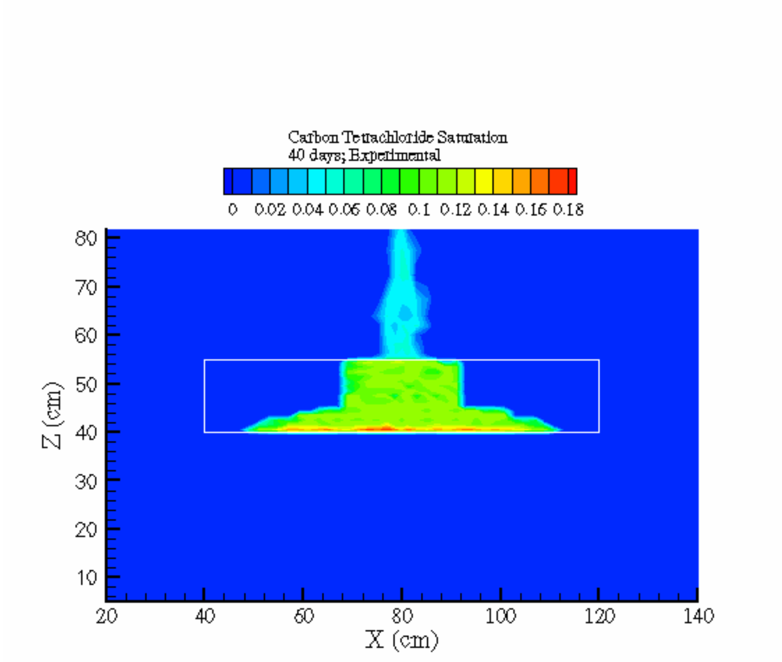

Figure 3. Measured carbon tetrachloride saturations of situation depicted in Fig. 2.

A theoretical description of the residual saturation formation process was provided by Lenhard et al. (2004). The theory was incorporated in to the STOMP simulator by White et al. (2004). Initial testing of the theory was provided by a comparison of predicted and measured fluid saturation of intermediate-scale column experiments. The results show that the new theory is capable or predicting the measured saturation well. The standard nonhysteretic theory consistently under predicts NAPL saturations because it allows continuous drainage. An example of this behavior is shown in Fig. 4. where the measured and predicted dodecane saturations are shown for a column experiment where a slug of this NAPL was allowed to infiltrate and drain into a partially saturated meter-scale column.

4. Acknowledgements

Pacific Northwest National Laboratory (PNNL) is operated by the Battelle Memorial Institute for the Department of Energy (DOE) under Contract DE-AC06-76RLO 1830. A portion of the research described in this paper was performed in the Environmental Molecular Sciences Laboratory (EMSL), a national scientific user facility sponsored by the DOE's Office of Biological and Environmental Research and located at PNNL. Information on the STOMP simulator can be obtained by contacting the senior author or by visiting http://www.pnl.gov/etd/stomp. For more information on the SFTEL, contact M. Oostrom at mart.oostrom@pnl.gov.

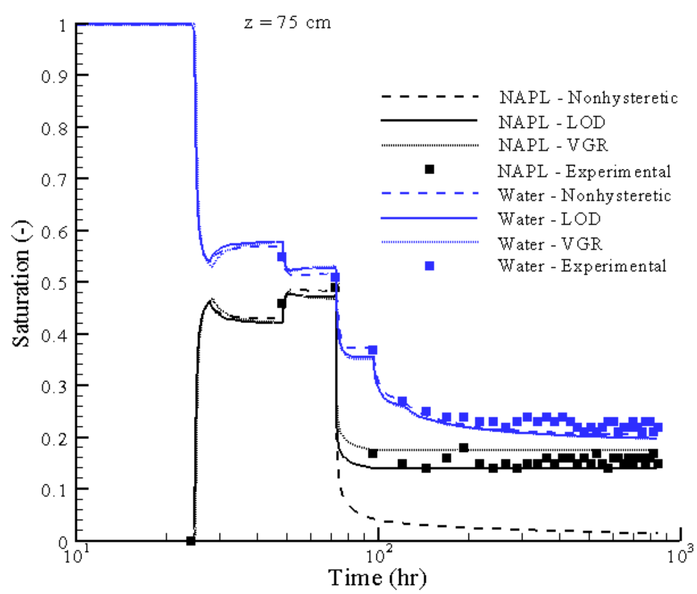

Figure 4. Predicted and measured dodecane saturations.

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