TESTING MODELS OF FLOW AND TRANSPORT IN

UNSATURATED POROUS MEDIA

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ABSTRACT

The design of an intermediate-scale flow and transport experiment in an unsaturated porous media is described. This experiment will be conducted a 3-m-diameter by 6-m-long caisson filled with silica sand. The study has been designed to minimize uncertainties in the conceptual model governing flow and transport of nonreactive tracers and to evaluate predictions of field tracer migration using laboratory data. Data from laboratory characterization of hydraulic and geochemical properties of the sand and tracer were used in preliminary modeling and sensitivity analyses to determine optimal fluid flux, influent tracer concentrations, and locations of sensors and sampling devices. Alternative approaches to compare observed and predicted data for model evaluation are discussed.

INTRODUCTION

The Yucca Mountain Site Characterization Project (YMP) is responsible for characterizing and assessing the suitability of Yucca Mountain, Nevada, for the siting of a potential high-level nuclear waste repository. Regulations governing the repository licensing process require assessments of potential radionuclide transport from the repository to the accessible environment for time periods up to 10000 years. Performance assessment will use subsurface flow and transport models or an appropriate abstraction of them in conjunction with data collected during site characterization to determine if the potential repository can meet the regulatory requirements. The validation step in the modeling process will consist of comparing predictions made using an independent data set to experimental results. This step is necessary to ensure confidence in the computer codes used to support the license application for the potential high-level waste repository.

The term validation invokes a sense of certainty that is difficult to achieve with models of geological settings and the time frame required for repository assessment[1]. Generic validation or validating of a model that will predict correctly under all conditions is impossible to achieve given current knowledge in the geosciences. Consequently, various approaches including laboratory, intermediate and field-scale experiments, and information from analogues will be used by the YMP for model validation. Advantages and disadvantages are associated with each approach, and no single one can satisfy all of the requirements for validation; however by employing all of the above approaches, it is possible to evaluate the models and their associated computer codes using data from a wide variety of conditions and achieve more accurate prediction.

This paper describes the status of an intermediate-scale experiment to test models of flow and transport through unsaturated porous media. The design described below was selected because porous material emplacement and boundary conditions can be well controlled, and point and total system tracer responses can be measured simultaneously. Models for both reactive and nonreactive tracers will be tested.

The intermediate-scale design was selected for nonreactive tracers because the underlying conceptual model governing water flow and tracer transport is a porous media continuum, which we have assured by constructing the porous medium and thus avoid introducing discontinuities that require consideration of

alternate conceptual models. Our objective is to reduce uncertainties associated with the model structure. The two sources of error that cannot be controlled are these associated with measurement and parameter heterogeneity. Measurement errors will be bounded by the choice of the instruments and procedures; and parameter heterogeneity will be introduced by packing the caisson with sand. The description of this experiment that follows discusses how parameter heterogeneity will be characterized.

Behavior of reactive tracers will be used to discriminate among alternate sorption models including K_d, or linear sorption, nonlinear sorption, ion exchange, and surface complexation models. Parameters for these models will be obtained using geochemical characterization described in a companion paper.[2] Predictions of tracer migration based on data from laboratory experiments will be compared to the results of this intermediate-scale experiment.

The highly controlled conditions and scale of this experiment provide an opportunity to evaluate predictive performance of transport models for conditions where three-dimensional flow and heterogeneities occur. These results are a valuable prerequisite for evaluating

field tests because better prediction of tracer migration cannot be expected when model structure is unknown and parameter heterogeneities and measurement errors are present. The measures of proposed criteria comparing predicted versus observed tracer migration from this study provide a foundation for gauging results from more complicated experiments.

DESCRIPTION OF THE ACTUAL WORK

Background

This intermediate-scale experiment will be carried out in a caisson, a corrugated, galvanized metal highway culvert set in a concrete base that is 6-m high by 3-m diameter[3]. Access ports are located at six different depths 75-cm apart to allow horizontal placement of instruments (Figure 1).

Siegel et al.[4] presented an initial design for this experiment and indicated that the final design would require several iterations of preliminary calculations, simple laboratory experiments and prototype constructions. This initial design consisted of a relatively chemically inert material, Wedron 510 silica sand (Wedron Silica, Wedron, IL 60557), as the background

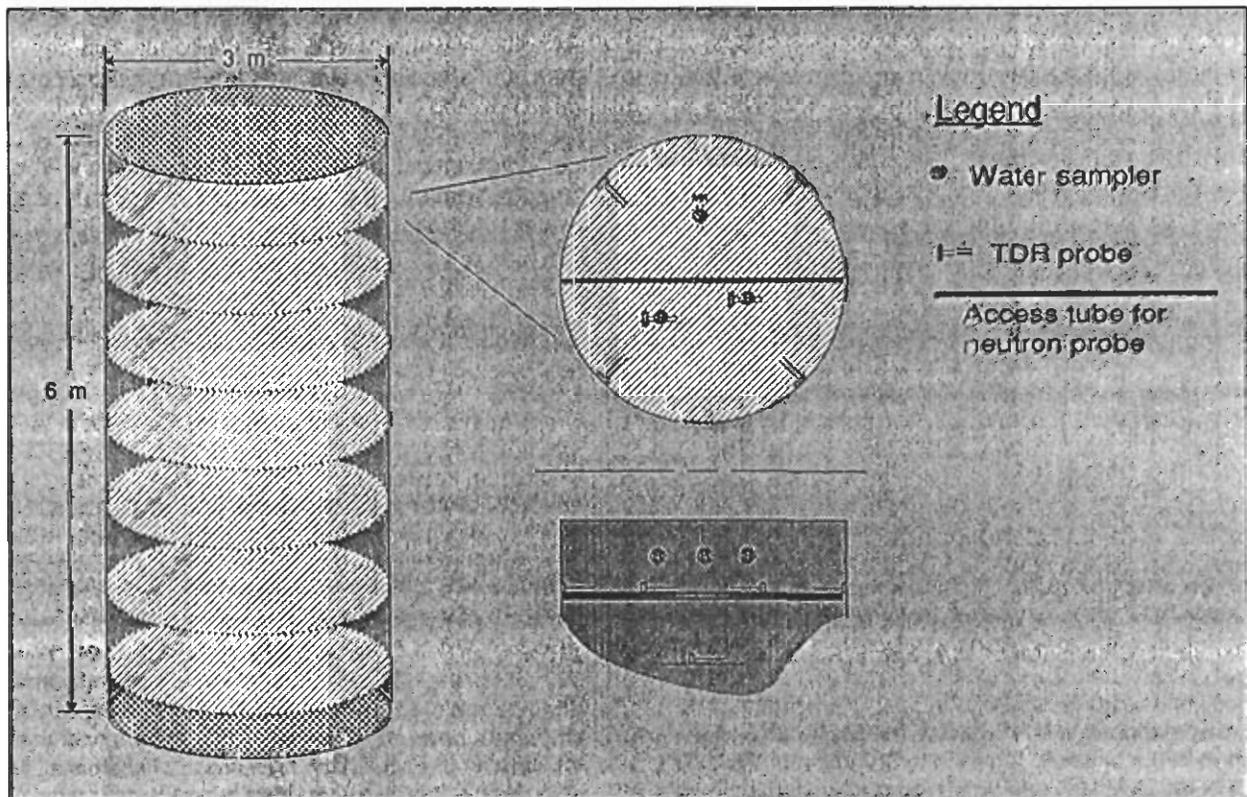## Caisson Geometry and Instrumentation

Figure 1. Schematic of the caisson showing locations of instrument.

porous matrix and a sorbing layer containing limonite. Characterization of the hydraulic properties of the porous medium and a description of transport calculations used in the design are presented below. A brief description of the chemical properties of the porous matrix is also given; details of the geochemical characterization studies are provided in a companion paper[2].

Studies of the hydraulic properties of the porous materials to be used in the caisson were carried out in laboratory columns and two-dimensional slabs filled with sand. The slab chambers are 60 x 30 x 1-cm so that high-resolution measurements can be made of moisture content or tracer concentration using either light transmission or x-ray. The two-dimensional slab chambers also can be used to develop scaling laws for effective media properties including the small-scale layering and cross-bedding that are expected to result from packing the caisson.

The geochemical properties of sand and limonite, which are important to tracer transport, were characterized to determine their suitability for the caisson experiment.[2] Samples of the limonite (Wards Earth Science) were characterized by x-ray diffraction and wet chemical analysis. The surfaces of the sand grains were examined using a combination of techniques including potentiometric titration, acid leaching, optical microscopy, and scanning electron microscopy with energy-dispersive spectroscopy. Chemical analyses of bulk compositions and acid-leachable components were made of several particle size fractions of the Wedron 510 sand. Small-scale column experiments were carried out to study the changes in the pH and composition of leachable components of the sand in dilute NaCl electrolyte as a function of time and water saturation. For the purpose of caisson design, experiments were conducted to examine the most important factors controlling sorption of each tracer. Nickel solubility as function of pH, and the adsorption of nickel (Ni), lithium (Li), and bromide (Br) on sand, limonite, and sand/limonite mixtures were measured using batch techniques. The batch experiments were conducted to bracket the ranges of solution pH values, tracer concentrations, and concentrations of competing anions and cations expected in the caisson experiment. The preliminary data were used for calculation of K_ds'; additional data will be collected and analyzed within the conceptual framework of nonlinear isotherms and surface-complexation models. In addition, interactions between the tracers and solution sampling equipment were studied to estimate uncertainties in measurements of the time-dependent tracer concentration profiles within the caisson.

Several models will be used to predict tracer transport in the caisson. The LEHGC version of the HYDROGEOCHEM[5] coupled transport/geochemical code was used in this and the previous[4] papers. Two three-dimensional flow and transport codes, TRACR3D[6] and FEHMN[7] will be used for tracers that exhibit nonlinear sorption.

A sensitivity analysis using LEHGC to simulate transport with linear sorption was performed to determine optimal flow conditions, solution sampler placement, and tracer input concentration. The response variable was depth of penetration of the solute front at concentrations 10 times current detection limits. Hydraulic parameters and sorption parameters from laboratory experiments were also used in this sensitivity analysis.

The experiment provides opportunities for development of criteria for testing model predictions against observed data, which has traditionally employed goodness-of-fit methods with a statement indicating the relative merit of the result. This experiment offers the advantage of point measurements from within the caisson and an integrated system response via the lower-boundary condition. Comparisons using these point and integrated data are being investigated for application to this experiment.

Bottom Boundary Condition

In previous caisson experiments[8,9], the bottom boundary was free drainage through a 15-cm diameter pipe connected to the central access caisson. This design is considered inadequate because of additional mixing caused by the convergence of the stream tubes and the presence of a locally saturated condition in the vicinity of the outlet. A design to provide a constant pressure boundary and to resolve the spatial distribution of solute transport has been developed. This design will use 268 ceramic porous cups that are vertically oriented and mounted on a plastic base. The tubing from each porous cup will be connected to a manifold that will allow an individual cup to be sampled, and suction will be supplied either by vacuum pumps or through a hanging water column arrangement.

Instrumentation

Figure 1 shows the location and type of measuring and sampling equipment that will be used.

Water content will be measured using both the neutron probe and time-domain reflectometry (TDR). Aluminum access tubes will be installed across the caisson at each caisson access port so that neutron probe measurements can be made. Neutron probe measurements are difficult to make near the air-soil interface because of the sphere of influence of the probe, and usually measurements are made at least 15 cm from any surface, so that measurement of the water content in the outer annulus region of the caisson with the neutron probe is prevented. This annular region is important because of potential wall effects or channeling of water at the boundary. TDR rods will be installed in the annular region to provide data on water content.

Solution samplers will be made from either ceramic cups or hollow-fiber porous membranes. Previous

caisson experiments[3] identified interactions between the ceramic material of the porous cups and the sorbing tracers, and the hollow-fiber bundle was developed to circumvent problems related to tracer uptake and consequential errors in estimated breakthrough times. The solution samplers are actuated by applying a suction to the tubing and collecting a sample after a given number of sample system volumes have been discharged. One of the constraints on the hollow-fiber sampler, however, is the amount of suction that can be applied without collapsing the filter.

RESULTS

Hydraulic Properties

Laboratory studies have been conducted to determine the hydraulic properties of the Wedron silica sand that was selected as the porous medium. Using the two-dimensional glass slab chambers (60 x 30 x 1 cm), the Wedron sand was emplaced with a filling extension containing randomized screens.[10] Although a homogeneous fill was desired, the pack did contain small-scale sinusoidal microlayering (Figure 2).

Transient moisture content of the slab for a series of wetting and drying events was followed using the light transmission technique.[11] Figure 3 shows the moisture profiles for the secondary wetting curve (Figure 3A) and primary drainage curve (Figure 3B). Moisture content was measured at 300 points across the width (30 cm) of the chamber at 0.1 mm resolution in height. Average moisture content was calculated for each height, which resulted in values for the average primary drainage, average secondary wetting, and drainage saturation and pressure relations given in Figure 4.

The sand exhibits substantial hysteresis and increased air entrapment within the material after primary wetting and drainage. This increased trapping caused a reduction in "saturated" moisture content of about 10% (from 0.34 to 0.31) and a reduction in "saturated" conductivity of about 65% (from 0.87 to 0.31 cm/min). These observations imply that it will be important to achieve a constant saturation throughout the caisson. The steady state flux rate will be established by filling the caisson with the background electrolyte solution from the bottom so that air can vent through the surface. After the caisson has been saturated, the lower-boundary condition will be applied and the surface-boundary flux, applied by a rotating spray system, will be adjusted to achieve a steady-state flux rate. In order to minimize the hysteresis effects, this procedure may be repeated three times.

The moisture characteristic relation, $\theta(\psi)$, can be described by any number of relationships. The equation for the moisture characteristic from Brooks and Corey[12] is

$$\theta(\psi) = \epsilon \left[\frac{|\psi_{ao}|}{|\psi|} \right]^{\lambda} \left[1 - \frac{|\psi_{ao}|}{|\psi|} \left[1 - \frac{\theta_s}{\epsilon} \right] \right], \psi < \psi_{ao} \qquad (1)$$

$$\theta(\psi) = \theta_s, \psi > \psi_{ao} \qquad (2)$$

where ϵ is the porosity; ψ_{ao} is the air entry or bubbling pressure; λ is the Brooks-Corey fitting parameter; and θ_s is the sand at saturation including entrapped air. Siegel et al.[4] used the method of Haverkamp and Parlange[13] and the Brooks-Corey relation to calculate the primary drainage curve of the Wedron sand from the grain size
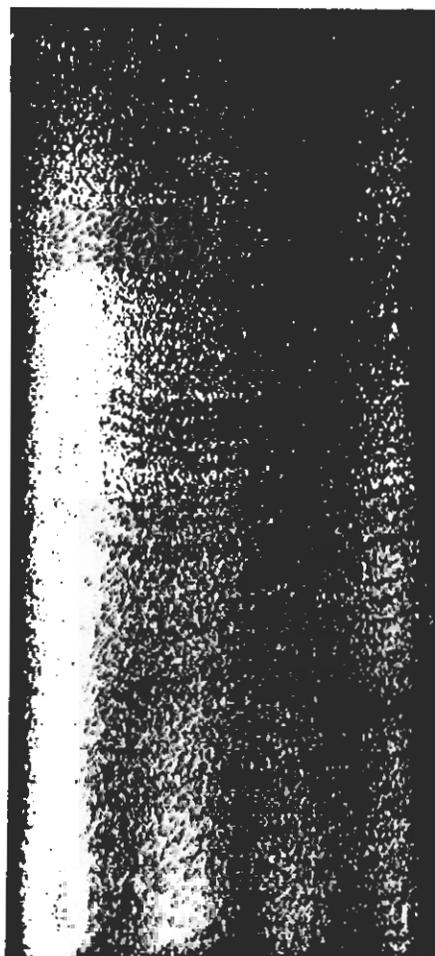

Figure 2. Heterogeneity field within the "uniform" pack of Wedron 510 sand within a thin (1 cm) 30 x 60 cm slab chamber as illuminated with the light transmission technique. Dark zones denote regions of higher bulk density and smaller mean grain size; light zones denote regions of lower bulk density and larger mean grain size. Sinusoidal microlayers of about 0.1 to 1.0 cm in thickness run from side to side in the chamber. Also note light and dark bands that run from top to bottom of the chamber cross-cutting the microlayering.
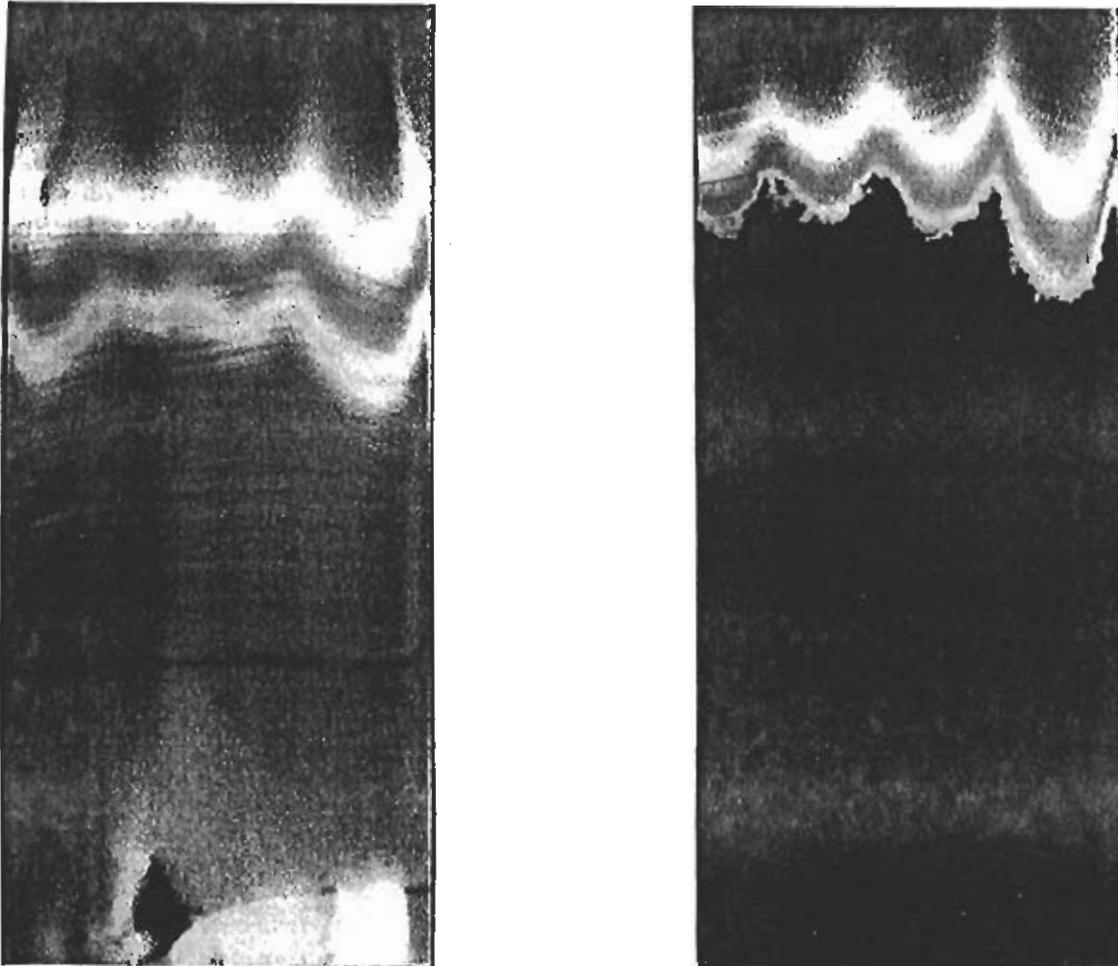
Figure 3. Equilibrium moisture content fields for the Wedron 510 sand pack shown in Figure 2 as illuminated with the light transmission technique. Images are black and white reproductions of false colored saturation images so that dark and light bands do not reflect absolute values of moisture content. Image a) depicts the saturation field that determined the primary drainage curve and b) that for the secondary wetting curve. Note that the atmospheric pressure position was approximately the bottom of the image for a) and approximately 20 cm above the bottom of the image for b). Bands of light and dark running across the chamber denote zones of nearly the same moisture content. Figure 3 demonstrates the effects of both the microlayering and vertical heterogeneities shown in figure 2 to vary the local air entry values for the sand pack on the order of 25% of its average value. Higher percent changes are expected in the permeability field due to these variations. An additional feature to note is the entrapment of an air pocket seen the zone at the bottom of the chamber in figure 3b. The air pocket was entrapped after the chamber was drained to field capacity and resaturated by capillary rise as depicted in figure 3b. This demonstrates the effect of local heterogeneity in combination with boundary conditions to create structures that will add to larger scale macrodispersivitiy. Higher rmacrodispersivity for the entire system is expected than will be measured on small diameter core samples taken during the post mortem phase of the caisson experiment.

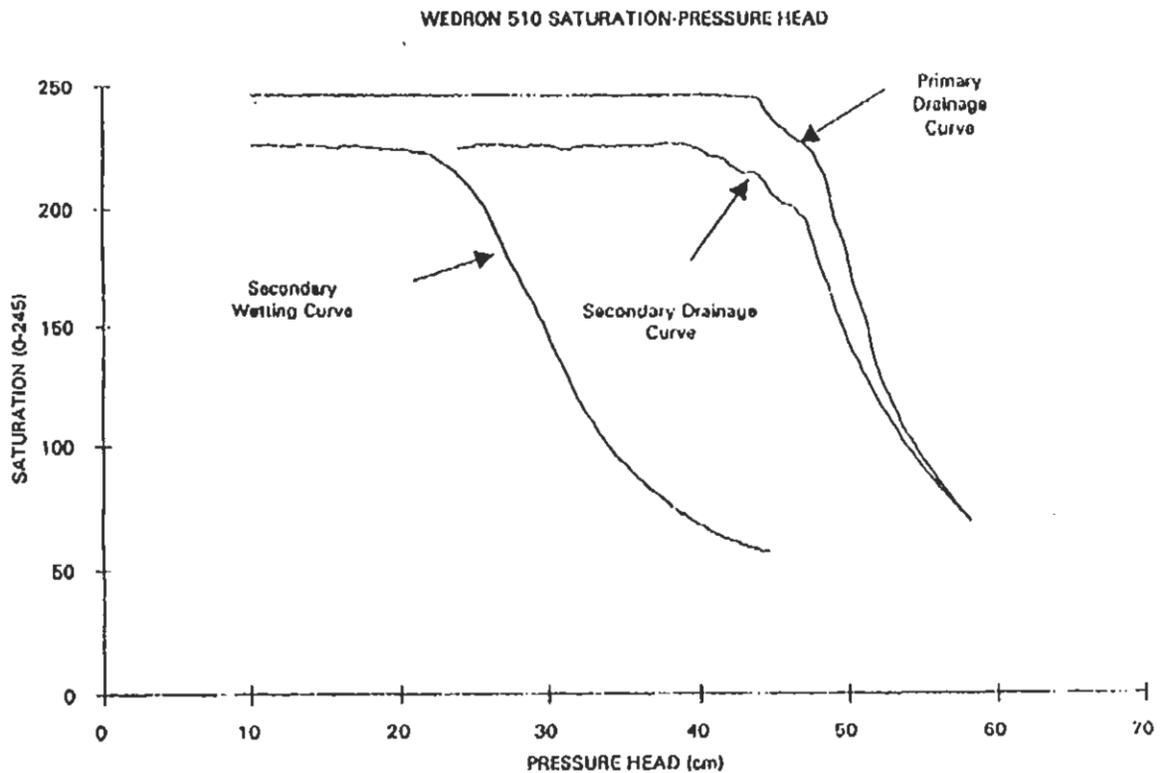WEDRON 510 SATURATION·PRESSURE HEAD

Figure 4: Measured moisture characteristic curves for the Wedron 510 sand. Primary wetting and secondary wetting and drainage curves were obtained from images such as those shown in figure 3a and 3b. Curves were averaged over the chamber to yield effective relations.

distribution and bulk density. A comparison of this calculated curve with the measured primary drainage curve from the slab can be seen in Figure 5 and the predicted parameters for Equation 1 are given in Table 1. From Figure 5, it can be seen that the predicted ψ_{ae} from the approximation of Haverkamp and Parlange[13] is reasonable, but the slope of the predicted drainage curve at higher values of pressure head reveal how poorly λ was estimated. Equations 1 and 2 were fitted to the primary drainage curve data (BC Fit in Figure 5) and the parameters are given in Table 1. The values for ψ_{ae} from the estimated and fitted curves are in reasonable agreement, but the difference between λ values is substantial.

Equations 1 and 2 are not continuous at the air entry value, ψ_{ae}; therefore the moisture characteristic relationship from van Genuchten[14] can be used to make numerical solution easier. The van Genuchten equation is

$$\theta(\psi) = \left[\frac{\theta_s - \theta_r}{(1+|\alpha\psi|)^n} \right]^m + \theta_r, \psi < 0; \qquad (3)$$

$$\theta(\psi) = \theta_s, \psi \geq 0; \qquad (4)$$

where θ_r is the residual moisture content; and α and n are the van Genuchten fitting parameters with $m = 1 - 1/n$. The van Genuchten fit to the primary drainage curve and to the Brooks-Corey curve from the particle-size mapping[4] can also be seen in Figure 5. Parameters for the van Genuchten fit can be found in Table 1. Again, note the agreement in the α values and the substantial change in the slope parameter, n, for the estimated and fitted Equation 3.

Geochemical Characterization of the Porous Matrix and Tracers

Limonite. Mineralogical and wet chemical analysis of three splits of the limonite revealed that the crushed 600-pound sample delivered for the caisson experiment (hereafter referred to as the "caisson limonite") contained much less iron than the limonite sample described in Siegel et al.[4] (referred to as Wards limonite #1). The caisson limonite contained a mixture of goethite-coated sand, goethite grains, detrital aluminosilicates and amorphous phases. Leaching studies in near-neutral solutions yielded significant amounts (>1 ppm) of Ca, Mg, and Ni. Acid leaching studies indicated the presence of an acidic component; the pH of deionized water in contact with the samples (solution:solid = 20:1) was 3.0 - 3.3. Repeated rinsing and boiling with deionized water failed to remove the acidic component.
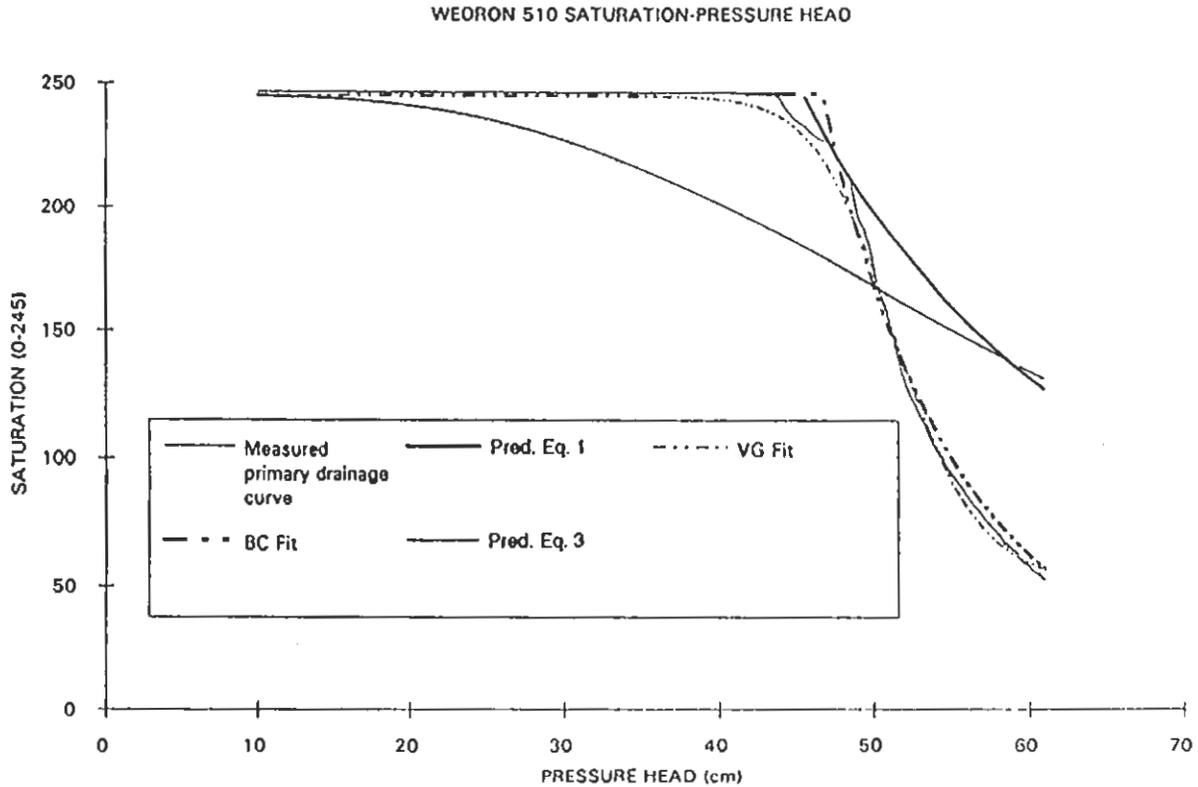
WEDRON 510 SATURATION-PRESSURE HEAD

Figure 5: Moisture characteristic curve prediction. The primary drainage curve for the Wedron 510 sand is compared to the predictions based on the theory of Haverkamp and Parlange[9] which maps grain size distribution and bulk density into the pressure saturation relation. Best fits to the data using the Brooks and Corey form (BC fit) and the van Genuchten form (VG fit) are also shown.

Table 1. Parameters for the Brooks-Corey[12] and van Genuchten[14] moisture characteristic relationships for the Wedron 510 sand using the method of Haverkamp and Parlange[13] to estimate based on particle size distribution and fitting these equation to the observed data for the primary drainage curve.

	Brooks-Corey		van Genuchten	
	Ψ_{ae} (cm)	λ	α (cm^{-1})	n
Estimated	40.75	2.09	.02	3.2
Fitted	42.0	5.0	.0125	18.0

Batch studies of sorption of Ni using mixtures of sand and Ward's limonite #1 (3% limonite: 97% sand) were carried out.[4,15] Sorption ratios (K_ds) ranged from 4 ml/g to 175 ml/g over the pH range 6 - 7.75, respectively. The presence of the acidic component in the caisson-limonite, however, prevented similar experiments with this material at neutral pH. At the low pH characteristic of batch systems containing the caisson-limonite, Ni

sorption was negligible[4,15], therefore layers containing mixtures of the caisson-limonite and sand would not appreciably sorb nickel. The above characteristics of the caisson limonite introduced a level of complexity into the chemical component of the experiment that could compromise the attempt to develop a baseline for comparisons of the predictive capabilities of alternate sorption models. In addition, because the amount of limonite that was available was limited to 272 kg (600 lbs), the "sorbent" sand-limonite mixture would contain only 3% limonite and the difference in magnitude of sorption by the sorbent layer versus the "inert" sand layers would be relatively small. The required effect of a sorption layer might not be seen; therefore, the sand-limonite layer was removed from the experimental design.

Bulk and Surface Properties of Wedron 510 Sand.
According to its bulk chemistry and mineralogy, Wedron 510 sand is nearly pure quartz, but characterization of some of its surface properties as described in Siegel et al.[2] suggests that it contains at least four components: quartz, carbonate, iron oxyhydroxide, and kaolinite. A

titration curve for Wedron 510 sand obtained under CO_2-free conditions over the pH range 2.75 to 11 showed development of a buffering plateau, suggesting slow dissolution of a conjugate base, tentatively identified as a carbonate. Measurements of dissolved Ca and Mg leached from the sand in batch systems are consistent with a concentration of 0.75 - 0.80 μmoles of carbonate per gm sand. Significant Fe (0.575 μmoles/g) and trace amounts of Na and K were also leached. Studies of thin-sections under an optical microscope revealed that the sand grains are coated with a yellowish material; therefore the majority of the Fe is likely to be present as a thin coating of hematite and/or goethite on the sand grains. The bulk analysis of the finest size fraction and the analyses of the surface coatings of all size fractions suggest that the finest size fractions contains fragments of sand grain coatings composed of calcite, dolomite, iron oxyhydroxide and clay minerals.

The results of the leaching experiments are described in Siegel et al.[2] (their Figure 2). The column effluent contains measurable concentrations of the major components of the sand (Ca, Mg, Si) and of the tracers (Ni, Br, Li) that will be used in the caisson experiment. Under unsaturated conditions, the effluent pH dropped to a steady-state range of 7.2 to 7.5 after elution of approximately 5 pore volumes. In preliminary saturated-condition experiments (not shown), the pH of the effluent was initially neutral, rose to >8.7, and then reached a steady-state value of approximately 8.2 after elution of 6 pore volumes.

The above results suggest that significant amounts of the Li, Ni, and Br will not be leached from the sand and interfere with the tracer tests. The studies indicate, however, that the sand will not be chemically inert during the caisson experiment. The chemical composition of pore fluids within the caisson will be controlled by the following: dissolution of quartz, carbonate cement, diffusion of atmospheric CO_2 into pore fluids, and hydrolysis of silanol (SiOH) and FeOOH surface groups. It is anticipated that chemical conditions within the caisson will initially be transient, but a steady-state chemical profile may develop after elution of 6-10 pore volumes. The flow velocity, the dissolution rate of carbonate cement, and the CO_2 diffusion rate will control the approach to a steady-state pH profile in the caisson; the amount of carbonate cement will determine the lifetime of the steady-state profile.

Sorption of Tracers by Wedron 510 Sand. Ni sorption has been measured in batch experiments equilibrated for at least 48 hours at solid:solution ratios of 1:1 over the pH range 5 to 9.5. The pH - percent sorption curve (Siegel et al.[2], Figure 5) shows a distinct sorption edge at pH ~6.5, reaching maximum sorbance at pH 8.0, followed by slowly declining sorbance at higher pH. The equivalent K_ds range from approximately 0 ml/g (at pH = 5.5) to 80 ml/g (at pH = 8).

Batch studies of sorption of Li and Br by Wedron 510 sand were also carried out[2]. Bromide sorption by sand was negligible over the pH range studied (6.2 - 7.3). Lithium K_ds' range from 0.16 to 0.89 ml/g over the same pH range; the Li concentration was 0.1 to 10 ppm and the solution:solid ratios were 1 - 10 ml/g. Data from lithium sorption measurements are summarized in Table 4 in [2] and show that the K_d depends on both the tracer concentration and the solution:solid ratio.

Instrumentation

Point measurements will be made throughout the caisson to complement the data provided by the exit samplers.

Appendix A in [2] describes studies of the adsorption of the tracers by hollow fiber and ceramic samplers in static and flowing systems. The results indicate that the hollow fiber samplers do not adsorb any of the tracers, but the ceramic samplers may be sources for trace Li and Br contamination and will adsorb nickel. The ceramic samplers cannot be used for Ni sampling because at least 1 l of solution must pass through the sampler before the Ni concentrations of the influent and effluent become equal. Appendix A in [2] also describes studies of the adsorption of Ni as a function of pH by several kinds of labware that could be used for sample collection tubes, syringes, and centrifuge tubes. These include low-density polyethylene, polypropylene, polyallomar, and polycarbonate. The results indicate that each of the plastics become a significant sorber of Ni under neutral and alkaline conditions; this effect will have to be accounted for in sorption and solubility experiments. These studies also indicate that all of the plastics are suitable for use as geochemical sampling vessels for acidified solutions.

Temperature fluctuations that might affect chemical reactions can be expected during the experiment so temperature will be measured at two depths in the caisson with a series of four thermocouples located across the caisson at each depth. Instrumentation will be emplaced during the filling of the caisson. To ensure good contact between instruments and porous material, instrument location is being optimized by preliminary modeling described below. The estimated depth of penetration of the tracers will be used to locate the samplers where they will be the most effective. For example, the hollow-fiber samplers should be located in the upper 300 cm of the caisson to track nickel migration rather than at the lower depths.

Preliminary Transport Modeling

The laboratory-scale hydraulic and geochemical data will be used to make predictions with several different flow and transport computer codes.

The flow problem was solved using the LLUVIA-II code.[16] This version of the code uses the method-of-lines technique to solve the Richard's equation for two-dimensional, isothermal flow of liquid water through partially saturated porous media. Hydraulic properties were measured in the laboratory experiments described in the previous section. All simulations assumed a steady-state water flow that was specified by the pressure applied to the lower boundary and the parameters in either Equation 1 or 3. The computed moisture content and Darcy velocities were written to an external file for use in subsequent transport calculations.

The preliminary transport calculations were made using the LEHGC version of the HYDROGEOCHEM[5] code. Transport of reactive tracers assuming linear sorption was simulated by scaling the capacitance and dispersion/diffusion terms of the transport equation by the retardation factor defined as

$$R = 1 + \frac{P_b K_d}{\theta} \qquad (5)$$

where P_b is the bulk density, K_d is the equilibrium sorption coefficient; and θ is the water content. The range of K_d values were obtained from the batch sorption experiments. The θ values were supplied by the LLUVIA-II calculations. A dispersivity of 5 cm was assumed for the LEHGC calculations.

The effect of flux rate and sorption were examined in this series of simulations. The sensitivity studies in Siegel et al.[4] assumed a ten-day pulse of tracer, a flux rate of 56 cm/day, and a lower-boundary condition of pressure head of -56 cm. From a logistics standpoint, the amount of water applied to the experiment is a consideration particularly if operations are maintained throughout the year. Therefore, a series of simulations were carried out assuming a -75 cm pressure head for the lower-boundary condition, reducing the steady-state flux to 12 cm/day, a ten-day pulse for lithium ($C_o = 1$ mg/l), and a constant source for nickel ($C_o = 0.1$ mg/l). Figure 6 shows the spatial distribution of Li and Ni with two different K_d values for several specified times. The calculations predict that the lithium concentration pulse breaks through at the outlet of the caisson after 25 days, and the nickel will migrate less than 200 cm at concentrations above the detection limit (1 ppb) in 200 days. Differences in transport of the lithium and the nickel are substantial and are expected given the difference in the K_d's. The case where the K_d for nickel is 30 ml/g with a 56 cm/day water flux can be seen in Figure 7. This situation is more favorable, allowing detection of nickel near 300 cm at 100 days (Figure 7). This information along with other data on experimental duration and time constraints will determine the final water-flux rate.

Model Comparisons

Model testing will involve comparison of numerical predictions, such as those presented in Figures 6 and 7, with observed data. The design of the caisson experiment will allow collection of both point data from individual samplers and integrated total system response by the exit samplers. This provides an opportunity to test our ability of using point samples to estimate the total system response.

The microlayering in two-dimensional glass slab experiments (Figure 2) suggest that heterogeneity and variability will be present in the caisson; and the less controlled filling of the caisson will only exacerbate this situation. In the previous saturated flow caisson experiment (Fuentes et al.[9]), variability of solute breakthrough in the horizontal plane increased with transport distance. All pre-experiment calculations, such as those reported in this and the previous[4] paper, assume homogeneous conditions. The uncertainty in solute migration can be addressed using a stochastic approach that requires assumptions about the mean, variance, and spatial correlation of the caisson hydrologic properties that control flow, and geochemical parameters controlling reactive transport. In a field experiment, the parameter statistics are obtained by sampling locations near the test volume and assuming a stationary random field. The test volume is minimally disturbed in this process, and this results in a minimum effect on the flow within the test region. The caisson presents a different problem because the variability in properties is a product of the filling process and is difficult to describe a priori. Disturbance of the test volume by introducing vertical or horizontal discontinuities through sampling may substantially alter the caisson flow and transport system. Post experiment sampling is the alternative that has been selected for characterizing the caisson. In this step, a number of samples will be removed, and their hydraulic and chemical properties will be characterized. By knowing the spatial location of the sample, the statistics that describe these parameter fields can be obtained. This information will be used for a new round of predictions to see if any improvement has been attained by the additional sampling.

Various objective functions for comparing observed and predicted hydrologic or chemical state variables can be defined. In many cases, these have been defined for model calibration exercises where goodness-of-fit is used to select a parameter set, boundary or initial condition, or source/sink term for a model. Other functions besides goodness-of-fit include comparing the temporal and/or spatial moments of predicted and observed concentration field. This approach has the advantage of providing an integrated quantity with a smoother response than the individual concentration measurements.
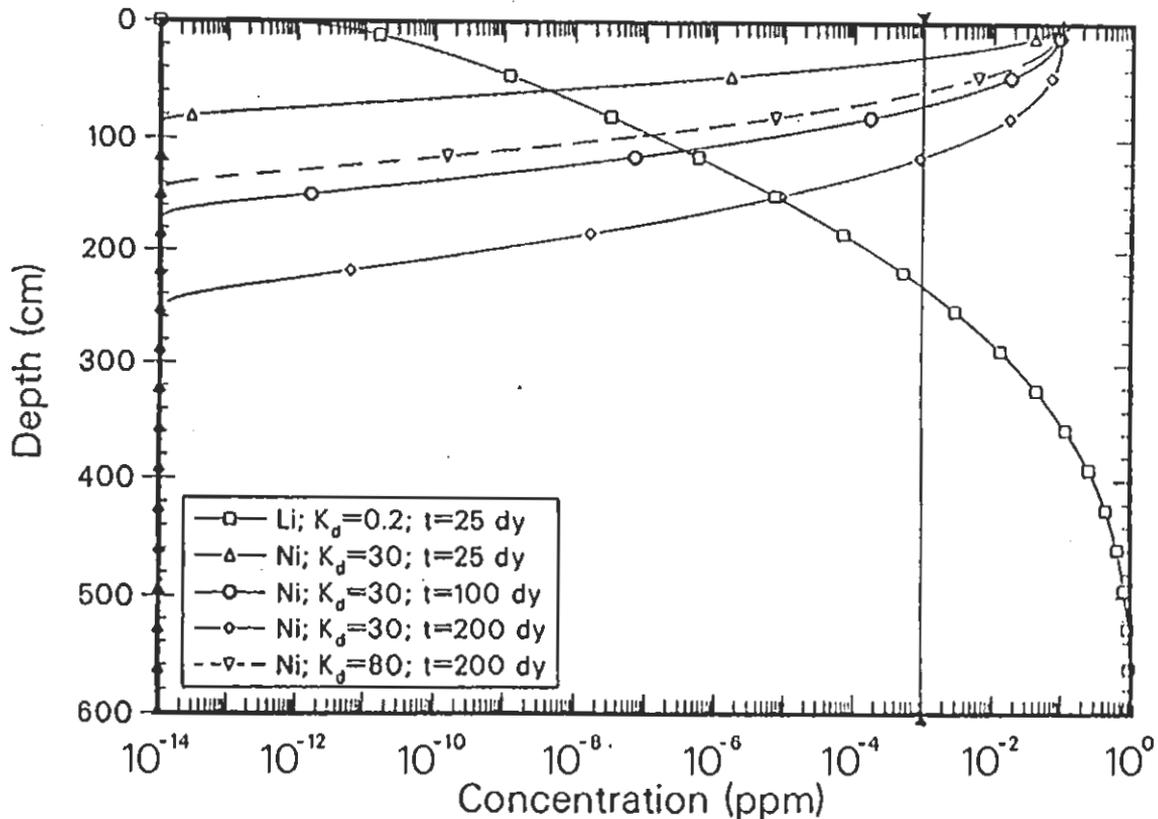
Figure 6. Concentration profiles for lithium and nickel for a water flux rate of 12 cm/day, a 10-day input pulse of lithium at an initial concentration of 1 mg/ℓ, and a continuous input of nickel with an input concentration of 0.1 mg/ℓ. The vertical line represents the detection limit for nickel.

The use of solute flux[17] rather than breakthrough curves represents another mode of comparison. This approach was used in analysis of previous caisson experiments by Dagan et al.[18,19] and Nguyen et al.[20] In these studies, the method was used to determine parameters for a stochastic model of the caisson under saturated conditions.[18,20] Cvetkovic et al.[17] derived the moments of the solute flux and cumulative solute flux for stratified, two-dimensional, and three dimensional porous media, and their results indicated that the solute flux is a robust state variable that can reduce uncertainty.

CONCLUSIONS

Model validation, or testing, is a critical issue for performance assessment of high-level nuclear waste disposal facilities. The proposed intermediate-scale experiment offers the advantage of exhibiting reduced uncertainty in the conceptual model governing water flow and nonreactive tracer transport, while using a porous medium that will be heterogeneous in physical and chemical properties. This combination will provide optimal conditions for prediction of flow and transport in a three-dimensional, heterogeneous system. The results of the transport experiments with these tracers and minerals will have implications for other studies in the YMP. Lithium is an important tracer because of its potential use in field experiments. Nickel is representative of radionuclides released from the structural components of the spent fuel rods in high-level waste and is considered an important waste element due its relatively low retardation in Yucca Mountain tuffs.[21] The component minerals of the caisson sand (quartz, calcite and iron oxyhydroxides) are present in fractures in Yucca Mountain tuffs.

The initial experimental design[4] was modified by adding a constant-pressure lower boundary and eliminating the sorbing layer. The new boundary condition eliminates additional mixing caused in the past by the convergent flow at the point discharge boundary, and eliminating of the sorbing layer reduces uncertainty in geochemical behavior.

Characterization of the hydraulic properties has shown the difficulty in using surrogates such as particle size to predict hydraulic properties. Further comparisons
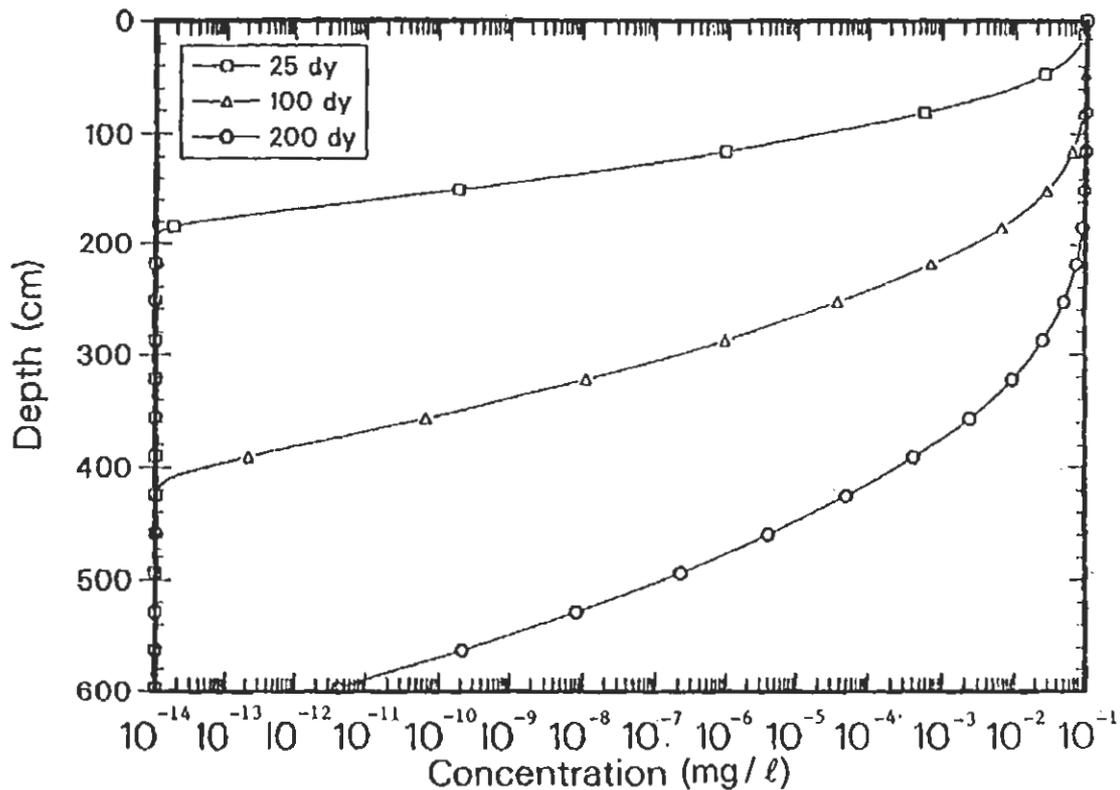
Figure 7. Concentration profiles of nickel in Wedron 510 sand for a water flux rate of 56 cm/day and a continuous input of nickel at a concentration of 0.1 mg/t.

will be made with samples collected from the caisson following the tracer tests.

Criteria to evaluate models represents a crucial element in this study. Questions such as, "What constitutes an adequate or defensible comparison between predicted and observed values?" must be addressed before moving on to field experiments.

Although far from complete, the caisson experiment has fostered the development of tools and procedures that will be needed in field experiments. This test does not, nor is it expected to, represent all of the conditions that will be found at Yucca Mountain. The results can temper expectations from any field experiments proposed for a high-level waste disposal site and provide a more pragmatic evaluation of predictive capability.

ACKNOWLEDGMENTS

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