DESIGN OF AN INTERMEDIATE–SCALE EXPERIMENT TO VALIDATE UNSATURATED-ZONE TRANSPORT MODELS

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ABSTRACT

An intermediate-scale experiment is being carried out to evaluate instrumentation and models that might be used for transport-model validation for the Yucca Mountain Site Characterization Project. The experimental test bed is a 6-m high x 3-m diameter caisson filled with quartz sand with a sorbing layer at an intermediate depth. The experiment involves the detection and prediction of the migration of fluid and tracers through an unsaturated porous medium. Pre-test design and materials characterization require estimation of physical properties of the porous medium such as the relative permeability, saturation/pressure relations, porosity, and saturated hydraulic conductivity as well as geochemical properties such as surface complexation constants and empirical K_d's. The pre-test characterization data will be used as input to several computer codes to predict the fluid flow and tracer migration. These include a coupled chemical-reaction/transport model, a stochastic model, and a deterministic model using retardation factors. The calculations will be completed prior to elution of the tracers, providing a basis for validation by comparing the predictions to observed moisture and tracer behavior.

I. INTRODUCTION

Validation of models of flow and contaminant transport in the unsaturated zone is an essential part of performance assessment and site characterization activities related to nuclear waste disposal. An intermediate-scale experiment is being carried out by Sandia National Laboratories and Los Alamos National Laboratory for the Yucca Mountain Site Characterization Project to demonstrate a model validation strategy that integrates a variety of hydrological and geochemical conceptual models, field measurements, and experimental approaches. The experiment involves the detection and prediction of the migration of fluid and tracers through a 6-m high x 3-m diameter caisson filled with a porous medium.

The experiment is being designed to ensure that the system will be simple enough to model from the pre-test characterization data, yet contains a sufficient number of chemical and hydrological phenomena such that the insights gained from this experiment will be useful in the planning of future field validation studies.

The caisson experiment will involve several distinct stages. These are:
1. Design phase,
2. Detailed pre-test characterization phase,
3. Predictive modeling phase,
4. Flow and transport test phase, and
5. Post-test phase.

The focus of this paper is the design phase; an overview of the detailed pre-test characterization also will be given. Predictive modeling using the pre-test characterization data will begin in early spring 1992. The actual flow and transport test will begin in late spring 1992 and will continue until fall of the same year. The post-test phase will involve comparison of model predictions to observed flow and transport, as well as some additional characterization of the porous media within the caisson. This will occur in late 1992 and early 1993.

The activities of the design phase must resolve a variety of issues:

1. Selection of the primary porous medium, based on hydraulic, dispersion, and geochemical properties;

2. Definition of the macroscopic system geometry;

3. Selection of the reactive substrate, and reactive solutes;

4. Selection of techniques for measurement of concentration, pressure, moisture content, and flux; and

5. Definition of boundary conditions, flow rates, and experimental sequence including the timing of tracer addition.

II. DESIGN CRITERIA

The caisson experiment is being designed to satisfy two sets of criteria: 1. requirements related to hypothesis testing and process definition/characterization; and 2. site-specific practical constraints. As part of the first set of design criteria, several hydrological and geochemical questions that are important to transport model validation will be addressed in this experiment. The most important of these are:

1. Can hydraulic properties and solute dispersivity at the caisson scale be predicted from small-scale measurements through the use of scaling laws which integrate over heterogeneity structures expected in the caisson?

2. Can geochemical transport properties (e.g., solute retardation factors) for porous media composed of mixtures of minerals be predicted from the batch sorption studies of the constituent minerals?

3. What criteria should be used to evaluate the validity of process-based flow and transport models that provide the theoretical basis for the total system models used in performance assessment?

Practical concerns contribute the second set of design criteria for the experiment. Such constraints are related to specific conditions of the test site and to available technology for monitoring the fluid flow and tracer migration, and include:

1. The need to ensure that the test will not present a hazard to the environment;

2. The requirement that the travel times for the water and conservative tracers are long enough to allow detection with sufficient resolution for the model comparison (approximately 1 to 4 days);

3. The requirement that the travel times for the reactive tracers are short enough to allow detection before winter weather makes monitoring impossible (approximately 4 to 6 months); and

4. The requirement that the chemical and moisture sensors do not significantly disrupt the flow field.

In addition to these model-validation questions and site-specific criteria, the design is constrained by instrumental considerations such as detection limits and precisions of analytical techniques used to measure tracer concentrations as well as the mechanical properties of materials (sampling devices and suction plates) used in the caisson. All of the above criteria are being evaluated simultaneously during the design process. The design process involves several iterations of preliminary calculations, simple experiments and prototype constructions. The final design will represent a compromise among the often conflicting demands of all of these factors.

III. GENERAL CAISSON DESIGN DESCRIPTION

The experiment will be carried out at the Experimental Engineered Test Facility at Los Alamos National Laboratory. Descriptions of the design of the caisson cluster, water-solute distribution system, and solute blending/storage system at the facility are given by Polzer et al.[1] The tracers will be introduced at the top of the caisson through an assembly of fluidic wafer switches, each of which is fitted with 24 outlet ports. The solutions will be withdrawn from the bottom of the caisson through a honeycomb of suction samplers. A schematic diagram of the caisson design for this experiment is given in Figure 1.

During the initial stages of design, several different materials were considered for the porous matrix. These included pure quartz sand, zeolites mined from three commercial deposits, quartz sand coated in the laboratory with iron oxyhydroxide, and powdered goethite. The preliminary design assumes that the caisson will be filled with quartz sand, with a 0.5 m thick sorbing layer at an intermediate depth. Quartz was chosen because it is relatively inert geochemically; the sorbing layer will be composed of quartz sand doped with minor amounts of a highly sorbent mineral. It is intended that the reactive tracers will be strongly retarded in this layer, while the non-reactive tracers pass through unretarded.

Several different particle-size distributions for the porous matrix were considered in combination with hydraulic boundary conditions (applied flux and suction). Hydraulic and dispersion properties of the filling material will be uniform within certain bounds throughout the caisson. This will be accomplished by using a narrow (but not unnatural) grain-size distribution devoid of silt- and clay-size particles. Microlayering and cross-bedding will occur while the caisson is filled and packed. Production of these features will be minimized, but no attempt to destroy them will be made once the sand is emplaced. Scaling laws for effective hydraulic and dispersion properties that integrate these small-scale heterogeneities will be developed as part of the detailed pre-test characterization phase outlined below.
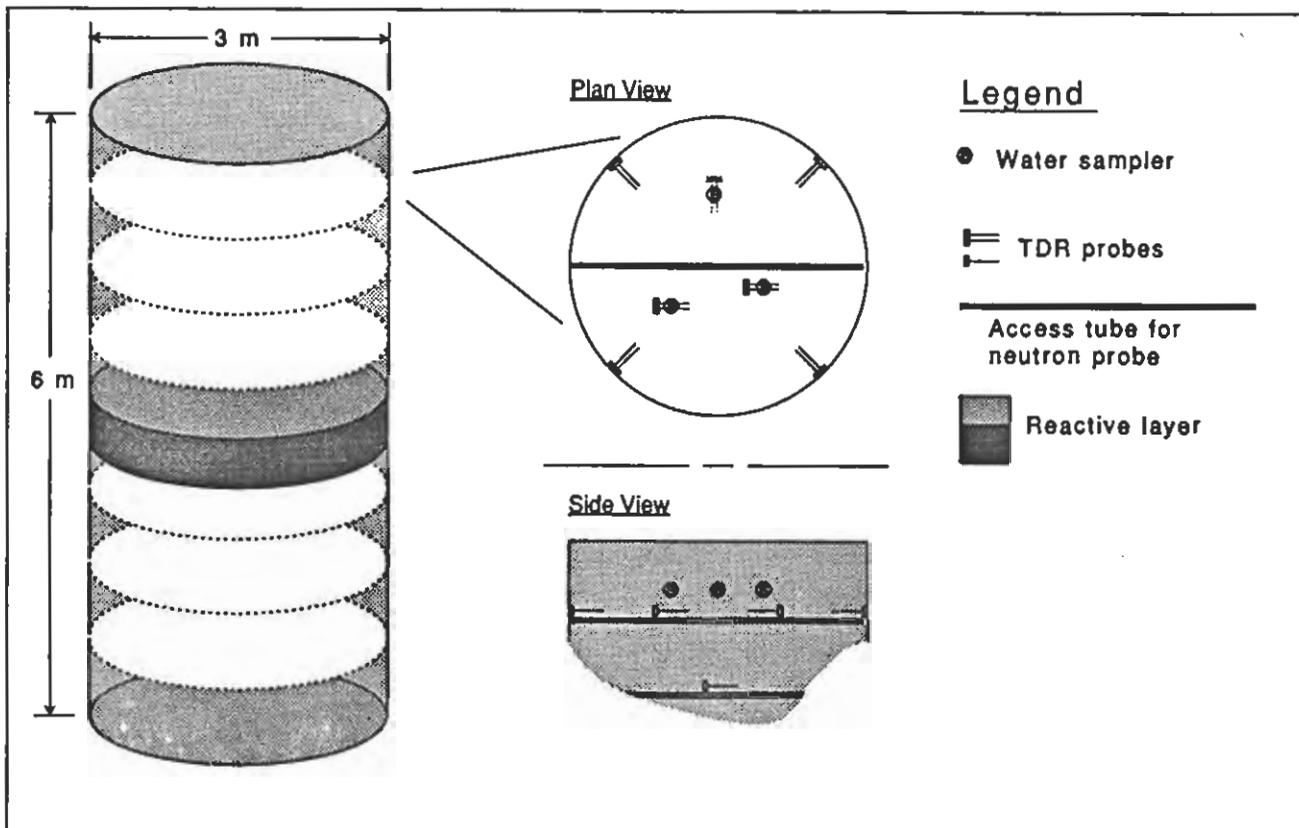
Fig. 1. Schematic diagram of caisson showing possible stratigraphy and placement of TDR probes, neutron probe access tubes and water samplers. Location of reactive layer (shown here in the middle of the caisson) will be determined after additional experimentation and sensitivity analyses. Instrument layers are on 75-cm centers and are shown in detailed plan and side views.

As discussed in detail later, the properties of the homogeneous sand have been estimated from candidate grain-size distributions and porosities, and have been used to calculate flow rates consistent with different boundary conditions and saturation states. The grain-size distribution and porosity (and hence the hydraulic and dispersion properties) in the sorbing layer will be matched to those of the quartz sand layers.

A desired retardation factor for reactive tracers has been calculated from the estimated flow rate and desired tracer travel time (40 to 100 days). Currently, a suite of conservative and reactive tracers is being chosen from a number of candidate solutes. The amounts of candidate sorbents required to achieve the desired retardation factor are being calculated from solute distribution coefficients (K_d's — the ratio of sorbed solute per unit mass sorbent divided by the aqueous solute concentration, commonly expressed in units of ml/g) of the candidate reactive tracers.

The experimental sequence will be as follows. At the start of the experiment, the sand will be saturated with the background electrolyte (0.01 M NaCl solution) by pumping tracer-free solution into the bottom of the caisson. After

the sand is saturated, bulk saturated permeability, moisture content, entrapped air content, and porosity will be determined. Subsequently, constant-flux boundary conditions will be established at the top of the caisson, and the sand will drain under suction to a desired steady-state unsaturated flow field and moisture content profile. This method of establishing the steady-state unsaturated flow field has been chosen to avoid wetting/drying hysteresis effects and reduce the likelihood of generating gravity-driven fingering.[2] A 10- to 20-day-long pulse containing a suite of non-reactive and reactive tracers will be added after steady-state hydraulic conditions have been achieved.

The progress of the wetting and drainage fronts will be monitored via time-domain reflectometry (TDR) and neutron probes. The placement of TDR probes and the neutron-probe access tubes in the caisson are shown in Figure 1. Pore solutions will be sampled by ceramic suction samplers to monitor tracer migration. The placement of samplers in the caisson also is shown in Figure 1.

IV. PRIMARY SELECTION OF CAISSON MATERIALS

The choice of porous media for the caisson filling material was based on estimation of average water travel time and hydraulic properties from sand grain-size distribution and porosity. The sand for the caisson was designed to have hydraulic properties that meet a 4-day water-travel-time criteria.

The water travel time t_w is given by:

$$t_w = L \cdot \theta / q \qquad (1)$$

where θ is the steady state moisture content, q is the system flux, and L is the system length. The moisture content at a point in the system is related to the pressure head (ψ) by the moisture characteristic relation, $\theta(\psi)$. (Note that the pressure head in unsaturated systems is intrinsically negative (suction) for water-wettable media.) The steady-state pressure-head distribution within the caisson is a function of the steady-state flux through the caisson, the applied suction at the bottom of the caisson and the relative conductivity relation, $K(\psi)$. Continuity of mass, under steady-state conditions, gives

$$\nabla \cdot q = 0 \qquad (2)$$

where the flux, q, is given by the extension of Darcy's law to unsaturated systems:

$$q = -K(\theta) \frac{dH}{dz} \text{ , and} \qquad (3)$$

$$H = \psi + z . \qquad (4)$$

where H is the total head and z is the height above the bottom of the caisson (positive upward).

Given steady-state conditions and an estimate of the material properties, the appropriate applied flux at the top boundary and suction at the lower boundary that will result in an acceptable travel time for a conservative tracer can be calculated. Due to the nonlinearities of the system, an initial assumption of unit head gradient was made in order to approximate the flux and suction. The resulting applied flux and suction then were used to define the boundary conditions necessary to solve numerically for the flow field of the caisson and to verify the desired travel time of 4 days. This design process may be repeated easily with changes in hydraulic properties.

For preliminary design calculations, the hydraulic properties of the sand were estimated from the grain-size distribution and an expected average bulk density. Both the moisture-characteristic relation and the relative-permeability relation are functions of the pore-size distributions and pore-network structure. For a granular material, the pore-size distribution is related to the grain-size distribution and the packing density (bulk density and porosity).

The method of Haverkamp and Parlange[3] was used to map the grain-size distribution and bulk density of candidate sands into the moisture-characteristic relation $\theta(\psi)$ given by a Brooks and Corey[4] form:

$$\theta(\psi) = \varepsilon \left[\frac{|\psi_{ae}|}{|\psi|} \right]^{\lambda_{ac}} \left[1 - \frac{|\psi_{ae}|}{|\psi|} \left(1 - \frac{\theta_s}{\varepsilon} \right) \right] , \; \psi < \psi_{ae} ; \quad (5)$$

$$\theta(\psi) = \theta_s , \qquad\qquad\qquad \psi \geq \psi_{ae} ; \quad (6)$$

where ε is the porosity of the sand, ψ_{ae} is the air entry or bubbling pressure, θ_s is the moisture content of the sand at saturation, including entrapped air (satiation), and λ_{ac} is the Brooks-Corey fitting parameter.

Equations 5 and 6 are not continuous at the air entry value ψ_{ae}; therefore, for ease of numerical solution, the moisture-characteristic relations of van Genuchten[5] (Equations 7 and 8) were fit to Equations 5 and 6.

$$\theta(\psi) = \frac{\theta_s - \theta_r}{(1 + |\alpha\psi|^{\beta})^{-\lambda}} + \theta_r , \qquad \psi < 0 ; \quad (7)$$

$$\theta(\psi) = \theta_s , \qquad\qquad\qquad \psi \geq 0 ; \quad (8)$$

where θ_r is the residual moisture content, α and β are the van Genuchten fitting parameters and $\lambda = 1 - 1/\beta$. The fitting was done by adjusting the values of the van Genuchten α and β parameters until the moisture characteristic curve calculated using Equations 7 and 8 was similar (as determined by visual inspection) to that calculated using the Brooks-Corey relationship (Equations 5 and 6).

Finally, the relative hydraulic conductivity relation $K(\psi)$ was estimated from the moisture characteristic relation using the method of Mualem[6] to give:

$$K(\psi) = \frac{K_s}{(1+|\alpha\psi|^{\beta})^{\lambda/2}} \left[1 - \left[\frac{|\alpha\psi|^{\beta}}{1+|\alpha\psi|^{\beta}} \right]^{\lambda} \right]^2 , \; \psi < 0 ; \quad (9)$$

$$K(\psi) = K_s , \qquad\qquad\qquad \psi \geq 0 , \quad (10)$$

where K(s) is the saturated conductivity and the other terms have been defined above. Values of the parameters in Equations 5 to 10 are summarized in Table 1.

Table 1. Parameters used in preliminary design calculations.

Symbol	Definition	Value
Parameters measured on Wedron 510 or similar materials:		
ε	porosity of sand	0.38
K_s	saturated conductivity	0.607 cm/day
θ_s	moisture content at saturation	0.34
θ_r	residual moisture content (assumed)	0.0
Parameters resulting from calculations:		
ψ_{ae}	Brooks-Corey air entry value	-40.75 cm
λ_{BC}	Brooks-Corey lambda	2.09
α	van Genuchten alpha	2.0 m
β	van Genuchten beta	3.2
q	steady-state flux	20.3 cm/day
ψ_L	suction head at bottom of caisson	-67.3 cm
θ	average (steady-state) moisture content	0.142
θ/ε	saturation	41.6 %

Through exercise of this design process on a number of commercially available silica sands, the Wedron 510 sand (Wedron Silica, Wedron, Ill. 60557) was chosen for the caisson material. Figure 2 shows the grain size distribution for this sand and several perturbations to this distribution which will be discussed in a later section. Parameter values for the sand for use in Equations 5 - 10 are given in Table 1. Figure 3 compares the moisture characteristic relation described by the Brooks-Corey[4] equations (Equations 5 and 6) with that of van Genuchten[5] in Equations 7 and 8.

V. DESIGN OF REACTIVE SUBSTRATE AND TRACER

A. Methods

The objective of geochemical characterization studies in support of design is to select tracer/sorbent combinations that will result in the desired tracer travel times. Preliminary sorption studies for proposed reactive tracers are being carried out in the proposed background electrolyte (0.01 M NaCl) with candidate sorbents. A suite of reactive tracers that spans a variety of chemical behaviors will be chosen. The kinds of tracers that are being considered include: 1. transition metals representative of waste elements released from the structural materials in the high-level waste (e.g., Ni, Co); 2.

conservative tracers that can be used to track the movement of water introduced into the site during construction of the repository or Exploratory Studies Facility (e.g., B, Br); and 3. nonradioactive isotopes of waste elements or analogs for transuranics (e.g., U). The suite of tracers that is chosen for the final design will depend on predicted travel times, suitability of sampling and analytical techniques, and environmental hazard considerations.

The physical and chemical properties of several sorbing porous materials for potential use in the sorbent layer have been examined. These include: three varieties of commercially-available clinoptilolite (Teague Minerals and St. Cloud Zeolites), goethite (Ward's Earth Sciences) and quartz sand (Wedron 510) coated with synthetic iron oxyhydroxide. Preliminary batch reaction studies in which the rock and water are pre-contacted for several days have been completed. The composition of the resulting solutions after contact with the solids provides some information about the probable composition of water in the caisson during the experiment. Surface areas of the quartz sand, clinoptilolite and goethite have been measured with the BET (Brunauer, Emmett and Teller) method as a function of particle-size distribution.

Preliminary batch nickel sorption measurements have been carried out with the zeolites, goethite, Wedron 510 quartz sand, and the ceramic solution samplers that will be placed in the caisson. In these experiments, the rock and 0.01 M NaCl solutions were pre-contacted for several days before introduction of the tracer. Experiments were carried out over a range of pH values at solution/solid ratios ranging from 1:1 to 10:1 with initial aqueous nickel concentrations of 100 ppb.

Scoping studies of nickel solubility in 0.01 M NaCl solutions also have been completed. The experiments were carried out by preparing solutions with initial nickel concentrations of 1 ppm at pH values of approximately 4. Solutions then were brought to final pH by addition of NaOH. Final concentrations were measured after equilibration periods ranging from 12 to 48 hours. Nickel concentrations in solutions from both solubility and sorption studies were measured on a Perkin-Elmer atomic-absorption spectrophotometer equipped with graphite-furnace atomization.

B. Results

Initial characterization of the iron-coated quartz sand suggested that high amounts of nitric acid were trapped in pores of the coating during synthesis; thus, this material was eliminated from further consideration.
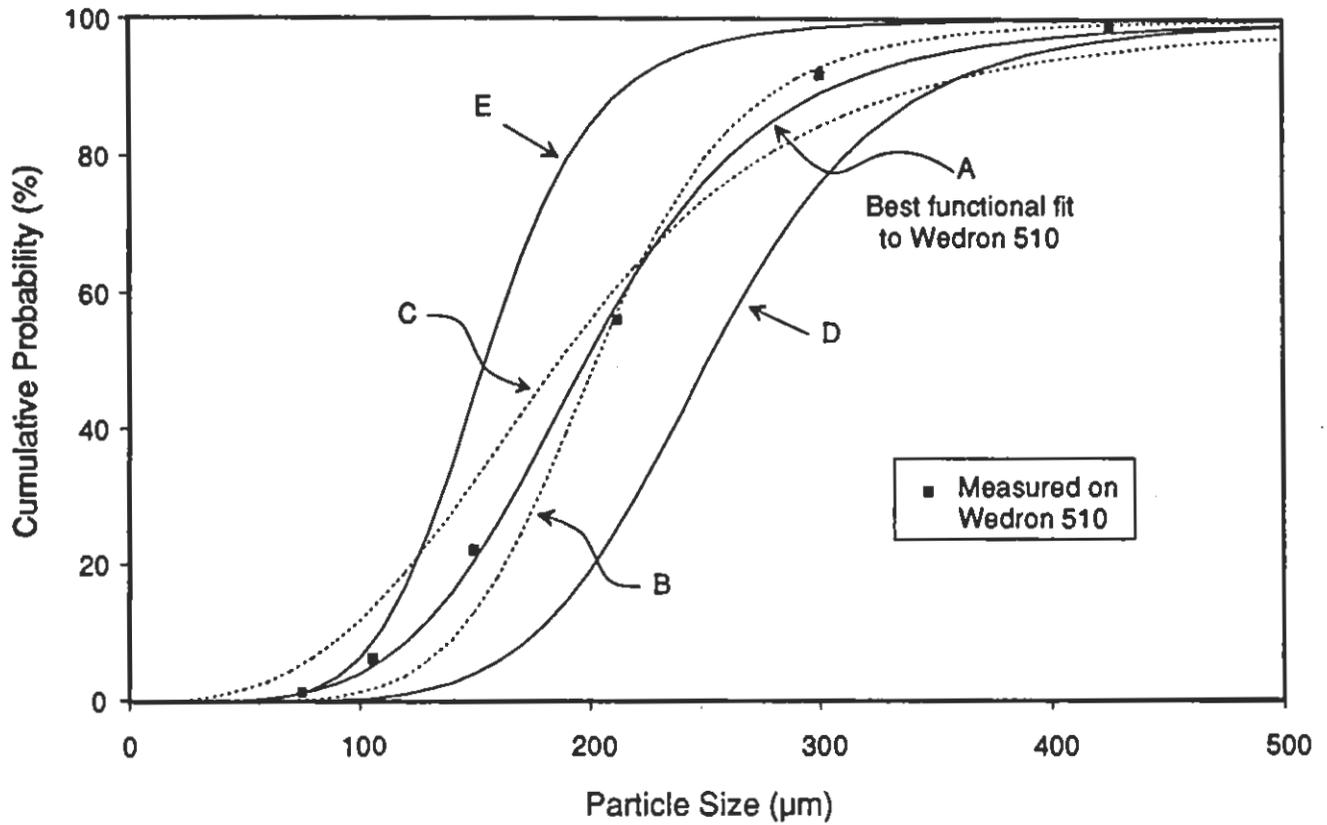
Fig. 2. Sand particle size distribution for Wedron 510 and perturbations expected due to microlayering and cross-bedding heterogeneities created during caisson filling and packing.
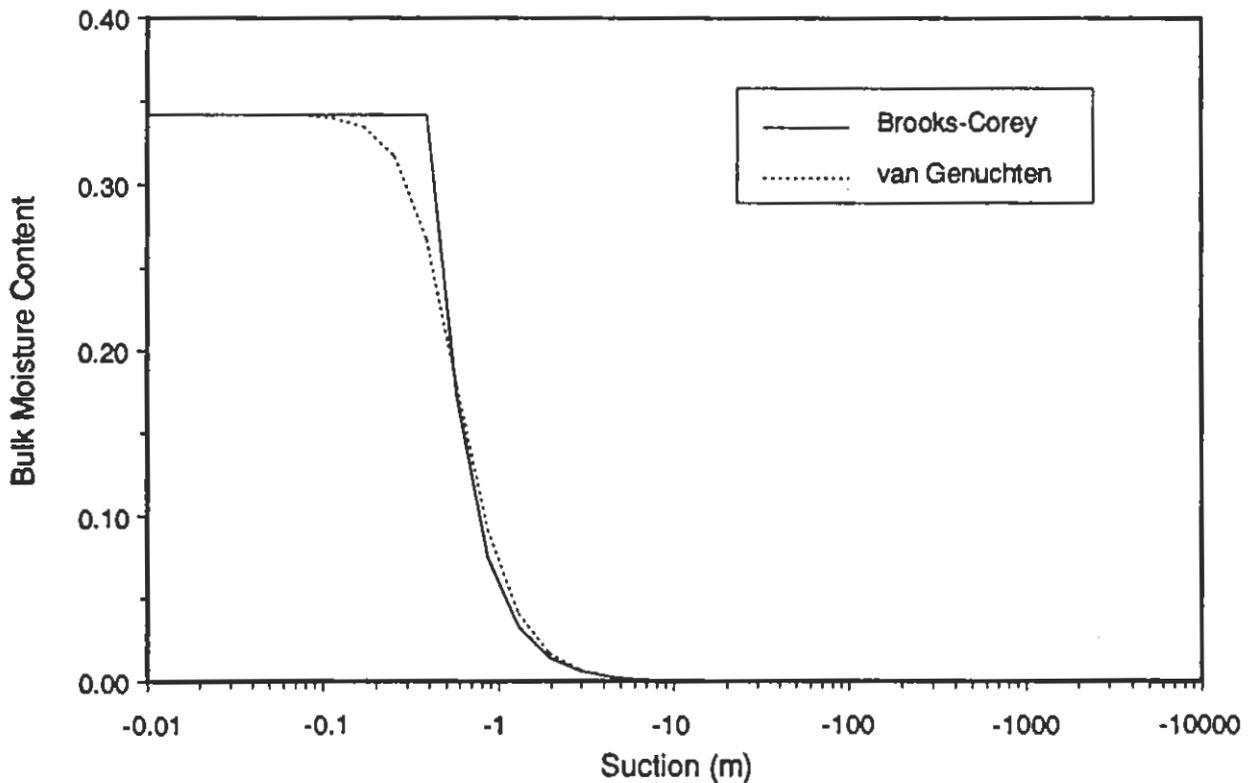

Fig. 3. Comparison of Brooks-Corey and van Genuchten forms of the moisture-content characteristic relation for Wedron 510 sand.

Investigations with the zeolites suggested that they were mechanically unsuitable for the experiment. In addition, column measurements of the hydraulic properties of quartz sand/zeolite mixtures and the results of the nickel sorption studies indicated that a sorbent layer containing zeolite would not have the desired hydraulic conductivity or retardation factor. For these reasons, the zeolites were eliminated from further consideration.

Results of the preliminary nickel sorption experiments for the quartz sand, goethite and ceramic sampler are shown in Figures 4 and 5. Figure 4 shows all of the data collected over the pH range 5 to 12. The contrasting sorption affinities of nickel for the three materials are evident in this figure. The nickel sorption edge (pH range wherein sorption increases dramatically) occurs for goethite at about pH 6.5. The large scatter in the data at the higher K_d's is due to relatively high analytical uncertainty at Ni concentrations near the limit of detection (0.5 - 1.0 ppb). The uncertainty in the K_d values increases non-linearly (quasi-exponentially) with K_d and decreases non-linearly with the solution/solid ratio. The error is distributed asymmetrically about the measured K_d value. For goethite (solution/solid ratio = 10:1), measured K_d's of 800, 100, and 10 ml/g correspond to K_d ranges of 440 to 4100 , 90 to 112 and 9.8 to 10.2 ml/g respectively. For sand, (solution/solid ratio = 1:1), measured K_d's of 80, 10, and 5 ml/g correspond to K_d ranges of 57 to 135, 9.4 to 10.6 and 4.8 to 5.2 ml/g respectively.

Figure 5 shows the data over a more limited range of pH and K_d relevant to the caisson conditions. Based on the results of preliminary sand/water equilibration studies (in vials closed to the atmosphere), water in the caisson is expected to be neutral to slightly alkaline. In the near-neutral pH range (pH = 6.8 to 7.5), the nickel K_d values range from 5 to 70 ml/g, from 10 to 80 ml/g, and from 600 to 1350 ml/g for the sand, ceramic sampler and goethite respectively. At slightly lower pH (6 to 6.5), Ni sorbs very weakly to the sand and ceramic sampler (K_d = 5 to 15 ml/g). At pH 6.5, Ni sorbs strongly to the goethite (K_d = 75 ml/g), but no data are available for the pH range 6 to 6.5.

The results of the preliminary studies of nickel solubility in 0.01 M NaCl (Figure 6) suggest that use of a total nickel concentration of 100 ppb in the sorption experiments avoided precipitation of nickel solids over the pH range (5 to 8) of interest. Final aqueous concentrations of nickel are well below the apparent Ni solubility limit. Future solubility experiments will use water pre-equilibrated with the porous matrix to be used in the caisson instead of 0.01 M NaCl solutions. Similar studies of solubility and sorption of other reactive tracers being considered for this experiment (B and U) are currently in progress.

The preliminary nickel sorption data suggest that only limited Ni transport will occur in the caisson during the course of the experiment. An approximation of the transport distance is required to determine the feasibility of using Ni as a reactive tracer and to guide placement of sampling devices within the caisson. These calculations are described in the next section.

VI. PRELIMINARY TRANSPORT MODELING

The hydraulic and geochemical characterization data described above are being used as input to several computer codes to predict the fluid flow and tracer migration. The flow problem is solved independent of the transport problem using the LLUVIA-II version of the LLUVIA computer code[7]. This version of the code uses the method-of-lines technique to solve the Richards' equation for the two-dimensional, isothermal flow of liquid water through partially saturated porous media. The user may employ the model of choice for describing the functional dependence of moisture content, capacitance, and conductivity on pressure head via user-defined subroutines. The computed moisture content and Darcy velocities, either steady-state or transient, are written to an external data file for use in the solution of subsequent transport calculations.

As discussed previously (Section IV), the hydraulic properties of the sand given in Table 1 were used to compute the applied flux at the top of the caisson and the suction at the bottom necessary to achieve the desired travel time for a conservative tracer. A calculated (downward) flux of 20.3 cm/day and pressure head (suction) of -67.3 cm were used as boundary conditions to LLUVIA-II to confirm the 4-day travel time. Under these conditions at steady state, the saturation of the sand of the caisson is approximately 41 percent.

Preliminary transport calculations are being made using the LEHGC version of the HYDROGEOCHEM[8] code. In the first set of calculations, the code was used to solve a system of transport equations which describe advection, dispersion/diffusion, and sources and/or sinks. In this exercise, the moisture content and Darcy velocities are taken from the solution computed by LLUVIA-II. Transport of reactive tracers is modeled by appropriately scaling the capacitance and dispersion/diffusion terms of the transport equation by a retardation factor. Alternatively, LEHGC can be used to model chemical
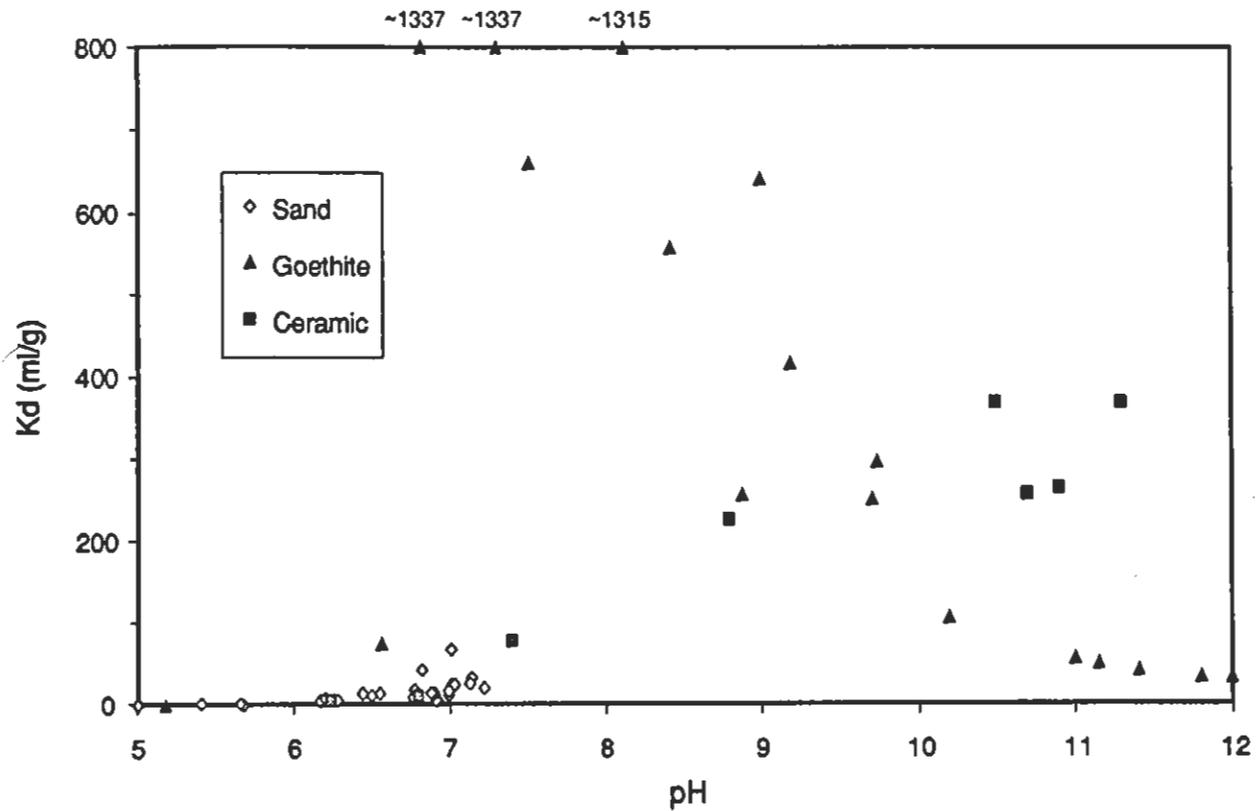
Fig. 4. Nickel sorption (K_d) by caisson-filling materials and ceramic samplers as function of pH. For K_d values exceeding 800, final aqueous Ni concentrations were equal to or less than the analytical detection limit of 0.75 ppb, so the detection limit was used instead in the K_d calculations.
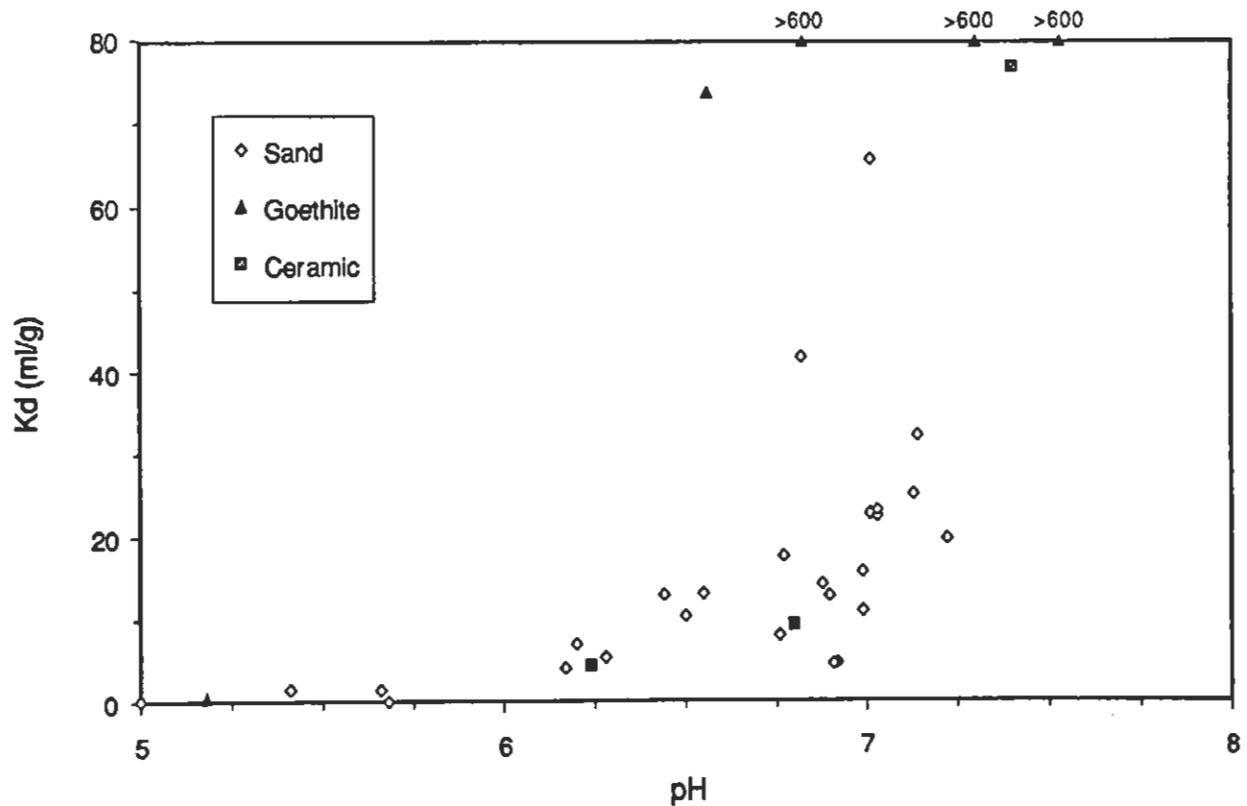

Fig. 5. Nickel sorption (K_d) by caisson-filling materials and ceramic samplers over pH range possible for caisson.
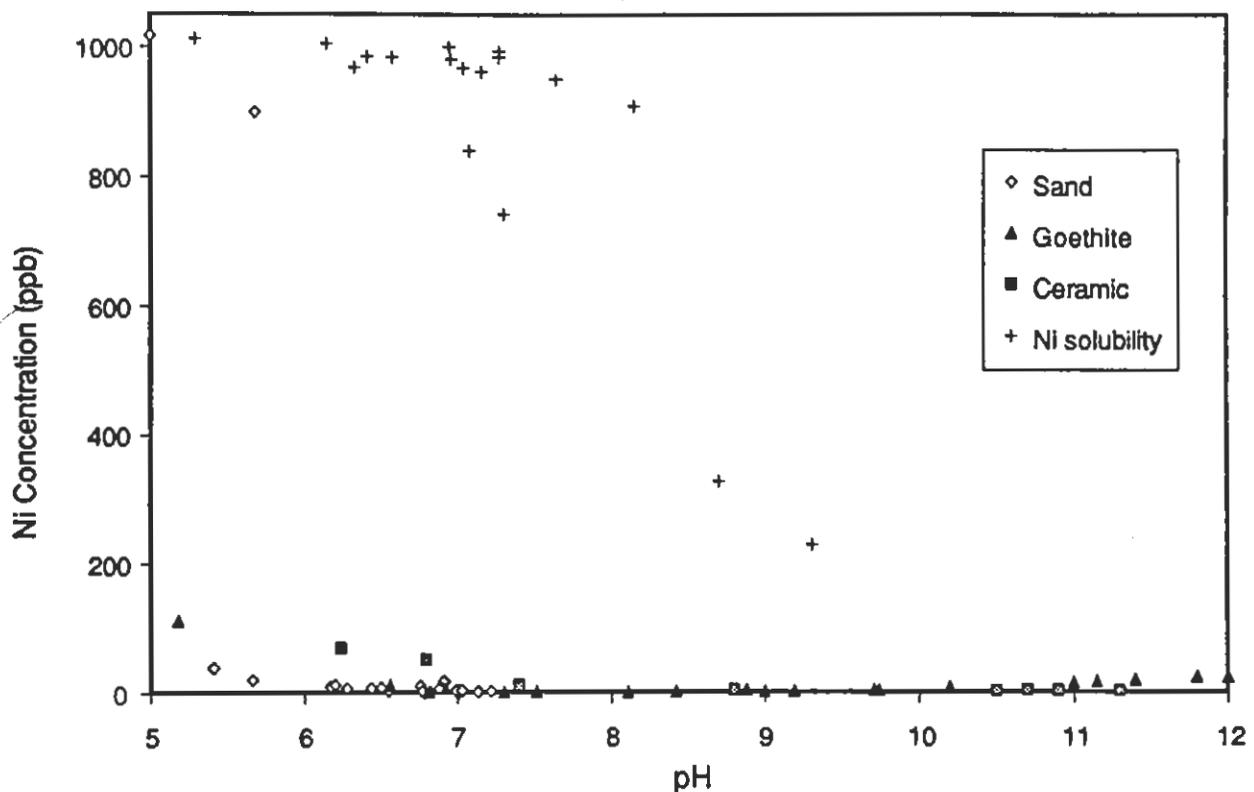
Fig. 6. Comparison of aqueous nickel concentrations in sorption and solubility experiments. Initial aqueous nickel concentrations for solubility and sorption experiments were 1000 ppb and 100 ppb respectively.

speciation and sorption using equilibrium constants. The code will be used in this latter manner in the *predictive modeling phase* of the experiment as described in a later section.

Figure 7 shows transport of Ni through the top 50 cm of the caisson as simulated by the LEHGC code in the first set of calculations. The steady-state (downward) flux and average moisture content were 20.3 cm/day and 0.142, as described above. A dispersivity of 5 cm was assumed based on dispersivity studies of similar sands; molecular diffusion was assumed negligible compared to the mechanism of advection. For this simple calculation, a retardation factor R was estimated in order to scale the appropriate terms of the transport equation:

$$R = 1 + \frac{\rho K_d}{\theta} = 1 + \frac{1.5 \text{ g/m}l \cdot 46 \text{ m}l/\text{g}}{0.14} \cong 500 \qquad (11)$$

where the value of the bulk density ρ was estimated from the particle-size distribution and the value for the K_d was chosen to represent conditions under which sorption of Ni by the sand was appreciable but not worst-case (see Figure 5).

In the calculations, a 10-day-long pulse of Ni at a

concentration of 100 ppb was introduced at the top of the caisson at t=0. Results were calculated over a 100-day interval. Figure 7 shows the Ni penetration profile at 3 computed times. The first is at 10 days, when the Ni pulse is terminated. At 60 and 100 days, the peak of the pulse has penetrated to depths of nearly 17 and 29 cm, respectively.

The analytical limit of detection for Ni is about 0.5 ppb. Figure 7 suggests that solution samplers should be placed at a depth of 28 cm for detection at 60 days or at 42 cm for detection at 100 days if the water travel time is 4 days and if Ni sorption by the sand is consistent with the K_d chosen for this calculation. Currently, additional sorption and rock/water equilibration studies are under way to allow more accurate estimation of the Ni retardation factor. If a retardation factor higher than that assumed in the simulation is predicted, hydraulic conditions could be set to decrease the water travel time or the solution composition could be altered to lower the retardation factor. Similar experimental studies and sensitivity analyses also will be carried out for other candidate tracers.
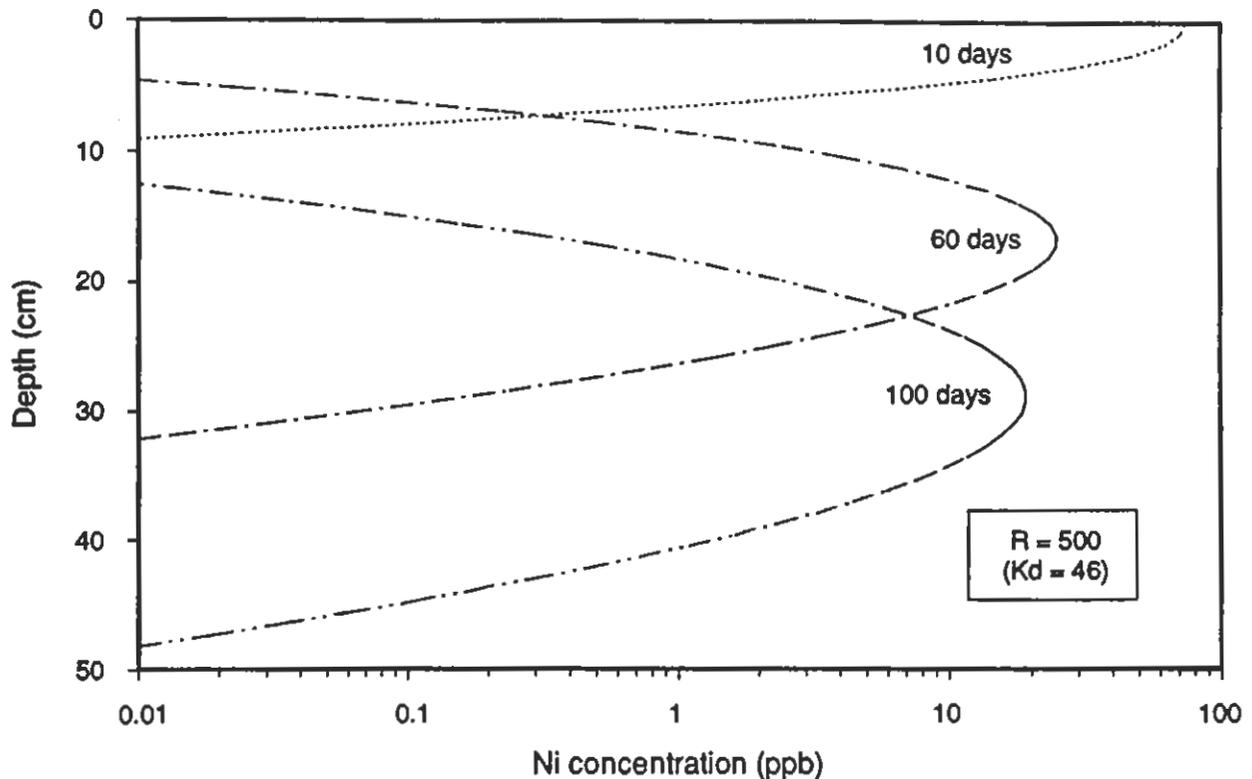
Fig. 7. Nickel concentration profiles in top 50 cm of caisson as simulated by LEHGC code for 10, 60, and 100 days. See Table 1 for parameter values. Note that depth is measured from top of caisson (where z = 600 cm).

VII. OVERVIEW OF DETAILED PRE-TEST CHARACTERIZATION AND PREDICTIVE MODELING PHASES

In order to more precisely predict fluid flow and tracer migration in the caisson experiment as part of a model validation exercise, detailed characterization of the hydraulic and dispersion properties and the geochemical properties of the porous medium must be carried out.

A. Hydraulic and Dispersion Studies

Hydraulic and dispersion studies during the pre-test characterization phase of the caisson experiment will include: 1. measurement of hydraulic and dispersion properties of the homogeneous sand and homogeneous mixtures of the sand and sorbent in laboratory columns and 2-D slabs; and 2. development of scaling laws for effective media properties which integrate over small-scale microlayering and cross-bedding heterogeneities expected in the caisson.

1. Measurement of hydraulic and dispersion properties of the homogeneous sand and sorbent layer. Experiments will be conducted to measure the hydraulic and dispersion properties of Wedron 510 sand and mixtures of sand and sorbents in 5-cm-diameter columns in which a

gamma densitometer can be used. Other studies will use 60 x 30 x 1-cm slab chambers where 2-D full field measurement techniques can be used. As described by Tidwell and Glass,[9] high-resolution data (both spatial and temporal) for moisture-content and tracer-concentration fields within thin slabs can be obtained using dyes and either x-ray or light transmission techniques. Studies will be carried out with 3 homogeneous sands that differ only in their grain-size distribution as described in the next section and in Figure 2 (curves D and E). Experiments with the sands and sorbents will be used to refine the hydraulic parameter values predicted from the material properties and will be used in a second round of water-flow and solute-transport numerical calculations.

2. Experiments to develop scaling laws that integrate over small-scale microlayering and cross-bedding. The caisson porous matrix will contain small-scale heterogeneities (0.001 to 0.3-m layering and cross-bedding) that will influence hydraulic and dispersion processes. Examples of these heterogeneity structures in thin slabs revealed with the light-transmission technique are shown in Figure 8.[10,11] From preliminary studies, perturbations to the grain-size distribution for Wedron 510 sand shown in Figure 2 are expected. The three central curves depict the Wedron 510 (A), a slight narrowing (B)
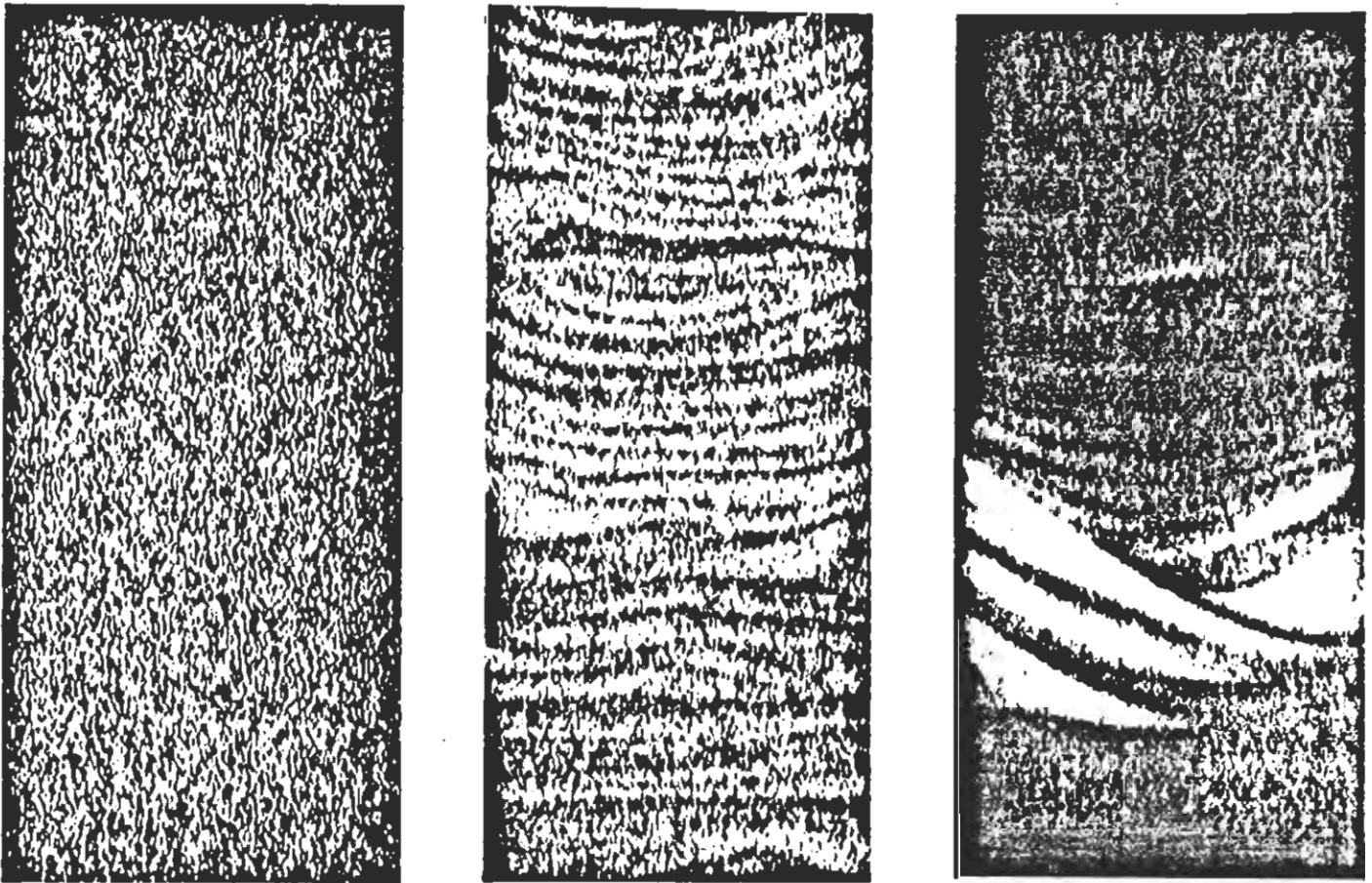
Fig. 8. Visualization of heterogeneity structure in thin sand slabs. Variations in porosity can be visualized using light or x-ray transmission techniques. The slab on the left was created using a chamber-filling technique developed to generate homogeneous thin slabs. Slabs in the middle and on the right were created using techniques that generate different types of heterogeneity structure. Light areas are regions of higher porosity, dark areas are lower porosity. Taken from the work of Glass and Tidwell.[10,11]

and a slight widening (C) of the grain-size distribution. Distributions between B and C are expected within the 78 tons of sand that will fill the caisson. In addition, as the caisson is filled and packed, modulations in mean grain size and narrowing of the size distribution will occur due to the grading process and will create microlayering and cross-bedding. The curves (D and E) are combinations of expected values of the mean grain size with the narrow distribution (B) resulting from the grading process. The second-order effect of variable bulk density also will contribute to caisson heterogeneity.

A scaling law that models the effects of these heterogeneity structures on hydraulic properties must be developed so that effective hydraulic and dispersion properties at a larger scale (0.3 to 1 m) may be defined. Such a scaling law will be developed through a series of column and slab experiments wherein the type, frequency, and strength of the heterogeneity will be varied systematically and the hydraulic and dispersion properties measured. Column experiments will be conducted on several lengths ranging from 0.05 to 1 meter. Slab experiments will be conducted at two scales: 30x60 cm and about 100x100 cm. The resulting effective properties as a function of microlayering will be used for a third round of numerical calculations for fluid flow and solute transport.

B. Geochemical Studies

After the tracers and sorbents for the caisson have been chosen from preliminary K_d measurements, more systematic sorption experiments will be carried out. Batch sorption data will be obtained over ranges of solution pH and concentrations of solids, tracers, and competing cations and anions and varying sand/sorbent ratios. In addition to obtaining data that can be used to calculate K_d's and isotherms, data that can be analysed within the conceptual framework of surface-complexation modeling will be obtained. In this approach, tracer sorption is considered to involve complexation with functional groups that exist on the surface of the minerals. The functional groups in this study are surface hydrolysis sites including

aluminol (Al-OH), silanol (Si-OH) and iron-oxyhydroxyl (Fe-OH) groups. Surface hydrolysis constants will be obtained by potentiometric titrations as described by Kent et al.;[12] specific adsorption constants for the reactive tracers and background electrolytes will be obtained by batch sorption techniques as described in Tripathi.[13]

Sorption data will be obtained for both pure sand, pure goethite and for mixtures of the two components in various proportions. These data will be used to determine if it is possible to predict the bulk sorption behavior of a mixture of minerals from the properties of the individual components. A basic assumption of surface complexation models is that different adsorption sites compete for adsorbate metals in a manner analogous to the competition among complexing ligands for metals in solution. The validity of this assumption will be assessed by using the complexation constants obtained for individual minerals to model aqueous speciation and sorption properties of mineral mixtures with the HYDRAQL[14] computer code. The predicted sorption properties of various mixtures will be compared to measured K_d's for the same mixtures. The same approach will be used to model reactive transport in the sorbent layer in the caisson using the LEHGC version of the HYDROGEOCHEM code as discussed below.

C. Predictive Transport Modeling

The pre-test characterization data will be used as input to predict the fluid flow and tracer migration using LLUVIA-II and the LEHGC version of HYDROGEOCHEM. The scaling laws developed through the systematic column and slab experiments described above will be tested against the observed flow field. The LEHGC code will be used as a coupled reaction/transport simulator to solve a system of both transport and geochemical equilibrium equations. The chemical processes, assumed to occur under conditions of local equilibrium, will include aqueous complexation, adsorption (surface complexation), ion exchange, and precipitation/dissolution. Transport predicted using retardation factors measured on bulk mixtures will be compared to the tracer behaviour predicted using surface complexation constants measured on the individual minerals.

The data sets from the pre-test characterization will be made available for use in models from other research institutions. These will include stochastic models and deterministic models using retardation factors. These calculations will be completed prior to elution of the tracers, providing a basis for validation by comparing the model predictions to observed moisture and tracer behavior.

VIII. SUMMARY

An intermediate-scale transport-model validation experiment is being carried out by Sandia National Laboratories and Los Alamos National Laboratory at the Experimental Engineered Test Facility at Los Alamos National Laboratories. The experiment involves the detection and prediction of the migration of fluid and tracers through an unsaturated porous medium. The experimental test bed is a 6-m high x 3-m diameter caisson filled with quartz sand with a sorbing layer at an intermediate depth.

The overall objective of the caisson experiment is to implement a model-validation strategy in a simple system which requires the integration of a variety of hydrological and geochemical conceptual models, field measurements, and experimental approaches. More specifically, the test will attempt to determine if tracer velocities and concentrations in a field-scale transport experiment can be predicted from the hydraulic boundary conditions and the geochemical, hydraulic, and dispersion properties of the porous medium as determined in laboratory-scale experiments.

The focus of this paper is the design phase of the experiment. The design process involves several iterations of preliminary calculations, simple experiments and prototype constructions. Constraints related to hydraulic, geochemical, and instrumental criteria are evaluated simultaneously during this process. The final design will represent a compromise among the often conflicting demands of all of these factors.

Several different materials were considered for the porous matrix. Several different particle-size distributions for the porous matrix were considered in combination with hydraulic boundary conditions (applied flux and suction). Preliminary geochemical characterization of candidate caisson-filling materials included pH measurements of solutions in contact with the sand and sorbents and measurement of empirical sorption distribution coefficients (K_d's). The preliminary design currently being used for sensitivity analysis assumes Wedron 510 sand with a sorbing layer composed of a mixture of sand and powdered goethite. Currently, a suite of both non-sorbing and reactive tracers is being chosen from a number of candidate solutes including Ni, B, Br, and U.

Once the reactive substrate, solution compositions and tracers have been chosen and the caisson filled and

instrumented, more detailed pre-test material characterization will begin. During several stages of the experiment, hydraulic and geochemical characterization data described above will be used as input to several computer codes to predict the fluid flow and tracer migration. These codes include LLUVIA-II which solves the Richards' equation for the two- dimensional, isothermal flow of liquid water and the LEHGC code, a coupled reaction/transport simulator which models chemical speciation and sorption using equilibrium constants. Predictive modeling using the pre-test characterization data will begin in the early spring 1992 and should be completed before the start of the actual flow and transport test in late spring 1992, thus providing a basis for validation by comparing the predictions to observed moisture and tracer behavior.

This collaborative effort integrates the laboratory research program developed for the Model Development and Validation Task for the Yucca Mountain Project[10,11] with an intermediate-scale experiment. The research described here demonstrates the necessity of a laboratory program focused on process definition and understanding. The current intermediate-scale experiment is a rehearsal for future field studies in tuff at Yucca Mountain, studies in which systematic laboratory research also will be required to interpret the measured field-scale system behavior.

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