Coupled Processes Influencing the Transport of Uranium over Multiple Scales

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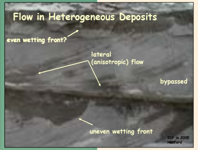
Waste storage tanks at Hanford


Flow in Heterogeneous Deposits

PROBLEM: Leaking radioactive waste has entered the subsurface at DOE sites. Prediction of contaminant transport is complicated by geological heterogeneities, resulting in scale-dependence of transport parameters.

GOAL: To provide validated scaling strategies which can be applied to existing contaminant distributions and migration scenarios at Hanford and similar sites

OBJECTIVES:

1. **Layer Scale:** Separate quantification of hydraulic, geochemical, and mineralogical factors influencing U(VI) transport
2. **Up-Scale:** Apply numerical, composite medium, and fractal approaches to compute effective coupled hydraulic and reactive transport parameters
3. **Validate:** Apply up-scaled parameters to U(VI) transport through progressively larger scales of intact samples that encompass both lateral and vertical U(VI) transport

FUTURE WORK:

1. Repeated measurements on unconsolidated granular material to determine precision of URC method.
2. Determination of hydraulic properties of individual layers of large intact Hanford sediment sample.
3. Determine applicability of composite medium model to transient system
4. Validate model with measurements at different scales
5. Extend model from monofractal (2 materials) to multifractal (many materials)
6. Extend uncertainty analysis to large-scale models

LAYER SCALE:

The ULTRA ROCK CENTRIFUGE (URC) measures water content as a function of pressure (θ(ψ)) and predicts hydraulic conductivity (K(S)). Solution is collected from centrifuged, originally saturated samples to generate a production curve (Fig. 1). The slope of the production curve is used to calculate relative permeability (Fig. 2) using the following expressions (Christiansen, 2002; Hagoort, 1980):

$$k_{rw} = \frac{\mu_w \varphi L}{\Delta\rho \alpha k} \frac{dQ_w}{dt}$$

$$S_w = 1 - Q_w + t\frac{dQ_w}{dt}$$

where μ_w viscosity, L length of sample, φ porosity, α average centrifugal acceleration, Δρ density difference between displaced and invading fluid, k intrinsic permeability, k_{rw} relative permeability, S_w saturation, Q_w cumulative water production, t time since centrifuge started.

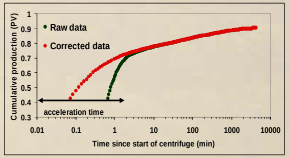


Fig. 1. Production versus time at constant angular velocity of 9000 RPM. Raw data is corrected during early times in which RPM < 9000.

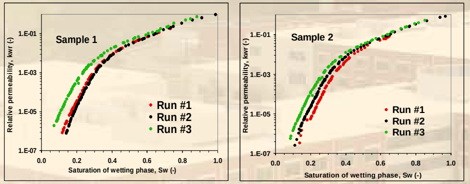


Fig. 2. Relative permeability versus saturation for two small diameter Berea Sandstone test cores. The measurement was repeated 3 times.

Berea Sandstone test cores were repeatedly measured to study the degree of consistency of the laboratory method (e.g., Fig. 2). At high saturations, the precision of the k_{rw} measurements appears to be higher than at low saturation, where the relative k_{rw} can vary up to 0.5–1.5 order of magnitude for any given saturation value.

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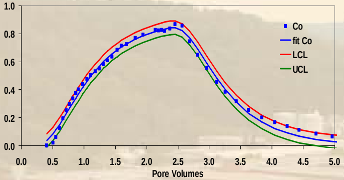
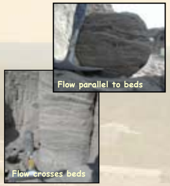
INTERMEDIATE SCALE:

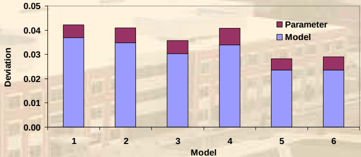
In order to reliably estimate transport parameters, analyses of uncertainty and sensitivity are performed to determine the contributions of model and parameter estimation errors. Previous work involved the transport of Br, Co, and U(VI) in intact Hanford sediment cores in which flow is parallel or perpendicular to bedding (Mayes et al., 2003, in prep; Pace et al., 2003, 2007). Six different parameter combinations using convective-dispersive equation (Parker and van Genuchten, 1984) for simultaneously fitting nonreactive tracer Br and reactive tracer Co are attempted (Fig. 3).




Flow parallel to beds

Flow crosses beds

Fig. 3. Fitting velocity (V), dispersion (D), retardation (R), pulse, and decay constant (μ) to flow-bedding-parallel Co data in Hanford Coarse (HC) sediment showing 95% confidence limits. The fit corresponds to Fit Combination #5 (Fig. 4).



Fit Combinations
1. D, R
2. V, D, R
3. V, D, Pulse, R
4. D, R, μ (decay)
5. V, D, R, μ
6. V, D, Pulse, R, μ

Fig. 4. Combined uncertainty due to model fitting error and parameter uncertainty for 6 combinations of fitting parameters.

Fig. 4 shows that uncertainties due to model fitting errors exceed those due to parameter estimation errors. Errors decrease with increasing estimated parameters for combinations 1-5. Combination 6, however, did not improve the fit and therefore can be eliminated.

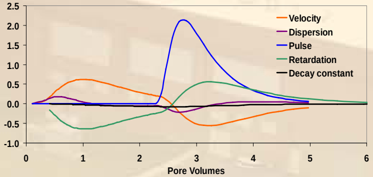


Fig. 5. Sensitivity of fitting parameters during experimental progress.

Fig. 5 demonstrates the sensitivity of parameters to the concentration measurements.

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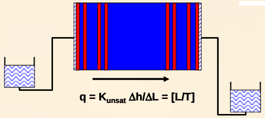
LARGE 2D SCALE:

Effective hydraulic parameters of unsaturated layered sediments were estimated using a physically-based Cantor Bar model to represent interbedded layers of coarse (blue) and red (fine) sediments (Tang et al., accepted).



A direct averaging approach, the composite medium model, is used to scale the hydraulic parameters from individual layers to a composite system (e.g., Mualem, 1984; Yeh et al., 1985; Pruess, 2004).

This approach has been criticized because it ignores variances in the hydraulic gradient (dh or Δh) (Khaleel et al., 2002). Therefore we tested the sensitivity of the model to variations in hydraulic gradient over scales of 10-100 cm.

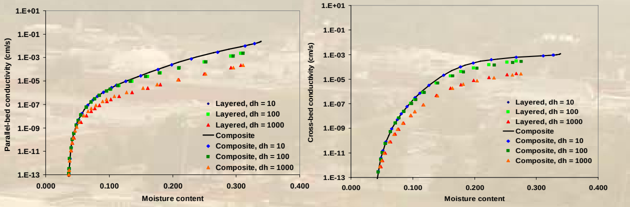


Fig. 6. Comparison between layered and effective (composite) parameters at a scale of 10 cm for parallel-bed and cross-bed conductivity for a range of hydraulic gradients (dh).

The hydraulic conductivity calculated from layered and effective parameters were similar regardless of the gradient (Fig. 6). The model works well the for steady-state 1D case. Results were similar for length scale of 100 cm (not shown). Anisotropy is not sensitive to the gradient (Fig. 7).

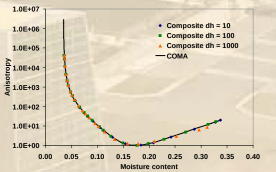


Fig. 7. Anisotropy as a function of gradient (dh).

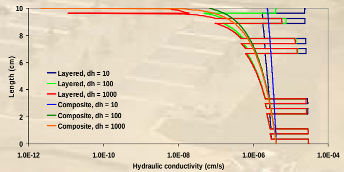


Fig. 8. Simulation of 2D layered system .

The variance in conductivity at high gradients causes the difference between layered and composite cases (Fig. 8). The hydraulic conductivity of the composite case is close to the harmonic mean of the layered case (Fig. 8), meaning that the composite medium approach is valid for these conditions.

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