Quantifying Flow and Reactive Transport in the Heterogeneous Subsurface Environment: From Pores to Porous Media and Facies to Aquifers

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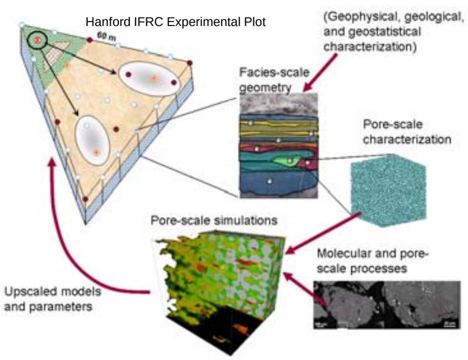
Introduction and Objective

Reactive transport simulation models serve as a critical integrating element among the various experimental elements of the SFA, in particular integrating information from fundamental process scales (molecular, sub-pore, and pore) to phenomenological prediction scales (column, field). Our work is focused on developing pore-scale models of processes critical to uranium transport at the Hanford IFRC, and integrating information from the pore-scale models into a field-scale model (in cooperation with IFRC researchers). Our approach will provide a means for systematically incorporating information from SFA experiments at smaller scales into the field scale model. Currently there remains a gap between scientific understanding at fundamental scales and simulation at field scales that leads to an unsatisfactory degree of model and parameter empiricism in many field-scale simulations. Quantitatively linking across this wide range of physical scales with minimal empirical calibration is an ambitious goal that is being enabled by strong coordination of SFA research and ongoing computational advances.

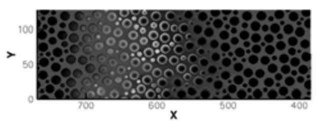
Approach

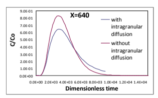
We have proposed a general strategy for predictive modeling that is based on the combination of pore-scale modeling of fundamental processes, upscaling to define hydrofacies-scale processes and properties, and application of advanced field-scale characterization methods to define the spatial distribution of hydrofacies. This strategy is described schematically in the diagram below, and will be tested at the Hanford IFRC site. This approach requires the following critical elements: 1) development and validation of high-performance codes for simulation of relevant pore-scale processes (SFA collaboration with Chongxuan Liu); 2) development and application of pore-to-facies-scale upscaling methods (external collaboration with Brian Wood, OSU); 3) integration of field geophysical data from the Hanford IFRC (SFA collaboration with Andy Ward).



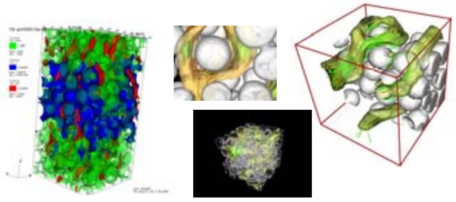
Pore-Scale Model Development and Validation

At the Hanford IFRC, surface complexation and intragranular diffusion are two critical pore-scale mass transfer processes. We have incorporated a model of intragranular diffusion into our parallel 3D smoothed particle hydrodynamics (SPH) code.

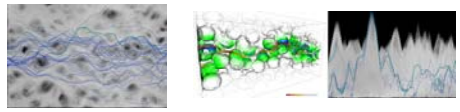




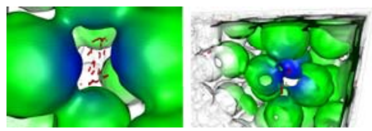
The parallel 3D code has been ported successfully to the Franklin computer at NERSC and increasingly large simulations have been performed (up to 14 million computational particles)



The graphics below illustrate insights that can be gained from visualizations of pore-scale simulations. Visualizations by Chad Jones (UC Davis). Top panel: 2D mapping of 3D pathlines showing target pathline in teal (left); 3D visualization of target pathline indicating distance between pathline and grain surfaces (center); velocity variations along pathlines with target pathline in teal (right).

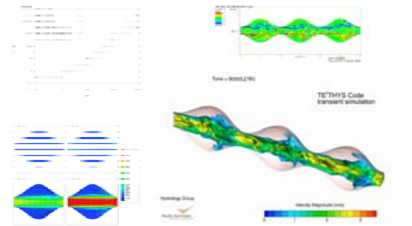


Right: Two views of "trapped" fluid particles that do not leave the local pocket in 150 simulation time steps.

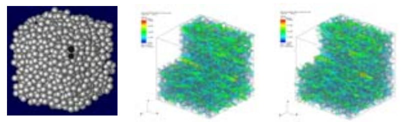


Upscaling Methods – Development and Application

We have successfully implemented numerical volume averaging (VA) in the TE^{2}THYS code and performed tests using the "Wavy Tube" model. The test results (graph in upper left below) demonstrate that the VA method reproduces dispersivity values calculated by particle tracking (PT).   We have performed a suite of simulations at variable Reynolds numbers (Re) that represent laminar and transient conditions. Surprisingly, dispersion is smaller for the fully transient solution compared to the equivalent average (steady) velocity field. Upon further inspection, this occurs because the transient flows move particles into and out of recirculation zones more effectively, reducing their impact on particle spreading.



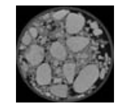
We are performing pore-scale simulations of oscillatory flows, representative of conditions at the Hanford 300 Area where river stage fluctuations induce groundwater flow oscillations. These oscillations may play a significant role in uranium transport by providing a mechanism for transporting solutes in the direction opposite to the average advective flow. (Please see video clip on laptop for animation of oscillatory flow simulations).



Summary and Conclusions

- We have developed and validated a novel capability for numerical upscaling using volume averaging, implemented in an efficient parallel code operating on high-performance platforms. We can now perform 3D simulations of transport incorporating intragranular diffusion, inertial flow, and oscillatory flow in domains large enough to approach representative elementary volumes (REVs).
- We have analyzed the effects of inertial flow regimes (transitory between laminar and turbulent) on dispersion.   Transitional unsteady flows decrease effective dispersion relative to laminar flow because of increased rates of mass transfer between advective and recirculation domains.
- Our CFD-based pore-scale code is extremely efficient for solving single-phase flow and non-reactive transport. Our SPH-based pore-scale code is well-suited to solving problems involving moving interfaces and multlicomponent reactions.

Future Research



Upscaling of Uranium Adsorption/Desorption:

The general objectives of our upscaling work will be to develop theory in support of the multi-scale experimental work being conducted by other SFA investigators. In particular, the following hypotheses are advanced to provide a framework for the upscaling efforts.

Hypothesis 1. **The effective rate of U adsorbtion / desorption in the meso-scale column system (< 2mm size fraction) can be explained on the basis of the measured kinetics from the microscale batch experiments and mass transfer. This microscale-meso-scale relationship can be explained quantitatively via upscaling using volume averaging.**
An effective transport model developed for this system via volume averaging should provide a quantitative explanation for the influence of geometry of the porous medium (including subgrain scale fissures) of the effective rate parameter for cases where the geometry is known. Such an approach has been used by previously to examine mass transfer limitations in biofilms in porous media (Wood et al., 2007), with significant success. The case of U transport in heterogeneous porous networks has many similarities to this problem, and it suggests that an approach similar to that outlined in Wood et al. (2007) will be a useful one. The practical outcome of this approach will be a model which incorporates a reaction rate effectiveness factor (e.g., Wood et al., 2007) that modifies the microscale reaction rate to account for the influence of mass transfer. Even for cases where the microscale geometry is not well characterized, this relationship will have significant value in its capacity to explain the physics and chemistry creating the reduced reaction rate.

Hypothesis **2. In field-textured materials, the multi-scale influence of convective-dispersive transport and mass transfer limitations can be determined quantitatively. This relationship can be determined by upscaling via volume averaging where both the microscale and mesoscale mass transfer, mass transport, and reaction processes are represented.**
A second level of upscaling (meso-scale to macroscale) can be accomplished via volume averaging by building off of the first level of upscaling generated as part of the research for hypothesis 1. In this case, the interaction between the microscale reaction and mass transfer limitations will be incorporated into a model that then also incorporates the larger-scale convective-dispersive mass transport processes. The result of this second level of averaging will be (i) an effective mass transport and reaction model for U transport in field-textured materials, and (ii) the effective parameters (effective rates of reaction, effective dispersion tensor, effective mass transfer coefficient) that applies to this scale. Such representations will have significant utility in making the link between the experimental results developed from the experimental micromodels and the field. Even for cases where the structure of the porous media is not explicitly known, such a development will provide a sound basis for understanding and correctly interpreting the combination of mass transfer, mass transport, and reaction processes in field textured materials.

Presentations

- Scheibe, T. D., A. M. Tartakovsky, and B. J. Palmer, "Pore-scale simulation and upscaling of solute transport in groundwater incorporating intra-grain mass transfer processes," *Eos Trans. AGU*, 90(52), Fall Meeting Supplement, Abstract H23D-0989, 2009.
- Scheibe, T. D., A. M. Tartakovsky, and B. J. Palmer, "Pore-scale simulation and upscaling of uranium transport in groundwater incorporating intra-grain mass transfer processes," poster presentation given by T. Scheibe at Migration '09, Kennewick, WA, September 20-25, 2009.
- Scheibe, T.D., "Quantifying Flow and Reactive Transport in the Heterogeneous Subsurface Environment: From Pores to Porous Media and Facies to Aquifers," Henry Darcy Distinguished Lecture (Invited) given at multiple locations through calendar year 2010. To date, this lecture has been given at: Florida State Univ. (1/7); Florida Int'l Univ. (1/8); UC Davis (1/21); Univ. of Arizona (1/27); UC Denver (2/2); Colorado St. Univ. (2/3); Michigan St. Univ. (2/11); Wright St. Univ. (2/17); Univ. of N. Carolina (2/19); LANL (3/2); USGS Tacoma (3/9); WA Dept. of Ecology (3/17 – See photo at right); and Univ. of Nevada (3/22).

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