Reactive Transport Modeling for the PNNL Science Focus Area

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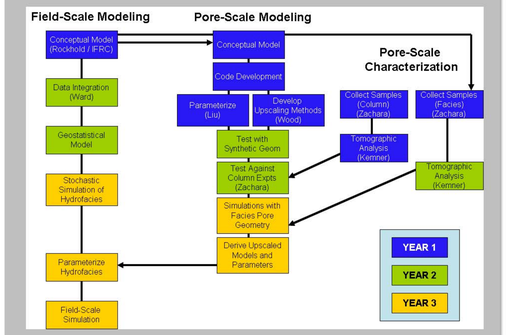
Project Role in PNNL SFA:



This project plays a central role in SFA integration by providing a quantitative framework for integrating data from the various SFA projects across scales to address key issues at the Hanford Integrated Field Research Challenge site.The chart above schematically represents the approach we are applying for quantitative integration across scales. Our PORE-SCALE MODELS (1) are being modified to incorporate NEW INFORMATION FROM MOLECULAR AND PORE-SCALE RESEARCH (2). Field-scale geophysical characterization provides a framework for modeling relatively large-scale features (hydrofacies) and their spatial distribution (including uncertainty). Within each hydrofacies class, samples will be subjected to 3D PORE-SCALE CHARACTERIZATION (3) as the basis for pore-scale simulations of critical geochemical reactions and mass transfer processes. Volume averaging and other UPSCALING METHODS (4) will be employed to define upscaled models and parameters for each hydrofacies class. The results will be integrated into the 300-AREA IFRC FIELD-SCALE MODEL (5) using the STOMP code to support design and interpretation of field experiments.

Fundamental Science Problems:

Scale issues are central to the SFA theme and hypotheses (microenvironments and transition zones are local-scale features that exert control on field-scale phenomena)

* Quantitative understanding of coupled flow, transport and reaction at the pore scale (mass transfer of uranium)
* Development and testing of rigorous continuum-scale models based on pore-scale simulations
* Prediction of laboratory- and field-scale experimental observations using upscaled, pore-scale, and hybrid models (with minimal "calibration")

(1) PORE-SCALE MODELING

Smoothed Particle Hydrodynamics

Smoothed Particle Hydrodynamics (SPH) is a particle-based (mesh-free) simulation method that is well-suited to pore-scale simulation. Our research group has applied this technique to simulation of multiphase flow and solute transport in porous media (Tartakovsky and Meakin, Vadose Zone Journal, 2005; Tartakovsky and Meakin, Phys. Rev. E., 2005; Tartakovsky et al., Physics of Fluids, 2007) and pore-scale mineral precipitation (Tartakovsky et al., Journal of Computational Physics, 2007, Tartakovsky et al., Water Resources Research, 2007 and 2008). Under this project, we have initiated the application of SPH to simulation of biofilm development in porous media at the pore scale (see images below from Tartakovsky et al, in press).

Under separate funding (SciDAC Science Application), we have developed a three-dimensional parallel SPH code for performing high-resolution pore-scale modeling in arbitrary 3D pore geometries. The images below visualize simulated flow velocities of fluid particles in a pore geometry measured by x-ray microtomography.

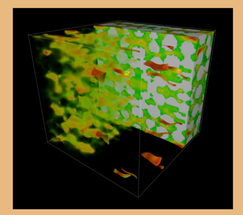
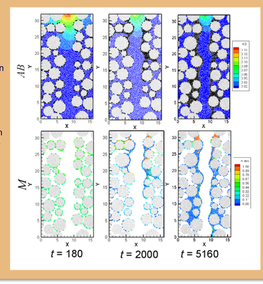


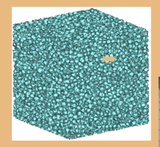
Image to right: Distribution of biomass simulated by the SPH model of biofilm growth, at three different dimensionless times. Gray particles denote soil grains. In the top row of figures, black particles represent fluid with a non-zero biomass concentration, and the color scale indicates the concentration product of electron donor and acceptor in the fluid with zero biomass concentration. In the bottom row of figures the color scale denotes the distribution of biomass.



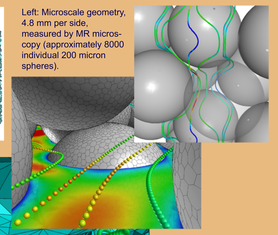
Finite Volume CFD Methods

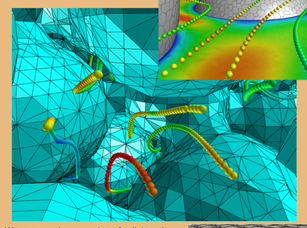
We are also applying grid-based finite volume methods drawn from the field of Computational Fluid Dynamics (CFD) to pore-scale simulation of flow and reactive transport. Grid generation issues for arbitrary and highly complex pore geometry are significant, particularly in the vicinity of grain-to-grain contacts. We have developed a method for automatically generating numerically accurate grids, and we have conducted 3D pore-scale flow simulations using both STAR-CD and a PNNL-developed code (TE2THYS) using local workstations, a Linux cluster, and the EMSL supercomputer Chinook.

These simulations are used both for direct simulation of pore-scale processes, and for solution of the upscaling closure equations for arbitrarily complex pore structures (see panel (3)).



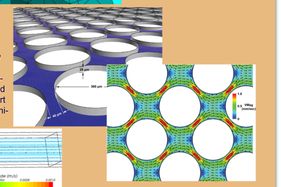
Left: Microscale geometry, 4.8 mm per side, measured by MR microscopy (approximately 8000 individual 200 micron spheres).





Above and left: Visualizations of simulated pore-scale flow through measured and synthetic grain packs. Colors indicate local flow velocity, with red being high and blue being low.

We are pursuing a number of collaborative efforts in which our pore-scale tools are being applied to support interpretation of microscale flow and reactive transport experiments. The images to the right show simulation of the velocity field within a quasi-2D microfluidic cell (experiments conducted by Werth and Valocchi at UIUC), and the image below represents colloid transport experiments performed at Oregon State University (Dragila and Ochiai)



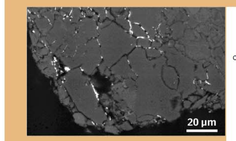
(2) INCORPORATE MOLECULAR AND PORE-SCALE INFORMATION

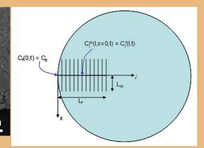
Example: Intragranular Mass Transfer of Uranium

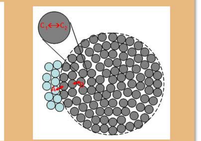
The image below shows a back-scattered electron SEM image indicating the intragrain distribution of U(VI) precipitates (white) within a feldspar grain (From Liu et al., *Water Resources Research,* 2006).

The diagram in the center indicates schematically the Liu et al. (2006) model of two-domain diffusion. Cf = concentration in primary fracture; Cm = concentration in secondary fracture (matrix); Cb = concentration in bulk pore water.

The diagram on the right shows the implementation of a two-domain diffusive mass-transfer model in SPH. Grey particles represent solid phase; blue represent fluid phase. Diffusive transfer between fluid and solid phase particles in primary porosity (i.e, A) and between two solid particles (i.e, B) is implemented using the standard SPH diffusion model. Diffusive transfer between the primary and secondary porosity domains (C1 and C2) occurs only within each SPH particle as denoted by the enlarged gray particle. This approach is being implemented in the 3D parallel SPH code to allow simulation of pore-scale transport of uranium including diffusion-limited mass transfer.

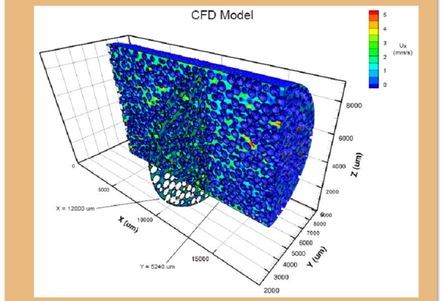




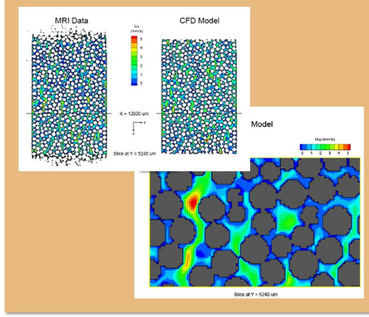


(3) PORE-SCALE CHARACTERIZATION

Example: Magnetic Resonance Imaging



Our collaborator at Montana State University, Dr. Joseph Seymour, has provided us with a 3D MRI dataset describing pore geometry and z-component of velocity in an experimental glass beadpack. The dataset is resolved in 40-micron voxels. We are using this dataset to test our 3D CFD and SPH models of pore-scale flow. Above: A 3D CFD simulation of flow in the experimental beadpack. Below: Comparison of MRI-measured velocities and simulated velocities in a 2D slice through the 3D model/data. Also shown is a close-up of a portion of the simulated velocity field, showing local high-velocity flow zones (preferential flow paths).



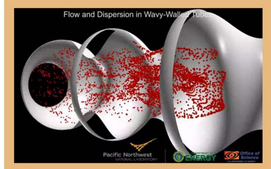
(4) UPSCALING METHODS

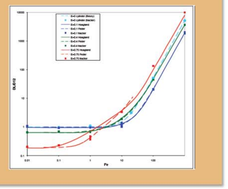
Example: Volume Averaging - Numerical Solution

Volume averaging is one approach to rigorous upscaling; i.e., definition of macroscopic process models (equations) and parameters based on detailed knowledge of microscopic processes. Most reported applications rely on analytical solutions that require restrictive assumptions (e.g., periodic monodisperse media with perfect cubic packing). However, volume averaging methods can be implemented for more general cases (e.g., arbitrary pore geometries) by solving the closure equations numerically. Such solutions take very similar form to the numerical solutions of the underlying flow equation, and therefore are amenable to application of CFD codes (see panel (1)). We have performed initial numerical studies using a 3D wavy tube model that approximates pore throats and pore bodies, to develop and test our numerical methods. Once validated, the codes will be applied to more generalized pore geometries.

Below left: 3D visualization of simulated Lagrangian particles moving through a 3D wavy tube, computed using grid-based CFD methods.

Below right: Axial dispersion in the wavy tube, calculated using our CFD model and compared with literature reports.





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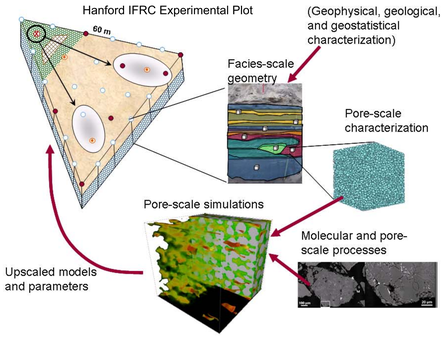

Publications:

Tartakovsky, A. M., P. Meakin, T. Scheibe, and B. D. Wood, "A smoothed particle hydrodynamics model for reactive transport and mineral precipitation in porous and fractured porous media," *Water Resources Research*, 43(5):Art. No. W05437, doi:10.1029/2005WR004770, 2007.

Golfier, F., B. D. Wood, L. Orgogozo, M. Quintard, and M. Buès, "Biofilms in porous media: Development of macroscopic transport equations via volume averaging with closure for local mass equilibrium conditions," *Advances in Water Resources*, 32: 463–485, 2009.

Tartakovsky, A. M., T. D. Scheibe, and P. Meakin, "Pore-scale model for reactive transport and biomass growth," accepted March 2008 for publication in *Journal of Porous Media*, in press.

Invited Presentation:

Scheibe TD, AM Tartakovsky, BJ Palmer, MC Richmond, BD Wood, I Battiato, and DM Tartakovsky. 2008. "Linking pore- and Darcy-scale models of reactive and non-reactive transport," *Eos Trans. AGU*, 89(53), Fall Meet. Suppl., Abstract

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About Pacific Northwest National Laboratory

The Pacific Northwest National Laboratory, located in southeastern Washington State, is a U.S. Department of Energy Office of Science laboratory that solves complex problems in energy, national security and the environment, and advances scientific frontiers in the chemical, biological, materials, environmental and computational sciences. The Laboratory employs 4,000 staff members, has a $760 million annual budget, and has been managed by Ohio-based Battelle since 1965.

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