

Report for 2003LA17B: Use of Synchrotron Microtomography and X-Ray Fluorescence to Better Understand Contaminant Diffusion in Reactive Barrier Systems

- Conference Proceedings:
 - Rahman, A., S. Hartano, D. Carlson, and C. Willson, 2003, Incorporating Uncertainty into High-Resolution Groundwater Supply Models, in Proceedings of the Probabilistic Approaches & Groundwater Modeling Symposium held during the World Water and Environmental Resources Congress in Philadelphia, Pennsylvania, June 24-26, 2003, edited by Srikanta Mishra, ASCE Pub.
- Other Publications:
 - Brown, G.E., S. R. Sutton, J.R. Bargar, D.K. Shuh, W.A. Bassett, P. M. Bertsch, J. Bisognano, W.F. Bleam, D.L. Clark, P. De Stasio, S.E. Fendorf, P.A. Fenter, E. Fontes, J. Hormes, K.M. Kemner, S.C.B. Myneni, P.A. ODay., K.H. Pecher, R.J. Reeder, A. Roy, S.J. Traina, C.S. Willson, and J.M. Zachara, 2003, Molecular Environmental Science: An Assessment of Research Accomplishments and Needs, EnviroSync Molecular Environmental Science White Paper, SLAC-R-704, Stanford University, Stanford, CA 94309, 66pp.
 - Hanson, B. C., C.S. Willson, R. Milner, D. Carlson, A. Rahman, and S. Hartano, 2003. Annual Report, Evaluation of Aquifer Capacity to Sustain Short-, Long-Term Ground Water Withdrawal From Point Sources in the Chicot Aquifer for Southwest Louisiana. Water Resources Division, Louisiana Department of Transportation and Development, June 2003.

Report Follows

SYNOPSIS

Title Use of Synchrotron Microtomography and X-Ray Fluorescence to Better Understand Contaminant Diffusion in Reactive Barrier Systems

Problem and Research Objectives

Sediment contamination can occur as a result from the deposition of pollutants from the water column, deposition of contaminated particles, or through the seepage of contaminated groundwater. Unless removed or “controlled”, contaminated sediments act as a continual source of pollutants to surface water bodies. This contamination may impact aquatic species and potentially render the water bodies unusable for recreation and/or drinking water supply.

This project will be among the first studies to non-destructively provide both spatial and temporal data on the migration of contaminants in porous media along with associated changes in the pore morphology. Contaminant concentration profiles will be quantified over time at the elemental level using X-ray fluorescence techniques. X-ray microtomography will be used to obtain high-resolution three-dimensional images of the pore structure. These images can be used to quantify the changes in pore morphology (e.g., tortuosity, porosity, etc...) over time due to physical and/or chemical processes. These data will allow for a better understanding of the pore-scale processes impacting the migration of contaminants in reactive barrier systems and for the validation or development of predictive models used to design capping systems.

Methodology

X-ray Fluorescence (XRF): Basic XRF has become a well-established multi-element technique, capable of yielding accurate quantitative information on the elemental composition of a variety of materials. The technique is well-suited for studying environmental science problems because it is non-destructive, relatively rapid, and solids can be analyzed with little or no sample preparation. Apart from light elements, all elements with atomic numbers greater than 11 can be detected. The method is sensitive down to microgram-per-gram level and the results are precise and accurate if matrix effects can be corrected.

White-light XRF experiments have been performed at the LSU Center for Advanced Microstructures and Devices (CAMD) synchrotron facility. To date, these scans have been on cores obtained from a pilot-scale experiment currently being conducted at the University of New Hampshire.

Synchrotron X-ray Microtomography (SXM): Synchrotron X-ray Microtomography has been developed over the past decade as a technique to non-destructively image the interiors of materials. Tomography deals with reconstruction of an object from its projections. Spatial resolutions on the order of ~ one to ten microns are possible because of the highly collimated and extremely bright X-rays produced by a synchrotron. These highly-parallel X-rays permit spatial resolution that is only limited by the optical

components used to image them. Furthermore, the ability to tune to a monochromatic X-ray energy allows elemental discrimination.

To date, all SXM experiments have been performed at the GeoSoilEnviroCARS (GSECARS) beamline at the Advanced Photon Source, Argonne National Laboratory. However, the tomography beamline at CAMD has recently been upgraded in order to improve our ability to image cores such as those proposed in this work.

Principal Findings and Significance

A series of XRF scans have been made over the past year. As mentioned above, all of the scans have been of cores obtained from the University of New Hampshire. However, the results from these experiments have demonstrated the effectiveness of CAMD's XRF facilities to obtain quantitative data from quasi-natural systems. The figure below shows one example of a diffusion profile obtained; in this case the diffusion profile is of Zn in a two sediment system where the negative values are in the contaminated Newton Creek sediment and the positive values are in a Great Bay sediment. Note the high resolution of the system (i.e., 0.5 mm) and the relatively small error bars, indicating good reproducibility.

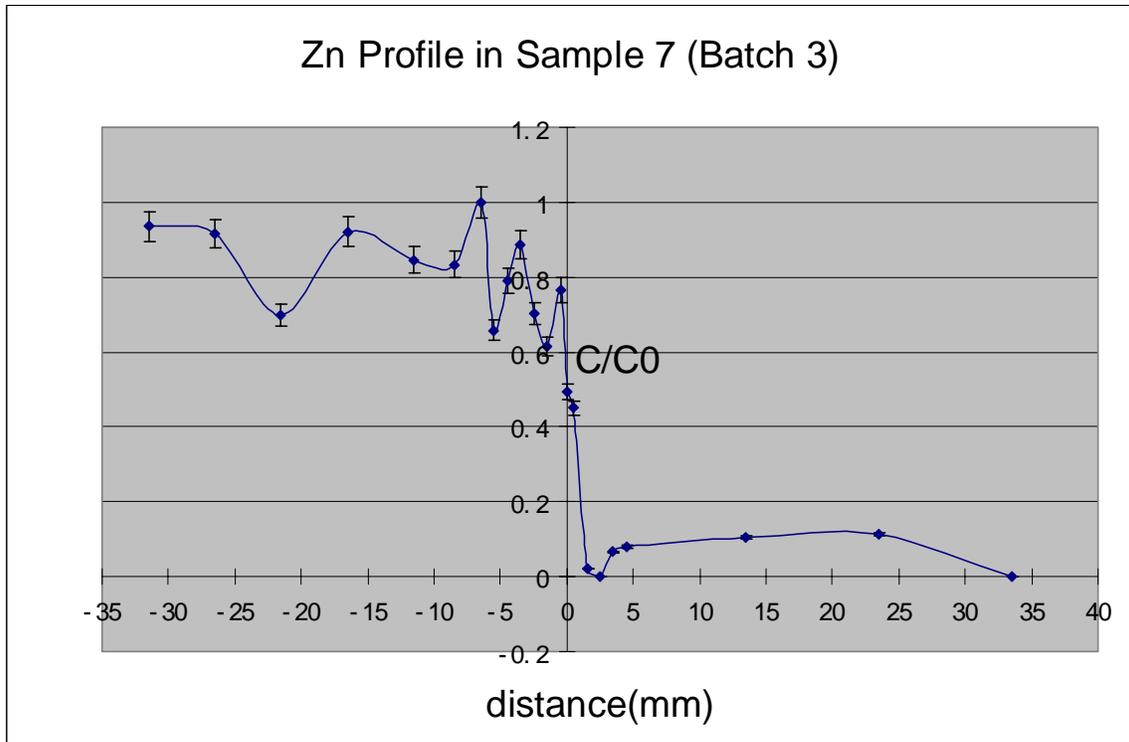


Figure 1: Zn Diffusion Profile

Based on the XRF data, we have been able to successfully fit a two-phase diffusion model and determine effective diffusivity values for a number of metals and sediment/barrier systems.

Synchrotron X-ray Tomography scans have been made on small tubes extracted from the larger cores from the UNH study and on tubes packed in our laboratory specifically for tomography experiments. The two figures below were made at the GSECARS tomography beamline at 12.54 micron-resolution and an energy of 33.0 keV. The first figure, a vertical cross-section through the column, shows the uneven interface between the sediment and the phosphate barrier system. Also note that even at this high resolution we are unable to distinguish individual sediment particles; this sediment primarily consists of fine sand, silt, and clay. The second tomography image is a three-dimensional image of a similar system. We are currently in the process of analyzing these systems for porosity, tortuosity, pore body/throat size distributions, etc...

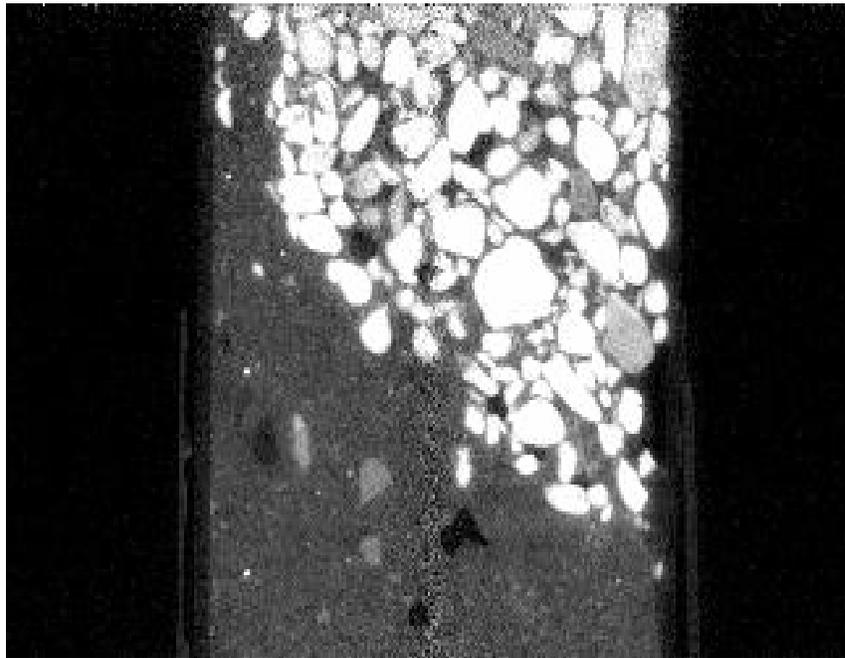


Figure 2: Vertical Cross-Section with sediment on bottom and phosphate barrier on top

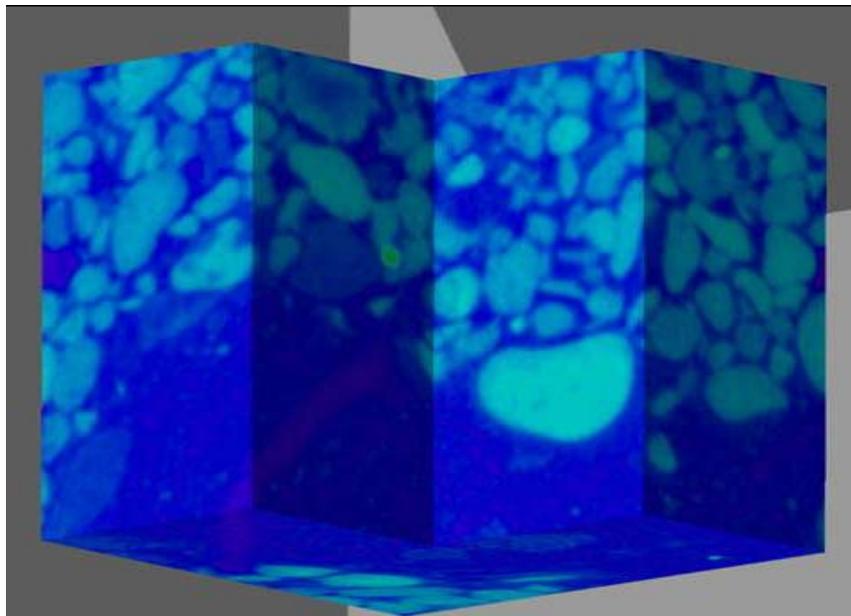


Figure 3: Three-dimensional image of sediment/phosphate barrier system

Based on the XRF data collected from the UNH project and the preliminary tomography images collected at APS, we have developed a detailed experimental plan to begin the small column XRF/SXM experiments this summer (i.e., 2004). We are now confident that we have the ability to obtain sufficiently high-quality data and quantify the important data. In brief, the procedure is as follows:

1. Packing of approximately 12 columns (5 mm i.d.). Two replicates of the following: Anacostia Sediment with no cap; Anacostia Sediment with “Florida” phosphate cap; Anacostia Sediment with “North Carolina” phosphate cap; Anacostia Sediment with sand cap; “spiked” sand system with no cap. The columns will be fully saturated and maintained at “ponded” conditions. The ponded water will be replaced at regular intervals.
2. SXM scan. From this scan, system (e.g., porosity, tortuosity, etc..) and pore-scale (e.g., pore body/throat sizes, connectivity, etc...) will be determined.
3. XRF scan. From this scan, diffusion profiles of the contaminants will be obtained.
4. Steps 2 and 3 will be repeated at 45 or 90 day intervals depending on the rate of diffusion and reactions.