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Simulation of Contaminant Flow in a Laboratory-scale Porous System

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Simulation of contaminant flow in a laboratory-scale porous system

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ABSTRACT

The microscopic movement of contaminants in a porous medium has been simulated in an experiment. The approach has been to study the microscale transport processes using a novel nonintrusive fluorescence imaging technique developed in our laboratories. The system studied consists of a packed porous column with a refractive index-matched fluid seeded with fluorescent tracer particles (for flow measurements) or an organic dye (for contaminant concentration measurements). Microscopic measurements of contaminant concentration, contaminant velocity, and pore geometry were obtained in a full three-dimensional volume of the test section at a good accuracy and a high resolution. 3D plots of these measurements show the complex geometry of the porous medium. It is also seen that near the contaminant front there is a significant correlation between the flow and the contaminant concentration. The goal is to use these and future results toward better understanding of contaminant flow and transport thorough natural porous media.

1. INTRODUCTION

Our research at Lawrence Livermore National Laboratory is a combined experimental and theoretical effort to improve our understanding of contaminant flow and transport in soils and other porous media by investigating the relationship between transport at the microscopic spatial scale and transport at the macroscopic scale. Many bulk phenomena associated with contaminant transport in porous media result from the flow behavior of the system at the microscopic spatial scale. However, very little is known about microscale contaminant/chemical transport in porous media.

There have been many experimental attempts to measure macroscopic variables and medium properties in porous media.¹⁻⁵ However, little research has been devoted to experimental observation and characterization of processes within the pore spaces themselves. Experimental observations of flow and transport behavior at the pore scale in three-dimensional systems are essentially nonexistent because of the difficulty of obtaining detailed dynamic measurements within the pore spaces of a solid medium in a nonintrusive fashion.

Despite the traditional difficulties involved in observing microscopic pore-scale processes, numerous theoretical approaches⁶⁻¹⁶ have been used to average pore-scale behavior to a more practical, operational level. They typically describe the problem on a macroscopic level in which the intricate structure of pore networks and grain boundaries are replaced with an effective continuum and an averaged set of balance equations and variables. While each approach presents a slightly different point of view, all require some assumptions that describe the physical or statistical structure of the medium as well as the spatial scales required to define the macroscopic quantities.

The present work is an effort to study microscopic contaminant flow and transport processes within porous media through direct measurements and observations. The goal is to provide the microscopic measurements of flow, concentration, and porosity using novel nonintrusive imaging techniques. Imaging within the pore-spaces

of the medium is made possible by constructing porous systems with solid and liquid materials having identical refractive indices. The resulting uniformity of refractive index eliminates refraction scattering from solid-liquid interfaces within the medium and allows clear imaging at any point within it. At the present time, our measurements are producing unique data¹⁷⁻²⁰ at spatial resolutions that have not been possible in the past. These measurements and the derived information are extremely valuable in understanding of contaminant transport through natural porous systems (e.g., soil).

2. EXPERIMENTAL FACILITIES AND MEASUREMENT TECHNIQUES

Figure 1 shows the general detail of the experimental facilities with a rectangular column for studying the contaminant transport in porous media. These facilities are equipped to two separate columns. One is a rectangular column for transport and bioremediation studies in aqueous systems and the other is a cylindrical column for transport investigations in nonaqueous systems.

Experiments in the aqueous system were performed in a clear Plexiglas rectangular packed column 3.0 cm in width and about 22.0 cm in length. The difficulty in performing experiments in aqueous system was to be able to find optically clear materials that have refractive indices close to water's refractive index of about 1.33 at 20.0°C and 514.5 nm. This task was accomplished after many searches and tests. We were able to find some natural minerals that can be found in nature or made as crystals in the laboratory with refractive indices close to that of water. These minerals are the crystals of fluoride salts that were made in the laboratory to our range of size specification for these experiments. Therefore, the column was filled with these natural mineral particles of nonuniform sizes with average diameters of about 0.15 cm. An aqueous solution was chosen as the fluid which matched the particles' refractive index at 20.0°C and a wavelength of 514.5 nm. The column was maintained at this temperature throughout all runs by being immersed in a circulating constant temperature bath. A constant volumetric flow rate of the above fluid was provided for each run covering a wide range of Reynolds number, 10^{-3} to 1 (Reynolds number is based on average pore velocity and average particle diameter).

Experiments in the nonaqueous system were performed in a clear polymethylmethacrylate (PMMA) cylindrical packed column 4.5 cm in diameter and about 23.5 cm in length. The column was filled uniformly with PMMA spherical beads of 3.1 mm. A mixture of silicone oils (Dow Corning 550 and 556) was chosen as the fluid which matched the beads' refractive index of about 1.49 at 19.8°C and a wavelength of 514.5 nm. The column was maintained at this temperature throughout all runs by being immersed in a circulating constant temperature bath. A syringe pump was used to provide a constant volumetric flow rate of the above fluid at 1.15 cm³/min and Reynolds number of about 10^{-3} (Reynolds number is based on average pore velocity and average particle diameter).

The experimental setups were designed such that at the test section, flow was free from any wall or entrance effects. The experiments were done with the refractive matched fluid seeded with fluorescent latex microspheres of 6.5 μ m in diameter (for velocity measurements) or an organic fluorescent dye (for concentration measurements). The column was illuminated by an Argon-ion laser (coherent) operated at 475 nm for velocity measurements and 488 nm for concentration measurements. A CCD camera was used to record the experimental runs. Since, the dye emission peaks around 514.5 nm, a band pass filter was used on the video camera to pass a narrow range of 514.5 nm $\pm$ 5 nm wavelength associated with the dye excitation. The video camera was placed perpendicular to the laser propagation beam on a remotely operated platform such that it moved with the beam in order to scan various cross-section of the column.

The refractive index-matching yielded a transparent porous medium, free from any scattering and refraction at the solid-liquid interfaces, thus allowing direct optical probing at any point within the porous system. In these experiments, a neutrally buoyant dye was steadily introduced into the column and its concentration was imaged by sequentially scanning the concentration fields in vertical cross-sections. The video camera recorded fluorescence images at every vertical plane location while sweeping back and forth (with the illumination plane) across the column at every minute.

Figure 1. Experimental setup and measurement techniques.

The experimental runs were recorded through the video camera by a computer controlled VCR (Sony, EVO-9650) in Hi-8 mm format. The VCR was a frame accurate model that produced high quality still images of specified frames. It was computer controlled through an RS-232 interface for automated concentration analysis. In order to capture the recorded images for data analysis, a frame grabber board (ITI PC Vision Plus) with 640 by 480 pixel resolution was used in conjunction with an IBM compatible 486-33 computer. The image analysis was done using the OPTIMAS (BioScan) software. Several detailed programs were developed as interface softwares for automated experimental run and analysis softwares for velocity, concentration, and porosity measurements. In order to evaluate the uncertainty in the measurements a test run was performed. The uncertainty in the values of concentration was about 5% at 95% confidence level for 200 frames analyzed (see Rashidi and Banerjee²¹, for detail uncertainty analysis of particle tracking techniques).

3. RESULTS AND ANALYSIS

In our laboratories at LLNL, we have developed novel nonintrusive imaging techniques to observe pore-scale behavior at high resolution and high accuracy.^{17, 18} As part of this research, we have applied these techniques to study contaminant transport within nonaqueous and aqueous porous systems. This paper concentrates mainly on some of the results in the nonaqueous homogeneous porous system.

The experimental approach involves measurement and analysis of the interstitial velocity fields, concentration fields, and pore geometry within a refractive index-matched transparent porous system. As seen from Figure 1, the system consists of a porous column packed with natural mineral particles (for aqueous experiments) or plastic spherical beads (for nonaqueous experiments) and a refractive index-matched fluid. By illuminating the flow within the column with a planar sheet of laser light, details of flow and transport through the porous regions is observed microscopically, and qualitative and quantitative in-pore transport information is obtained at a good resolution and accuracy. This information is accumulated over a three dimensional volume within the column by scanning the illumination plane across that region. Experimental results have proved our capability in successfully measuring various geometric, flow, and concentration quantities within the porous system. These include local and volumetrically averaged porosities, velocity and concentration fields, microscopic and volumetrically averaged dispersive fluxes, and estimates of the dispersivity and the dispersion coefficient. Rashidi et al.^{19, 20} provide the complete results and the analysis of the experimental measurements.

Experiments were carried out, in which fluid was seeded with microspheres for velocity measurements and a dye front was injected into the porous system for concentration measurements. Microscopic measurements of flow, contaminant concentration, and pore geometry were collected via the camera as the column was scanned with the laser sheet. Figure 2 shows the typical scanned concentration slices in the middle of the column. As seen here, the column is getting saturated with the organic dye (contaminant) as the experiment continues. Figure 3 shows the typical overlaid slices of contaminant velocity and concentration within the porous column. This figure shows the detailed microscopic measurements that can be obtained with our technique in a porous medium. Our new experimental setup has the capability of collecting up to 360,000 measurements at each given time.

These slices were later assembled together and averaged in order to obtain information on the breakthrough curve and dispersive fluxes (see Rashidi et al.¹⁹ for more detailed information on these results). Figure 4 shows the breakthrough and the dispersive flux curves averaged for the entire column. The dispersive fluxes describe the mechanical dispersion of the contaminants as they move through the medium. As seen from this figure, the dispersion of the contaminant peaks at about 28 min into the experimental run and becomes negligible as the column gets saturated.

In order to investigate the detail of the microscopic processes of contaminant flow, pore-scale values of velocity, concentration, and medium's geometry were plotted at several cross-sections within the porous medium. Figure 5 shows the distributions of concentration, velocity, and pore geometry at several

(a)

(b)

(c)

low concentration high

Figure 2. Typical scanned concentration slices in the column showing the regions of high and low contaminant concentrations at the pore-scale. (a) beginning; (b) mid; (c) late in the run.

Figure 3. Typical slice of velocity and concentration fields in the central region of the porous column.

Figure 4. Distributions of average dimensionless concentration- breakthrough curve (a) and average axial dispersive flux (b) as a function of time plotted for the entire porous column.

Figure 5. 3D distributions of (a) axial velocity deviations (mm/sec), (b) dimensionless concentration, and (c) pore geometry as a function of several cross sections within the porous medium. Here, flow direction is from bottom to top, velocity values are deviations from the average velocity, and concentrations are non-dimensionalized by saturated concentration values. The horizontal cross sections are representative of the beginning, middle and end sections of the test section from the bottom to the top and are at the same locations in all the plots.

corresponding cross-sections in the column. As seen here, the microscopic movement of contaminant can be observed and analyzed at a very high resolution. These plots show that the contaminant moves faster through the higher porosity regions. It is also observed that near the contaminant front the microscopic velocities and concentrations are highly correlated. In other words, the high velocity regions are seen to be correlated with the regions of high contaminant concentration and vice versa. (see Rashidi et al.²⁰ for detailed analysis of the flow and concentration correlations) These 3D microscopic plots provide an unprecedented insight to the problem of contaminant migration through natural porous media.

4. CONCLUSIONS

The transport of contaminant in porous media has been simulated experimentally in a laboratory-scale porous system via novel imaging techniques. The test section consists of a porous column packed with optically clear particles and a refractive index-matched fluid. The system is automated to image through the porous medium for collecting microscopic measurements of concentration, velocity, and porosity. Measurements were obtained in a full three-dimensional volume of the test section at a good accuracy and a high resolution. 3D plots of these measurements show the complex geometry of the porous medium. It is also seen that near the contaminant front there is a significant correlation between the flow and the contaminant concentration. These results show the importance of micro-scale observations and measurements in understanding of contaminant migration in soil or other natural porous media. These experimental tools are extremely valuable when dealing with bioremediation problems where microscopic particle-contaminant-bacterium interactions are the key to understanding and optimization of these processes.

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6. REFERENCES

1. Schwartz, C.E., and J.M. Smith, "Flow distribution in packed beds," *Ind. Eng. Chem.* **45**, 1209-1218 (1953).
2. Harleman, D.R.E., and R.R. Rumer, "Longitudinal and lateral dispersion in an isotopic porous medium," *J. Fluid Mech.* **16**, 385-394 (1963).
3. Han, N.W., J. Bhakta, and R.G. Carbonell, "Longitudinal and lateral dispersion in packed beds: Effect of column length and particle size distribution," *AIChE J.* **31**, 277-288 (1985).
4. Bories, S.A., M.C. Charrier-Mojtabi, D. Houi, P.G. Raynaud, "Noninvasive measurement techniques in porous media," *Convective Heat and Mass Transfer in Porous Media*, S. Kakac et al. (eds.), Kluwer Academic Publishers, Netherlands (1991).
5. Wan, J., and J.L. Wilson, "Visualization of the role of the gas-water interface on the fate and transport of colloids in porous media," *Water Resour. Res.* **30**, 11-23 (1994).
6. Whitaker, S., "Diffusion and dispersion in porous media," *AIChE J.* **13**, 420-429 (1967).
7. Whitaker, S., "Advances in theory of fluid motion in porous media," *Ind. Eng. Chem.* **61**, 14-28 (1969).

8. Whitaker, S., "The transport equations for multiphase systems," *Chem. Eng. Sci.* **28**, 139-147 (1973).
9. Whitaker, S., "Flow in porous media I: A theoretical derivation of Darcy's law," *Transport in Porous Media* **1**, 3-25 (1986).
10. Slattery, J.C., "Flow of viscoelastic fluids through porous media," *AIChE J.* **13**, 1066-1071 (1967).
11. Slattery, J.C., *Momentum, Energy, and Mass Transfer in Continua*, McGraw-Hill, New York, NY (1972).
12. Bear, J. *Dynamics of Fluids in Porous Media*, Elsevier, New York, NY, (1972).
13. Garg, S.K., and J.W. Pritchett, "On pressure-work, viscous dissipation and the energy balance relation for geothermal reservoirs," *Advances in Water Resources* **1**, 41-47 (1977).
14. Gray, W.G., "A derivation of the equations for multiphase transport," *Chem. Eng. Sci.* **30**, 229-233 (1975).
15. Hassanizadeh, S. M., and W. G. Gray, "General conservation equations for multi-phase systems: Averaging procedures," *Advances in Water Resources* **2**, 131-144 (1979).
16. Tompson, A. F. B., and W. G. Gray, "A second-order approach for the modeling of dispersive transport in porous media, 1. Theoretical development," *Water Resour. Res.* **22**(5), 591-600 (1986a).
17. Rashidi, M. and F. Milanovich, "3D Imaging of Chemical Transport in Porous Media," *AIChE National Meeting (PTF)*, Denver, CO, August 14-19 (1994).
18. Peurrung, L., M. Rashidi, and T. Kulp, "Measurement of porous medium velocity fields and their volumetric averaging characteristics using particle tracking velocimetry," *Chem. Eng. Sci.* , accepted (1995).
19. Rashidi, M., L. Peurrung, A. Tompson, and T. Kulp, "Experimental Analysis of Pore-Scale Flow and Transport in Porous Media," *Advances in Water Resources*, in review (1995a).
20. Rashidi, M., A. Tompson, T. Kulp, and L. Peurrung, "Microscopic Measurements of Chemical Flow and Transport in Porous Media," *ASME J. Fluids Eng.*, in review (1995b).
21. Rashidi, M., and S. Banerjee, "Turbulence structure in free surface channel flows," *Phys. Fluids* **31**, 2491-2503 (1988).