Microscale Transport in Porous Media

Mehdi Rashidi
Lawrence Livermore National Laboratory
&
Robert Rinker
Chemical Engineering Department
University of California, Santa Barbara

This paper was prepared for submittal to
The proceedings of the 5th World Congress of Chemical Engineering
San Diego, CA - July 14-18, 1996

April 1996

This is a preprint of a paper intended for publication in a journal or proceedings. Since changes may be made before publication, this preprint is made available with the understanding that it will not be cited or reproduced without the permission of the author.
Microscale Transport in Porous Media

Mehdi Rashidi
Lawrence Livermore National Laboratory
University of California

April 1996

This paper was prepared for submittal to:
5th World Congress of Chemical Engineering
San Diego, CA
July 14-18, 1996
DISCLAIMER

This report was prepared as an account of work sponsored by an agency of the United States Government. Neither the United States Government nor any agency thereof, nor any of their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by trade name, trademark, manufacturer, or otherwise does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government or any agency thereof. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government or any agency thereof.
DISCLAIMER

Portions of this document may be illegible in electronic image products. Images are produced from the best available original document.
**Microscale Transport in Porous Media**

Mehdi Rashidi  
Lawrence Livermore National Laboratory  
University of California, Livermore, CA 94550  

Robert Rinker  
Chemical Engineering Department  
University of California, Santa Barbara, CA 93106

**ABSTRACT:**

In-pore transport processes in homogeneous and heterogeneous porous media have been investigated using novel 3D imaging techniques. The experimental system consists of a clear column packed with clear particles and a refractive index-matched fluid seeded with fluorescent tracers and an organic solute dye. By illuminating the porous regions within the column with a planar sheet of laser beam, flow and transport processes through the porous medium can be observed microscopically, and qualitative and quantitative in-pore transport information can be obtained at a good resolution and a high accuracy. Fluorescent images are captured and recorded at every vertical plane location while sweeping back and forth across the test section. These digitized transport images are then analyzed and accumulated over a 3D volume within the column. This paper reports on pore-scale observations of velocity, chemical concentration, and fluxes.

**INTRODUCTION:**

Traditionally, there have been three different approaches to the modeling of flow and transport in porous media. The first approach is a statistical method treating the porous media as a random structure. The second is a geometrical approach assuming a similar geometry to the porous media under study in order to solve the conservation equations. The third approach (continuum) is a volume averaging procedure that allows one to describe porous medium transport in terms of macroscopic rather than microscopic scale variables. All three approaches result in undetermined parameters that must be evaluated from experiments. However, the third approach (volume averaging method), if done correctly, provides the most general macroscopic conservation equations for transport in porous media. The volume averaging or continuum approach is the basis for the most recent theoretical investigations. Here, the general microscopic equation of transport in porous media

\[
\frac{\partial \rho \psi}{\partial t} + \nabla \cdot (\rho \psi \mathbf{v}) - \nabla \cdot \mathbf{J} - \rho \mathbf{f} - \rho \mathbf{G} = 0
\]

where,

- \( \rho \) is the mass density function,
- \( \psi \) is a typical intrinsic thermo-mechanical property,
- \( \mathbf{v} \) is a velocity vector,
- \( \mathbf{J} \) is a flux term,
- \( \mathbf{f} \) is an external supply term, and
- \( \mathbf{G} \) is the net rate of production.

is volume averaged over a representative elementary volume (REV) of the medium. As shown in Figure 1, the intricate structure of pore networks and grain boundaries are replaced with an effective continuum and an averaged set of variables and balance equations. These equations are derived from their pore-scale counterparts by the method of volume averaging. The works of Whitaker [1], Slattery [2], Bear [3], Hassanizadeh and Gray [4], Gray et al. [5], and Rashidi et al. [6] provide some insights of the current approach in this field. While each model presents a slightly different point of view, all require some assumptions about a specific medium behavior that must be confirmed and parameterized by detailed experimental work.

Previous experimental investigations (Schwartz and Smith [7], Harleman and Rumer [8], Hassinger and Von Rosenberg [9], Klotz et al. [10], and Han et al. [11]) have provided some valuable information...
on the macroscopic behavior of the flow and transport in porous media. Even though, many bulk transport phenomena result from the flow behavior at the microscopic spatial scale, until recently, little work has been done on the microscopic characterization of processes at the pore-scale. This can be attributed to the experimental difficulty of nonintrusively measuring flow and transport at high resolutions within the solid matrix.

Recent experimental improvements have allowed some investigations of pore-scale processes. These include studies using certain forms of noninvasive optical techniques (PIV, MRI) in and above packed beds for velocity and porosity measurements (Stephenson and Stewart [12], Bories et al. [13], Saleh et al. [14], Li et al. [15], Derbyshire et al. [16], and Shattuck et al. [17]) and studies in surrogate media composed of two-dimensional etched glass or capillary network micromodels (Soll et al. [18] and Wan and Wilson [19]). The present study is part of an extensive research in our laboratories to understand the nature of the microscopic flow and transport processes within porous media using 3D imaging techniques. The overall objective is to use our results toward gaining an understanding of chemical flow and transport in a porous medium and, as a result, provide the basis for improved modeling and treatment of these processes.

**EXPERIMENTAL FACILITIES & MEASUREMENT TECHNIQUES:**

Experimental facilities and measurement technique are shown in Figure 2 (Rashidi et al. [6] & [20]). These facilities are equipped to two separate columns. One is a rectangular column for chemical transport and bioremediation studies in aqueous heterogeneous systems and the other is a cylindrical column for flow and transport investigations in nonaqueous homogeneous systems.

Experiments in the aqueous systems [21] 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 an aqueous system was 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. Some minerals were discovered that are 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 (NaF & LiF) that were made in the laboratory to our size specification for these experiments. Therefore, the column was filled with these natural mineral particles (LiF) 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 refractive indices of the particles and the solution were matched by adjusting the temperature and a small addition of some sugar (dissolved) to the aqueous fluid. The column was maintained at this temperature throughout all runs by being immersed in a circulating constant temperature bath. A series of experiments were performed 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 systems 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 0.31 cm. 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 cm^3/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 (3 or 6 μm 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. As shown in Figure 3, 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 ± 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. As seen from Figure 4, the refractive index-matching yields a transparent porous medium, free from any scattering and refraction at the solid-liquid interfaces, thus allowing direct optical probing at various vertical planes within the porous system.

In these experiments, first the velocity measurements were taken by sequentially scanning the flow field in vertical cross-sections. Velocity measurements were obtained by tracking the seeded microspheres (particle density = 10 particles/mm³) in the fluid. This provided velocity measurements of the longitudinal and one transverse velocity components at a large number of microscopic locations within the cylindrical test section. Then, a neutrally buoyant organic dye (molecular diffusivity, $D_m = 10^{-5}$ cm²/sec) was steadily introduced into the column and its concentration was imaged within the same segment of the column, where the velocity field was determined. The video camera recorded fluorescence images on a series of vertical planes while sweeping back and forth. This allowed interstitial chemical concentrations to be determined as a function of time in the same planes where velocity measurements were taken. Roughly ten seconds were required to sweep through all vertical planes within the column. Sequential sweeps were separated by thirty-second intervals. Measurements from each sweep were collected into aggregate blocks of data and used to approximate a snapshot of the system at a single point in time. Since the flow was slow enough and the plane to plane time delay was significantly shorter than the time between complete sweeps through the volume, concentration images of vertical planes in one sweep could be considered to be made simultaneously. Dye concentration measurements also allowed the pore geometry on each plane to be measured. These images were recorded through a high resolution video camera by a frame accurate VCR and then analyzed rigorously using a microcomputer. The experimental results presented in this article are mainly for the case of the nonaqueous system. In these experiments, the uncertainty in the values of axial velocity and concentration were about 3% and 5%, respectively, at 95% confidence level for 200 frames analyzed (see Rashidi and Banerjee [22], for detail uncertainty analysis of particle tracking techniques).

RESULTS AND ANALYSIS:

Microscopic measurements of flow, chemical concentration, and pore geometry were collected using the CCD camera as the column was scanned with the laser sheet. Figure 5 represent the typical scanned region of a slice showing the microscopic velocity, concentration, and pore geometry within the homogeneous porous column. This figure shows the detailed microscopic measurements that can be obtained with our imaging techniques in a porous medium. Our experimental setup has the capability of collecting more than 2 million measurements (120 x 120 x 140) at each given time. These microscopic measurements of flow, transport, and pore geometry are used to obtain a dynamic 3D insight on the transport processes in porous media.

The scanned slices were later assembled together and averaged in order to obtain information on the breakthrough curves and dispersive fluxes. The microscopic (deviatoric) flux, $j$, describes the mechanical dispersion of the chemicals as they move through the medium (see Rashidi et al. [6], for more detailed information on these results). The axial component of microscopic deviatoric fluxes are evaluated from the values of axial velocity deviations and concentration deviations:

$$j = v' c'$$

where, $v'$ and $c'$ are the deviation velocity and the deviation concentration, respectively (see Whitaker [1] and Rashidi et al. [6]). The macroscopic average of this flux is the traditional macroscopic dispersive flux. Figure 6 shows the axial component of the averaged dispersive flux curves measured at different time during the experiment for the entire axial length of the test section. As seen from this figure, on the average the dispersion of the chemical peaks at about 30 min into the experimental run and becomes negligible as the column gets saturated.

In order to investigate the detail of the microscopic processes of chemical flow, pore-scale values of velocity, concentration, and deviatoric flux were plotted for time of 30 minute at several corresponding cross-sections within the porous medium. Figure 7 shows the distributions of microscopic velocity, concentration, and flux for $t = 30$ minute at several corresponding cross-sections in the column. As seen here, the chemical transport can be visualized and analyzed at a very high resolution and accuracy. These plots show that the chemical zone moves faster through the higher porosity regions. It is also observed that near the chemical front the microscopic velocities and concentrations are highly correlated. In other words,
Figure 1. Volume averaging of the microscopic equations.

Figure 2. Experimental facilities.

Figure 3. Experimental setup and measurement techniques.

Figure 4. Refractive index matching between solid particles and the liquid. Left picture: not matched; Right picture: matched.

Figure 5. Representative regions of a scanned data slice.

Figure 6. Distribution of axial dispersive fluxes as a function of time plotted for various horizontal cross section in the column.
the high velocity regions are seen to be correlated with the regions of high chemical concentration and vice versa. (see Rashidi et al. [20], for detailed analysis of the velocity and concentration correlations). Furthermore, it is observed that there is no such a thing as uniform flow/transport front within the porous regions. Due to the complexity of pore structure, chemical transport becomes very complex phenomenon. As early as minute 30, one can observe saturated regions at the end of the test section while the average chemical front is half way in the test section. These 3D microscopic plots provide an unprecedented insight to the problem of contaminant migration through natural porous media. They help in formulating physically sound models for the complex problem of transport in porous media.
CONCLUSIONS:

Chemical transport in porous media has been studied experimentally in a laboratory-scale porous system via novel imaging techniques. The system is automated to image through the porous medium for collecting microscopic measurements of pore geometry, velocity, concentration, and fluxes. 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 chemical front there is a significant correlation between the flow and the chemical concentration. Furthermore, it is shown that the chemical front moves faster through the higher porosity regions overpassing the average flow front in some regions during the experiment. These results show the importance of micro-scale visualization and measurement in understanding the complex problem of transport in porous systems.

ACKNOWLEDGMENTS:

The authors would like to thank U. S. Department of Energy for funding of this project, under OHER contract # W-7405-ENG-48 and LDRD contract # 95-LW-026. The authors would also like to acknowledge Dr. F. Milanovich (LLNL), Dr. F. Hoffman (LLNL), Dr. A. Tompson (LLNL), and Professor S. Whitaker (UC Davis) for their valuable assistance in this research.

REFERENCES: