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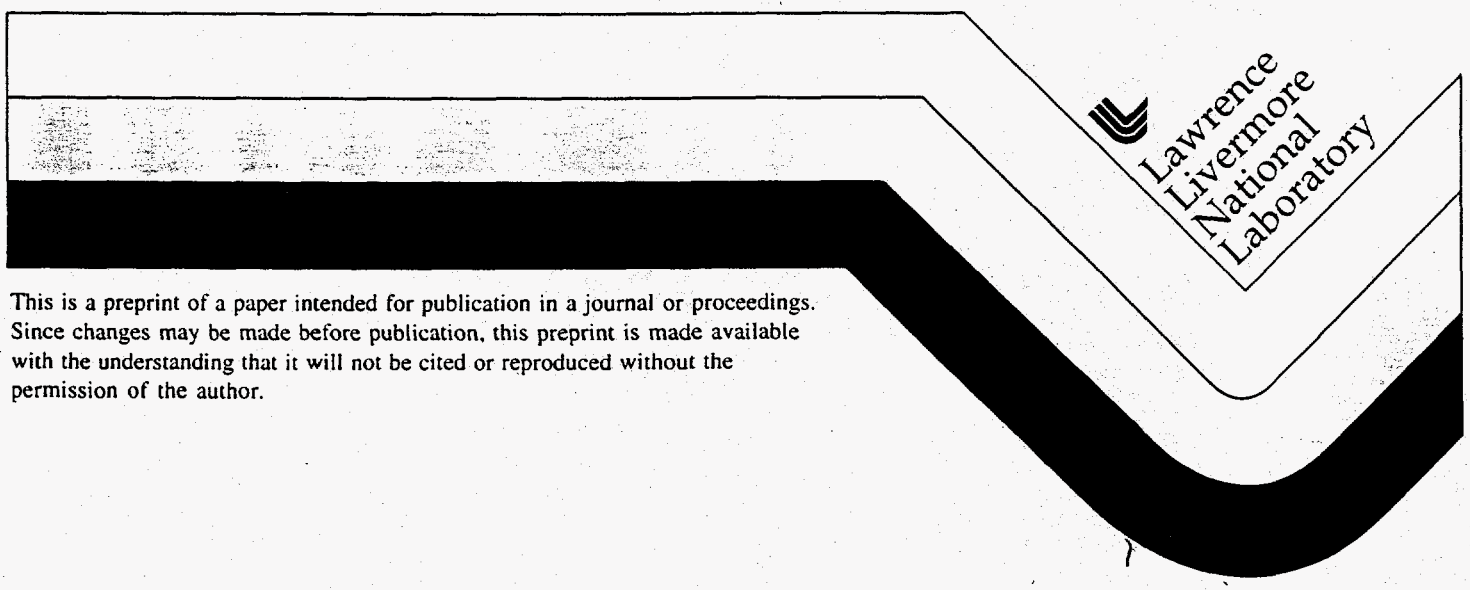

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SMALL SCALE FLOW PROCESSES IN AQUEOUS HETEROGENEOUS POROUS MEDIA

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ABSTRACT

Small scale flow processes in aqueous heterogeneous porous systems have been studied experimentally via novel nonintrusive fluorescence imaging techniques. The techniques involve 3D visualization and quantification of flow fields within a refractive index-matched transparent porous column. The refractive index-matching yields a transparent porous medium, free from any scattering and refraction at the solid-liquid interfaces, as a result allowing direct optical probing at any point within the porous system. By illuminating the porous regions within the column with a planar sheet of laser beam, flow 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 good accuracy. A CCD camera is used to record the fluorescent images at every vertical plane location while sweeping back and forth across the column. These digitized flow images are then analyzed and accumulated over a 3D volume within the column. Series of flow experiments in aqueous, refractive index-matched, porous systems packed with natural mineral particles have been performed successfully in our laboratories.

INTRODUCTION

Many bulk phenomena associated with transport in porous media result from the flow behavior of the system at the microscopic spatial scale. Recent experimental studies have provided some macroscopic results but limited microscopic data on the chemical flow and transport in porous media. Experiments of Schwartz and Smith (1953), Murphy (1967), and Musser (1971) in packed beds have shown the development of a peak in velocity profile very close to the wall. However, these results were not characterized conclusively as a function of the flow Reynolds number and the pore geometry. Flow visualization and transport studies of Joll and Hanratty (1966 and 1969) revealed some flow and mass transfer information near spherical particles. Joll and Hanratty (1966) examined the path line of flow through a packed bed of spherical particles using dye visualization. They observed a transition from laminar to turbulent flow at Reynolds numbers of 268 to 366 (based on pore velocity and particle diameter). Furthermore, they observed a smooth transition from steady to unsteady mass transfer at approximately the same Reynolds numbers.

Experimental studies of Harleman and Rumer (1963), Hassinger and Von Rosenberg (1968), Klotz et al. (1980), and Han et al. (1985) have provided some macroscopic estimates of longitudinal and lateral dispersion coefficients. Han et al. (1985) studied the effect of column length and particle size distribution on the two components of the dispersion coefficients. They showed that for the case of uniform size particles, the longitudinal dispersion coefficients were a strong function of axial position in the bed unless the dispersion length satisfied a constraint dependent on the value of the Peclet number. Furthermore, it was seen that for the case of the wide size particle distribution, longitudinal dispersion coefficients were larger (up to 3 times) than in the uniform size particle distribution case, and a longer dispersion length was required in order to obtain a constant dispersion coefficient. However, due to the limited amount of data, they could not provide a quantitative measure of the size of this length scale.

Laser anemometry and flow visualization study of Dybbs and Edwards (1984) for liquid flows in several porous structures has shown the existence of four flow regimes as a function of flow Reynolds number (based on particle

diameter and pore velocity). These include a Darcy flow regime (Re < 1), an inertial flow regime (1 < Re < 150), an unsteady laminar flow regime (150 < Re < 300), and a turbulent flow regime (Re > 350). Their results also showed an increase in velocity profile in about one to two particle diameters from the wall similar to the findings of Schwartz and Smith (1953).

Although these experimental efforts have provided some valuable information on the macroscopic behavior of the flow and transport in porous media, 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 renewed investigation of pore-scale processes. These include studies using certain forms of noninvasive imaging techniques (PIV, MRI) in and above packed beds for velocity and porosity measurements (Stephenson and Stewart 1986, Bories et al. 1991, Shattuck et al. 1991, Saleh et al. 1993, Li et al. 1994, and Derbyshire et al. 1994) and studies in surrogate media composed of 2D etched glass or capillary network micromodels with known pore geometries (Soll et al. 1993, Soll and Celia 1993, and Wan and Wilson 1994.)

Theoretical descriptions of flow and transport in porous media have been generally derived from simpler "bulk" equations of mass and momentum balance or from more systematic approaches in which pore-scale behavior is rigorously averaged over representative elementary volume (REV) of the medium. The works of Whitaker (1967), Slattery (1967, 1972), Bear (1972, 1979), Gray (1975), Hassanizadeh and Gray (1983), and Gray et al. (1993) are representative 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 microscopic experimental work.

The present work is part of an extensive research in our laboratories to understand the nature of the microscopic flow and transport processes within porous media. A novel nonintrusive imaging approach has been used to observe the pore-scale flow and transport behavior at a high resolution and a high accuracy. Previously, we applied these techniques to study flow and solute transport within a non-aqueous homogeneous porous system (Rashidi et al. 1996a and Rashidi et al. 1996b). This paper reports on the successful application of our experimental imaging techniques to flow and transport studies in aqueous heterogeneous porous systems. Furthermore, it provides some results on 3D flow experiments performed in our unique refractive index-matched aqueous systems with natural packing minerals of similar shape and sizes as to soil particles. The overall objective is to use these findings toward gaining a physical understanding of small scale chemical/contaminant flow and transport in heterogeneous porous systems and, as a result, provide the basis for improved modeling of these flow processes.

EXPERIMENTAL FACILITIES & TECHNIQUES

Figure 1 shows a picture of the experimental facilities and measurement techniques (Rashidi et al. 1996a & 1996b). 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. Some natural 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 and LiF) that were made in the laboratory to the 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 refractive index of the aqueous solution was matched to the refractive index of the particles by adjusting the temperature and small addition of dissolved sugar (sucrose) to water. The refractive index matching was critical up to third digit after decimal point for the refractive indices of the packing particles and the aqueous solution. 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, $\sim 10^{-3}$ to 1 (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 3 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. 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 (see Figure 2).

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/mm^3) in the

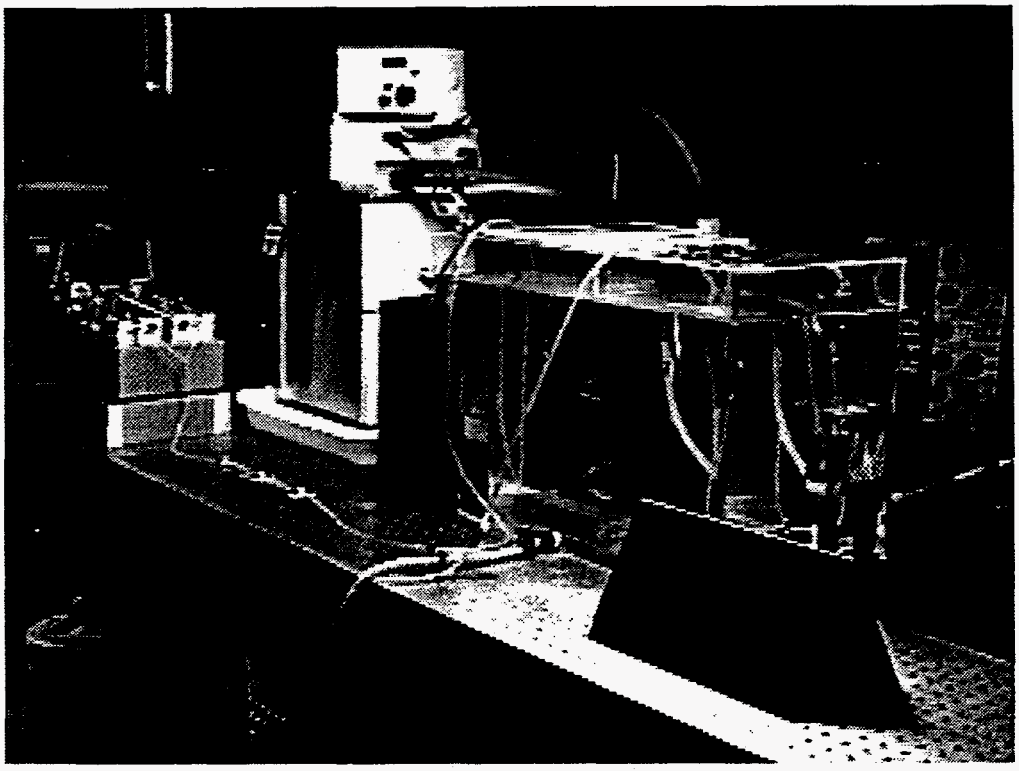


Figure 1. Experimental facilities.

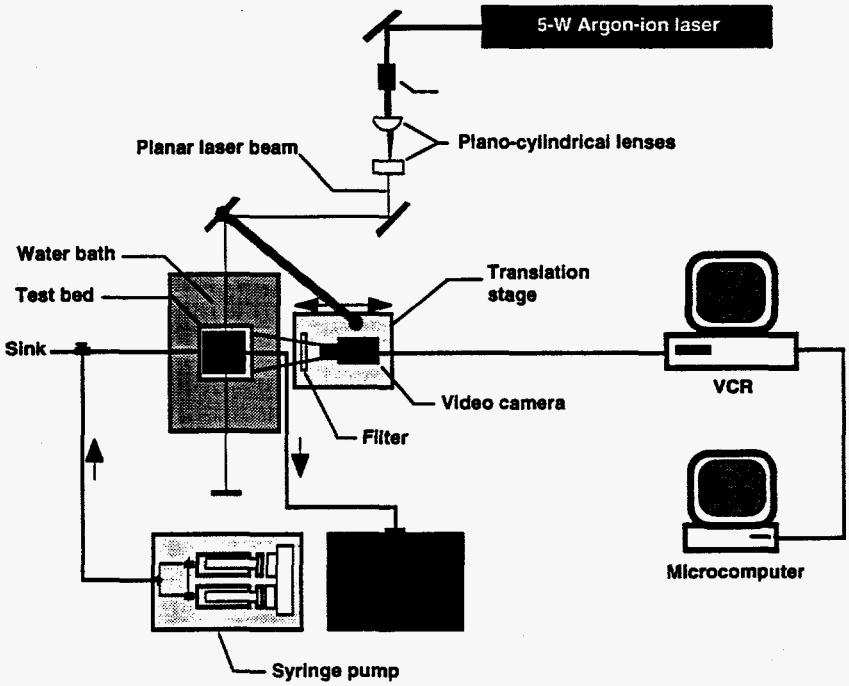


Figure 2. Experimental setup and measurement techniques.

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, $D_m \approx 10^{-5}$ cm^2/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 (with the illumination plane) across the column once every minute. 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 60 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 an IBM compatible computer.

A test run was performed to evaluate measurement uncertainties. The uncertainty in the values of average axial velocity was about 2% at 95% confidence level for 200 frames analyzed (see Rashidi and Banerjee 1988, for details on uncertainty analysis of particle tracking techniques). Similarly, the concentration and porosity measurements revealed an uncertainty estimate of about 2% at 95% confidence level. Rashidi (1996) provides more details on the image analysis techniques. In particular, the three velocimetry techniques of Particle Imaging Velocimetry (PIV), Particle Streak Velocimetry (PSV), and Particle Tracking Velocimetry (PTV) have been compared in details (see Figure 3). It is shown that because of the desire for rapid, automated, and dynamic data collection, the present technique (PTV) is more advantageous in studying the flow and transport in porous media. The present technique also provides an improvement in measurement uncertainties and resolution over our previous experiments.

RESULTS AND ANALYSIS

The experimental runs were conducted in the described facilities. The details of the experimental conditions are summarized in Table I. Experiments were performed at seven different Reynolds (and Peclet) numbers with the Reynolds numbers being based on the average packing particle diameter and pore velocity. As a result, a wide range of flow conditions were studied in these experiments.

As previously described, the camera and laser illumination sheet were placed on a platform that moved back and forth at a controlled speed for scanning the column. Microscopic values of velocity, concentration, and pore-space geometry were obtained at a 80 by 60 cell resolution per slice. During each experimental run, 288,000 individual measurement points were collected each minute. The given resolution is based on a measurement grid spacing of (0.5 x 0.5 x 0.5) mm^3 In our experimental techniques, this resolution can be easily increased to (0.25 x 0.25 x 0.25) mm^3 which in turn increases our measurement capability to more than 2 million measurement points per given time. This results in an enormous amount of data within our porous medium.

Table I. Experimental conditions.

Run	T (°C)	Average Pore Velocity $\bar{v}$ (cm/s)	Average Particle Diamter d_p(cm)	Reynolds Number Re ($\bar{v}\, d_p/\nu$)	Peclet Number Pe ($\bar{v}\, d_p/D_m$)
1	20.0	0.00106	0.15	0.0042	4.8
2	20.0	0.0027	0.15	0.010	12
3	20.0	0.0053	0.15	0.021	24
4	20.0	0.0106	0.15	0.042	48
5	20.0	0.027	0.15	0.10	121
6	20.0	0.06	0.15	0.20	251
7	20.0	0.11	0.15	0.40	482

Figure 4 shows some of the vertical slices that are scanned during the experiments. As seen here, these images show the detail heterogeneity and structural complexity of the porous medium. The matching of fluid and particle refractive indices has allowed us to probe into various regions of our porous system to gather detail microscale flow and transport information. Figure 5 shows a comparison of several runs at various corresponding time during the experiments. As expected, the slower runs get saturated at a longer time than the runs with higher Reynolds numbers. In particular, column saturation happens in less than 11 minutes for Run 7 as opposed to about 11 hours for Run 1. These images also show the porosity distribution at the middle of our porous column. It is interesting to note that at lower flow rates flow does not follow the same path in these experiments and is dependent on Reynolds numbers.

The present experiments not only shed light on the general modeling of flow processes in porous media but also helps us in extending our unique aqueous experimental capabilities in studying such important problems of bacterial transport, filtration, surface deposition, bioremediation, contaminant retardation, and contaminant-bacterium-particle interactions in natural porous systems.

CONCLUSIONS

Pore-scale flow processes have been studied experimentally in an aqueous heterogeneous porous column using unique imaging techniques. Experiments were performed at seven Reynolds (and Peclet) numbers covering a wide range of flow conditions. Microscopic values of velocity, concentration, and pore geometry have been obtained at a measurement resolution of more than 280,000 measurement points per given time. These experiments provide dynamic new tools for qualitative and quantitative analysis of flow processes in

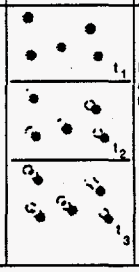
Technique	Imaging method	Image	Advantages	Disadvantages
Particle Imaging Velocimetry (PIV)	Double exposure; velocity calculated by correlating particle displacements		Large particle density possible; good for fast-moving particles	Poor dynamic range; poor velocity resolution with video
Particle Streak Velocimetry (PSV)	Long exposure, pulses at both ends to show particle stayed in plane; velocity = length/time		Good dynamic range, fair velocity resolution; valid streaks easily identified visually	Low particle density necessary; tracks which leave plane discarded; difficult to automate valid streak recognition
Particle Tracking Velocimetry (PTV)	Video recording of particle motion; velocity calculated by tracking each particle from frame to frame and recording position history		Better dynamic range *and resolution*; easy to automate	Velocities limited to *several* video pixels per frame

Figure 3. Comparison of three approaches in particle velocimetry techniques.

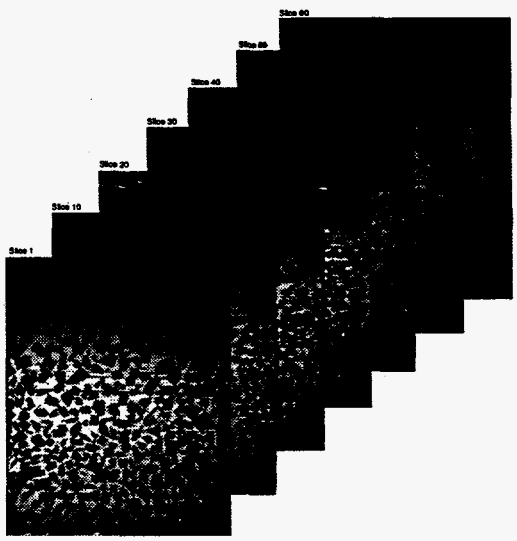


Figure 4. Typical imaged slices within the porous column.

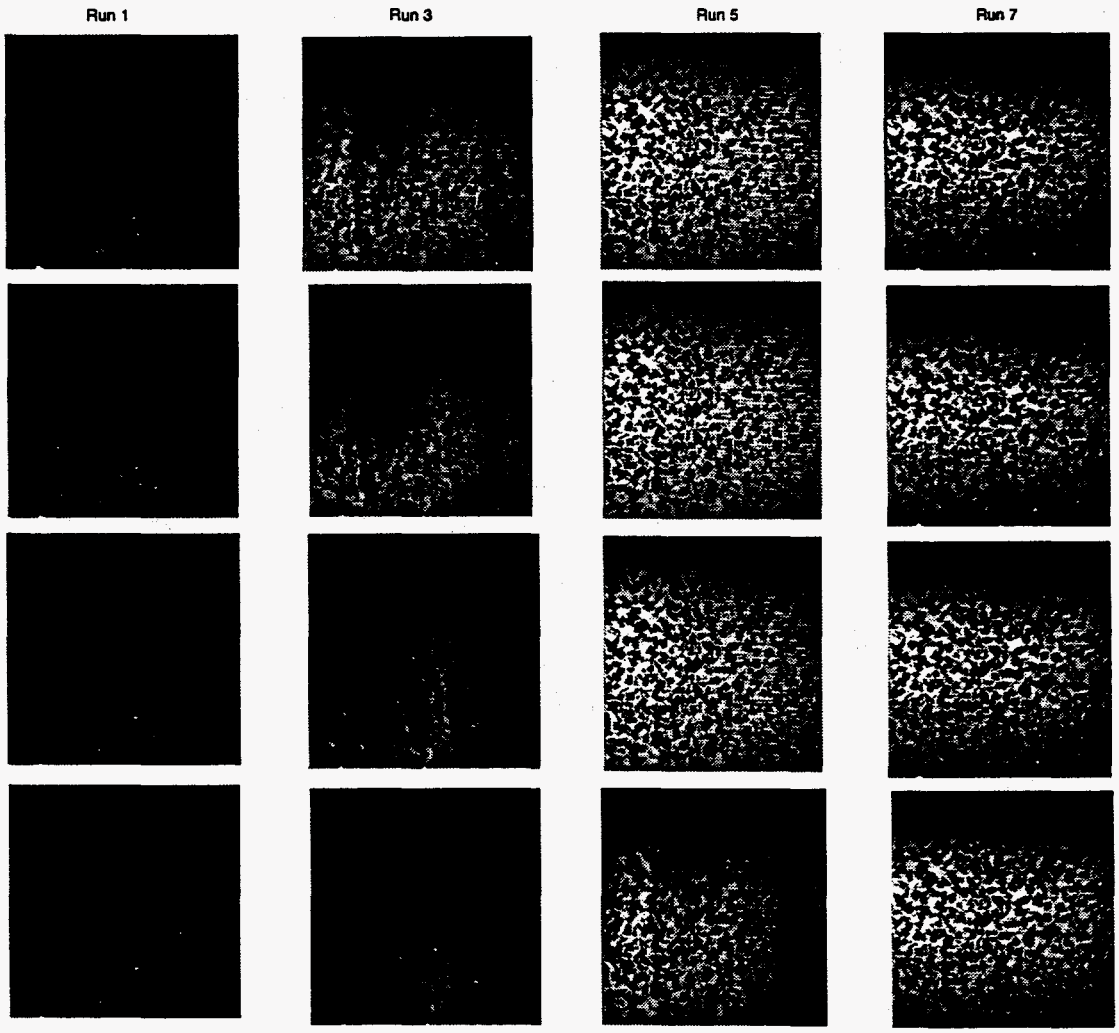


Figure 5. Comparison of small scale flow processes in the central region (slice 30) of our aqueous heterogeneous porous system. Time = 2, 5, 8, and 11 minute from bottom to top for each run.

the aqueous heterogeneous porous media and as a result, improve our understanding of theoretical modeling and natural treatment (bioremediation) of these processes.

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