

Gravity-destabilized nonwetting phase invasion in macro-heterogeneous porous media: Experimental Observations of Invasion Dynamics and Scale Analysis

Robert J. Glass, Stephen H. Conrad, and William Peplinski

Flow Visualization and Processes Laboratory

Sandia National Laboratories, Albuquerque, NM 87185 USA

January 17, 2000

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Abstract

We designed and conducted experiments in a heterogeneous sand pack where gravity-destabilized nonwetting phase invasion (CO_2 and TCE) could be recorded using high resolution light transmission methods. The heterogeneity structure was designed to be reminiscent of fluvial channel lag cut-and-fill architecture and contain a series of capillary barriers. As invasion progressed, nonwetting phase structure developed a series of fingers and pools; behind the growing front we found nonwetting phase saturation to pulsate in certain regions when viscous forces were low. Through a scale analysis, we derive a series of length scales that describe finger diameter, pool height and width, and regions where pulsation occurs within a heterogeneous porous medium. In all cases, we find that the intrinsic pore scale nature of the invasion process and resulting structure must be incorporated into our analysis to explain experimental results. We propose a simple macro-scale structural growth model that assembles length scales for sub-structures to delineate nonwetting phase migration from a source into a heterogeneous domain. For such a model applied at the field scale for DNAPL migration, we expect capillary and gravity forces within the complex subsurface lithology to play the primary roles with viscous forces forming a perturbation on the inviscid phase structure.

1. Introduction

Spilled solvents have created pervasive groundwater contamination problems because of their ubiquitous use, their toxicity and persistence in the environment, and the difficulty of recovering them from the subsurface. Because organic solvents are more dense than water and immiscible with water, they are commonly referred to as DNAPLs (dense non-aqueous phase liquids). They migrate below the water table downward and laterally under the influence of gravity, capillary and viscous forces. Because DNAPLs are generally the nonwetting fluid, variations in media texture that the DNAPLs encounter as they migrate can have a profound influence on the migration path. This capillary control induced by textural heterogeneities complicates the migration of the DNAPLs and makes it difficult to predict the locations in the aquifer where the spilled DNAPLs may ultimately reside. Uncertainties in the region of solvent contamination translate into higher remediation costs because the remedial system must be designed in light of these uncertainties.

Several experiments have shown the importance of heterogeneous texture in affecting the migration of DNAPLs in two phase systems in laboratory sand packs [Kueper et al., 1989; Illangasekare et al., 1995, 1996] and in micromodels [Conrad et al., 1989, Wilson et al., 1990]. Similarly, 2D, heterogeneous, glass bead pack experiments have been performed for the upward migration of a gas [Ji et al., 1993], also the nonwetting fluid. Additionally, two field experiments of DNAPL migration below the water table have been conducted at the Borden site [Kueper et al., 1993; Brewster et al., 1995]. Not surprisingly, all these studies have produced irregular spatial distributions of the nonwetting phase. Experimental and modeling studies of DNAPL dissolution processes in heterogeneous systems [e.g.,

Anderson et al., 1992; Powers et al., 1998] have shown that this irregular distribution of DNAPLs has significant impacts on the rate of dissolution. Of course, it also has implications for remedial strategies that attempt to improve pump-and-treat by enhancing dissolution, e.g., surfactants or co-solvents, or oxidizing the DNAPL in situ, e.g., potassium permanganate.

The complicated pattern and dependence on small scale detail places nonwetting invasion in a separate class from wetting invasion where combinations of film flow and multiple neck pore filling facilitation create macroscopic fronts [e.g., Lenormand and Zarcone, 1985; Blunt and Scher, 1995; Glass and Yarrington, 1996]. Modeling nonwetting displacements with current two-phase continuum-scale porous media approaches that integrate over a large number of pores in an effort to define effective behavior at an REV scale must be considered as questionable for many situations. Alternative modeling approaches that better embody the underlying physics make use of modified forms of invasion percolation (MIP). While forms of MIP have been applied in the past to yield phenomenological understanding in small random pore networks, our desire is to develop such models to be predictive of system behavior at much larger scales [e.g., Kueper and McWhorter, 1992; Glass et al., 1995; Ioannidis et al., 1996; Ewing and Berkowitz, 1998]. While still in their infancy, such approaches show great promise. In order to develop these models as well as to test current porous continuum approaches, experiments are required that capture invasion dynamics within large heterogeneous pore networks under the combined influence of capillary, gravity, and viscous forces.

In this paper we present and analyze three experiments designed to quantitatively visualize the evolving invasion structure during gravity destabilized, nonwetting phase invasion within macro-heterogeneous porous media. We built our media to create a series of capillary barriers and evoke lithology reminiscent of fluvial channel lag cut-and-fill architecture within translucent sands. Using light transmission techniques, we collected invasion data rapidly at near pore scale resolution. This data allows us to track the invasion front with exceptional clarity and analyze the unsteady dynamics of the phase invasion process both during initial invasion and upon redistribution. Our experiments demonstrate a characteristic alternation between gravity stabilized and destabilized invasion growth controlled by the sequential capillary barriers within the heterogeneous field. Under gravity destabilized growth periods, pore scale fingering is organized away from the source, while under gravity stabilized periods, invasion coalesces into macroscopic horizontal fronts that grow backwards, overwriting the original finger structure. Behind the invasion front, we find rich dynamics with multiple cycle pulsation before, within, and beyond capillary barriers and along the path of fingers, yielding highly non-monotonic saturation behavior in time and space.

Through a scale analysis using local force balances, we derive a series of length scales that control invasion structure and dynamics such as finger diameter, the height of pools above capillary barriers, and the location of zones of nonwetting fluid pulsation. In all cases, we find that the intrinsic pore scale nature of the invasion process and resulting structure must be incorporated into our analysis to explain experimental results. Analysis also demonstrates that while global viscous forces are small, the pore scale nature of capillary barrier breaching and fingering can cause them to be locally significant and on the order of

capillary and gravity forces. Such results suggest significant difficulties in the application of standard porous continuum approaches. We propose an alternative modeling approach that assembles these length scales within the context of a macro-scale 'structural' growth model akin to upscaled MIP. Such a model yields directly the nonwetting phase migration pathway evolution, its geometry of fingers and pools, and the locations of zones of pulsation within complex subsurface lithologic heterogeneity. Inadequacies of continuum scale models can thus be avoided and the structural growth model combined easily with high resolution Monte Carlo simulation of the controlling subsurface geology to yield probabilistic prediction of subsurface DNAPL contamination.

2. Experimental Design

We designed experiments within a thin but extensive sand chamber where we could use quantitative light transmission techniques to record invasion dynamics in time. A reproducible heterogeneity structure was placed within the chamber and three constant flux, gravity destabilized, nonwetting phase invasion experiments were conducted where we varied the interfacial tension, density difference, and viscosity using two fluid/fluid systems: CO₂/water and TCE/water.

2.1. Chamber Construction

The chamber was constructed of two plates of hydrophilic glass (1.25 cm thick) held apart by two aluminum spacers (1 cm thick) that ran down the long edges of the glass. The internal dimensions of the chamber were 1 cm thick (~10-40 pores/grains) by 26 cm wide by 60 cm tall. O-rings embedded in the edge spacers provided a seal with forces applied by an external clamp that ran the length of the chamber sides. At the top and bottom, a screened manifold and a porous plate, respectively, were sealed to the chamber allowing for the inflow and outflow of water. Steel bars were clamped to the outside of the glass plates at the top and bottom to minimize chamber bowing under the pressures created by filling the chamber with sand and fluids. A port for injecting the nonwetting phase was laterally centered in the front glass plate, within 3 cm of one edge.

2.2. Sand Packing

The chamber was filled using a sand filler designed to create reproducible 'facies-like' geometric structures. A computer controlled positioner was mounted on a platform, which could be raised and lowered manually. Attached to the positioner was a sand hopper, funnel, tube, and vibration source that delivered sand to the chamber at specified locations. Horizontal position, the falling distance from the end of the delivery tube to the sand already in the chamber, and the mass of the particular sand delivered, was controlled. Features intended as analogues to channel cut-and-fill facies were created by keeping the positioner stationary while sand was being added to the chamber. Continuous layers were created by programming the positioner to travel back and forth across the chamber while delivering sand at a uniform rate.

The heterogeneity structure, shown in **Figure 1**, was designed to represent a qualitatively realistic sequence of three lithologic units of different texture. Three commercially available, translucent, hydrophilic silica sands with narrow, non-overlapping grain size distributions were chosen to comprise the three units (Accusand 12-20, a coarse sand; 30-40, a medium sand; and 50-70, a fine sand; see **Table 1**). We created unit shapes reminiscent of those seen in channel cut and fill deposits but with vertical exaggeration. Fine sand emplaced along the edges of the chamber removed possible preferential flow of the nonwetting phase along the chamber boundaries.

We also packed an additional chamber where we maintained the continuity of the thin layer of fine sand stretching across the chamber about 2/3rds of the way toward the outlet from

the injection port (see **Figure 1**). We refer to this pack as the secondary hetero-structure while the other we refer to as the primary hetero-structure. We used this small, yet critical, difference between the primary and secondary hetero-structures to demonstrate the sensitivity of TCE migration behavior to the lateral continuity in capillary barriers.

In each of the experiments, the chamber was oriented so that the invading nonwetting phase was introduced in a density-unstable manner. That is, TCE (more dense than water) was introduced from the top, while CO₂ (less dense than water) was introduced from the bottom. For the sake of comparison, both nonwetting fluids advanced in the same direction through the heterogeneous structure. That is, TCE advanced downward through the structure just as shown in **Figure 1**, while CO₂ advanced upward, with the structure oriented upside-down to that shown in **Figure 1**.

After filling and orienting the chamber, we settled the sand with capillary forces and localized vibration. Beginning with an air-dry sand-filled chamber, we displaced air with CO₂ through the bottom manifold. Gas flooding was followed by gravity and viscous stabilized flooding with degassed, de-ionized water until the chamber was saturated. We then drained the chamber through the bottom porous plate under suction, air entering from above. We continued pulling filtered dry air through the chamber and porous plate using a vacuum pump until the sand in the chamber was dry. The front of the chamber was then tapped with a hard plastic cylinder (a screwdriver handle) to enhance further settling with localized vibration. This CO₂/water/drainage/drying/tapping cycle was repeated several times causing the heterogeneous packed sand structure to settle. Digital images of

transmitted light through the chamber showed that the vast majority of the settling was achieved after two cycles.

Close comparison of the settled primary and secondary hetero-structures shows small variations in shape and lens interface orientation between the two packs. Additionally, because the chamber was disassembled between experiments for cleaning, re-assembly slightly changed the distance between the two glass plates thus causing the entire hetero-structure to 'scale' slightly in the vertical direction.

2.3. Material Properties

Pressure/saturation curves for each of the three sands were measured within the chamber for the air water fluid system using the light transmission method. The chamber was filled with a single sand type following the homogeneous filling method of Glass et al. [1989b]. A sequence of filling and drainage experiments to equilibrium points yielded primary wetting and drainage curves for the media [e.g., Norton, 1995] (See Figure 2). Porosity and saturated permeability were also measured during this characterization sequence and are presented in **Table 1**.

Fluid properties are given in **Table 2**. Interfacial tension (IFT) measurements were made using a Fisher Model 20 surface tensiometer following the manufacturer modified ASTM D 971-91 method. **Fluids were equilibrated for two minutes before IFT was measured.** Fluid viscosity for the TCE was measured using a Cannon-Fenske viscometer

following procedures recommended by the manufacturer. TCE density was measured with a pycnometer. Because we added Oil-Red-O dye (0.9 g/L, Fisher Scientific) to the TCE in our experiments, we measured the properties of the dyed liquid. The presence of dye was found to decrease interfacial tension of the TCE.

2.4. Invasion Experiments

We conducted three invasion experiments (see Table 3). In our first experiment we injected CO₂ from the bottom into the water saturated primary hetero-structure. After conclusion of the experiment, the CO₂ was dissolved away with deaired water and the chamber inverted. We then performed our second experiment by injecting TCE from the top. After emptying, cleaning, and repacking the chamber with the secondary hetero structure, we then conducted our final experiment with TCE injected from the top. Each invasion experiment started from the fully water saturated state verified with the light transmission method. Outflow occurred from the side opposite injection and was connected to a tube directing the water to a reservoir on a scale with a drip point 1.5 cm above the top of the chamber. Prior to starting the CO₂ invasion experiment, we displaced the degassed, de-ionized water in the chamber with CO₂ saturated water. This minimized the dissolution of the free phase CO₂ into the pore water as CO₂ invasion progressed.

An 18-gage needle (0.84 mm ID) was used to inject the non-wetting fluid into the coarse sand. For the TCE experiments, liquid was injected using a syringe pump, while for the CO₂ experiment, gas was delivered to the chamber from a pressure bottle through a

metering valve and flow meter. Injection rates were designed to be ~1 ml/min. for all experiments (see Table 3) so that average viscous forces would be small. The mass of water leaving the chamber was measured every 30 seconds with a computer monitored electronic balance to yield the displaced volume as a function of time. For the TCE/water experiments, Oil-Red-O dye (0.9 g/L) was added to the TCE to enhance visibility. For the CO₂/water experiments, changes in fluid saturation were easily measurable without dyes due to the very different refractive indices between gas and water. Fluid injection was terminated when the leading edge of the nonwetting phase had reached the far side of the chamber. Redistribution of the nonwetting phase was monitored for the TCE experiments over a several day period.

2.5. Image collection and processing

The experimental chamber was placed between a controlled output diffuse light source and a digital camera that measures the intensity of light transmitted through the chamber as a function of space and time. Digital images were acquired at a rate of one image every 20 seconds (or ~1 image per 0.33 milliliter of injected fluid) with a 12-bit shuttered digital camera (1317x1035 pixels, 4096 gray levels) using a 60 mm lens and **~ 0.1 sec exposure time** to record intensity fields from a distance of 4.3 meters within a dark room. Camera resolution was ~0.5 mm per pixel (0.25 mm²/pixel).

We processed digital images to yield saturation fields as a function of space and time and then used these images to create 'composite' fields that summarize 1) the movement of the nonwetting invasion front in time, and 2) cyclic temporal variations in saturation behind the front. Preliminary to this, images were first preprocessed to adjust the intensity field for small fluctuations of the light source using a photographic constant-density-stepped-wedge (included in every image) and to re-align the image for occasional small shifts between the camera and the test cell (tolerance of 0.05 pixels).

Saturation fields were calculated using modifications of techniques presented by Tidwell and Glass [1994] for both refraction dominated and absorption dominated energy transmission within homogeneous media. For the CO₂/water experiments where light passage is dominated by refraction, we used their theoretical development to calculate point-wise the number of pores (their k) from the saturated and dry images and then used this 'k field' to calculate the transient saturation fields. For the TCE/water systems where the refractive index of the fluids are nearly the same, saturation was calculated using energy absorption by the Oil-Red-O dye analogous to their x-ray transmission. However, because a fully saturated Oil-Red-O dye image was not obtained, we substituted the low intensity dry image and scaled it to yield saturation values within pools of the coarse sand near the expected value of 0.83 (see Figure 2). This approach, while approximate for sands other than the coarse, maintained excellent spatial definition of the saturation field. Other inaccuracies were also noted in the experiments. Inspection of the saturation fields showed highly variable saturation when the nonwetting phase did not fully span the thickness of the sand, i.e., the saturation is not uniform from the front wall to the back wall of the chamber. Additionally, where high

nonwetting phase saturation regions were next to completely water saturated regions, the scatter of light caused blurring of the sharp transition. To estimate the gross error associated with our saturation fields, we compared volumes of injected nonwetting phase as calculated from saturation fields against the known injected volumes. Total nonwetting volume error was a maximum of 12% across the three experiments. While these gross errors can be reduced through additional calibration, error within the field where the nonwetting phase does not fully span the thickness of the pack and where high and low saturation regions are juxtaposed will remain. However, for the tracking of the invasion front and subsequent analysis of saturation dynamics, saturation errors are of secondary importance.

To track the progression of the advance of the nonwetting phase, we used the individual saturation images taken over time to create a composite order image that shows the advance of the invasion front. This order of invasion image is built by tracking the first nonwetting saturation increase **above system noise** at each point within the field. In building a composite order image, some pore scale detail is lost **as we ignore the degree of saturation**. However, using this method, we are able to define the extent of the invading phase with excellent precision as a function of time throughout an entire experiment in a single image.

To analyze the local monotonicity of saturation in time within the field, we compared sequential saturation images and counted the number of cycles from increasing to decreasing saturation **above system noise** at each pixel in time. A cycle image shows locations where saturations increase and decrease. Values in the cycle images of one and

above indicated that a location behaved non-monotonically as the experiment progressed.

We note that while cycle images give an indication of internal dynamics behind the initial invasion front, they weight early invasion regions higher than later invasion regions.

Additionally, they will not catch all cycles if the period of the pulsation is less than our 20 second acquisition interval between images.

3. General Invasion Results

Use of the light transmission technique allows us to observe, record and analyze the invasion process with unprecedented clarity. In this section we present general invasion results through a series of images that show 1) nonwetting phase structure at conclusion of fluid injection, 2) development of this structure in time, 3) phase saturation nonmonotonicity as this structure evolves, and 4) subsequent redistribution of the nonwetting phase. Note that all images are oriented such that flow is in the same direction to facilitate comparison. That is, the CO₂ invasion is rotated 180 degrees with gravity now acting upward in the image.

Figure 3 shows saturation structures when the CO₂ or TCE had just spanned the system and injection was terminated. We see that the nonwetting fluid is confined mainly to the coarse units and is composed of a number of 'pools' behind capillary barriers (CBs) and 'fingers' that penetrate CBs and connect the pools together along a pathway. The continuity of the fine unit in the secondary hetero-structure (**Figure 3c**) creates a significant TCE pool ~2 times deeper than found in the primary hetero-structure. **Saturation within the coarse unit pools is near uniform and at the nonwetting satiated value, consistent with our calibration.** The local scatter of light blurs the saturation field around sharp transitions at the edges of pools and thus shows artificial nonzero nonwetting saturations in the surrounding capillary barriers. Overestimation of the saturation is particularly noticeable in the fine CB (broken in the primary hetero-structure) as it is both thin and has nonwetting fluid on its top and bottom. Also note in the secondary hetero-structure that maximum saturations within the medium sand in the pool above the fine CB

are underestimated due to our lack of a fully TCE saturated image for saturation calculations.

The saturation within fingers is much more variable than within pools. This is further shown in magnified and contrast enhanced images of the final saturation structures at and beyond CB4 for CO₂ and TCE in the primary hetero-structure (**Figure 4**). The highly variable appearance of the saturation along a finger demonstrates that the fingers are not smooth macroscopic features. We see that the appearance of fingers in the coarse unit as a series of near disconnected regions of high saturation is due to the ‘pore scale’ nature of the fingers amplified by the imaging technique. The finger is composed of a string of nonwetting clusters connected by single pores and thus does not uniformly fill the pore network between the front and the back of the chamber. **This structure corresponds closely to the link/node/blob picture of a percolating backbone (e.g., Stanley, 1977) except that it is directionally oriented by gravity.** When the finger is in pores near or at the front of the chamber, we see it as a discrete, connected pore structure. As the finger meanders further behind the chamber front, it becomes increasingly blurred by the scatter of light. As seen in **Figures 3 and 4**, the CO₂ invasion shows a more “slender” pathway with pools of lower height and narrower connecting fingers than that for the TCE invasions. Additionally, detailed evaluation at high magnification shows that all CBs were breached by a single finger for the CO₂ invasion while multiple fingers extend through each CB for both TCE invasions.

Composite order images showing the invasion sequence for each experiment are presented in **Figure 5**. As discussed earlier, the fundamental pore scale behavior is less obvious in

these images as we simply track increases in nonwetting saturation above zero. The order images clearly demonstrate the heterogeneity structure to impose an oscillation between modes of gravity-destabilized and gravity-stabilized invasion. Near vertical fingers grow away from the source during the gravity-destabilized invasion mode while flat horizontal fronts grow back toward the source during the gravity-stabilized mode. This dynamic is clearer at higher resolution as shown in **Figure 6** composed of magnified and re-colored zones of the order images containing CB1 and CB2.

In general, the gravity-destabilized finger growth mode occurs in all units until a CB is encountered. Once encountered, CBs then cause gravity-stabilized pool growth behind until they are breached. During this mode, the earlier gravity-driven fingering is partially or in some cases completely ‘over written’ by the backward growing horizontal front. As the stabilized growth period progresses, if units of intermediate texture are encountered (i.e., lower entry pressures than the primary CB), they are entered if the appropriate pressure is exceeded. This behavior is particularly well demonstrated in the secondary hetero-structure TCE invasion where the continuity of the fine textured CB was maintained. Refer to the green and yellow regions in the composite order image (**Figure 5c**) while also looking back to the hetero-structure image (**Figure 1b**). Notice that medium units and coarse-textured units were being filled concurrently, the medium units at lower elevations than the coarse units.

As nonwetting phase pressure builds behind a CB, entry of the barrier begins at a series of points along the interface from which small fingers grow. When one or more of these fingers reach the far side of the barrier, the barrier is breached, and the fingers continue

growing into the underlying layer. For the CO_2 invasion, only the leading finger that is first to enter the coarser layer ever continues growth. However, for both TCE invasions, several of the lagging fingers always make contact with the underlying coarser layer and generate separate fingers beyond. For TCE invasion, the pool height behind the capillary barrier continues to rise during this period and stops after the final finger spans the capillary barrier. However, for CO_2 , pool height stops increasing as soon as the first finger spans the barrier. Close study of **Figure 4** shows that the initial growth points in the primary hetero-structure are identical for both the CO_2 and TCE experiments, however, for TCE, all three fingers continued to grow.

Animation of sequential saturation images for all experiments clearly shows that the nonwetting phase saturation does not increase monotonically in time at all locations along the invasion pathway. The spatial structure of this non-monotonicity up to the conclusion of injection is shown in **Figure 7a and b** for the TCE and CO_2 invasions in the primary hetero-structure. Color represents the number of increase-decrease cycles that occurred as invasion proceeded. For the CO_2 experiment, we see pulsation through nearly the entire gas invasion structure. Maximum cycles reached 18 within a small region beyond the breach of CB1. For TCE invasion in the same pack, we find pulsation concentrated directly beyond CB3 where it reached values of 15. A second region within the fingering zone below CB4 has undergone a single cycle by the time of system breakthrough. Pulsation in the secondary hetero-structure TCE invasion (not shown) was similar but more subdued reaching a maximum value beyond CB3 of 4. Again, we note that cycle counts could have been higher than we measured as our acquisition interval was only 20 seconds.

Close examination of animations and magnifications of the cycle images (see **Figure 8**) show pulsation to occur at two different scales: pore scale and pool scale. In pore scale pulsation, individual pores and clusters of pores are seen to 'drip' in time. Such pulsation can be seen within one CB (e.g., CB1 in CO₂ injection) and within the coarse unit directly below CBs and along fingers. In pool scale pulsation entire pools 'throb' in time. High cycle values are found both in the large region at the top of the pool and along the edges of the pool where pores repeatedly fill with wetting and nonwetting phases in concert. This larger scale throbbing is seen within the pools behind all CBs in the CO₂ invasion but is not detected within the TCE invasion structure. A glimpse at the temporal complexity of pulsation within our experiments is shown in **Figure 9** where nonwetting fluid saturation is plotted as a function of time for a series of small ~25 pixel regions located along the CO₂ invasion structure. We see that while the traces may look generally periodic, they are not. At each location, both the magnitude of the saturation swing and the local period are different from one pulse to the next, the small 'hot spots' within and below capillary barriers showing a more erratic pulsation than the pools.

When the invading fluid reached the distal end of the chamber, nonwetting injection was terminated and the redistribution phase of the experiment began. For the CO₂ invasion, negligible redistribution occurred. However, for the TCE, redistribution lasted several hours (see **Table 3**). A fine layer at the bottom of the chamber kept the TCE within the chamber so simultaneous drainage and invasion could be visualized. **Figure 10** shows the difference between the beginning and end of the redistribution stage for the TCE invasions into the primary and secondary hetero structures; dark zones indicate where TCE has left, bright where it has moved to, and gray where no change occurs. More fluid redistributed

for the secondary hetero-structure than the primary structure as can be seen by the depth of pooled TCE at the bottom of the cell. Visual inspection of the drained zones of pools and fingers within the coarse unit show the TCE to be entirely trapped in a series of blobs/ganglia at a residual saturation of ~ 0.2 to 0.3. Still, a majority of the mass is held in the pools at high saturations of ~ 0.85 to 0.9. During redistribution, we also find significant pulsation to occur, much greater than during the initial invasion. Figure 7c shows a cycle image built over the redistribution period for the primary hetero experiment. Pulsation is found as TCE moves through the coarse run between CB3 and CB4 and beyond CB4 as we saw in the CO₂ experiment. An additional finger below CB4 also formed and pulsated below CB4, the main pulsation point just overlapping that for the CO₂ experiment beyond this barrier.

In summary, we see:

- CBs create a pool and finger invasion structure. Pore scale fingers occur as interconnected clusters of nonwetting phase resulting in highly variable macro-scale saturation. Pools occur as macro-scale zones of high saturation. The vast majority of the non wetting phase mass is held in the pools.
- Gravity driven fingers always occur as invasion proceeds away from the source. Gravity stabilized invasion can overwrite the fingers when a CB is encountered and a pool forming horizontal front grows backwards toward the source.
- Pulsation occurs within fingers and within pools at different scales of expression for each. Pulsation increases as flow rate diminishes, and in the TCE experiments, as redistribution commenced.

- Redistribution after termination of injection occurred for TCE but not for CO_2 . As we will show in the next section, this difference results from the importance of viscous forces at the flow rates imposed to increase TCE pool heights during invasion over their equilibrium or final static levels.

In the next section, we focus on defining a suite of length scales that describe the evolving and dynamic invasion structure generated by the nonwetting phase within a heterogeneous porous medium.

4. Length Scales for Invasion Structure and Dynamics

Invasion structure and dynamics within the granular pack can be analyzed by considering local differential pressures within the invading phase, ΔP_i , associated with capillary, gravitational, and viscous forces (subscripts c, g and v, respectively) written as:

$$\Delta P_c \sim 2\sigma \left(\frac{1}{R_{\min}} - \frac{1}{R_{\max}} \right) \quad \Delta P_g \sim \Delta \rho g h_g \quad \Delta P_v \sim \frac{Q \mu h_v}{kA} \quad (1)$$

where σ is the interfacial tension, $\Delta \rho$ the density differential (positive) between fluids, g the gravitational constant, h_g a local macroscopic vertical distance, Q the flow rate, μ the viscosity of the invading fluid, h_v a local macroscopic length in the direction of invasion, k the permeability, and A the local cross-sectional area of the invading fluid filled porous media. Note that k applies to the structure being considered, i.e., a string of pores within a finger or a 'matrix' of pores within a pool and will be approximated by the intrinsic permeability of the media within A. R_{\min} and R_{\max} represent radii of curvature for the pore scale fluid/fluid interface typical of the minimum and maximum encountered locally such as to consider variability in capillary forces within a unit, between units, or between wetting and nonwetting entry, depending on the situation of interest. As written, ΔP_i are all positive quantities.

Considering fluid properties alone (see Table 2), we see that CO₂ has slightly larger ΔP_c

and ΔP_g than TCE (within factor of 2 to 3) while TCE has significantly larger ΔP_v (factor of 39). Ratios of the ΔP_i yield dimensionless numbers such as the Bond Number (gravity/capillary) or the Capillary Number (viscous/capillary) which can be used to define the regime in which our experiments lie. For a heterogeneous system such as ours, these numbers will vary as a function of time and space and thus are less defining than in homogeneous media.

Here we use simple local scale analyses that balance the local pressure differentials, i.e.,

$$\Delta P_c + \Delta P_g + \Delta P_v = 0 \quad (2)$$

to derive approximate relations for a set of local macroscopic length scales that control phase invasion structure and dynamics within the heterogeneous system. The sign of each ΔP_i in (2) depends on its action with respect to the situation of interest. We consider length scales for finger diameter, finger pulsation, maximum pool height, pool pulsation, and minimum pool height, and evaluate derived formulations with respect to experimental observations. In all cases, we find that the intrinsic pore scale nature of the invasion process and resulting structure must be incorporated into our analysis to explain experimental results. In this, our analysis departs from standard scale analysis for continuum scale flow in porous media where assumed governing equations (usually PDEs for conservation of mass, momentum and energy) are used to develop such relations [e.g., Began, 1985]. Given the pore scale control within our problem, such macroscopic continuum approaches applied at a typical REV scale to the porous medium are not fully

applicable.

4.1. Finger diameter:

As described earlier, fingers are composed of a series of clusters connected or 'strung' together by single pores yielding a highly variable finger diameter (see Figures 4 and 6). These clusters form because of the random nature of the capillary pressure required to fill individual pores within the network. Additionally, we observe that a finger grows through a series of 'bursts' where a small capillary finger moves forward rapidly across a single pore at or near the finger tip and then stops followed by local 'back filling' to create a wider cluster. Therefore, we conclude that capillary pressure oscillates between a low value somewhere within the backfilling cluster (R_{\max}) and a high value just before the next burst occurs as the single 'critical' pore is entered (R_{\min}). Observationally, we can also conclude that due to the 'stringed cluster' nature of the finger, viscous forces acting against flow along the finger are controlled primarily within a single to several pore cross-sectional area comprising the primary conductive backbone of the nonwetting finger. **Along the backbone we consider k to be ~ the intrinsic permeability of the granular media.**

To yield an approximate macroscopic length scale for finger diameter we consider a local balance of ΔP_c that creates the clusters we associate with the diameter to ΔP_g - ΔP_v that drives directional finger growth. At the scale of the finger diameter, d , we take

$h_g = h_v = d$ and find:

$$d \sim \frac{2\sigma}{\Delta\rho g - \frac{Q\mu}{k_f A_f}} \left[\frac{1}{R_{nw\min}} - \frac{1}{R_{nw\max}} \right] \quad (3)$$

where $R_{nw\min}$ and $R_{nw\max}$ are the average minimum and maximum radius sampled during finger growth and subscript f refers to the finger. Here we consider A_f to be independent of d as d is controlled by the average cluster size along its length while A_f is restricted to the conductive backbone within the finger. $R_{nw\min}$ is a function of the pore size distribution and its connectivity within the sand thus yielding a dependence of d on pore size distribution such as found in the analysis of Glass and Yarrington (1996). Additionally, $R_{nw\min}$ will be dependent on gravity and viscous forces active in the media, gravity forces increasing and viscous forces decreasing $R_{nw\min}$ sampled. We note that (3) has a similar form to that of Glass et al. [1989a, b], derived through a combination of dimensional analysis and experimentation for gravity driven wetting fingers. There, however, the macroscopic sorptivity evaluated between wetting entry and the saturation (or pressure) within the finger was used to represent the influence of finger widening capillary forces. Additionally, for wetting invasion, fingers are macroscopic and so A_f is tied to the finger diameter. Note that for nonwetting invasion considered here, capillary forces oppose finger widening and thus finger diameter above that of a single pore is due to the invasion process within a network of variable pores.

Because of the pore scale nature of fingers, experimental diameters are difficult to measure.

However, we estimate them below CB4 for the CO₂ and TCE experiments to be ~ 0.7 and 1.2 cm, respectively (see Figure 4). Additionally, below CB4, we note that three fingers formed for both TCE experiments. In order to compare (3) to these estimates, we must obtain values for the various parameters within the relation. We take R_{nwmax} as the air entry value of the drainage pressure-saturation curve for the coarse sand of ~ 0.0192 cm, assuming contact angles for all systems are the same. R_{nwmin} can also be estimated from the drainage pressure-saturation curve at a representative fluid saturation for the finger. Neglecting the gravitational or viscous dependency of R_{nwmin}, we first consider an average effective fluid saturation derived from standard percolation theory. Considering a random, spatially uncorrelated network of pores within a cubic lattice, the percolation threshold is found to be ~ 0.3 (Stauffer and Aharony, 1992, pg. 17). Estimating the effective nonwetting saturation, S_{enw}, by this value (assuming all pores to have the same volume) yields R_{nwmin} ~ 0.0173 cm for the coarse sand from the drainage pressure-saturation curve. Neglecting viscous forces in (3) yields finger diameters of ~1.0 and 0.68 for CO₂ and TCE, respectively. If we now consider the experimentally measured saturations within the fingers for CO₂ of ~0.15-0.20 and TCE ~0.35-0.40, R_{nwmin} ranges of ~0.0178-0.0176 and ~0.0169-0.0166 cm, with corresponding finger diameters of ~0.73-0.85 and ~0.84-0.96, respectively. For CO₂, our prediction is now quite close. Assuming that each TCE finger carries a third of the flow, including viscous forces with experimental parameters for k and Q, if we take A_f equivalent to ~ 3 pores (~ 3 δ^2 where δ is the mean grain size for the unit), d increases to ~ 1.2-1.3 cm for TCE (no change for CO₂ for a single finger carrying the full supplied flow). Thus finger width can be matched reasonably well with (3) if we consider the finger's pore scale nature.

4.2. Pulsation within a finger:

As a finger lengthens, we find the wetting fluid re-invades selected pores a distance behind the finger tip. This happens as hydrostatic gravity forces act against viscous and hysteretic capillary forces along the vertical length of the finger. Here, the differential capillary pressure of interest is between the nonwetting invasion at the finger tip and the wetting invasion behind. If we consider a local balance of differential pressures from the finger tip backwards, we find that as the finger grows vertically, gravity allows the re-invasion of pores with the wetting fluid when the finger reaches an average vertical tip length, h_t :

$$h_t \sim \frac{2\sigma}{\Delta\rho g} \frac{Q\mu}{k_f A_f} \left[\frac{1}{R_{nw}} - \frac{1}{R_w} \right] \quad (4)$$

where R_{nw} and R_w are average radii for nonwetting and wetting invasion within the unit, respectively. This process was first described by Glass et al. [1989c], for gravity driven wetting fluid fingers. As we will see, for the nonwetting case considered here, the stringed cluster nature of the finger and the lack of connecting films once pores are re-invaded with the wetting phase imposes pulsation while only a single oscillation is found for the wetting case.

The small critical pores connecting clusters along the finger form the critical links within the conducting backbone of the finger. Based on our earlier description of finger burst growth,

these linking pores are also likely to be the smallest pores sampled and thus most likely to be re-invaded with the wetting phase. When they are re-invaded with the wetting fluid, the pathway is broken. Pressure within the upstream nonwetting phase must rebuild ~ 2 times wetting entry (see hysteretic curves in Figure 2) to re-invade a critical pore. After re-invasion with the nonwetting fluid, h_t , given by (4), is in time achieved again, the wetting fluid enters the critical pore, and the cycle begins again. Because gravity forces reduce the total pressure back along the finger, other non-critical 'ancillary' pores within the upstream cluster with slightly larger R also join the pulsation, first filling with the nonwetting phase until the critical pore is 'opened' and then emptying until it is 'closed'. Thus, the upstream cluster grows and then shrinks, amplifying the pulsation induced and controlled by the downstream critical pore. As a finger grows beyond a length of h_t it will contain a number of critical pores along its length where h_t is achieved and thus the nonwetting phase fragments into a series of steady 'residual' clusters connected by pulsating critical and ancillary pores. When a finger reaches a CB, the invasion mode switches from gravity destabilized to gravity stabilized invasion. As the pool grows back toward the source, pulsation is curtailed to a distance h_t from the edge of the growing pool back along the supplying finger.

While we found that finger diameter given by (3) is quite sensitive to the saturation within the finger and thus choice of the R values, (4) is not. If we estimate R_{nw} and R_w by an average value of S_{nw} and S_w of 0.3 from the pressure-saturation curves ($R_{nw} \sim 0.0172$ cm; $R_w \sim 2 R_{nw}$), calculation of h_t with (4) for the coarse layer neglecting viscous forces yields values of ~ 5.1 and 3.4 cm for CO₂ and TCE, respectively. Experimentally, we find pulsation during the CO₂ experiment throughout the entire finger run between CB3 and the

pool above CB4 as well as below CB4, thus supporting the inviscid value given by (4) for CO₂. For TCE, however, only the two zones directly below CB3 and CB4 show pulsation, each > 15 cm above the maximum pool height behind CB4 and the bottom of the chamber, respectively. Including viscous forces increases the TCE value calculated by (4) to ~15 cm for measured k and Q, *if* we take A_f of ~3.3 δ², with again, no influence for CO₂.

Considering that 3 fingers form in the TCE experiment, this corresponds to ~1.1 δ² per finger. We note that when viscous forces are important, (4) is very sensitive to the value of A_f and that to match the experimental results for TCE, it must be reduced from ~ 3 δ² per finger found earlier for a growing finger tip with (3). This result further emphasizes the importance of the conducting backbone and reflects the decrease in its cross-sectional area from several pores at the finger tip down to a single pore controlling pulsation at h_c.

4.3. Maximum pool height:

We can obtain a simple relation for maximum pool height behind a CB, h_{pmax}, by once again balancing local pressure differences for capillary, gravity, and viscous forces. The differential capillary force of interest is that required to enter pores within the coarse unit behind the barrier versus that to enter and span the barrier. Viscous forces must now incorporate both losses within the pool and the barrier. We find:

$$h_{p\max} \sim \frac{2\sigma}{\Delta\rho g - Q\mu \left(\frac{f_b}{k_b A_b} + \frac{f_p}{k_p A_p} \right)} \left[\frac{1}{R_{nwb}} - \frac{1}{R_{nwp}} \right] \quad (5)$$

where $h_{p\max}$ is defined as the distance from the far edge of the capillary barrier to the top of the pool, f is the fraction of $h_{p\max}$ that is within each unit, and subscripts b and p apply to the barrier and pool, respectively. Note that both f_b and f_p are a function of $h_{p\max}$ and thus (5) is not explicit in $h_{p\max}$ and requires iterative solution.

Because the R values in (5) are for different materials, both of which have sharp pressure saturation curves in our experiment, (5) is not very sensitive to the value of the saturation that we use to take corresponding R's. Neglecting viscous forces, and taking R's corresponding to an average S_{nw} of ~ 0.3 from the drainage curves in Figure 2 for the coarse to medium CB ($R_{nwb} \sim 0.0085$ cm, $R_{nwp} \sim 0.0172$ cm), $h_{p\max} \sim 10.8$ cm for the CO_2 invasions. Comparison to a measured mean $h_{p\max}$ for CO_2 of ~ 7 cm shows the estimates based on the pressure-saturation curves to be high by $\sim 50\%$ of the measured value. There are at least four possible explanations for this discrepancy. The first is that the internal porous geometry of the units, especially that composing the CB, were different from that where the properties were measured. This is probable as the hetero-structure and the homogeneous packs were filled and packed with different methods. The second is that the thinness of the CBs allowed them to breach at a lower pressure. All the CBs were thin, ~ 1 cm (~ 20 pores), and laterally extensive, $\sim 10 \times 1$ cm ($\sim 200 \times 20$ pores) and thus the probability of breaching the barrier at a lower pressure should be enhanced. The third is that our assumption of invariant contact angles for all fluid-fluid systems is incorrect and

thus the R distributions are different. Visual inspection of entrapped nonwetting phase, however, showed very little if any deviation from a contact angle of zero between the systems. Finally, it is possible that the higher pressures required to breach the CB material were capable of moving some grains of sand such as to create an, on average, larger set of pores spanning the barrier.

Since all of these possibilities work to modify the bracketed term in (5) primarily by increasing R_{nwb} , we scale it to match the CO₂ data (factor of 1.2) yielding h_{pmax} for TCE of ~4.7 cm without viscous forces. For the TCE injection, mean h_{pmax} (only measurable before CB4) attained ~ 11 cm in the primary hetero-structure. Taking measured values for k and Q, and estimating A within the pool to be ~ 6 cm², if we take $14\delta^2$ for the backbone of the conducting finger network within the barrier, (5) matches the measured value of ~11 cm for TCE. Similar calculations for the fine CB of the secondary hetero-structure yields h_{pmax} of ~20 cm neglecting viscous forces and ~40 cm including them. The distance between the fine CB and CB3 above is ~24 cm and we see in the experiment that the pool comprises the full coarse structure between. Thus for both TCE experiments, we again find that when viscous forces are important, the most critical data is the cross sectional area within the backbones of the conducting finger network, here spanning the CB, and this value must once again be small and on the pore scale to match the pool height data.

Similar relations are easily written for the pool heights at which intermediate units are entered such as seen in our experiment within the secondary-hetero structure. Additionally, while not explored in our experiments, we note that viscous forces will also influence pool geometry as units become wider by causing the breakthrough of capillary barriers before

the entire pool unit is laterally spanned and filled to a uniform pool depth. For a 2D pool such as we have in our experiment, we can approximate the horizontal expansion distance, h_{phor} , by balancing ΔP_v for flow horizontal within the pool unit against the total pressure required for breaching the barrier to yield:

$$h_{phor} \sim h_{p\max} (1 - f_b) \frac{\Delta \rho g k_p A_p}{Q\mu} \quad (6)$$

where A_p now is an approximate average vertical cross-sectional area per unit thickness of the pool and $h_{p\max}$ is given by (5). For the coarse-medium transition and our flow rates, if we take A_p as $0.5 h_{p\max}$, we calculate an h_{phor} of ~ 30 cm for TCE and ~ 1.8 m for CO_2 , both wider than our system width. We can see the beginnings of horizontal viscous force influence in Figure 5b where more distant coarse regions fill at the same time as higher more central regions in the pool above CB4 (see yellow in image). We also note that if the nonwetting fluid can find a way around the CB within the horizontal distance h_{phor} , the CB will not be breached by a vertical finger.

4.4. Pulsation within a pool:

When a finger spans a CB, it encounters a lower capillary pressure in the unit beyond. The pores within the CB and the pool behind, thus, can experience a depressurization. If their pressure decreases to the wetting entry values for pores in the pool or the CB, they will be re-invaded with the wetting fluid. For the pool, pores at the top of the pool will be re-

invaded and the pool height will drop. Additionally, smaller pores along the pool edges are also likely candidates for re-entry. For the CB breached by fingers, the smallest pores which join the clusters of a finger are the most likely to be re-invaded with water.

To consider the re-entry of pool pores with the wetting fluid after a CB is breached, we balance viscous forces across the pool/barrier system against capillary and gravity forces to yield h_{tpool} :

$$h_{tpool} \sim \frac{2\sigma}{\Delta\rho g - Q\mu \left(\frac{f_b}{k_b A_b} + \frac{f_p}{k_p A_p} \right)} \left[\frac{1}{R_{nw2}} - \frac{1}{R_{w1}} \right] \quad (7)$$

where subscripts 1 and 2 denote the unit before and beyond the CB respectively. As with (5), (7) is not explicit in h_{tpool} . Evaluation of this relation with values used previously yields $h_{tpool} \sim 5.2$ cm for the CO_2 and ~ 9.7 cm for TCE. Because the pool height (~ 7 cm) is greater than h_{tpool} for the CO_2 experiment, pores within the pool are immediately re-invaded with wetting fluid and the pool height begins to drop as soon as the CB is spanned. For TCE, the same should occur but it does not. We believe that this difference is likely due to our simplified treatment of viscous forces within the barrier. If A_b is reduced slightly from 14δ to 12δ , (7) matches the h_{pmax} of ~ 11 cm. Thus the reinvasion of a very small number of pores with water can increase the viscous drop across the barrier and yield an h_{tpool} the same as h_{pmax} such that the pool height will not decrease until flow stops and redistribution begins.

Pool height will decrease until the critical pore within the CB is re-invaded with the wetting fluid. For negligible viscous forces and assuming the influence of the growing finger below the CB is curtailed at the CB, this minimum pool height, $h_{p\min}$, is given by:

$$h_{p\min} \sim \frac{2\sigma}{\Delta\rho g} \left[\frac{1}{R_{wb}} - \frac{1}{R_{wp}} \right] \quad (8)$$

where subscripts b and p refer to the barrier and pool, respectively. Taking R_{wb} and R_{wp} as ~ 2 times their nonwetting values yields $h_{p\min}$ of $\sim 0.5 h_{p\max}$ or ~ 3.5 cm. Comparison to the CO_2 experiment where we find $h_{p\min}$ of ~ 3.0 cm shows (8) to fit our data reasonably well. Once the CB closes, h_p must once again achieve $h_{p\max}$ before it reopens thus causing the \sim factor of 2 unit scale pool throbbing seen in the CO_2 invasion. The only pool that does not throb throughout the invasion period is behind CB1. There we see that the finger through the CB itself pulsates, something we do not detect in any of the other CBs. This pulsation suggests a pathway through the barrier with large backbone pores which may be re-invaded at much lower pressures thus curtailing large excursions in pool height.

5. Basis of a 'Structural' Growth Model for nonwetting phase migration

The results of our experiments and analysis point to the critical pore scale control of the invasion process. Incorporating such pore scale control into standard porous continuum approaches is problematic. However, sub-structures within the heterogeneous experiments, including where pulsation will occur, can be reasonably well predicted with the set of length scales derived through scale analysis. We use this result as the foundation of a 'structural' growth model for nonwetting phase migration within a heterogeneous aquifer. By structural growth model, we mean a model that produces the evolution of nonwetting phase structure, the resulting finger and pool spatial configuration, and the zones where dynamic pulsation occurs. Considering first the situation where viscous forces are negligible, for a given heterogeneity structure with known pressure saturation curves for each unit, we can predict the growth of the nonwetting phase straightforwardly by assembling features defined by our scale analysis derived length scales. The size of fingers, the height of pools, the penetration of CBs, and the regions where pulsation will occur can all be delineated from the point of injection to the boundaries of the field. With further knowledge of Q , the permeability for each unit, and relations for the critical conducting backbone areas for fingers both within and below CBs, non-negligible viscous forces could then be included as a perturbation of the inviscid structure yielding increased pond heights and finger widths. Finally, if important at the scale of application, the influence of both pulsation and viscous forces to generate divergent pathways (i.e., multiple and divergent fingers) could be considered and superimposed on the core model results to yield the final nonwetting phase structure within the domain.

In order to develop confidence in such a structural growth model, our experiments suggest that we must understand in order of importance: 1) the interplay between capillary and gravity forces in context of inter and intra facies lithologic variation; 2) the added influence of viscous forces; and 3) the possible additional complication imparted by pulsation. Obviously, facies-scale or macro lithologic variation controls the core structure of the nonwetting phase migration and thus is of highest importance. However, we also expect that the migration process will be affected by the ever-present intra facies microlayering found in subsurface sediments with the internal contrast and length scales (vertical, horizontal) of the subscale structure critically controlling nonwetting invasion just as we see in our current macro heterogeneous experiments. The importance of this structure will increase as both its contrast and length scales increase. In general, we expect greater meandering parallel to the microlayering (usually horizontal) within a given unit. While for wetting fluid advance such microlayering yields a wider macroscopic structure that may be consistent with unit scale anisotropic effective properties, we are skeptical that a similar approach will be valid for nonwetting invasion. However, the greater horizontal meandering should cause a greater randomness within the invasion structure because a larger number of units can be sampled as invasion progresses.

When viscous forces are non-negligible, prediction of finger diameter and pool height and width within a heterogeneous formation now requires knowledge of both Q and A_f comprising the conducting backbone of the finger network that forms. It is likely that a relationship between Q and A_f will exist such as to only require knowledge of Q . Unfortunately, since A_f is not the total cross-sectional area occupied by a finger, the results

of linear stability theory applied to continuum equations typically used to develop such a relation is insufficient. Therefore, we must resort to a combination of experimentation and alternative analysis. Such an approach could use the work of Held and Illangasekare [1995a, b] as a starting point.

Pulsation such as seen in our experiments, has been found in rough-walled fractures for both wetting [Nicholl et al 1993, Glass and Nicholl, 1996] and nonwetting [Glass and Nicholl, 1995] gravity driven invasion under imposed constant flux conditions. Because of its basic similarity to the dripping faucet dynamic, pulsation could be chaotic, as suggested for fractures by Nicholl et al [1993]. Regardless of whether it is chaotic or periodic, pulsation will generate different nonwetting phase fluid pathways in time such as we see below CB4 in both the TCE and CO₂ experiments. Thus on a small scale, we hypothesize pulsation to impart a rich spatial and temporal variability superimposed on the basic structural form selected by simple gravity and capillary forces within the heterogeneous field.

While the effects of viscous forces and pulsation are required to completely explain the migration behavior at small scale, will these added complexities matter for prediction of DNAPL migration at the field scale? The critical influences are not those that influence local saturations but that influence the large scale pathway of DNAPL migration. Viscous forces will cause greater pool depths which can cause the selection of a pathway that spills over before the local CB is breached. However, viscous forces can also inhibit lateral migration over and around a CB and thus contribute to its breaching. These two influences will compete with each other and so the combination may yield only a small influence in

pathway choice, however, it is difficult to develop a generic rule of thumb for whether viscous forces contribute to lateral migration or inhibit it. Both viscous forces and pulsation also cause multiple finger formation and thus the possibility of divergent pathways. However, at the field scale this influence should be small unless the large scale pathway is sensitive to small changes in connection via either multiple fingers or pulsation. Thus, while further research is required, we believe the aspects of multiple finger formation and pulsation to be of second or even third order with respect to prediction of nonwetting migration at the field scale.

Finally, we note that the 'structural' growth model outlined above is simply a further upscaling of macro MIP models originally suggested by Glass et al. [1993], for the migration of DNAPL at the field scale. This approach has significant advantages over two-phase continuum models as it allows several orders of magnitude increased geological detail to be incorporated into the problem domain. As we have shown in our experiments, this detail critically controls the invasion structure. Unfortunately, at the majority of spill sites, we may never know enough geological detail for deterministic modeling approaches to be more than marginally useful in finding DNAPLs. However, probabilistic modeling approaches, such as Monte Carlo analysis, show promise in aiding DNAPL source zone characterization efforts. And fortunately, unlike two-phase continuum models, MIP models carry a light computational burden and are thus well-suited to Monte Carlo analysis. Borchers et al. (1997) showed that Monte Carlo simulation of DNAPL migration through 3-D geological fields could be used to produce a probability map of potential DNAPL source zone location. In an example problem, such simulation was used to create many realizations of plausible heterogeneous lithologies based on what was known about the

system, while still reflecting the degree of remaining uncertainty. A MIP model was then used to simulate DNAPL migration on each realization and the results of the ensemble compiled into a probability map of where the DNAPL resides. Using such an approach, uncertainty about the distribution of geological features and the disposal history at the site can be propagated through the probabilistic modeling effort to reflect the degree of uncertainty in the extent of DNAPL migration. The probability map can then either be used directly, or used in conjunction with decision tree approaches, to suggest favorable sampling and/or recovery locations.

6. Concluding Remarks

Our experiments clearly demonstrate nonwetting invasion to occur as a sequence of gravity-stabilized and -destabilized displacements within the heterogeneous media. Pore scale gravity-destabilized fingers occur in all units as they are entered, but they are overwritten by macroscopic gravity-stabilized displacements behind capillary barriers that create backward growing macroscopic pools. The extent of the overwriting depends on the pool height before breakthrough of its controlling capillary barrier. This is illustrated in the secondary hetero-structure, where similar gravity-driven fingering as seen in the primary hetero-structure was completely overwritten in the top 2/3 of the chamber. Such overwriting of fingers suggests that the destabilized signature of the flow will often be missed in experiments without capturing appropriate time history data, especially for experiments conducted in the field.

Our experiments show the majority of the mass is held in high saturation regions commonly referred to as pools. A much smaller portion of the mass is held at residual saturation either in drained fingers or in the drained portion of pools. While this is not at all a startling revelation, we point out that experiments and analyses founded on an assumption of nonwetting phase held predominantly as a residual saturation may have limited applicability to situations where mass is held predominantly in pools. Failure to consider the presence of DNAPLs held in pools during remediation design may lead to decreased performance or inadvertent downward mobilization. For example, in designing a surfactant flood to enhance aqueous solubility, using the results of laboratory column studies or trapping number calculations based on the assumption of residual saturation may

significantly underestimate the potential for mobilization of mass held in pools. On a more positive note, we consider that the filling of pools provides the primary mechanism for lateral spreading and thereby the opportunity for detecting DNAPL source zones through the placement of vertical boreholes. This approach may prove profitable in cases where geological knowledge is sufficient to identify potential locations of lithologic traps and thus hypothesize where the pools are likely to be. (That is, provided that sufficient care is taken to prevent the breaching of the capillary barrier present on the underside of the pool and inadvertently creating a downward migration pathway allowing its drainage.) Indeed, explicitly identifying the locations of pools may provide the opportunity to pump organic liquids from them prior to attempting more exotic remediation strategies.

Our experiments also clearly show the existence of pulsation behind the invasion front both at the pore scale within fingers and at the pool scale behind capillary barriers. This dynamic is due to the combination of gravitational forces and hysteretic capillary forces with viscous forces working to suppress pulsation as flow rate increases. To our knowledge, this is the first time such dynamics have been clearly observed and explained. At present, we expect pulsation to occur under most circumstances as long as viscous forces are 'low enough'. It is interesting to note that viscous fingering, which creates extreme randomness under conditions of high flow rate has, at low flow rate, a gravity / capillary controlled counterpart that also increases randomness in pathway formation. A more thorough understanding of the pulsation phenomena is required to fully consider this effect and its

repercussions for predictability; however, we expect that for field scale migration problems, its influence is minimal.

Comparison of scale analysis derived length scales and experimental measurements show nonwetting invasion structure and dynamics to be controlled at the pore-scale. The competition between local random capillary forces, gravity forces and viscous forces acting along the narrow conducting backbone creates the dynamic that yields the stringed cluster structure of gravity-driven fingers. We find that this pore-scale finger structure can also control the pressure at which a capillary barrier breaches. Viscous forces acting along the conducting backbone of the fingers penetrating the capillary barrier can become significant and on the same order as capillary and gravity. Thus, analyses of the importance of viscous forces for a particular problem must properly account for finger structure.

The results of our experimental work have implication for two-phase modeling approaches currently in use. Given the intrinsic pore-scale nature of finger formation and propagation, together with the local nature of the failure of capillary barriers, we question whether two-phase, porous continuum-scale modeling would be able to capture the unstable, lithology-driven migration behavior found in our experiments. And to the extent that we believe our laboratory-constructed lithology provides a reasonable analogue to a field situation, we question the suitability of this class of models to field applications as well. Our experiments and those presented by Glass and Nicholl [1996] for gravity destabilized wetting fluid invasion in heterogeneous systems, also question the relevance of approaches based on stability analysis in homogeneous media to predict details such as finger width, the number of fingers, or finger velocity in heterogeneous media. In the field, heterogeneities and

capillary barriers will focus flow into a series of 'zones' or 'points' rather than a distributed source such as analyzed through simple linear stability theory. While knowledge of system stability derived from such an analysis is important, quantitative predictions based on such an approach are highly speculative.

As an alternative, we propose a simplified approach based on our length scale analysis as a more appropriate means to model nonwetting phase movement under many natural gradient conditions similar to our experiments. In principal, knowledge of the heterogeneous porous network structure and critical unit properties of saturated permeability and pressure-saturation curves allow us to predict the structure of the invasion pathway directly.

Beginning with a core structure formed by capillary and gravity forces, and incorporating effects imposed by viscous forces or pulsation as needed, allows us to generate phase structure growth with a set of physically based rules. Such a structural growth model is a very different approach to prediction than the standard two-phase continuum approach used by the vast majority of researchers, and forms a class of upscaled MIP. However, implementation of upscaled MIP not only allows, but requires sufficient geological detail at the scale controlling migration behavior, that is, the scale where significant textural contrasts occur. To the extent that deterministic data at that scale will seldom be available, we recommend probabilistic modeling of migration through multiple geological fields that capture the essence of the lithologic architecture.

Acknowledgments

This research was partially supported by Sandia National Laboratories Research and Development Program, and the Department of Energy's Environmental Science Management Program and Basic Energy Science Geoscience Program under contract DE-AC04-94AL85000. John Gilletly helped conduct the experiments and Eric Webb gave advice on the design of the heterogeneous sand pack. We thank Martin Blunt, Toby Ewing, and Clinton Willson for thier constructive reviews of the manuscript.

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Figure Captions

Figure 1. Digital images of the heterogeneous sand packs used in the invasion experiments.

In these images the coarse sand transmits the most light and hence is the lightest; the medium sand appears gray; and the fine sand appears dark. The injection port and four major capillary barriers (labeled CB1 through CB4) are identified for the primary heterogeneous structure. These capillary barriers occur where medium sand separates coarse facies from one another. These same features can be found on the secondary heterogeneous structure as well. The difference between the primary and secondary structures can be seen in the fine sand layer extending across the column about 2/3 of the way from the top. In the primary structure, this layer is discontinuous, while in the secondary structure this layer is continuous and forms one additional capillary barrier. The primary structure was used for experiments 1 and 2, and the secondary structure was used for experiment 3. Notice that small variations in facies location and shape exist between the two sand packings. These small differences had negligible effect on the behavior of the migrating fluids.

Figure 2. Hysteretic pressure-saturation curves (air-water) for the three sands used within the heterogeneous sand packs. The experimental chamber was packed uniformly with each of the three sands and the light transmission method was used to measure equilibrium saturation profiles to yield the primary drainage curve (fully water saturated initial condition) and the main wetting curve (residual nonwetting air phase initial condition). The main wetting curve for the fine sand was not measured.

Figure 3. Nonwetting saturation fields at the time the invading non-wetting phase spanned the length of the chamber just before fluid injection was terminated: a) CO₂ in the primary hetero-structure; b) TCE in the primary hetero-structure; c) TCE in the secondary hetero-structure. Color indicates the local nonwetting phase saturation. Note that the local scatter of light blurs the saturation field around sharp transitions at the edges of pools and within finger zones. Also note that maximum saturations within the medium sand in c) are underestimated.

Figure 4. Magnification of non-wetting saturation structure at and beneath CB4 in the primary hetero-structure: a) CO₂; b) TCE. Saturation fields have been contrast enhanced to bring out the underlying pore scale nature of the fingers as a string of clusters connected by single pores. Note that the two stalled fingers within CB4 marked for CO₂ continued growing for TCE and spanned the capillary barrier. Boxes shown are representative finger widths of 0.7 and 1.2 cm for CO₂ and TCE, respectively.

Figure 5. Composite order images of nonwetting invasion: a) CO₂ in the primary hetero-structure; b) TCE in the primary hetero-structure; c) TCE in the secondary hetero-structure. Color indicates the order of filling. In c) initial data was lost resulting in the black region at early times at the top of the chamber.

Figure 6. Magnification of the composite order images of nonwetting invasion in the vicinity of CB1 and CB2 in the primary hetero-structure: a) CO₂; b) TCE. Color indicates the order of filling.

Figure 7. Fluctuation cycle images for saturation of the non-wetting phase in the primary hetero-structure: a) CO₂ during invasion; b) TCE during invasion; c) TCE during redistribution. Color indicates the number of cycles of saturation increase then decrease.

Figure 8. Magnification of fluctuation cycle images in the vicinity of CB3 for the primary hetero-structure: a) CO₂; b) TCE. Color indicates the number of cycles of saturation increase then decrease.

Figure 9. Time series of saturation at various locations during non-wetting phase invasion for the CO₂ experiment illustrating chaotic fluctuations in saturation. Saturation data has been normalized by the mean at each location and shifted so that saturation histories may be compared.

Figure 10. Redistribution of non-wetting phase at the conclusion of invasion for the TCE experiments: a) TCE redistribution in the primary hetero-structure; b) TCE redistribution in the secondary hetero-structure. Dark zones indicate locations where TCE has drained. Light zones indicate locations to where the TCE has migrated.

Table 1 Sand Properties

| Sand | Sieve Size | Grain Size Mean-Max-Min mm | Porosity | Intrinsic Perm. cm ² |
|--------|------------|----------------------------------|----------|------------------------------------|
| Coarse | 12-20 | 1.1-1.7-0.84 | 34.8 | 6.32E-06 |
| Medium | 30-40 | 0.05-0.60-0.42 | 35.4 | 1.20E-06 |
| Fine | 50-70 | 0.26-0.30-0.21 | 36.2 | 2.97E-07 |

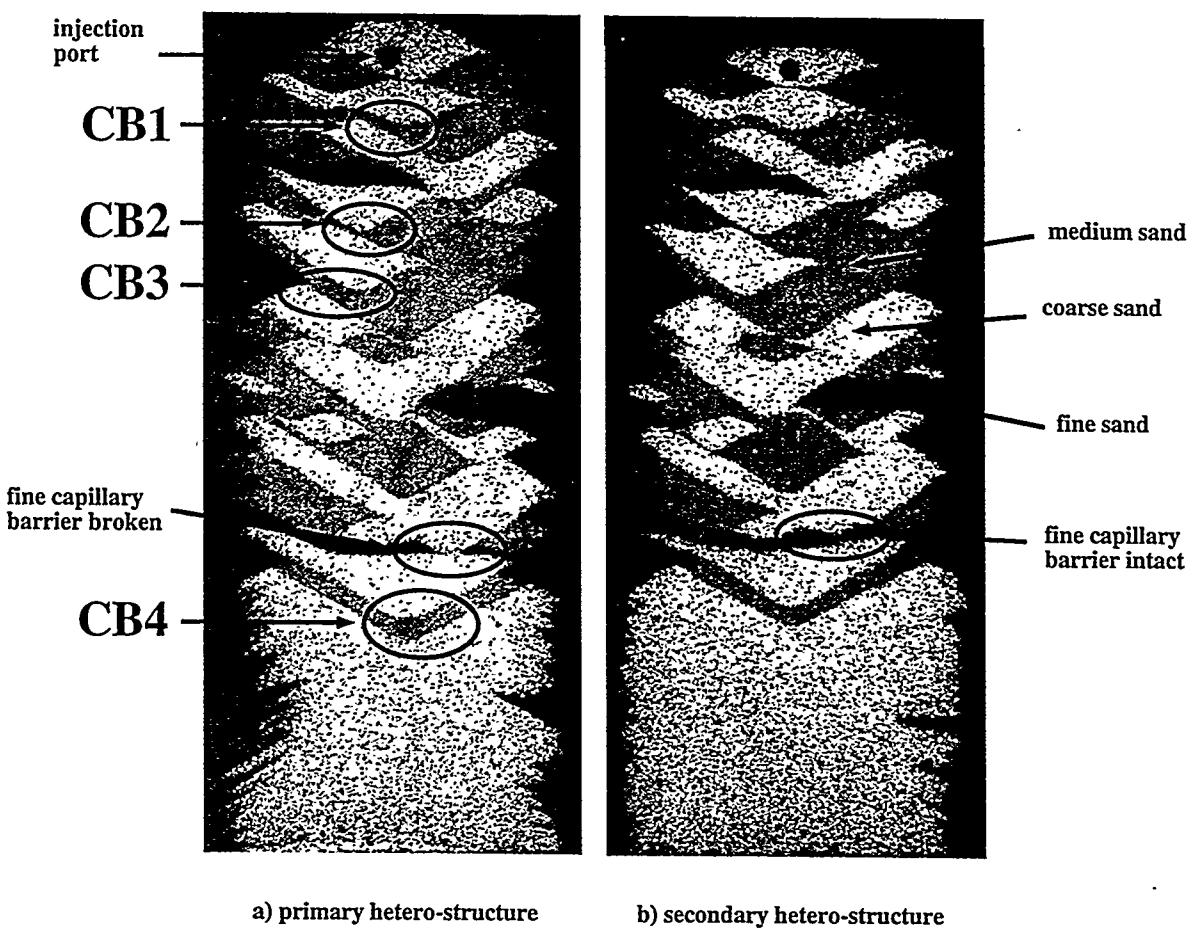
Table 2 Fluid Properties

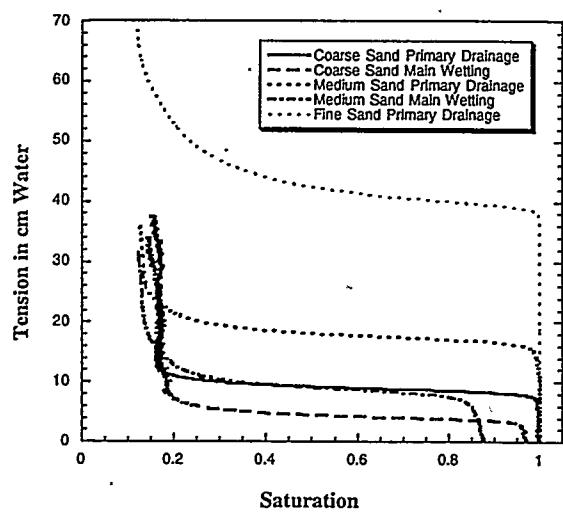
| Fluid | Density g/ml | IFT Dyne/cm | Viscosity poise |
|------------------|-----------------|----------------|--------------------|
| CO ₂ | 0.196 * | 71 | 1.48E-02 * |
| H ₂ O | 0.996 * | NA | 1.00E-02 * |
| TCE | 1.464 | 27 | 5.75E-03 |

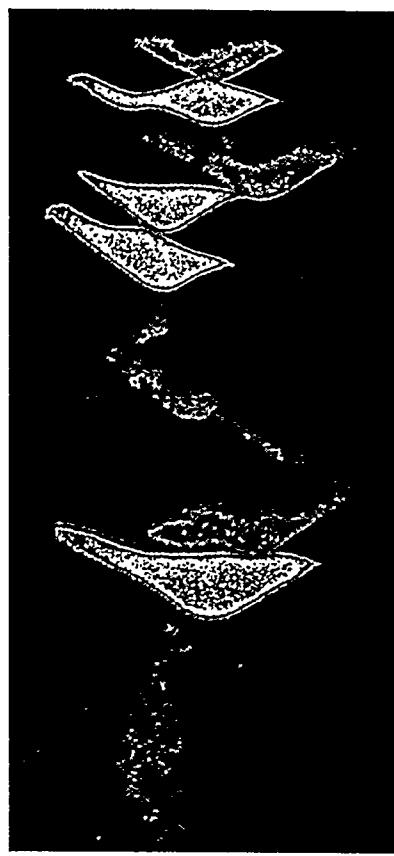
*Values from CRC Handbook of Chemistry and Physics 66th Edition, 1985, CRC press Inc., Robert C. Weast, editor.

Table 3 Experiments Conducted

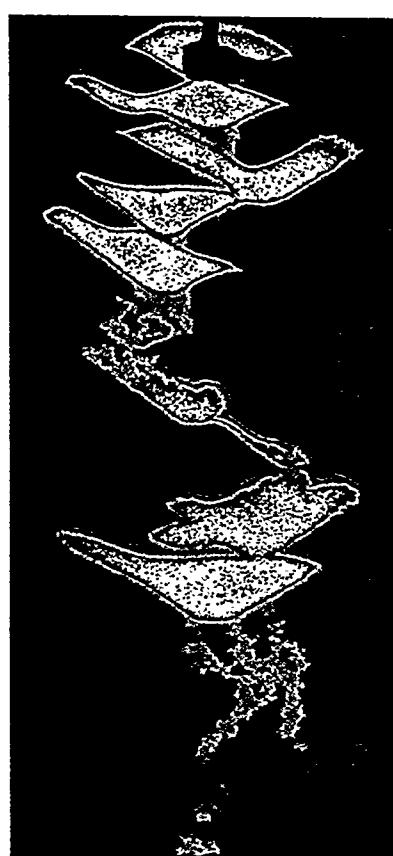
| Fluid/Fluid System | Heterogeneous Structure | Q mL/min | System Breakthrough mL | Redistribution End minutes |
|------------------------------------|-------------------------|----------|------------------------|----------------------------|
| CO ₂ / H ₂ O | Primary | 1.2 | 50.8 | NA |
| TCE / H ₂ O | Primary | 0.9 | 73.1 | 303 |
| TCE / H ₂ O | Secondary | 1.0 | 158 | 240 |





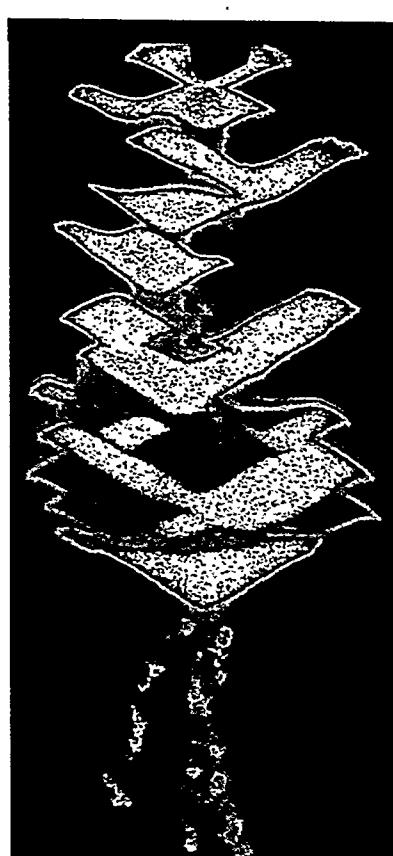


a) CO₂



b) TCE

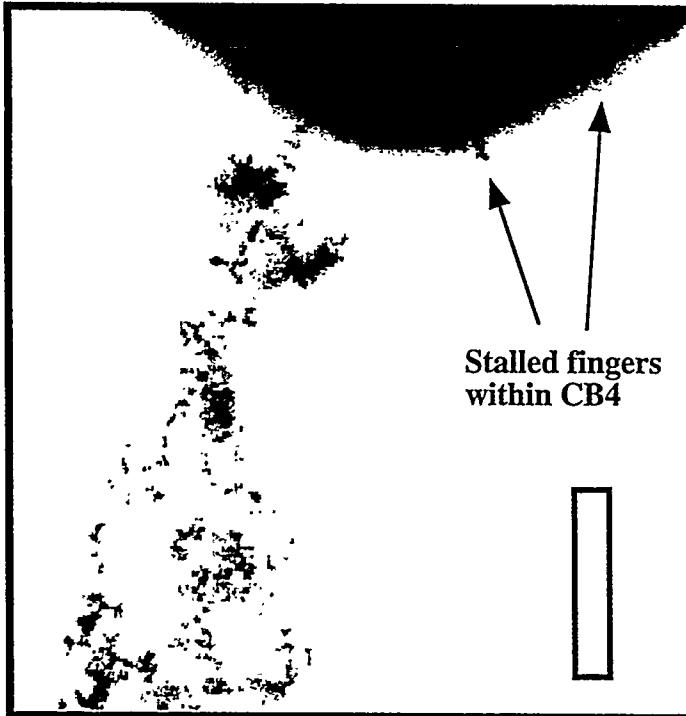
Primary Hetero Structure



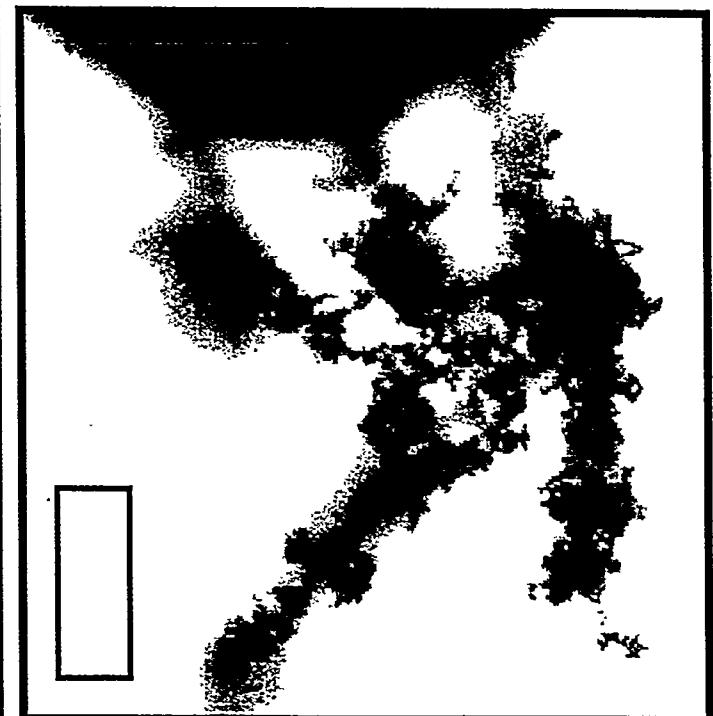
c) TCE

Secondary Hetero Structure





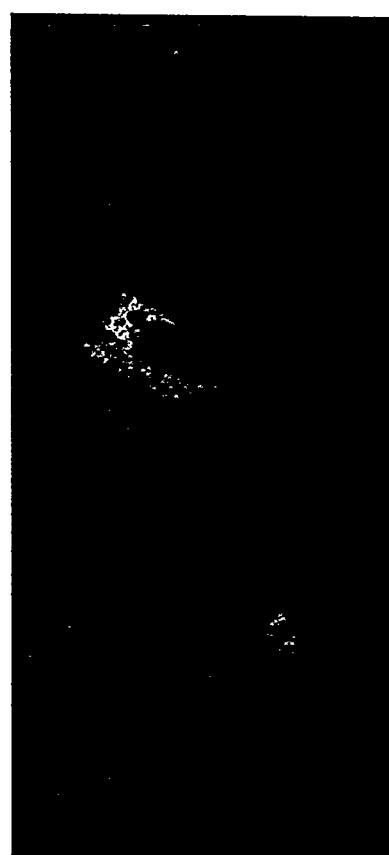
a) C02



b) TCE



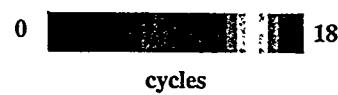
a) CO₂



b) TCE



c) TCE
Redistribution



cycles



a) CO₂

Primary Hetero Structure



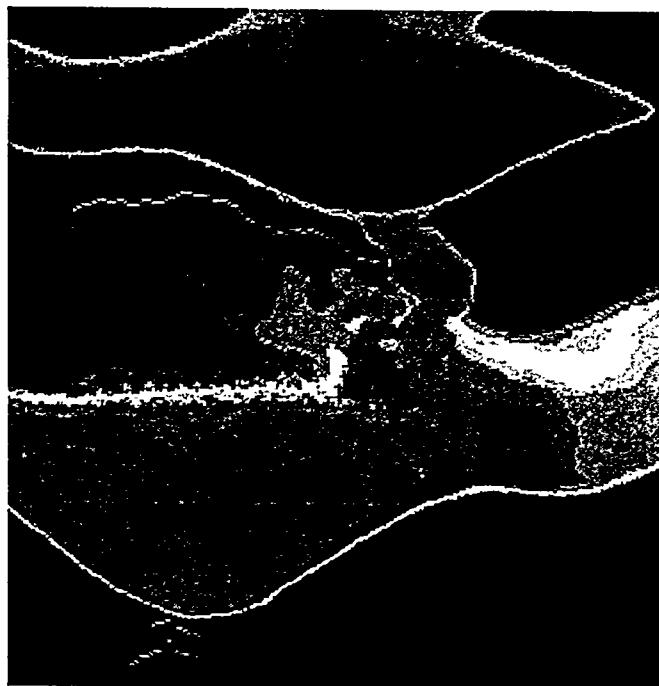
b) TCE



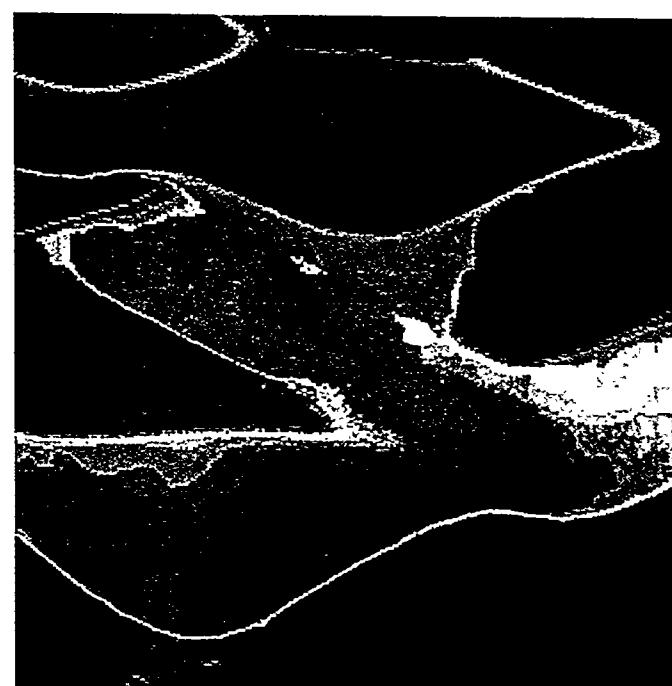
c) TCE

Secondary Hetero Structure

early  late
filling order



a) CO₂



b) TCE

early late
filling order



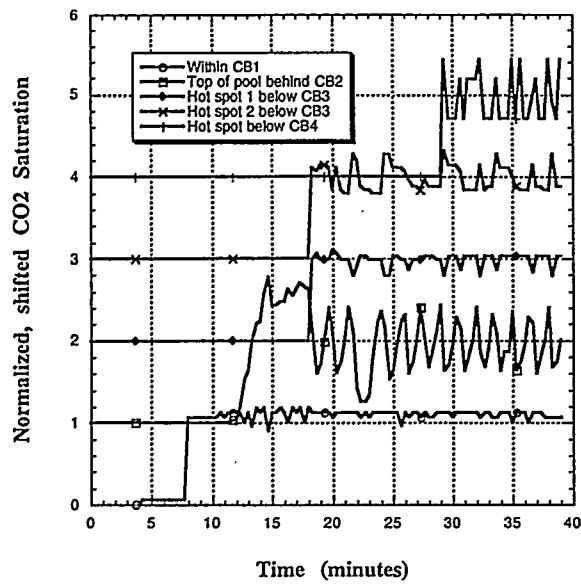
a) CO₂

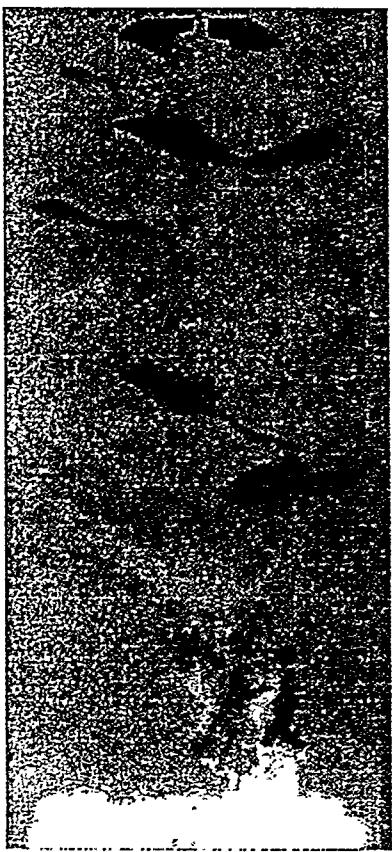


b) TCE



cycles





a) primary hetero-structure



b) secondary hetero-structure