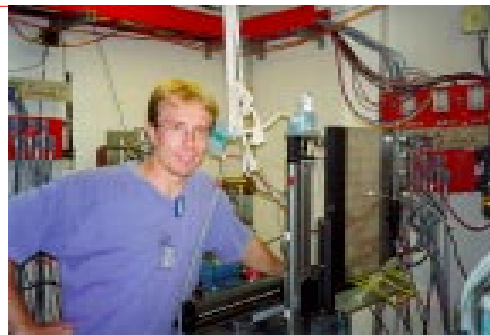


Rapid, Quantitative Tracking of Fluid Flow in Soils with Synchrotron X-rays

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Tim Bauters at the A-2 hutch at CHESS. Beside him is a plastic chamber (35 cm width x 55 cm height) representing a 2 dimensional layer of soil mounted on an x-z stage to move the chamber into the fixed X-ray path. This path can be visualized by connecting both X-ray detectors: the upstream X-ray detector is located beside Tim's left shoulder, while the downstream detector is partly visible on the right side of the picture. The tensiometers in the middle of the chamber are attached with tubing. The tensiometers measure the capillary pressure in the soil layer.

The building of the Agricultural and Biological Engineering Department is less than 50 meters from the synchrotron ring. Despite the close proximity there was very little communication except during a few summers when we played soccer together on the sport field above the ring. In 1982 with the help of Barnes Bierck we finally submitted a CHESS proposal. Initially the CHESS staff was reluctant to work with an agricultural engineer and civil engineer who wanted to do "BIG SCIENCE". Thus, a time slot from the day before Thanksgiving until the Monday after Thanksgiving was appointed. Apparently a good impression was made because the next year we were offered a longer time slot between Christmas and New Years Day. When David DiCarlo (a true physicist) joined our group, we became more than an interesting curiosity. With Dave's help we measured the very fast transient fluid content changes in three phase systems, which few in the world are able to do.

Introduction to soil filtration

Many people, when they think of the word soil, have in mind the basic loose material that nourishes and supports growing plants. Soil also has the desirable attribute of being able to filter out contaminants, and to support microorganisms that can digest contaminants and turn them into safe compounds.

To illustrate some basic principles, imagine rainwater on a field, which then infiltrates into the soil and moves towards the groundwater under gravity. As the water infiltrates the soil, the soil acts as a filter, cleansing out contaminants such as applied agri-chemicals (fertilizer, pesticides, herbicides, etc.) from the rainwater. If the rainwater moves through the soil as a horizontal front heading toward the groundwater, the resulting wetting front is termed "stable" (Figure 1a), and a maximum of the chemicals are retained in the soil where they belong and can be eventually degraded. But often the rainwater infiltrates in an irregular pattern, producing preferential flow paths or "fingers" (Figure 1b). Since preferential flow bypasses some or all of the filtering action of the soil, more contaminants can enter and poison the groundwater. This can be a major problem, since many communities rely on groundwater as a drinking water source.

The term preferential flow actually describes three different flow processes: macropore, funnel and fingered flow. Macropore flow refers to water passing through some preferred

path such as decayed roots and worm paths. Funnel flow occurs when the downward water flow gets funneled or diverted toward one side because of impermeable layers. Fingered flow occurs in a perfect homogeneous sandy porous media, where the wetting front breaks up like a flame front. The latter category of fingered flow in sandy soil was the initial focus of our research at the Cornell High Energy Synchrotron Source (CHESS).

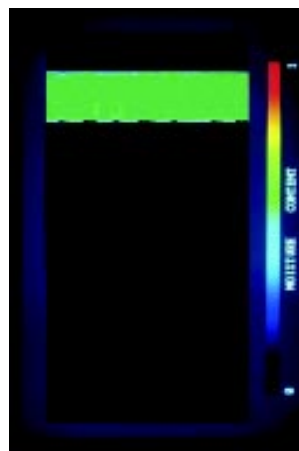


Figure 1a: The infiltration (above) is a stable infiltration (conventional theory).

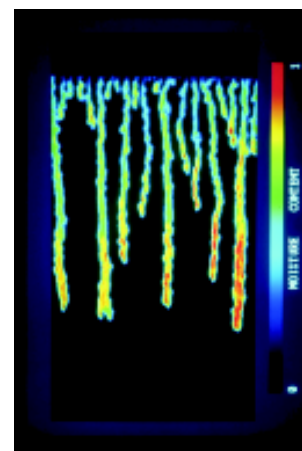


Figure 1b: Unstable pattern with finger flow. Both infiltration patterns are visualized with the light transmission technique.

Theory of fluid flow

The two relationships that characterize the physics and movement of water in soil are the pressure-saturation (P-S) curve and the conductivity curve (Figure 2). The P-S curve describes the relationship between the tension or capillary pressure by which the water is held in the porous medium (soil) and the water content (saturation) of the medium. The conductivity curve relates the rate of water movement (conductivity) through the medium and the water content of the medium.

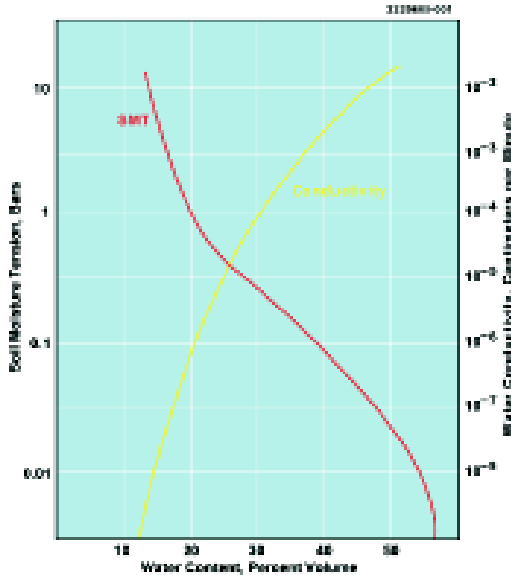


Figure 2: The two crucial soil-water relationships are the pressure saturation curve and the conductivity curve. The pressure saturation relationship gives the tension at which the water is held in the porous medium (soil) at a certain water content prevailing in the medium. The conductivity describes the rate of water movement (velocity) through the medium at a certain water content.

As fingering is essentially a fast transport phenomenon, rapid measurements of the saturation and fluid pressure in the soil are needed to understand the physics of the creation of the flow paths. Validation of the theoretical predictive fingering approaches formulated for water infiltrating air dry soil (two-phase or water and air) was attempted using the light transmission technique shown in Figure 1. A transparent chamber with a thin layer of soil between two walls is placed in front of a series of high fluorescent lights, and water is distributed uniformly on the surface. The more water in the sand, the greater the transmission of light, which allows us to quantify the water saturation and position as the fingers move downward in two dimensions.

Light transmission works only with coarse quartz sands in a slab chamber that is less than one-centimeter thick, and its accuracy is limited by light diffusion. In order to obtain higher accuracy and study different systems of fluids, high-energy synchrotron X-rays (courtesy of CHESS) were substituted for the fluorescent light rays. At CHESS, water saturations during fingering were studied using two-

dimensional soil slabs as shown in Figure 3. The slab chambers were mounted on an x-y moveable stage, which allowed us to specify locations. The distribution of the water and oil saturations over time were then monitored using monochromatic radiation. The attenuation of high energy X-ray radiation is linearly related to the material within the radiation's path [5]:

$$A = \ln(T/I) = -\sum_i U_i x_i$$

where A is the measured attenuation defined as the natural logarithm of the transmitted intensity T, divided by the incident intensity I. The sum is over each material in the radiation's path, where U_i is the attenuation constant and x_i is the cumulative thickness of each material. Substituting in the relevant materials for multiphase measurements in a porous media of total thickness x gives:

$$A = -U_{H_2O} \theta_{H_2O} x - U_{air} \theta_{air} x - U_{soil} \theta_{soil} x - U_c + D$$

Here, x is the thickness of the experimental chamber and the θ values are the volumetric saturations for each phase. Attenuation by the chamber walls and the air between the detectors is given by the constant U_c . The term D is a constant offset that occurs if relative, rather than absolute, intensities are measured. Attenuation by the air inside the chamber can be considered negligible. In two-phase flow situations where there is only one independent quantity, for example, air in partially saturated soil, the equation can be inverted to yield the fluid saturations in terms of measured attenuation,

$$\theta_w = -\frac{A - A_0}{U_w x}$$

where A_0 is the attenuation measured for the dry soil. Notice that only changes in the attenuation enter into the fluid determination, and knowledge of the individual contributions of D and U to the attenuation is unnecessary.

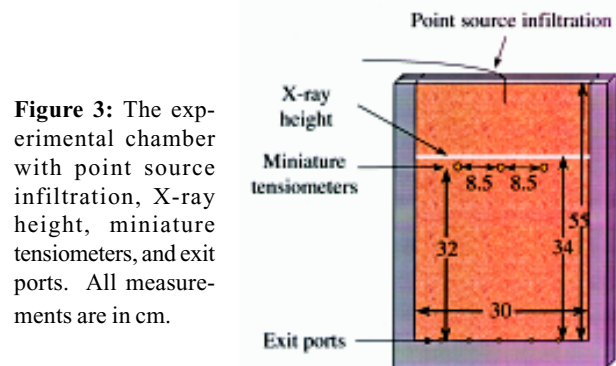


Figure 3: The experimental chamber with point source infiltration, X-ray height, miniature tensiometers, and exit ports. All measurements are in cm.

Initial work performed at CHESS on the two-phase systems resulted in the first confirmation of a key element of our theoretical model. Previously, using the standard light transmission technique, we had been unable to answer decisively if the tips of the fingers were saturated. At CHESS we proved that the tips of an infiltrating finger were indeed saturated. Another important discovery was that when collecting data for the pore saturation and capillary pressure simultaneously, we could derive the important conductivity

relationship for a soil within one minute. That was due primarily to the ability to acquire data on a one-second basis, compared with the labor-intensive alternative technique, which takes one month to deliver the same relationship.[8]

Dual energy setup

With the success of using CHESS to study water-air systems, we decided to apply the measurement technique to more complicated flow problems, which could not be studied using any other technique. This included three-phase flow, when a contaminating separate phase fluid (e.g. gasoline in the soil from a leaking service station) is simultaneously flowing, and also when the soil structure changes with water content, which occurs for many clay soil types. In both of these systems, where three components are variable (water, oil and air; or water, soil and air), another independent measurement is needed to characterize the system. Measuring the attenuation of radiation of a different energy will provide this (dual energy setup). In a sample in which saturations of water, oil, and air are variable, the fluid saturations can be determined from the attenuation of the high (A_H) and low (A_L) energy radiation [9]

$$\theta_w = \frac{U_{OL}(A_H - A_{HO}) - U_{OH}(A_L - A_{LO})}{(U_{WL}U_{OH} - U_{WH}U_{OL})x}$$

$$\theta_o = \frac{U_{WH}(A_L - A_{LO}) - U_{WL}(A_H - A_{HO})}{(U_{WL}U_{OH} - U_{WH}U_{OL})x}$$

where the subscripts H and L denote the high- and low-energy attenuation constant and A_{H0} and A_{L0} are the high and low attenuations, respectively, for the dry sand. These are the saturation equations for any dual-energy attenuation apparatus. Simultaneous phase selective capillary pressure measurements are needed to obtain the fundamental soil relationships between capillary pressures and soil pore saturation (P-S curves). Initially it seemed promising that results from the two-phase systems could be easily expanded to the three-phase systems. However, accurate monitoring of fast occurring flow instabilities within three mobile phases (such as water, light oil, and air) in porous media are inherently complex. Unfortunately, traditional dual gamma rays are unsuitable because of their low source intensities and non-tunable source energies, which are too slow. An alternate technique was developed, which uses synchrotron X-rays from CHESS to measure three-phase fluid saturations on the time scale of seconds. Using the harmonic content resulting from X-ray diffraction, we obtained a high intensity X-ray beam consisting of distinct tunable energies (dual energy X-rays, Figure 4). [6,8] This mode can be selected by the CHESS USER on stations with a double crystal monochromator by tuning to the peak of the rocking curve. Normally the crystals are detuned 50% to eliminate the harmonic.

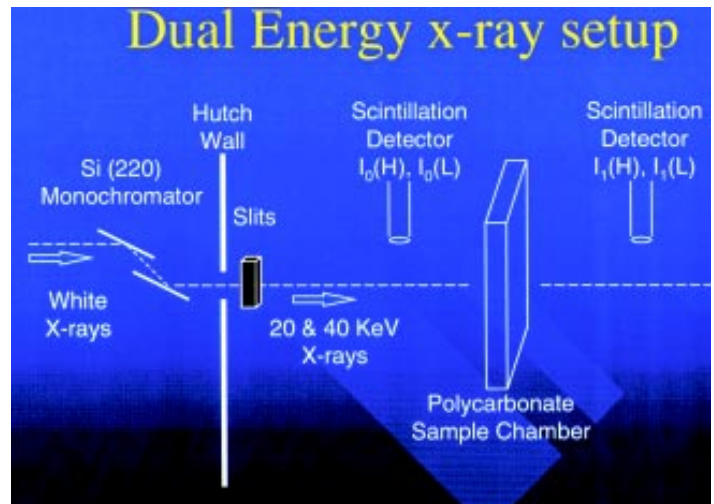


Figure 4: A schematic of the experimental set up at the F-2 hutch of CHESS. The monochromator transforms the white X-ray beam into a multiple-energy beam. Scintillation detectors measure the intensity of both energy X-rays by detecting air scattering from the main beam. The sample chamber is mounted on a movable platform allowing measurements from many different positions within the chamber.

Figures 5 and 6 give us an insight into three-phase flow and show the strength of using synchrotron X-rays. In the three-phase studies, it can be important to know which of the other 2 phases exits a region when a third phase enters the region.

Measurements were taken on a 5 second interval, at the same time 3 different volumetric fractions were measured: the newly introduced oil, the present water, and air. The oil and water phases were directly measured, and the air phase is the result of a subtraction since the total measured volume does not change over time. The sand particles remain in position during the experiment and thus a measurement before the oil infiltration gives us that volumetric fraction. The difference between the experiments is that the quartz sand used in Figure 6 is of a finer fraction (30-40 US sieve sizes) than the one in Figure 5 (20-30 US sieve sizes). While the oil infiltrating in Figure 6 causes no change in the present water content, it does cause a 5% decrease in volumetric water content of the water phase. Comparing both figures, it is obvious that with this particular synchrotron setup, very fast and small volumetric changes can be easily observed and documented.

Results

This experimental setup gave us the idea that we could study more complex soils than the idealized, homogeneous sands used previously. [4] We could perform a whole range of new experiments where the moisture content of the porous media might be varying. This was first tried with prewetted

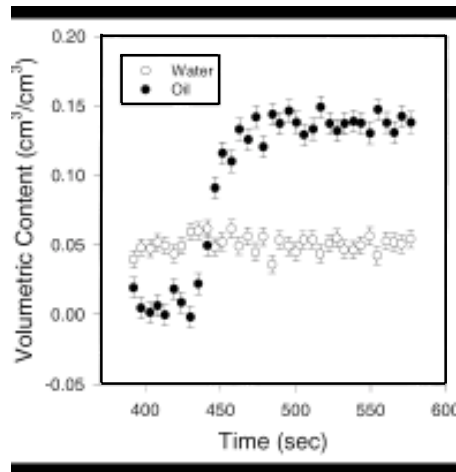


Figure 5: Synchrotron data from CHESS. Oil infiltrating in residual water at 5% volumetric water content, no drop in water content.

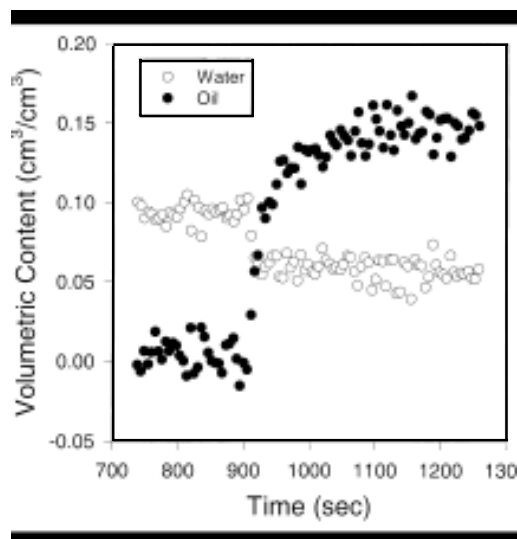


Figure 6: Oil infiltrating into $\pm 10\%$ water content with the passing of the water at time=900 s. to cause a drop in water content to 5% volumetric water content.

sands. [3] A series of experiments was conducted where water was infiltrated in a coarse sand with initial water contents of 0, 0.5, 1, 1.5, 2, 3, 4 and 4.7%. Both water content and capillary pressure were measured in order to determine the pressure-saturation curves. Water content was measured with synchrotron X-rays at CHESS and capillary pressure with fast responding tensiometers. The flow field changed gradually from unstable fingered flow at 0% initial water content to a stable wetting front at 4% and higher initial water contents. For unstable flow, the capillary pressure and water content decreased a short distance behind the wetting front. Except in the initially dry soil, the finger pattern began to widen immediately after infiltration. In general, this research in prewetted sands has shown that in coarse sandy soils, fingered flow occurred with both the capillary pressure and water content dropping behind the wetting front. [3] When stable fronts occurred, the capillary pressure potential and water content increased

(slightly) behind the wetting front (Figure 1).

Using water repellent soils produced different results than those found in previous experiments. [1] First of all, the organic matter present in the real soil determines in a large part the wettability of a soil and changes the surface tension of the infiltrating water. A 20% drop in the surface tension of the drainage water was noted when organic material is present in the soil. Second, the water-repellent soils used had a wide range of particle sizes, which created additional difficulty packing the soil in the chamber, due to separation. A square column was built to minimize those effects during experimentation.

Although it is generally well known that water repellent soils have preferential (fingered) flow patterns, the physics of this phenomenon is not well understood. We show that water repellency affects the wettability and this, in turn, has a distinct effect on the fundamental pressure-saturation curves during infiltration. Using these pressure-saturation curves, unstable flow theory developed for coarse-grained soils (two-phase systems, water-air) can be used to predict the shape and water content distribution for water repellent soils. [2]

Conclusion

We have tried to give an insider's view of the discoveries achieved in the field of fingered flow and soil science using powerful, fast synchrotron X-rays. The discoveries started with the early work on two-phase flow systems, where the researchers validated the theoretical assumption of a saturated finger tip and laid the basis for a quick measurement technique of the conductivity curve, which previously took up to a couple of months. From the two-phase systems we moved on to the three-phase systems and a new setup was devised and tested. The dual energy synchrotron X-ray setup gave us the ability to perform very fast (on the order of seconds) spatial and multi-phase measurements. This resulted in acquiring data where oil (NAPL) displacements were measured. The dual energy technique gave us a never before seen insight in prewetted soils, where the change from unstable preferential flow to stable flow was found by increasing the initial moisture content. Lastly, it enabled us to study naturally occurring soils with organic material, which gave us a more in-depth understanding of the physics behind water repellent soils.

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References:

- [1] Bauters, T.W.J., DiCarlo, D.A., Steenhuis, T.S. and Parlange, J.-Y. Preferential Flow in Water Repellent Sands. *Sci. Soc. Am. J.*, **62**, 1185-1190 (1998)
- [2] Bauters, T.W.J., Steenhuis, T.S., DiCarlo, D.A., Nieber, J.L., Dekker, L.W., Ritsema, C.J., Parlange, J.-Y. and Haverkamp, R. Physics of Water Repellent Soils. *J. Hydrol.* **231-232**:233-243 (2000)
- [3] Bauters, T.W.J., DiCarlo, D.A., Steenhuis, T.S. and Parlange, J.-Y. Soil Water Content Dependent Wetting Front Characteristics in Sands *J. Hydrol.* **231-232**:244-254
- [4] Charlie, W.A., Durnford, D. and Steenhuis, T.S. Rapid Density Profiling of Consolidating Clay Using Synchrotron Radiation. *ASTM Geotechnical Testing Journal* **20**(3):340-346 (1997)
- [5] Cullity, B.D. Elements of X-ray Diffraction, *Addison-Wesley Publishing Co., Reading, Mass* (1956)
- [6] DiCarlo, D.A., Bauters, T.W.J., Steenhuis, T.S., Parlange, J.-Y. and Bierck, B.R. High-Speed Measurements of Three-Phase Flow Using Synchrotron X-Rays. *Water Resources Research* **33**(4):569-576 (1997)
- [7] DiCarlo, D.A., Bauters, T.W.J., Darnault, C.J.G., Steenhuis, T.S. and Parlange, J.-Y. Rapid Determination of Constitutive Relations with Fingered Flow. Proceedings *International Workshop on Characterization and Measurement of the Hydraulic Properties of Unsaturated Porous Media*. Riverside, CA. October 22-24, 1997. pp. 433-440
- [8] Garnier, P., Angulo-Jaramillo, R., DiCarlo, D.A., Bauters, T.W.A., Darnault, C.J.G., Steenhuis, T.S., Parlange, J.-Y. and Baveye, P. Dual-Energy Synchrotron X-Ray Measurements of Rapid Soil Density and Water Content Changes in Swelling Soils During Infiltration. *Water Resources Research* **34**:2837-2842 (1998)
- [9] Oostrom, M.H., Dane, J.H., Missildine, B.C. and Lenhard, R.J. Error Analysis of Dual-Energy Gamma Radiation Measurements. *Soil Sci.*, **160**, 28-42 (1995)