GEOPHYSICAL RESEARCH LETTERS, VOL. 31, L08502, doi:10.1029/2003GL019282, 2004

Linking pressure and saturation through interfacial areas in porous media

J.-T. Cheng, Laura J. Pyrak-Nolte,[1] David D. Nolte, and Nicholas J. Giordano
Department of Physics, Purdue University, West Lafayette, Indiana, USA

Received 15 December 2003; revised 1 March 2004; accepted 24 March 2004; published 22 April 2004.

[1] Using transparent microfluidic cells to study the two-phase properties of a synthetic porous medium, we establish that the interfacial area per volume between nonwetting and wetting fluids lifts the ambiguity associated with the hysteretic relationship between capillary pressure and saturation in porous media. The interface between the nonwetting and wetting phases is composed of two subsets: one with a unique curvature determined by the capillary pressure, and the other with a distribution of curvatures dominated by disjoining pressure. This work provides experimental support for recent theoretical predictions that the capillary-dominated subset plays a role analogous to a state variable. Any comprehensive description of multiphase flow properties must include this interfacial area with the traditional variables of pressure and fluid saturation. *INDEX TERMS:* 1829 Hydrology: Groundwater hydrology; 1832 Hydrology: Groundwater transport; 1875 Hydrology: Unsaturated zone; 1899 Hydrology: General or miscellaneous. **Citation:** Cheng, J.-T., L. J. Pyrak-Nolte, D. D. Nolte, and N. J. Giordano (2004), Linking pressure and saturation through interfacial areas in porous media, *Geophys. Res. Lett.*, *31*, L08502, doi:10.1029/2003GL019282.

1. Introduction

[2] The problem of predicting and visualizing the interwoven distributions of multiple fluids in a porous medium is ubiquitous and crosses many disciplines. These include oil, gas and water production in the geosciences, ground water protection and contamination remediation in environmental science, chemical mixing and processing in chemical engineering, and in porous tissue applications of biology and bioengineering. In light of the crucial importance of multiple fluids in porous media, it is surprising that current research relies on work that has seen little improvement over 150 years. In 1856, Henri Darcy [*Darcy*, 1856] developed an empirical law for single-phase flow in a porous medium. Since that time, Darcy's law has been used in various extended forms to describe the flow of multiple immiscible fluid phases and applied to problems in diverse fields. Traditionally, Darcy's flow equation is written for each fluid phase in terms of fluid phase pressures and fluid saturations. Two constitutive equations are used to couple the two flow equations. One of the constitutive equations relates the pressure contrast between the fluid phases (capillary pressure) to saturation and the other equation relates relative permeability of a phase to the saturation of the phase. Saturation of each phase is the parameter that is common to both constitutive relations. Unfortunately, such descriptions of multiphase flow are known to fail badly in many cases [*van Genabeek and Rothman*, 1996; *Muccino et al.*, 1998].

[3] The failure of the extensions of Darcy's Law to multiple phases indicates that knowledge of the saturation of each phase is not sufficient to describe the state of the system. Several investigators [*Rapoport and Leas*, 1951; *Gvirtzman and Roberts*, 1991; *Bradford and Leij*, 1997; *Hassanizadeh and Gray*, 1990; *Gray and Hassanizadeh*, 1991, 1998a, 1998b] have recognized that an accurate description of multiphase flow in a porous medium must account for the thermodynamics and the geometry of the interfaces between the fluids and between the fluids and the solid phase. Fluid flow and the distribution of multiple fluid phases are controlled by the movement of the interfaces that results from changes in pressure and saturation. The physics of the interfaces is accounted for in these theories by explicitly introducing interfacial area per volume in combination with capillary pressure and saturation as an indicator of the thermodynamic energy state. Based on this hypothesis, the hysteretic relationship universally observed between capillary pressure and saturation for equilibrium conditions should be a unique and invertible relationship when interfacial area per volume is included.

[4] The experimental results described in this paper provide the first direct measurements of interfacial area, examines how it varies during multiphase imbibition and drainage, and verifies for the first time that interfacial area per volume does indeed lift the hysteretic ambiguity between capillary pressure and saturation. The experiments were performed on an artificial porous medium, often referred to as a "micro-model". A general review of micro-models and the associated scientific issues is given by *Giordano and Cheng* [2001].

2. Experimental Technique

[5] The micro-models we use are 2D porous media composed of a thin (1.08 microns) photo-sensitive polymer (photoresist) sandwiched between two glass slides. Regions of the plastic polymer are removed through photochemical reactions prior to making the sandwich, and these regions are accessible to the fluid(s) in our experiments. Details of the sample fabrication, as well as the measurement technique are given elsewhere [*Cheng*, 2002]. The fabrication process allows full choice of the geometry of the flow region. In our experiments we studied random porous geometries that are generated by distributing voids over a

---

[1] Also at Department of Earth and Atmospheric Sciences, Purdue University, West Lafayette, Indiana, USA.

0094-8276/04/2003GL019282$05.00

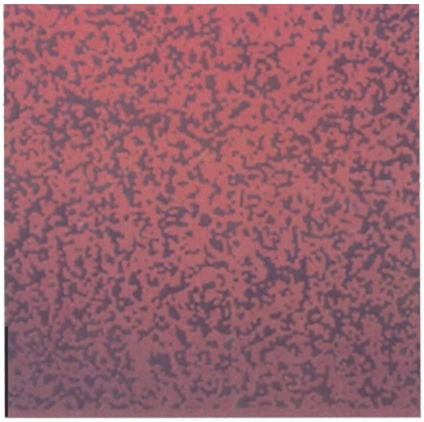

Figure 1. Photograph of a random micro-model filled with nitrogen gas (the bright red region) The solid, which is inaccessible to fluid, is the darker red (photoresist) region. The sample is approximately 0.6 mm on a side, and the height of the fluid filled regions is 1.08 micron. The porosity of the micro-model is 64%.

specified area [*Nolte and Pyrak-Nolte*, 1991]. The micro-models are completely transparent, which enables full visualization and observation of the fluid distributions. Figure 1 is a typical photograph (resolution 0.6 micron per pixel) of a random micro-model with a porosity of 64%.

[6] We measured fluid pressures while simultaneously recording photos of the sample so that the fluid distributions are known in detail. The experimental apparatus contained two pressure sensors to monitor the input and output pressures, and a video camera interfaced to an optical microscope to image the two-phase displacement experiments. This apparatus was used for simultaneous measurements of flow rate and optical characterization of the geometry of the various phases within the sample.

[7] To perform a displacement measurement on a micro-model, the micro-model is initially saturated with a fluid such as decane, which is inserted through the outlet region. A second fluid, nitrogen gas, is then introduced through the inlet region. All measurements are conducted at room temperature (temperature stability better than 0.5°C during a measurement), with the apparatus located within a clean bench environments. The measurement of interfacial area per volume (IAV) is accomplished with our video microscopy setup (CCD camera and microscope).

[8] To perform an experiment, the system was initially filled completely with decane. Nitrogen gas was then invaded into the micro-model by the application of pressure. This injection of gas was performed by increasing the pressure incrementally. After each pressure step the system was allowed to equilibrate. The pressure and a photograph of the fluid-filled micro-model were recorded, and then another pressure step was applied. Before the nitrogen reached the opposite side of the sample (breakthrough), the pressure was then reduced, again in a stepwise fashion, until most or all of the nitrogen gas retreated from the system.

3. Results

[9] Figure 2 shows the pressure-saturation history for a series of drainage-imbibition cycles. It is seen that the behavior is hysteretic, as the system does not follow a unique trajectory in terms of pressure and saturation. Even the drainage and imbibition branches are not unique, i.e., the system does not follow a unique trajectory during drainage and the same is true during imbibition. Note that during these measurements, the flow rate was zero, as the nitrogen did not extend fully from the inlet side of the sample to the outlet side. However, the strong hysteresis implies that knowledge of pressure and saturation does not fully specify the state of the system. As outlined above, there have been theoretical and numerical [*Reeves and Celia*, 1996; *Hassanizadeh and Gray*, 1993a, 1993b] assertions that interfacial area per volume is essential for a description of multiphase flow. The micro-models provide a unique opportunity to explicitly test this hypothesis.

4. Analysis

[10] The photographs at each stage in the imbibition and drainage were analyzed using codes developed with the IDL software package to compute the saturation level of decane, and the area of the interface per unit volume of sample separating nitrogen and decane. The interfaces between all three phases (wetting, non-wetting and solid (photoresist)) are identified. An example during the drainage phase of a random pattern is shown in part in Figure 3. The blue area is the wetting phase, and the red is the draining non-wetting phase. The interface identified between the non-wetting and the wetting phase is shown in white. and between the non-wetting phase and the photoresist in light red. The curvatures of the interfaces are calculated using level set methods [*Sethian*, 1985] and are histogrammed for the entire image in Figure 3b. The histogram for the full set of nonwetting interfaces is shown along with two subsets: the subset of capillary-dominated nonwetting-wetting interfaces (interfaces between bulk fluids) and the subset of nonwetting-solid interfaces. The striking feature that emerges in Figure 3b is the differentiation of the curvature into two distinct distributions. The subset of nonwetting-wetting interfaces has a nearly unique negative curvature (by convention), while the subset of nonwetting-solid interfaces has a broad distribution of curvatures, many of which are positive. Furthermore,

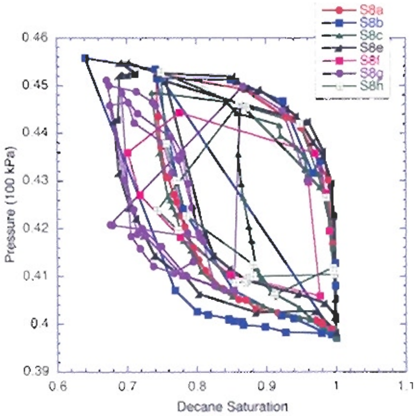

Figure 2. Pressure as a function of decane saturation for a series of drainage-imbibition cycles for the random sample shown in Figure 1. Clear history-dependent hysteresis in the Pressure-Saturation plane is observed.

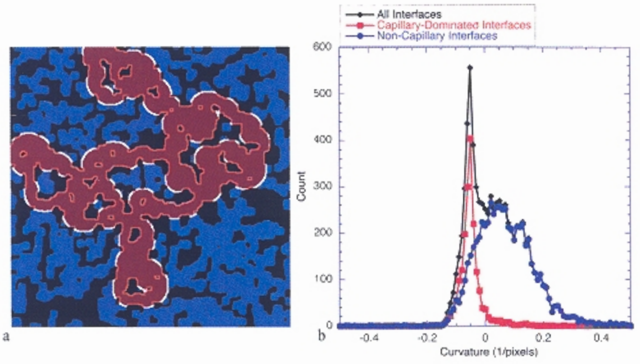

Figure 3. (a) An image of one-quarter of a micro-model sample partially saturated with decane (blue) and nitrogen (maroon) during drainage. Photoresist is represented by the black regions. The capillary-dominated interfaces are shown in white, and the disjoining-pressure dominated interfaces are shown in light red. (b) The histograms of the interface curvatures are for the all interfaces (black), for the subset dominated by disjoining pressure (non-capillary interfaces shown in blue) and for the subset that is capillary dominated (red).

the average curvature of the nonwetting-wetting interface agrees with measurements of capillary pressure.

[11] These results present an important finding of our micro-model and interface visualization work with regard to the critical issue of thin films and their role in porous media. Because we are invading into a pre-wet micro-model, a thin film of wetting phase remains between the non-wetting phase and the photoresist. Hence, there are strictly only two interfaces: between the non-wetting phase and the wetting phase which changes during imbibition-drainage cycles, and that between the wetting phase and the photoresist which is constant throughout the experiment. However, the lateral optical resolution (0.6 microns) of the imaging system prevents direct resolution of the thin film. In this context, it is evident that the striking difference between the curvature distributions presents a means of distinguishing between the physical processes that dominate the interfaces. For instance, the nonwetting-wetting interface can be divided unambiguously into two subsets, one that is dominated by capillary pressure (the narrow peak), and the other that is dominated by disjoining pressure (the broad distribution) associated with the proximity of the solid interface [*Deriagin et al.*, 1987]. The capillary-dominated interface is the interface between bulk fluids, while the disjoining-pressure-dominated interface is the interface between a bulk fluid and a fluid film.

[12] The capillary-dominated interfacial area per volume (IAV) values for the random pattern are shown in Figures 4 and 5 as functions of pressure (P) and saturation (S) along with all three 2D projections (Figure 5). Although each 2D projection (Figure 5) of the surface is hysteretic (P vs. S, IAV vs. S, and IAV vs. P), the surface is invertible: for a given degeneracy between two data points in P and S, the IAV differentiates between them. In other words, given any two measured parameters, the third can be found uniquely. Furthermore, the surfaces are unique for a given percolation pattern. When multiple data fall within small regions of the P-S plane, regardless from which of numerous cycles, we find they have similar IAV to within 5%. Different patterns will have different IAV surfaces, and hence the surfaces are not "universal", but the invertibility for each pattern provides a powerful new tool for the study of porous media.

5. Conclusions

[13] To the best of our knowledge, the results reported here are the first direct experimental quantification of the

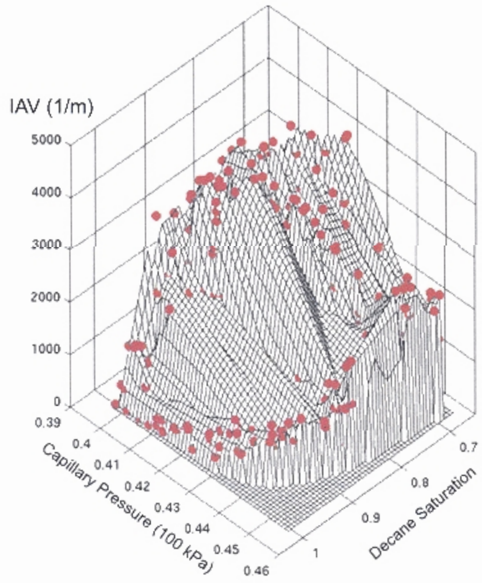

Figure 4. Interfacial area as a function of pressure and saturation. These results were obtained during the drainage-imbibition cycles shown in Figure 2. The surface is unique to within 5%.

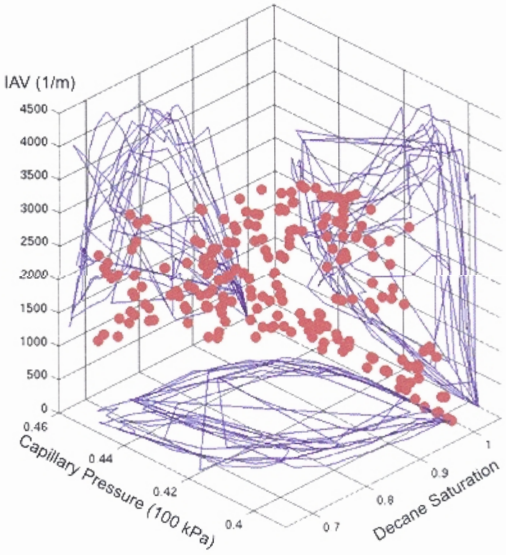

Figure 5. Interfacial area as a function of pressure and saturation. These results were obtained during the drainage-imbibition cycles shown in Figure 2. The data points are the same as those on shown in Figure 4 but the axes are rotated 220 degrees to show the strong hysteresis in all three projections.

interfacial area as a function of pressure and saturation for any random porous system. We have found that the interfacial area per volume of the capillary-dominated interfaces lifts the pressure-saturation hysteresis (and hence ambiguity) observed for most conditions of multiple fluids in porous media. Furthermore, the capillary-dominated IAV produces an invertible relationship among all three variables of IAV, P, and S. Hence, the results provide initial support for the theoretical and numerical arguments that IAV must be treated as a state variable in descriptions of multiphase flow. The work described here represents a fundamental advance in the field that will lead to important new insights into multiphase fluid flow in many types of random media across many disciplines.

[14] **Acknowledgments.** The authors wish to acknowledge the Department of Energy DE-AC26-99BC15207 Fossil Energy, NSF grant DMR-9970708 and Geosciences Research Program, Office of Basic Energy Sciences US Department of Energy (DE-FG02-97ER14785). LJPN and DDN wish to acknowledge University Faculty Scholar program at Purdue University.

[15] The authors wish to thank William G. Gray for very useful discussions.

References

Bradford, S. A., and F. J. Leij (1997), Estimating interfacial areas for multi-fluid soil systems, *J. Contam. Hydrol.*, *27*, 83–105.

Cheng, J.-T. (2002), Fluid flow in ultrasmall structures, Ph.D. thesis, Purdue Univ., West Lafayette, Indiana.

Darcy, H. (1856), Determination of the laws of flow through sand, in *Les Fontaines Publiques de la Ville de Dijon*, pp. 590–594, Victor Dalmont, Paris. (Translated and Reproduced in *Physical Hydrology*, edited by R. A. Freeze and W. Back, John Wiley, New York, 1983.)

Deriagin, B. V., N. V. Churaev, V. M. Muller, and J. A. Kitchener (1987), *Surface Forces*, translated from the Russian by V. I. Kisin, 440 pp., edited by J. A. Kitchener, Consult. Bur., New York.

Giordano, N., and J.-T. Cheng (2001), Microfluid mechanics: Progress and opportunities, *J. Phys. Condensed Matter*, *13*, R271–R295.

Gray, W. G., and S. M. Hassanizadeh (1991), Unsaturated flow theory including interfacial phenomena, *Water Resour. Res.*, *27*, 1855–1863.

Gray, W. G., and S. M. Hassanizadeh (1998a), Averaging theorems and averaged equations for transport of interface properties in multiphase systems, *Int. J. Multiphase Flow*, *15*, 81–95.

Gray, W. G., and S. M. Hassanizadeh (1998b), Macroscale continuum mechanics for multiphase porous-media flow including phases, interfaces, common lines and common points, *Adv. Water Res.*, *21*, 261–281.

Gvirtzman, H., and P. V. Roberts (1991), Pore scale spatial analysis of two immiscible fluids in porous media, *Water Resour. Res.*, *27*, 1165–1176.

Hassanizadeh, S. M., and W. G. Gray (1990), Mechanics and thermodynamics of multiphase flow in porous media including interphase boundaries, *Water Resour. Res.*, *13*, 169–186.

Hassanizadeh, S. M., and W. G. Gray (1993a), Thermodynamic basis of capillary pressure in porous media, *Water Resour. Res.*, *29*, 3389–3405.

Hassanizadeh, S. M., and W. G. Gray (1993b), Toward an improved description of the physics of two-phase flow, *Adv. Water Resour.*, *16*, 53–67.

Muccino, J. C., W. G. Gray, and L. A. Ferrand (1998), Toward an improved understanding of multiphase flow in porous media, *Rev. Geophys.*, *36*, 401–422.

Nolte, D. D., and L. J. Pyrak-Nolte (1991), Stratified continuum percolation: Scaling geometry of hierarchical cascades, *Phys. Rev. A*, *44*, 6320–6333.

Rapoport, L. A., and W. J. Leas (1951), Relative permeability to liquid in liquid-gas systems, *Pet. Trans.*, *192*, 83–98.

Reeves, P. C., and M. A. Celia (1996), A functional relationship between capillary pressure, saturation, and interfacial area as revealed by a pore-scale network model, *Water Resour. Res.*, *32*, 2345–2358.

Sethian, J. A. (1985), Curvature and the evolution of fronts, *Commun. Math. Phys.*, *101*, 487–499.

van Genabeek, O., and D. H. Rothman (1996), Macroscopic manifestations of microscopic flows through porous media: Phenomenology from simulation, *Annu. Rev. Earth Planet. Sci.*, *24*, 63–87.

J.-T. Cheng, N. J. Giordano, D. D. Nolte, and L. J. Pyrak-Nolte, Department of Physics, Purdue University, 525 Northwestern Avenue, West Lafayette, IN 47907-2036, USA. (ljpn@physics.purdue.edu)