Modelling two-phase flow and transport in industrial porous media

S. Majid Hassanizadeh
Department of Earth Sciences
Utrecht University
The Netherlands

Collaborators:
Simona Bottero; Utrecht University, The Netherlands
Jenny Niessner; Utrecht University, The Netherlands
Vahid Joekar-Niasar; Utrecht University, The Netherlands
Cas Berentsen; Delft University of Tech., The Netherlands
Rainer Helmig; University of Stuttgart, Germany
Michael A. Celia; Princeton University, USA
Helge K. Dahle; University of Bergen, Norway
Outline of presentation

Introduction:
  Darcy’s law for two-phase flow in porous media
  Capillary pressure in porous media
  Shortcomings of standard capillary pressure theory

Experimental evidences

New theory of capillarity

Significance of new theory

New theories of two-phase flow

Conclusions
INTRODUCTION

Theories of (multiphase) flow in porous media have been too empirical in nature.

They have been developed through “extensions” of simple relationships in ad-hoc fashion.

They are not necessarily valid for complex systems we are considering today.

Take Darcy’s law.
DARCY TESTED HIS FORMULA ON A COLUMN OF SAND IN A LABORATORY AND DETERMINED THE VALUE OF CONDUCTIVITY (K):

\[ Q = KA \frac{\Delta h}{L} \]
Simple System

Complex System
# METAMORPHOSIS OF DARCY’S LAW

| Darcy’s law was proposed for one-dimensional steady-state flow of almost pure incompressible water in saturated homogeneous isotropic rigid sandy soil under constant temperature | It is now used in the same form for three-dimensional unsteady flow of two or more compressible fluids, with lots of dissolved matter, in anisotropic heterogeneous deformable porous media under non-isothermal conditions |
Write the formula in terms of velocity \( q = \frac{Q}{A} \)
METAMORPHOSIS OF DARCY’S LAW

FOR ONE-DIMENSIONAL FLOW, Change $\frac{\Delta h}{L}$ to $-\frac{dh}{dx}$

$q \equiv -K \frac{dh}{dx}$
METAMORPHOSIS OF DARCY’S LAW

For unsteady state conditions: \( h = h(x, t) \)

\[ q = -K \frac{\partial h}{\partial x} \]
METAMORPHOSIS OF DARCY’S LAW FOR THREE-DIMENSIONAL FLOW

\[ q_i = -K \frac{\partial h}{\partial x_i} \]
METAMORPHOSIS OF DARCY’S LAW

FOR ANISOTROPIC MEDIA, REPLACE $K$ BY $K_{ij}$

$$q_i = - K_{ij} \frac{\partial h}{\partial x_j}$$
METAMORPHOSIS OF DARCY'S LAW FOR HETEROGENEOUS MEDIA

\[ K_{ij} \]

is assumed to be a function of position
METAMORPHOSIS OF DARCY’S LAW

FOR FLOW IN UNSATURATED ZONE:

\[ q_i = -K_{ij} \frac{\partial h}{\partial x_j} \]
METAMORPHOSIS OF DARCY’S LAW

FOR FLOW IN UNSATURATED ZONE:

\[ K_{ij} \]

is assumed to be a function of water content \( \theta \).
METAMORPHOSIS OF DARCY’S LAW

FOR TWO-PHASE FLOW (say WATER AND OIL):

\[ q = \frac{k}{\alpha} \]
METAMORPHOSIS OF DARCY’S LAW

FOR TWO-PHASE FLOW (say WATER AND OIL):

\[ q_i^w = - K_{ij}^w \frac{\partial h^w}{\partial x_j} \]
METAMORPHOSIS OF DARCY’S LAW

FOR TWO-PHASE FLOW (SAY WATER AND OIL):

\[ q_i^w = -K_{ij}^{w} \frac{\partial h^w}{\partial x_j} \]

\[ q_i^o = -K_{ij}^{o} \frac{\partial h^o}{\partial x_j} \]
METAMORPHOSIS OF DARCY’S LAW

FOR TWO-PHASE FLOW (say WATER AND OIL):

\[ K_{ij}^w = k_r^w K_{ij} \]

\[ K_{ij}^o = k_r^o K_{ij} \]
Capillary Pressure-saturation relationship

\[ h^o - h^w = \frac{p^c(S)}{\rho^o g} + \left( 1 - \frac{\rho^w}{\rho^o} \right) (z - h^w) \]

The form of Darcy’s Law remains unchanged:

\[ q^\alpha_i = - K^\alpha_{ij} \frac{\partial h^\alpha}{\partial x_j} \]

We are adding bells and whistles to a simple formula to make it valid for much more complicated situations!
“Extended” Darcy’s Law

\[ q = K \frac{\Delta h}{\Delta x} \]

The “extended” Darcy’s Law remains unchanged:

\[ q_i^\alpha = - K_{ij}^\alpha \frac{\partial h^\alpha}{\partial x_j} \]

We have added bells and whistles to a simple formula to make it valid for much more complicated situations!
Comparison of theories of Aristotle and Galileo for free fall of objects with hypothetical measurements (after French, 1990)
PhD RESEARCH PROJECT

Hypothetical plot of terminal velocity as a function of mass density for various shapes.
How many relative permeability-saturation curves have been measured during the past few decades!?
How many capillary pressure-saturation curves have been measured during the past few decades!? 

- **Imbibition scanning curves**
- **Drainage scanning curves**
Relative permeability is supposed to be less than 1.

Water and oil relative permeabilities in 43 sandstone reservoirs plotted as a function of water saturation.
Problems with current theories of multiphase flow

Pressure gradient and gravity are assumed to be the only driving forces. Effects of all other forces are lumped into coefficients.

In particular, interfacial forces and interfacial energies are not explicitly taken into account. This is unacceptable.

There is discrepancy between the theory and measurement.

Capillary pressure curves are measured under equilibrium conditions and then used under non-equilibrium (flow) conditions.

Capillary pressure is believed to go to infinity. Nonsense!
Forces acting within interfaces control fluid distribution within the porous medium.

\[ p^c = \frac{2\sigma}{R_M} \]
Mass transfer among phases occurs through interfaces.
Where are interfaces in porous media theories!?
Problems with current theories of multiphase flow

Pressure gradient and gravity are assumed to be the only driving forces. Effects of all other forces are lumped into coefficients.

In particular, interfacial forces and interfacial energies are not explicitly taken into account. This is unacceptable.

There is discrepancy between the theory and measurement.

Capillary pressure curves are measured under equilibrium conditions and then used under non-equilibrium (flow) conditions.

Capillary pressure is believed to go to infinity. Nonsense!
Capillary pressure-saturation curves for a given medium are obtained by measurement of saturation inside and fluid pressures outside the porous medium under quasi-equilibrium conditions. Time to equilibrium can be 24 hours to 7 days. Hydrophobic and hydrophilic membranes affect the results.

\[ p^c = p^a - p \]
Many industrial processes involve porous media flows.

Some examples are processes in fuel cells, paper-pulp drying, food production and safety, filtration, concrete, ceramics, moisture absorbents, textiles, paint drying, polymer composites, and detergent tablets.

There is a clear difference between industrial porous media and soils:
- fast flow
- significant mass transfer across fluid-fluid interfaces
- significant deformations
- wettability changes
- high porosity
Dewatering of paper pulp

The paper pulp layer is transported through the press aperture on a felt. The paper-felt sandwich is thus compressed, so that the water is pressed out of the paper, is absorbed by the felt, and then flows laterally away from the nips.

Pictures courtesy of Oleg Iliev of ITWM, Kaiserslautern
Simulation of dewatering of paper pulp

Capillary pressure curves are measured for felt under quasi-static conditions. It takes around 24 hours to obtain them.

They are then used to model a process that takes only seconds!
Air and oil flow in oil filters

Relative Permeability and Capillary pressure

Pictures courtesy of Oleg Iliev of ITWM, Kaiserslautern
Pc-Sw measurement of hydrophobic gas diffusion layer of a PEM fuel cell
Unsaturated flow in thin fibrous materials
Unsaturated flow in thin fibers
Unsaturated flow in pampers

Measurement of capillary pressure curves takes about 12 hours.
Unsaturated flow in pampers
Capillary pressure-saturation experiments (PCE and Water)

Experiments carried out at GeoDelft; Soil sample: 3 cm high and 6 cm in dia. Fluids: PCE and water; Primary drainage, Main drainage, Main imbibition; Quasi-static and dynamic experiments
PC-S curves based on pressures measured inside the soil sample.

Capillary pressure DOES NOT go to infinity!

Are these the curves we should use in our simulators?
Measurement of “Dynamic Pressure Difference” Curves

\[ P^n - P^w = \Delta P_{\text{dyn}} \]

\[ P^c \]

Hassanizadeh et al., 2004
Two-phase flow dynamic experiments (PCE and Water)

- Prim drain PN ~ 16kPa
- Main imb PW ~ 0kPa
- Main imb PW ~ 5kPa
- Main imb PW ~ 8kPa
- Main imb PW ~ 10kPa
- Main imb PW ~ 0kPa last
- Main drain PN ~ 16kPa
- Main drain PN ~ 20kPa
- Main drain PN ~ 25kPa
- Main drain PN ~ 30kPa
- Static Pc inside

Graph showing saturation (S) vs. Pn-Pw (kPa) with different lines representing various pressure conditions.
Experiments on the uniqueness of $p^c - S$ curves
(drainage experiments by Topp, Klute, and Peters, 1967)

These are all drainage curves
There is no unique $p^c$-S curve.

- Dynamic Drainage Curves
- Main Scanning Drainage Curves
- Main Drainage Curves
- Secondary Scanning Drainage Curves
- Primary Drainage Curve
- Primary Imbibition Curve
- Main Imbibition Curve
- Main Scanning Imbibition Curves
- Secondary Scanning Imbibition Curves
- Dynamic Imbibition Curves
There is no unique $p^c$-S curve.
A New Theory of Capillarity in Porous Media

1. Difference in fluids’ pressures are equal to capillary pressure but only at equilibrium.

2. Capillary pressure is not only a function of saturation but also of interfacial area.
Forces acting within interfaces control fluid distribution within the porous medium.

\[ p^c = \frac{2\sigma}{R_M} \]

\[ p^n - p^w = p^c \]
Consider two-phase flow in a single tube:

\[
p^n - p^w = \frac{2\gamma}{R_M} + B \left( \frac{\mu q}{r^2} \right)^A
\]
NONEQUILIBRIUM CAPILLARY EFFECT

Linear theory

Equilibrium capillary theory:

\[ p^n - p^w = p^c (S) \]

Under non-equilibrium conditions:

\[ p^n - p^w = p^c + f(S, \partial S/\partial t) \]

Linear approx.:

\[ p^n - p^w = p^c - \tau(S) \frac{\partial S}{\partial t} \]

where \( \tau \) is a damping coefficient.
Dynamic capillary pressure curves

\[ p^c = \left( p^a - p^w \right) - p^c(S) = -\tau \frac{\partial S}{\partial t} \]

This equation captures the behavior observed in the experiments.

Curves measured in the lab can be used to evaluate \( \tau \)
Two-phase flow dynamic experiments (PCE and Water)

\( \frac{dS}{dt} \) vs. \( S \) - all curves

\( \frac{dS}{dt} \) vs. \( S \) - all curves
Two-phase flow dynamic experiments (PCE and Water)

Determination of $\tau$
Primary drainage; S=70%

$y = 54.0x + 1.2$
$y = 61.4x + 0.7$
$y = 50.0x + 0.4$

Pair = 16 kPa
Pair = 20 kPa
Pair = 25 kPa
Dynamic coefficient, $\tau$

<table>
<thead>
<tr>
<th>$S_w [-]$</th>
<th>$\tau [\text{Pa.s}]$</th>
</tr>
</thead>
<tbody>
<tr>
<td>0.80</td>
<td>$9.83 \times 10^4$</td>
</tr>
<tr>
<td>0.70</td>
<td>$1.06 \times 10^5$</td>
</tr>
<tr>
<td>0.60</td>
<td>$1.33 \times 10^5$</td>
</tr>
<tr>
<td>0.50</td>
<td>$2.61 \times 10^6$</td>
</tr>
<tr>
<td>0.40</td>
<td>$3.62 \times 10^7$</td>
</tr>
</tbody>
</table>
Estimation of $\tau$ from literature data

(Hassanizadeh, Celia, and Dahle, *Vadose Zone Journal*, 2002)

<table>
<thead>
<tr>
<th>Reference</th>
<th>Soil Type</th>
<th>Type of experiment</th>
<th>Value of $\tau$ (kg/m.s)</th>
</tr>
</thead>
<tbody>
<tr>
<td><strong>For Air-Water Systems:</strong></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td><em>Topp et al.</em> (1967)</td>
<td>Sand</td>
<td>Drainage</td>
<td>$2 \times 10^7$</td>
</tr>
<tr>
<td><em>Smiles et al.</em> (1971)</td>
<td>Sand</td>
<td>Drainage</td>
<td>$5 \times 10^7$</td>
</tr>
<tr>
<td><em>Stauffer</em> (1978)</td>
<td>Sand</td>
<td>Drainage</td>
<td>$3 \times 10^4$</td>
</tr>
<tr>
<td><em>Nützmann et al.</em> (1994)</td>
<td>Sand</td>
<td>Drainage</td>
<td>$2 \times 10^6$</td>
</tr>
<tr>
<td><em>Hollenbeck &amp; Jensen</em> (1998)</td>
<td>Sand</td>
<td>Drainage</td>
<td>2 to $12 \times 10^6$</td>
</tr>
<tr>
<td><strong>For Oil-Water Systems:</strong></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td><em>Kalaydjian</em> (1992b)</td>
<td>Sandst./limest.</td>
<td>Imbibition</td>
<td>$2 \times 10^6$</td>
</tr>
</tbody>
</table>
Significance of new theory for unsaturated flow: Development of vertical wetting fingers in dry soil

Experiments by Rezanejad et al., 2002
Consequences of new theory for unsaturated flow: Development of vertical wetting fingers in dry soil

David A. DiCarlo, “Experimental measurements of saturation overshoot on infiltration” VOL. 40, 2004
Consequences of new theory for unsaturated flow: Development of vertical wetting fingers in dry soil

David A. DiCarlo, “Experimental measurements of saturation overshoot on infiltration” VOL. 40, 2004
Classical unsaturated flow equation

Flow equation:

\[ n \frac{\partial S}{\partial t} = \frac{\partial}{\partial z} \left( \frac{k_r(S)k}{\mu} \left( \frac{\partial p^w}{\partial z} + \rho g \right) \right) \]

Capillary pressure-saturation relationship:

\[ p^a - p^w = p^c(S) \]

Richards equation:

\[ n \frac{\partial S}{\partial t} = \frac{\partial}{\partial z} \left( D \left( \frac{\partial S}{\partial z} + \rho g \right) \right) \]
Stability analysis by Dautov et al. (2002) has proven that:

**Richards equation is unconditionally stable.**
- It does not produce any fingers, but a monotonically decreasing saturation profile toward the wetting front.

**Modified Richards equation (with dynamic effect) is conditionally unstable.**
- It is able to produce gravity wetting fingers.
Dynamic capillary pressure theory

Unsaturated flow equations:

Flow equation:

\[ n \frac{\partial S}{\partial t} = \frac{\partial}{\partial z} \left( k_r(S)k \left( \frac{\partial p^w}{\partial z} + \rho g \right) \right) \]

Capillary pressure-saturation relationship:

\[ p^a - p^w = p_{stat}^c (S) - \tau \frac{\partial S}{\partial t} \]
Development of vertical wetting fingers in dry soil; Simulations based on new capillarity theory

Dautov et al. (2002)
1. Difference in fluids’ pressures are equal to static capillary pressure but only at equilibrium.

2. Capillary pressure is not only a function of saturation but also of interfacial area.
Interfacial area and saturation are two independent properties

\[ r = R / 2 \]

\[ V_1 = \frac{4}{3} \pi R^3 \]
\[ p_1^c = \frac{2\sigma}{R} \]
\[ a_1^{wn} = \frac{3}{R} \]

\[ V_2 = V_1 = 8 \times \frac{4}{3} \pi r^3 \]
\[ p_2^c = \frac{2\sigma}{r} = 2p_1^c \]
\[ a_2^{wn} = 8 \times \frac{3}{r} = 16a_1^{wn} \]
CAPILLARY PRESSURE-SATURATION DATA POINTS
MEASURED IN LABORATORY
CAPILLARY PRESSURE-SATURATION DATA
POINTS MEASURED IN LABORATORY
New capillarity theory with no (need for) hysteresis

\[ p^c = p^c (S, a^{wn}) \]

where:

- \( p^c \) is capillary pressure
- \( S \) is wetting phase saturation
- \( a^{wn} \) is the specific interfacial area between wetting & nonwetting phases
$p^c - S - a^{wn}$ Relationships

Figures courtesy of Laura Pyrak-Nolte of Purdue University
Imbibition

Drainage

Figure courtesy of Laura Pyrak-Nolte of Purdue University
Comparison of Drainage & Imbibition Surfaces

Mean Relative Difference: -3.7%
Standard Deviation: 10.6%
Capillary pressure at macroscale

\[ a_{wn} = a_{wn}(S_w, p_c) \]
\[ a_{wn}(S_w, p_c) = (a_{00} + a_{10}S_w + a_{01}p_c + a_{20}S_w^2 + a_{11}S_w \cdot p_c + a_{02}p_c^2) \]

\[
\begin{align*}
    a_{00} &= -358 \ [1/\text{m}] \\
    a_{10} &= 5535 \ [1/\text{m}] \\
    a_{01} &= 0.085 \ [1/(\text{m} \cdot \text{Pa})] \\
    a_{20} &= -3937 \ [1/\text{m}] \\
    a_{11} &= -0.307 \ [1/(\text{m} \cdot \text{Pa})] \\
    a_{02} &= -5.2 \cdot 10^{-10} \ [1/(\text{m} \cdot \text{Pa}^2)]
\end{align*}
\]

(data obtained from Vahid Joekar-Niasar et al. 2007)
New theories of two-phase flow

Extended Darcy’s law:

\[ q^\alpha = -\rho^\alpha K^\alpha \cdot (\nabla G^\alpha - g) \]

where \( G^\alpha \) is the Gibbs free energy of a phase:

\[ G^\alpha = G^\alpha \left( \rho^\alpha, a^{wn}, S^\alpha, T \right) \]

Extended Darcy’s law:

\[ q^\alpha = -K^\alpha \cdot \left( \nabla p^\alpha - \rho^\alpha g - \sigma^{\alpha a}\nabla a^{wn} - \sigma^{\alpha S}\nabla S^\alpha \right) \]

Equation of motion for interfaces:

\[ w^{wn} = -K^{wn} \left[ \nabla \left( a^{wn} \gamma^{wn} \right) + \Omega^{wn} \nabla S^w \right] \]
New theories of two-phase flow

Volume balance equation for each phase

\[ n \frac{\partial S^\alpha}{\partial t} + \nabla \cdot q^\alpha = 0 \]

Area balance equation for fluid-fluid interfaces

\[ \frac{\partial a^{wn}}{\partial t} + \nabla \cdot \left( a^{wn} w^{wn} \right) = r^{wn} \left( a^{wn}, S^w \right) \]
Summary of two-phase flow equations

\[ n \frac{\partial S^\alpha}{\partial t} + \nabla \cdot q^\alpha = 0 \]

\[ q^\alpha = -K^\alpha \cdot \left( \nabla p^\alpha - \rho^\alpha g - \sigma^{\alpha \omega} \nabla a^{wn} - \sigma^{\alpha S} \nabla S^\alpha \right) \]

\[ \frac{\partial a^{wn}}{\partial t} + \nabla \cdot \left( a^{wn} w^{wn} \right) = r^{wn} \left( a^{wn}, S^w \right) \]

\[ w^{wn} = -K^{wn} \left[ \nabla \left( a^{wn} \gamma^{wn} \right) + \Omega^{wn} \nabla S^w \right] \]

\[ p^n - p^w = p^c - \tau \frac{\partial S^w}{\partial t} \]

\[ p^c = f \left( S^w, a^{wn} \right) \]
CONCLUSIONS

Darcy’s law is not really valid for complex porous media.

Difference in fluid pressures is equal to capillary pressure but only under equilibrium conditions.

Under nonequilibrium conditions, the difference in fluid pressures is a function of time rate of change of saturation as well as saturation.

Fluid-fluid interfacial areas should be included in multiphase flow theories.

Hysteresis should be modelled by introducing interfacial area into the two-phase flow theory.