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Water Transport in Gas Diffusion Layers

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Acknowledgements: ***Shawn Litster***
Bojan Markicevic
David Sinton

Hydrogen Fuel Cells Workshop, Ottawa, May 10-12, 2006

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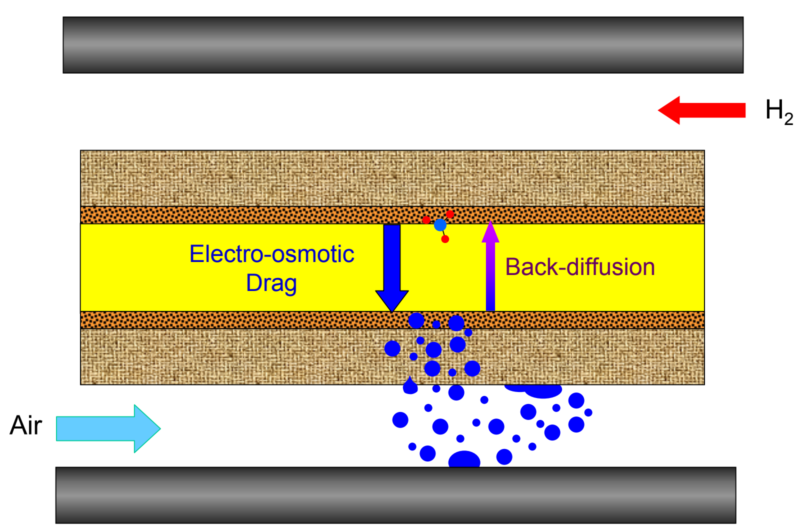
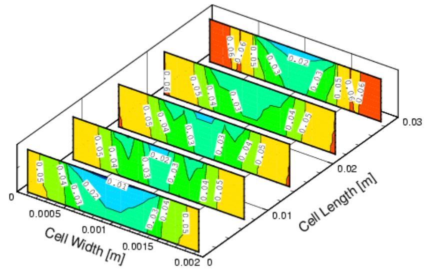
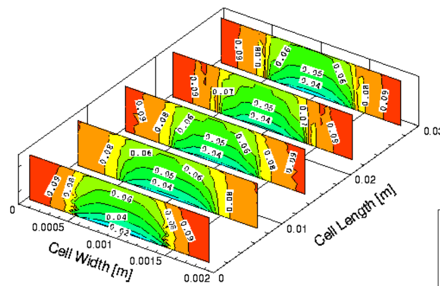
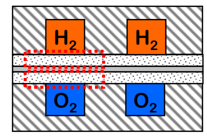
Water Transport



3D CFD Example: Saturation



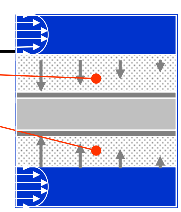




T. Berning and N.D,
J. Electrochem. Soc. Vol. **150**, no. 12, pp. A1589A1598

University of Victoria

Transport in GDL: Modelling Framework



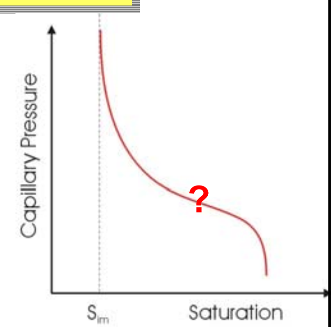
Flux in the GDL

$$\mathbf{u}_g = -\frac{k_p^g}{\mu_g}\nabla p_g = -(1-s)^n \frac{k_p^0}{\mu_g}\nabla p_g$$

$$\mathbf{u}_l = -\frac{k_p^l}{\mu_l}\nabla p_l = -s^n \frac{k_p^0}{\mu_l}\nabla p_g + s^n \frac{k_p^0}{\mu_l}\frac{\partial p_c}{\partial s}\nabla s$$

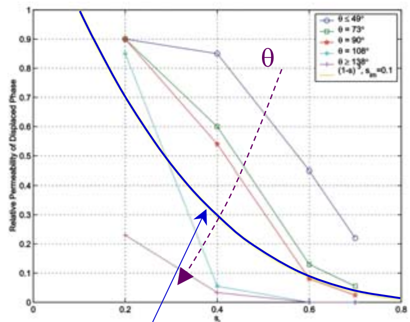
with $$p_c = \sigma\left(\frac{\varepsilon}{k_p^0}\right)^{1/2} f(s)$$

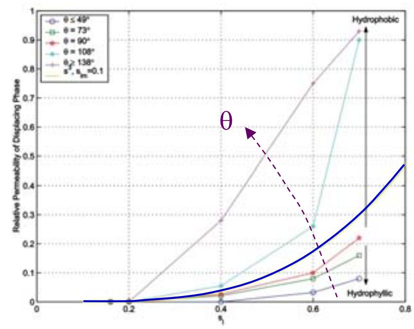
$$f(s) = 1.417(1-s) - 2.12(1-s)^2 + 1.263(1-s)^3$$



Relative Permeability & Hydrophobicity

k_{rg} of displaced phase (gas) | k_{rl} displacing phase (water)





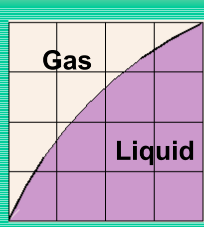
$(1 - s)^3$ and $s_{im,l} = 0.1$

Anderson, J. Petrol. Tech., 1453-14681987

Some Issues & Approach

- Complex geometry and interfaces
- Capillary pressure J-function for PEM GDLs
- Relative permeability
- Water transport mechanisms/dynamics

Dynamic VOF Simulations



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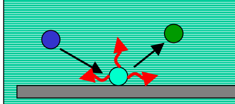
Fluorescence Microscopy Visualization



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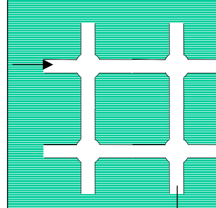
Pore Network Modelling



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Dynamic Simulations in Reconstructed GDL

Solutions are obtained using Fluent 6.2:

- Volume-of-Fluid (VOF) method
- Sub-models for surface tension and wall adhesion
- Re-constructed porous media (RPM) for computational geometry

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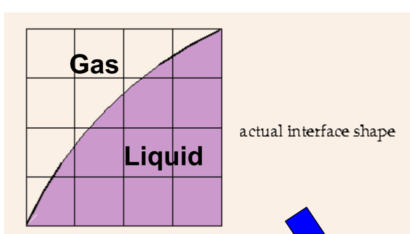
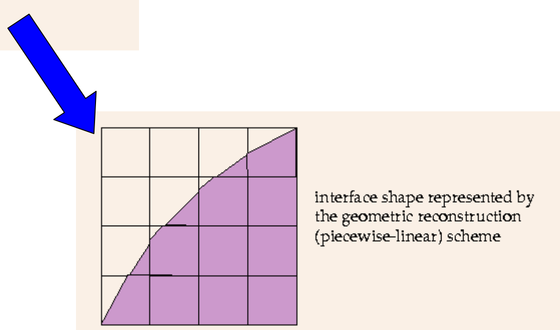
Volume-of-Fluid Model

Static-grid Eulerian representation





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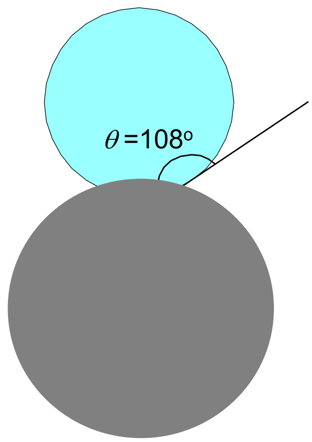
Surface Tension and Wall Adhesion



• Interface represented using a force density proportional to interface curvature [15]

→ Simplifies calculation of surface tension

• Three phase boundary is key feature

→ Contact angle condition is met by calculating the curvature of the interface

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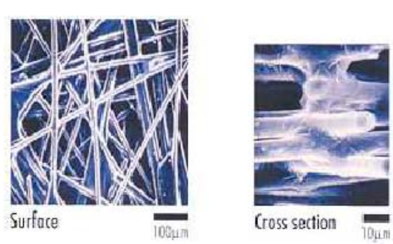
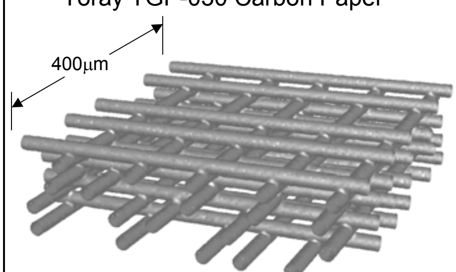
Reconstruction of the GDL

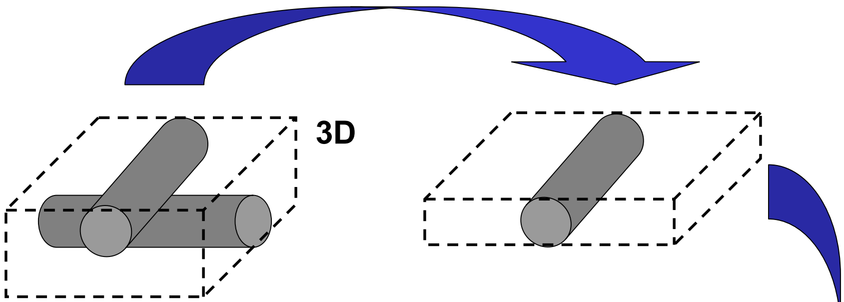
SEM Images of Toray TGP Series Carbon Paper*



Computational Model of Toray TGP-030 Carbon Paper

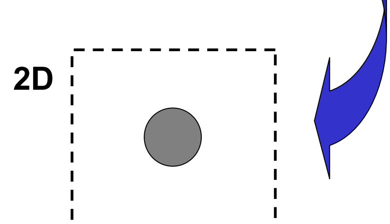


Geometry – Elementary Volumes

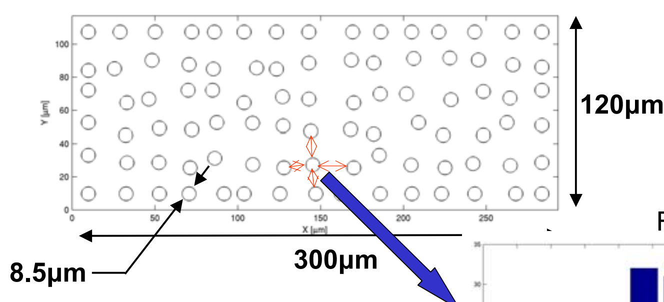


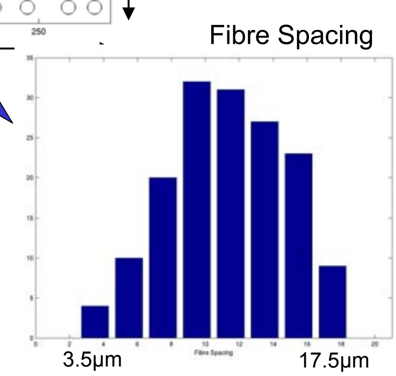
Transformation Constraints:

- Equal porosity.
- Equal surface area to volume ratio.
- Equal curvature.

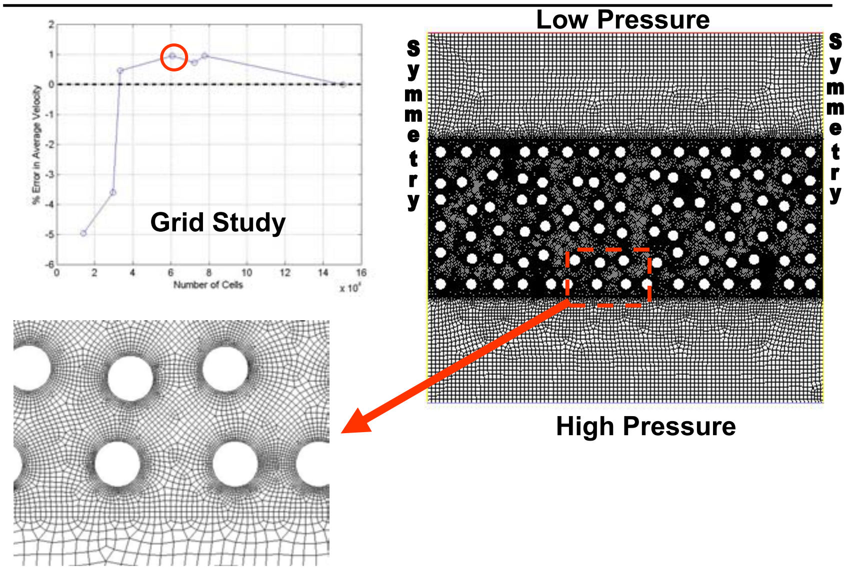


Fibre Array

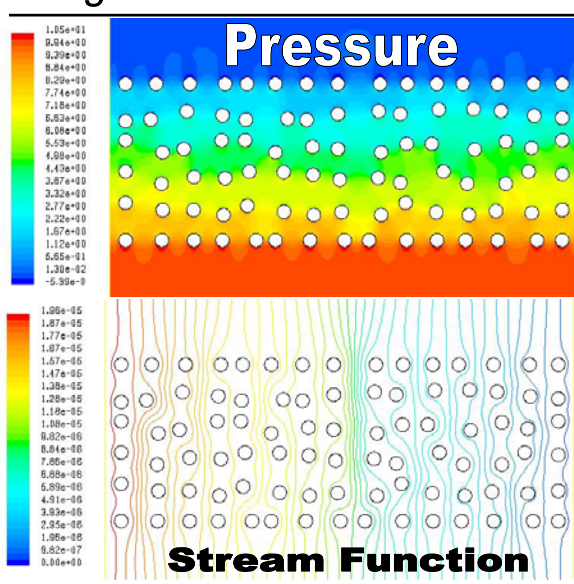




Mesh and BC's



Single Phase Flow ΔP = 10Pa



Experimental Permeability:

$$k = 5 - 10 \times 10^{-12}\ \text{m}^2/\text{s}$$

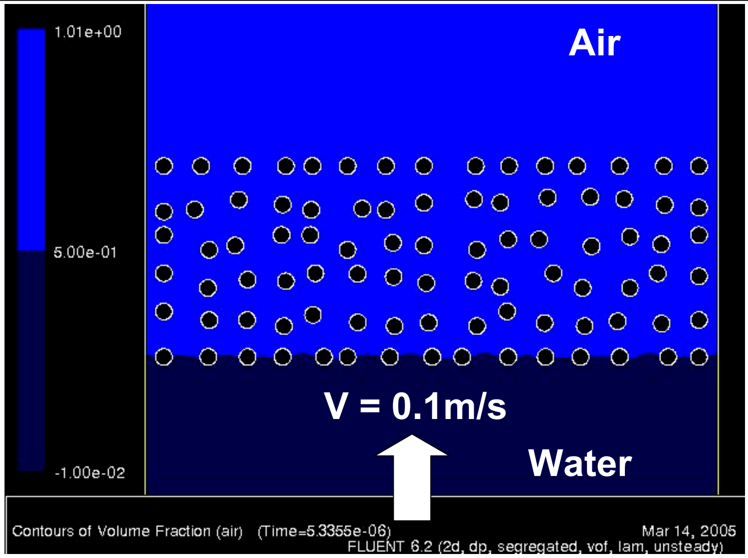
Mathias et al., *Handbook of Fuel Cells*, 2003.

Numerical Permeability:

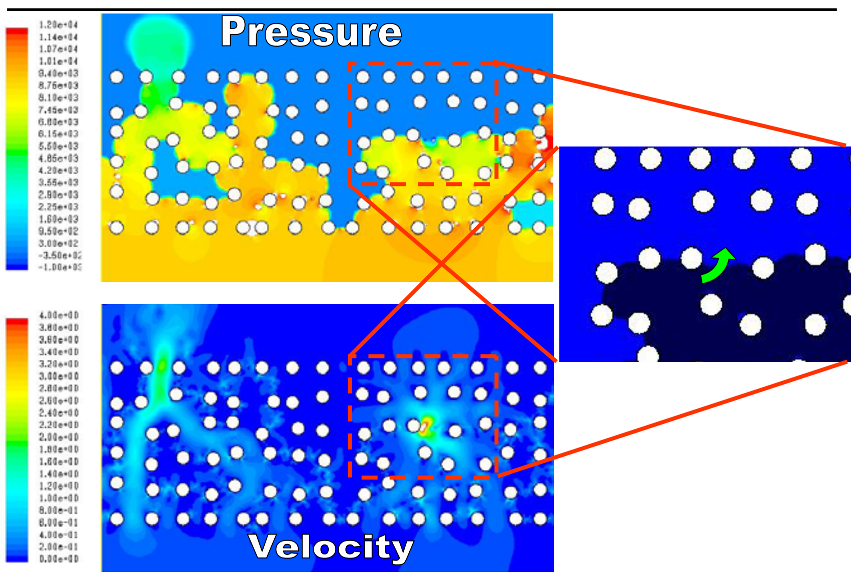
$$\bar{V} = 0.05467\ \text{m/s}$$

$$k = \frac{\mu L V}{\Delta P} = 10.75 \times 10^{-12}\ \text{m}^2/\text{s}$$

Two-Phase Flow $\theta = 108^o$



Two-Phase Flow, cont'd



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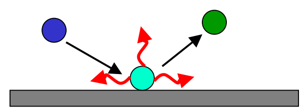
Visualization: Fluorescence Microscopy

- Scalar based flow visualization
- Molecules that emit a photon immediately after being excited by a light source.
- Three steps: Absorbance, dissipation, and emission.



- Increasingly employed in microfluidics for velocimetry and quantifying mixing.

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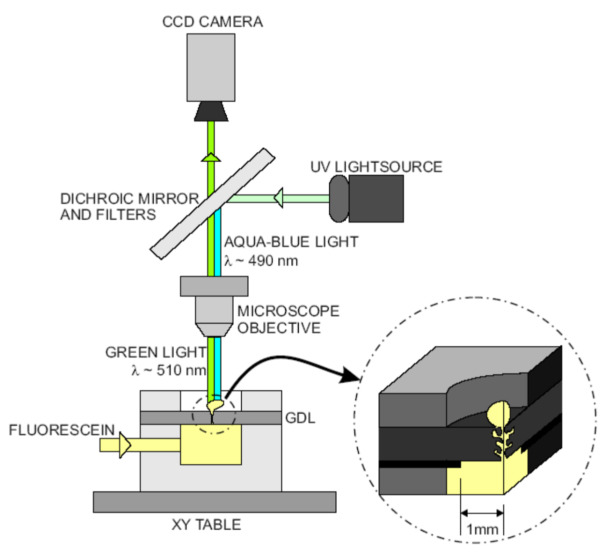
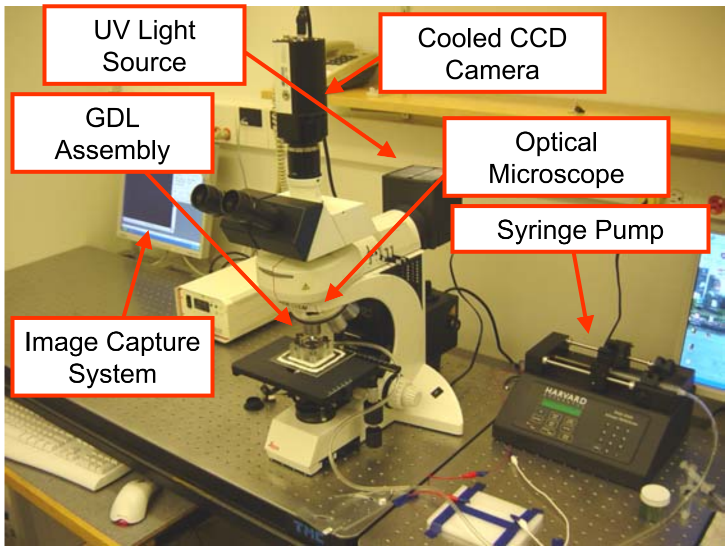
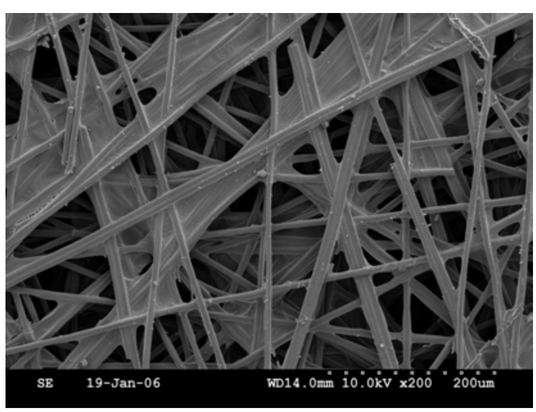
Fluorescence Microscopy



Experimental Set Up

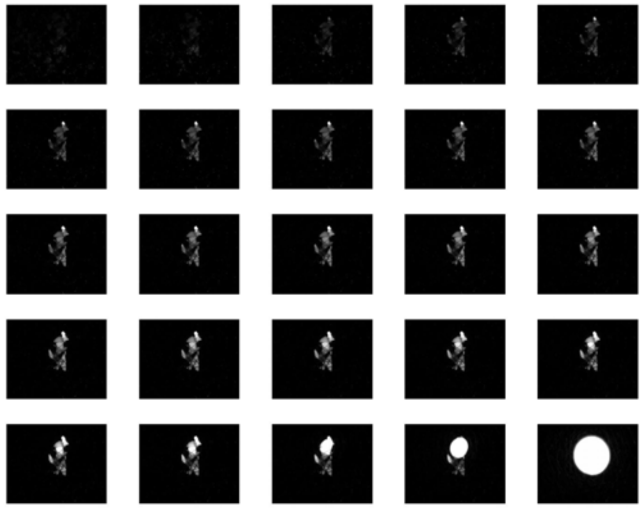


Gas Diffusion Layer



- Thickness = 190µm
- Porosity ~ 85%
- Mean Pore Dia. ~ 40 µm
- Fibre Dia. = 8.5µm

Raw Images



Enhanced Images

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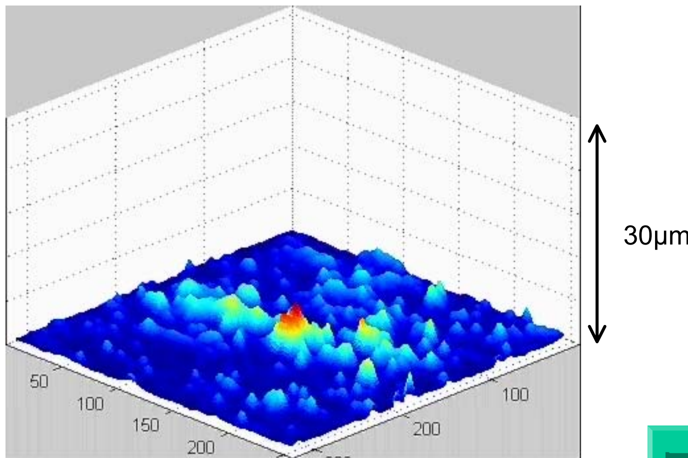
3D Imaging



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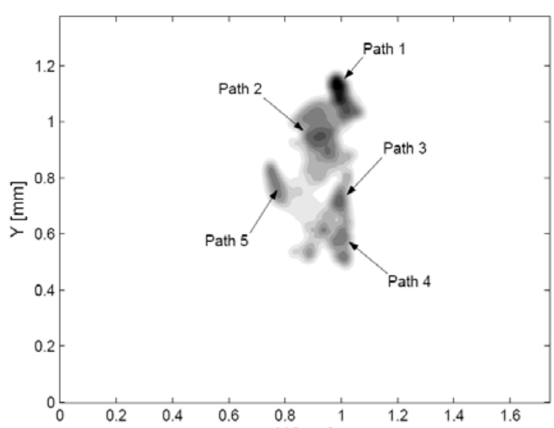
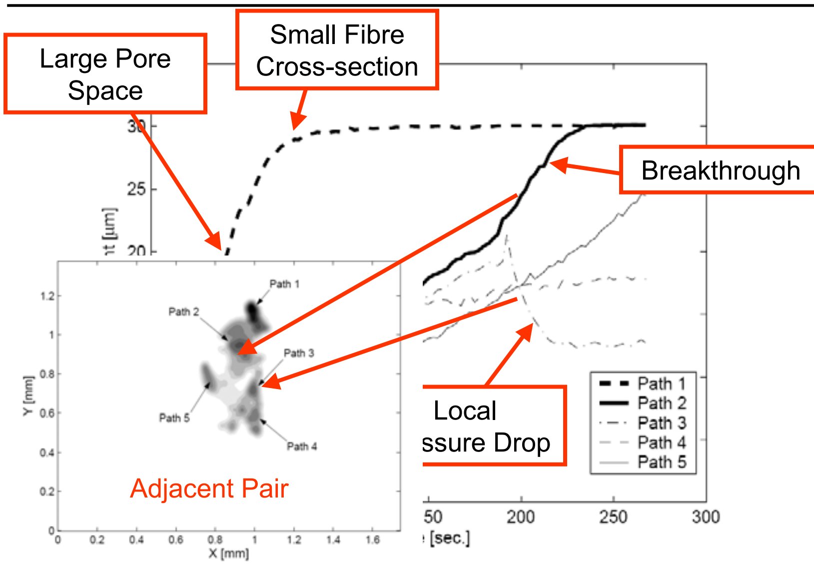
Distinct Flow Paths

- Identification of distinct pathways through GDL.
- Used for analysis of the transport mechanism.

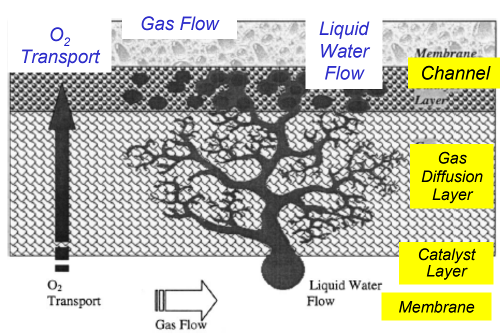


Interface Heights



Common Assumption

- Tree network with convergence of small capillaries into larger capillaries.



U. Pasaogullari and C.Y. Wang. *J. Electrochem. Soc.*, 151(3):A399-A406, 2004

⇨ Increasingly larger capillary near GDL surface does not agree with experimental observations.

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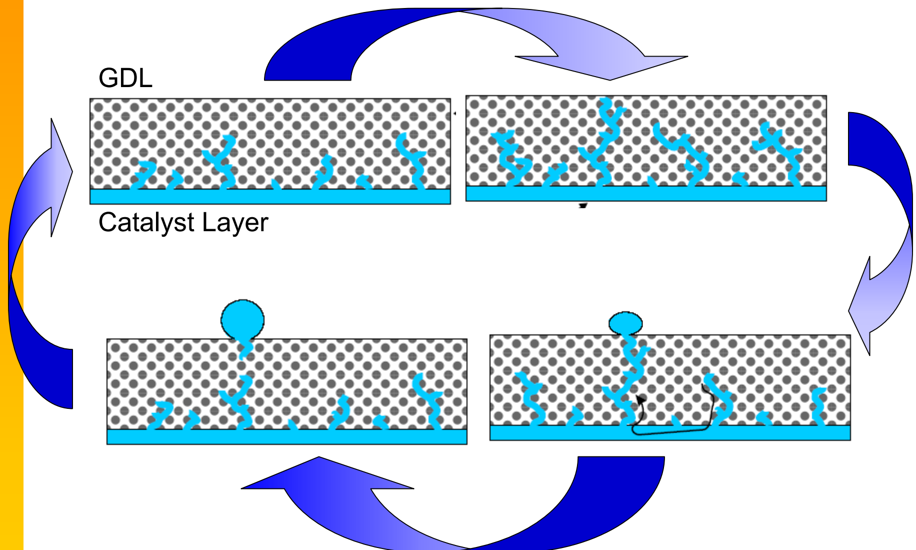
New Hypothesis



S. Litster, D. Sinton & ND, *J. Power Sources*, Vol. 154 (1): *95-105*, 2006

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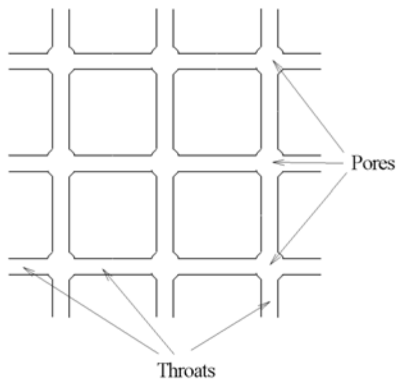
Insights: VOF + LIF

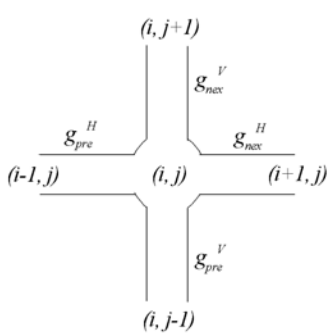
- Quantitative experimental and numerical visualization of liquid water transport within GDLs.
- Improved understanding two-phase flow in GDLs.
- New hypothesis for the liquid transport mechanism within GDLs.

Pore Network Model



- Porous media represented as a capillary network
- Pores are non-resistive elements providing fluid storage
- Throats are capillaries of arbitrarily shape and size
- each throat is occupied with one phase only.
- irregularity phase patterns due to local heterogeneities → random distribution (*rt*)

Pore Network: Method



- Total volume of pores (V_p) is equal to the void volume of the porous medium.

$$\varepsilon = \sum_i V_{pi} \Big/ V$$

- Permeability of network set by adjusting the throats appropriately.

Given ∇P & flow rate (Q)

$$K_{sp} = \mu \frac{L}{p_{inl} - p_{out}} \frac{Q}{A}$$

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Pore Network: Method (cont'd)

- In multiphase systems, transfer resistances and potentials differ

  → phase with the lower potential is displaced by other(s) phase(s) with higher potential

- Phase content quantified by the phase saturation (s).

- Permeability (K_i) of each phase calculated using Darcy's law (momentum)

$$\mathbf{u}_i = -\frac{K_i}{\mu_i} \nabla p_i$$

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Pore Network: Method (cont'd)

- relative permeability

$$k_{r,i} = K_i / K_{sp} = (Q_i / (\Delta p_i / L_i))((\Delta p / L)/Q)$$

(K_i) is phase permeability
(K_{sp}) single phase permeability

- Criterion for phase displacement: pressure difference exceeds threshold capillary pressure

$$\Delta P \; > \; p_c = 2\,\sigma / r_t$$

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Numerical Procedure

- randomly generated throat radii
- invading fluid enters the network at one side (inlet) and the invaded fluid flows out of opposite side (outlet).
- invading fluid occupies in a sequence of discrete steps throats with the lowest potential (largest throat radius).

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Numerical Procedure (cont'd)

- To compute (K_{sp}) and (K_i) pressure solutions within the network and the carrying backbone are required.

  → simultaneous solution of the balance equations over all pores within network/backbone.

$$\sum_{j=1}^{c} q_j = 0, \quad c = 4 \qquad (q_j) \text{ flow rate}$$

- coordination number (c) represents the number of throats belonging to each pore (≡ four for regular square 2-*D* network).

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Numerical Procedure (cont'd)

- Balance for one throat

$$q_j = g_j \Delta p$$ (g_j) throat conductance

- For a capillary of radius (r_t) and length (l) conductance is obtained from Poiseuille's law

$$g_j = \frac{\pi r_t^4}{8 \mu l}$$

- mass balance for each pore

→ linear system of equations for pressure ($\mathbf{p}$):

$$\mathbf{A\,p = b}$$

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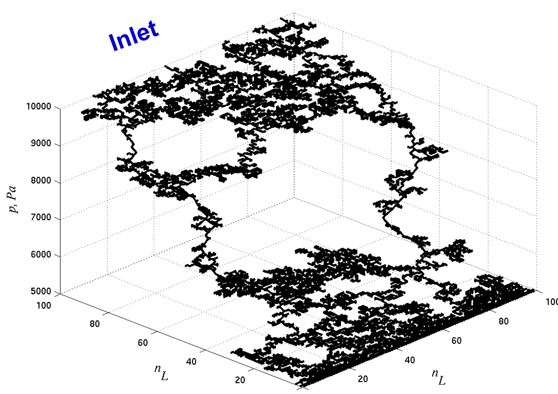
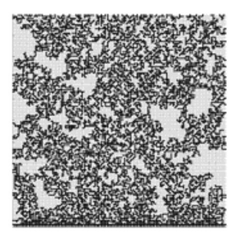
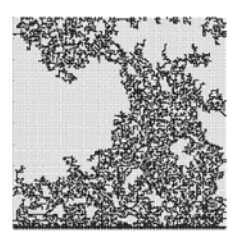
Simulation: One Mobile Phase

Set inlet and outlet pressure





High saturation



Low saturation

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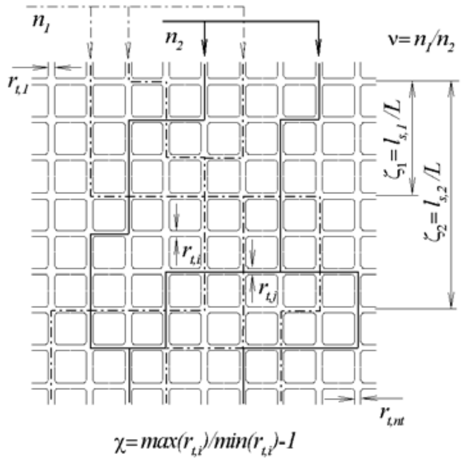
Two-Mobile Phases: Network/Flow Parameters.



Network rules:

- Phase carrying backbones transfer momentum
- Flow paths of the phases can intersect
- Only clusters of the originally present phase can be formed.

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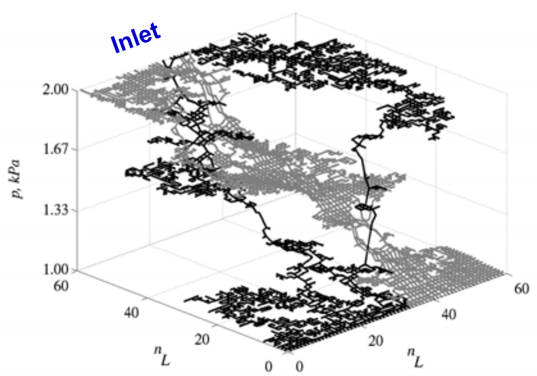
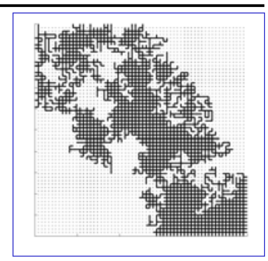
Simulation: Two Mobile Phases

Set inlet and outlet pressure

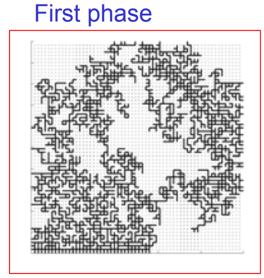


Phase flow rate

$$Q_i = \sum_j g_m (p_{in} - p_j)$$



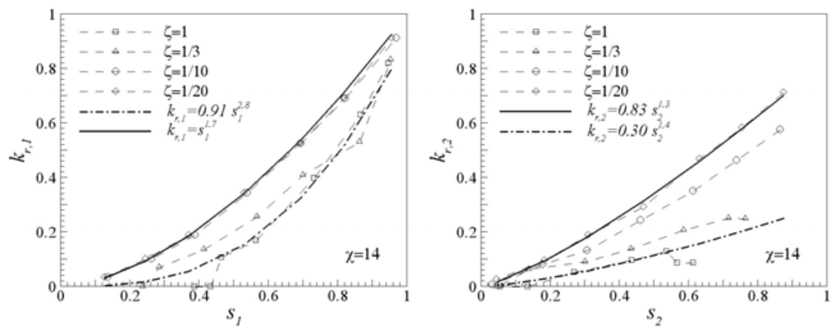
First phase



Second phase

Relative Permeability

Power law for the overall phase saturation



$1.7 \le \beta \le 2.8$ $\quad k_{r,i} = Bs_i^{\beta}$ $\quad 1.3 \le \beta \le 1.4$

Maximum value of the power is observed for capillary dominated flow (dash-dot line) due to the largest clusters.

Insights

- Saturation *range* in which both phases percolate can be altered
- Both ($k_{r,i}$) and (p_c) *vary* with heterogeneities and cluster size
- For power law ($k_r \sim s^n$), (n) *changes* with process extent, but *not* with heterogeneity

Further Reading

- Berning, T., and N. Djilali, "A 3D, Multi-Phase, Multicomponent Model of the Cathode and Anode of a PEM Fuel Cell", *J. Electrochem. Soc.*Vol. 150, No. 12, pp. A1589-A1598, December, 2003.
- Litster, S. and N. Djilali "Two-Phase Transport in Porous Gas Diffusion Electrodes" Chapter 5, in Transport Phenomena in Fuel Cells (Eds. M. Faghri & B. Sundén), pp. 175-213, WIT Press, Southampton UK, 2005
- Litster, S., D. Sinton and N. Djilali, "Ex situ Visualization of Liquid Water Transport in PEM Fuel Cell Gas Diffusion Layers", *J. Power Sources*, Vol. 154 (1): *95-105,* 2006.