

Development and Validation of a Two-phase, Three-dimensional Model for PEM Fuel Cells

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Sandia National Laboratories

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FC027

Overview

Timeline

- Project was started on 10/1/09
 - DOE Kickoff meeting held 9/30-10/1/09
- Project will end on 9/30/13
- Percent complete: ~13%

Budget

- Total project funding (over 4 years)
 - DOE share: \$4,292,000
 - Contractor share: \$1,200,000
- Funding received in FY09:
\$816,000
- Funding for FY10:
\$244,000

Barriers

- Barriers addressed
 - Performance
 - Cost
 - Durability

The validated PEM fuel cell model can be employed to improve and optimize the design and operation of PEM fuel cells and thus address these barriers.*

Partners

- Direct collaborations with Industry, University and other National Labs:
 - Ford, Ballard**
 - Penn State University**
 - LANL, LBNL.**
- Project lead: Sandia National Labs

* PEM refers to polymer electrolyte membrane

Relevance/Objectives

- To develop and validate a **two-phase, three-dimensional transport model** for simulating PEM fuel cell performance under a **wide range of operating conditions**.
- To apply the validated PEM* fuel cell model to **improve fundamental understanding** of key phenomena involved and to **identify rate-limiting steps** and **develop recommendations for improvements** so as to accelerate the commercialization of fuel cell technology.
- The validated PEMFC model can be employed to improve and optimize PEM fuel cell operation. Consequently, the project helps: **i) address the technical barriers on performance, cost, and durability; and ii) achieve DOE's near-term technical targets on performance, cost, and durability in automotive and stationary applications.**

* PEM refers to polymer electrolyte membrane



Approach

Our approach is both **computational** and **experimental**:

- Numerically, develop a **two-phase, 3-D, transport model** for simulating PEM fuel cell performance under a wide range of operating conditions.
- Experimentally, measure **model-input parameters** and generate **model-validation data**.
- **Perform model validation** using experimental data available from the literature and those generated from team members.
- Apply the validated transport model to **identify rate-limiting steps** and **develop recommendations** for improvements.

A staged approach will be adopted in model development and validation:
Single phase (dry) → Partially two-phase (dry-to-wet transition) → Fully two phase (wet)



Approach:

FY10 Milestones and Go/no-go decision and Their Current Status

Month/Year	Description of Milestone or Go/No-Go Decision
September/2010	Develop a 3-D, partially two-phase, single-cell model. Status: ~ 50% complete
September/2010	Measure model-input parameters and generate model-validation data for single-phase operating regime. Status: ~ 50% complete
September/2010	Go/no-go: determine whether or not we should proceed to develop a 3-D, fully two-phase, single-cell model. Status: not yet time to decide

Governing Equations Sandia National Laboratories

(Based on the conservations of mass, momentum, energy, species, and charge)

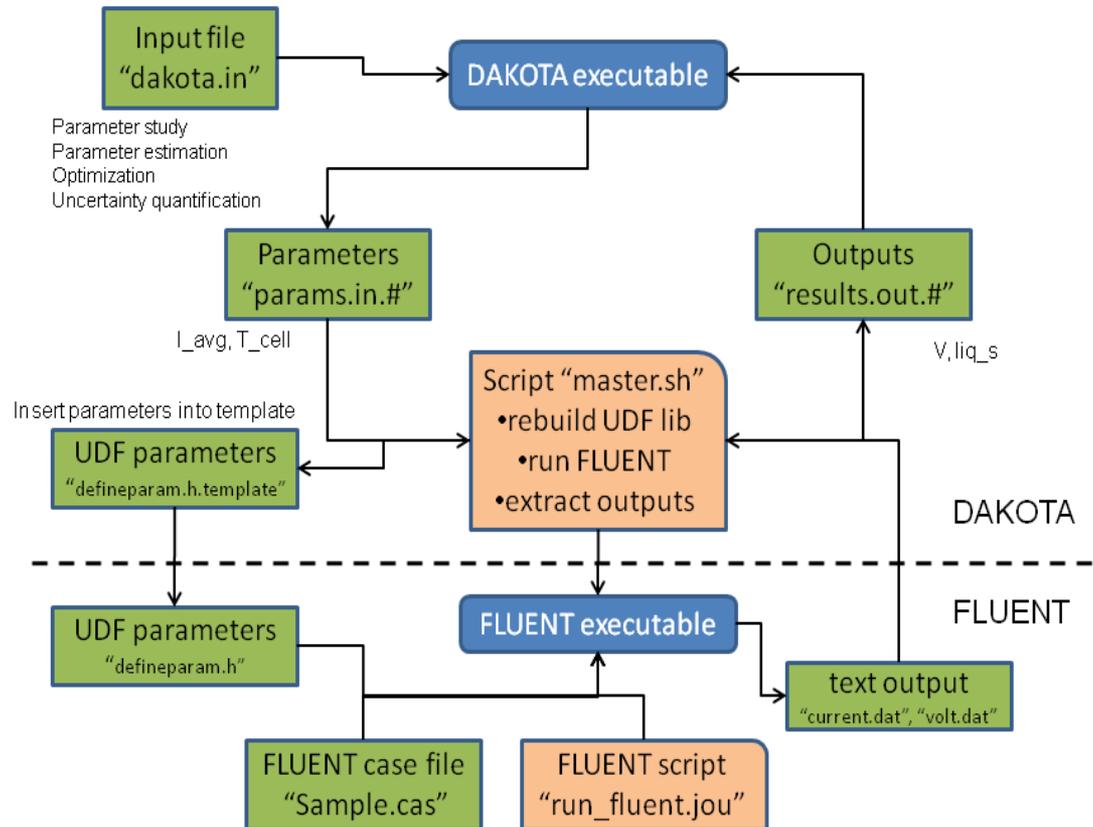
- Mass: $\nabla \cdot (\rho \vec{u}) = S_m$
- Momentum: $\frac{1}{\varepsilon^2} \nabla \cdot (\rho \vec{u} \vec{u}) = -\nabla P - \nabla \cdot (\rho \tau) + \frac{v}{K} (\rho \vec{u})$
- Energy: $\nabla \cdot (\gamma_T \rho C_p \vec{u} T) = \nabla \cdot (k_{eff} \nabla T) + S_T$
- Species: $\nabla \cdot (\gamma_c \vec{u} C^k) = \nabla \cdot (D_g^{k,eff} \nabla C_g^k) - \nabla \cdot \left[\left(\frac{mf_l^k}{M^k} - \frac{C_g^k}{\rho_g} \right) \vec{j}_l \right] + S_c$
- Charge (Electrons): $0 = \nabla \cdot (\sigma^{eff} \nabla \Phi_s) + S_{\Phi_s}$
- Charge (Protons): $0 = \nabla \cdot (k^{eff} \nabla \Phi_e) + S_{\Phi_e}$

References:

- 1) K. S. Chen, B. Carnes, F. Jiang, G. Luo, and C.-Y. Wang, "Toward developing a computational capability for PEM fuel cell design and optimization", ASME Proceedings of FuelCell2010, paper # FuelCell2010-33037 (2010).
- 2) Y. Wang and C. Y. Wang, "A Non-isothermal, Two-phase Model for Polymer Electrolyte Fuel Cells", *JES*, 153, A1193-1200 (2006).
- 3) U. Pasaogullari and C. Y. Wang, "Two-phase modeling and flooding prediction of Polymer Electrolyte Fuel Cells", *JES*, 152, A380 (2005).

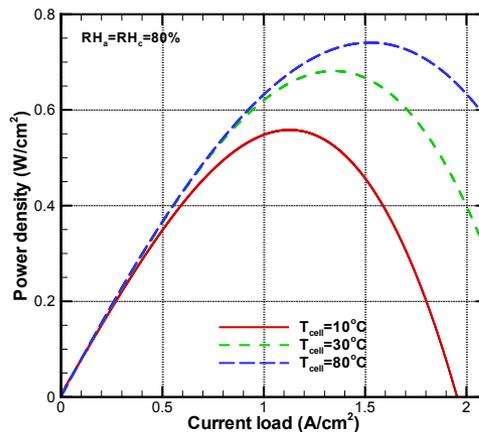
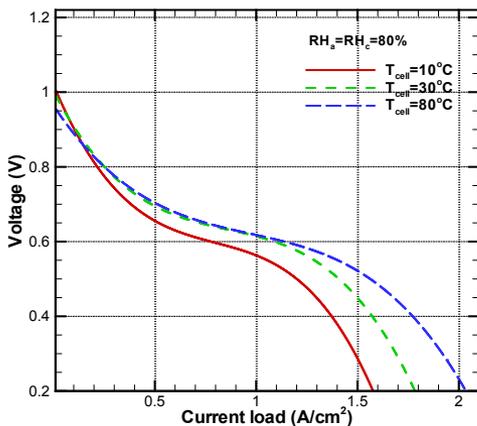
Enabling Advanced Fuel Cell Design Studies Using DAKOTA

- DAKOTA is a toolkit for design, optimization, and uncertainty quantification developed and being enhanced by Sandia National Labs
- We implemented a script to allow DAKOTA to run the fuel cell model (implemented in FLUENT using user defined functions or UDFs).
- Each evaluation
 - updates parameters,
 - rebuilds the UDF lib,
 - runs the model, and
 - extracts outputs (I,V)
- Parameter sensitivities
 - from finite differences
- Reuse of previous state
 - Improves robustness

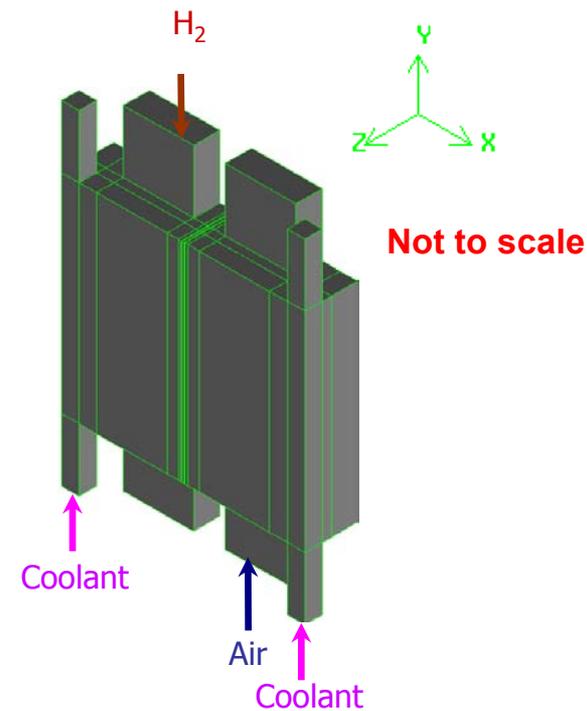
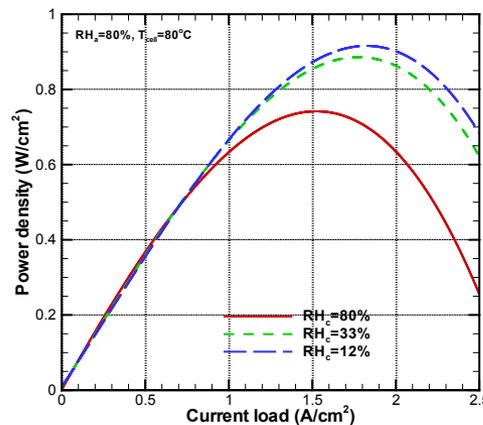
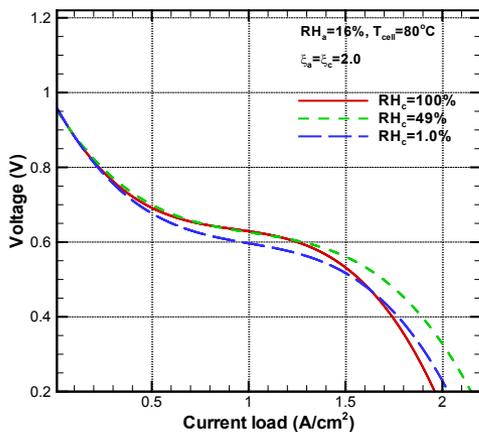


Technical Accomplishments and Progress: Example of model prediction – polarization curves

Computed Effect of Operating Temperature on PEM Fuel Cell Performance



Computed Effect of Cathode RH on PEM Fuel Cell Performance

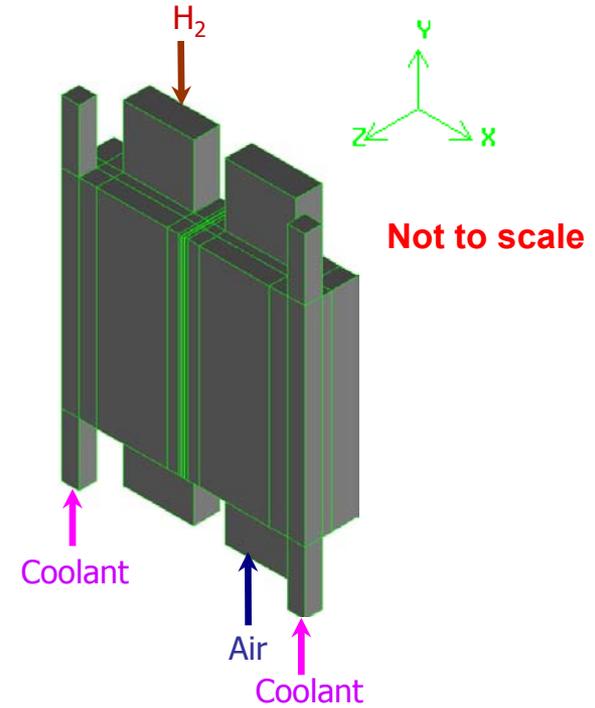
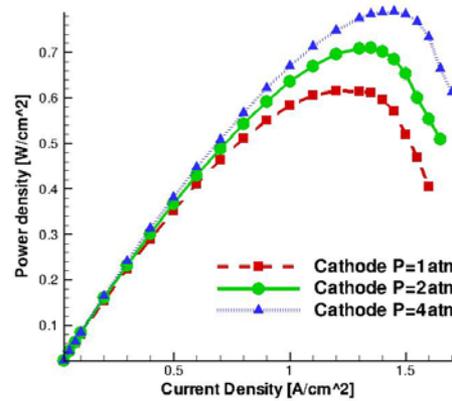
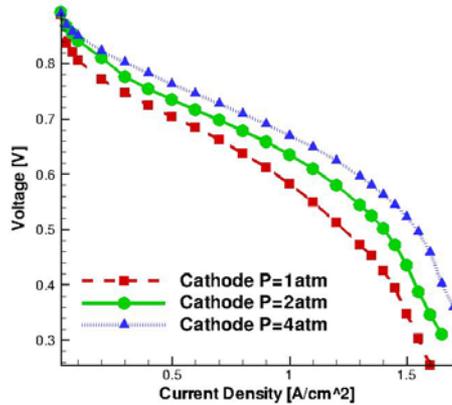


Thickness (x direction)
 Membrane: 30 micron CL: 10 micron
 MPL: 40 micron GDL: 160 micron
 GC: 1mm Land: 0.5mm
 Cell length (y direction): 0.3 m
 Channel height (z direction): 0.75mm
 Land height (z direction): 0.75mm
 Coolant channel: $0.5 \times 0.5 \text{ mm}^2$

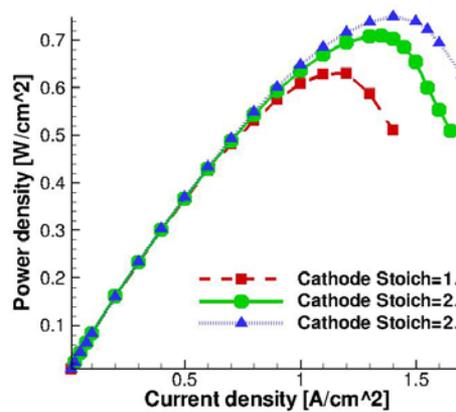
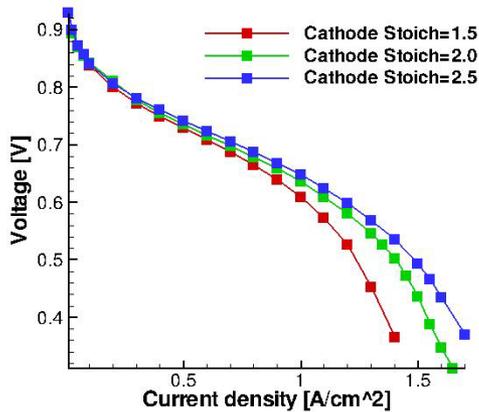
- Cell voltage or performance increases with operating temperature.
- Cell voltage increases with cathode RH when current density is relatively low.
- Cell voltage decreases with cathode RH when current density is sufficiently high.

Technical Accomplishments and Progress: Example of model prediction – polarization curves

Computed Effect of **Cathode Back-Pressure** on PEM Fuel Cell Performance



Computed Effect of **Cathode Stoic** on PEM Fuel Cell Performance

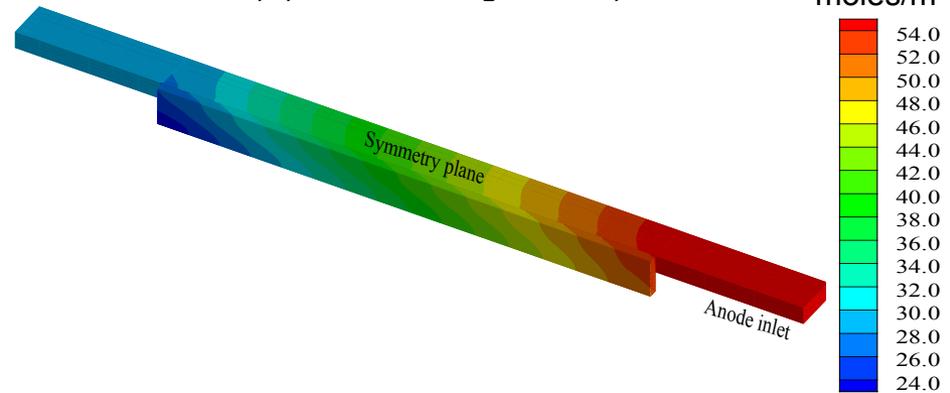


Thickness (x direction)
 Membrane: 30 micron CL: 10 micron
 MPL: 40 micron GDL: 160 micron
 GC: 1mm Land: 0.5mm
 Cell length (y direction): 0.3 m
 Channel height (z direction): 0.75mm
 Land height (z direction): 0.75mm
 Coolant channel: 0.5×0.5 mm²

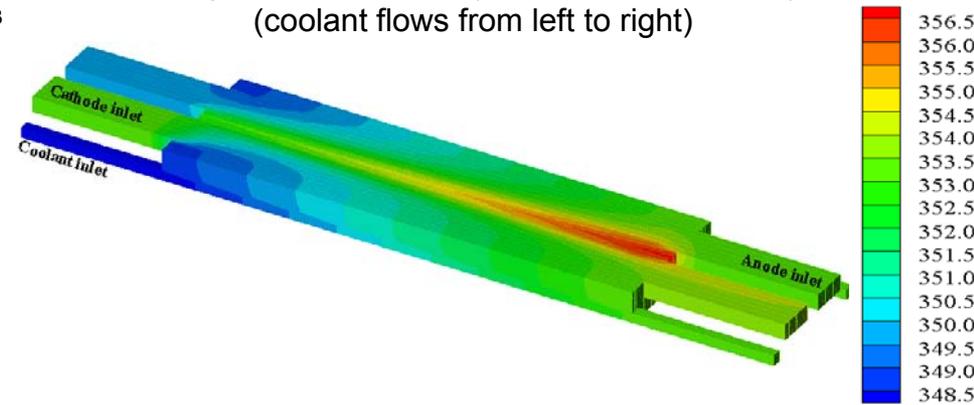
- Cell voltage or performance increases with increasing cathode back-pressure.
- Cell voltage increases with increasing cathode stoichiometric flow ratio.

Technical Accomplishments and Progress: Examples of model prediction – 3D contours plots

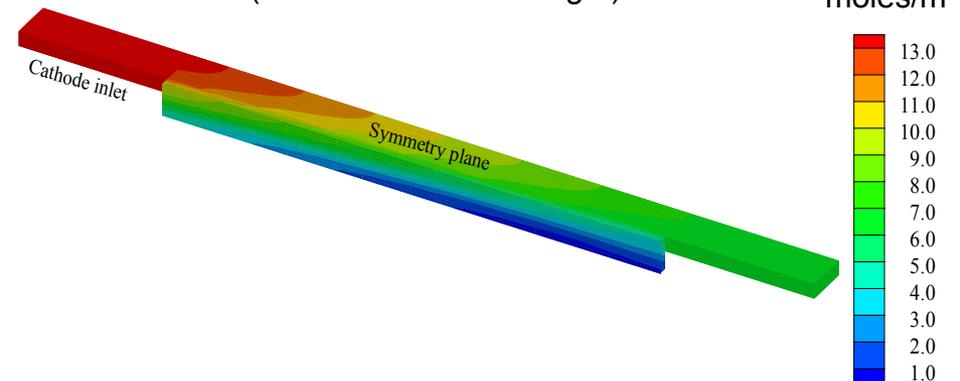
H₂ concentration contours in anode channel, GDL, CL
(H₂ flows from right to left)



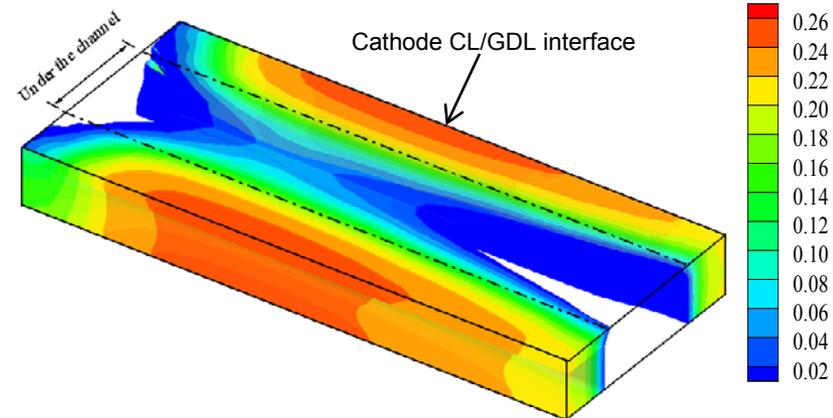
Temperature contours (with coolant channels)
(coolant flows from left to right)



O₂ concentration contours in cathode channel, GDL, CL
(air flows from left to right)



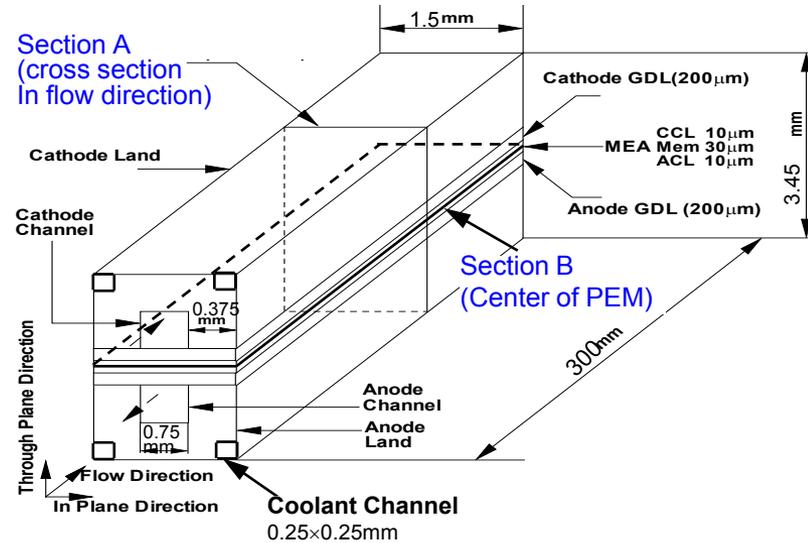
Liquid water saturation contours within cathode GDL



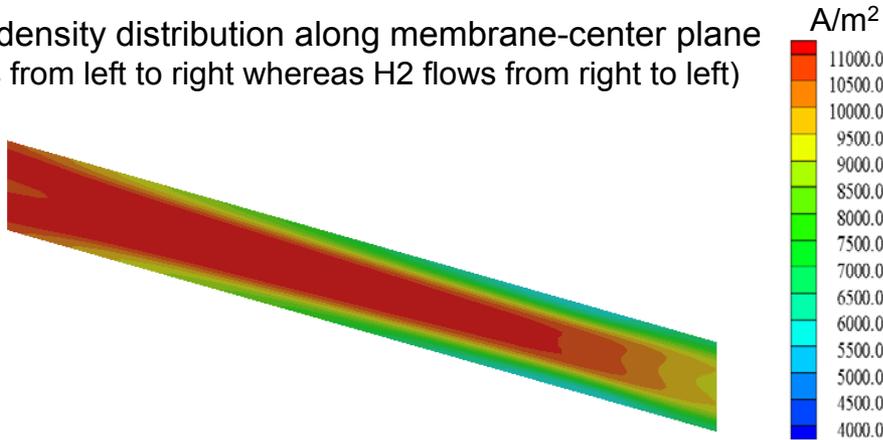
- Quantitative information on H₂ and O₂ concentrations, temperature, and liquid-water saturation, etc. can be readily obtained from model prediction.

Technical Accomplishments and Progress: Examples of model prediction – contour & vector plots

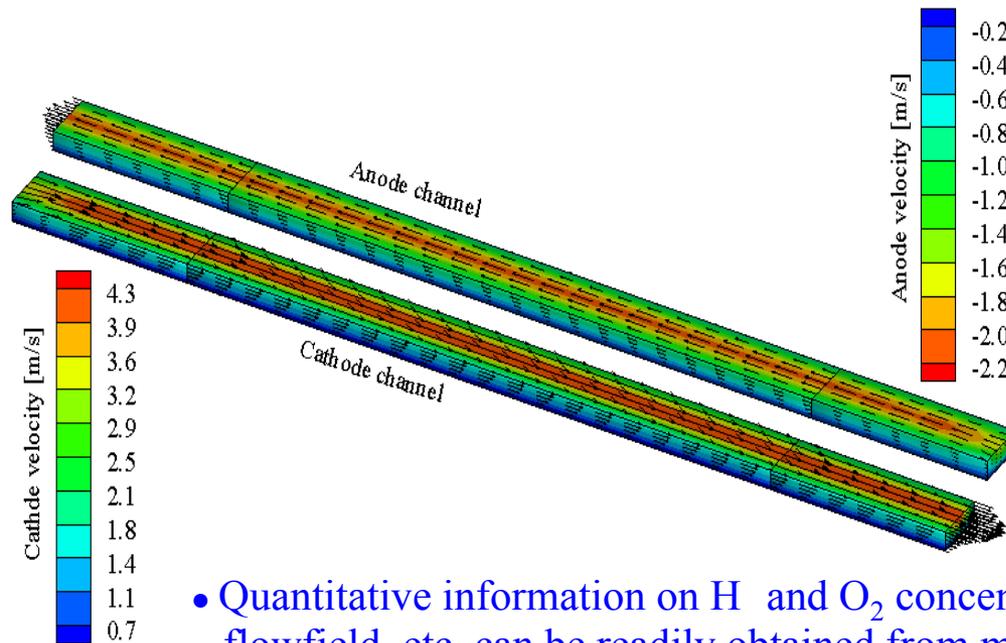
Model geometry and cross sections for 2-D contour plots



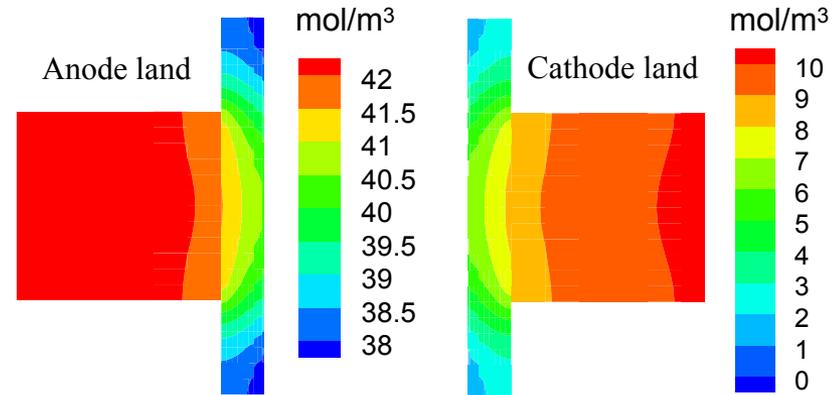
Current density distribution along membrane-center plane (Air flows from left to right whereas H₂ flows from right to left)



Flowfield (velocity vectors) in anode and cathode flow channels



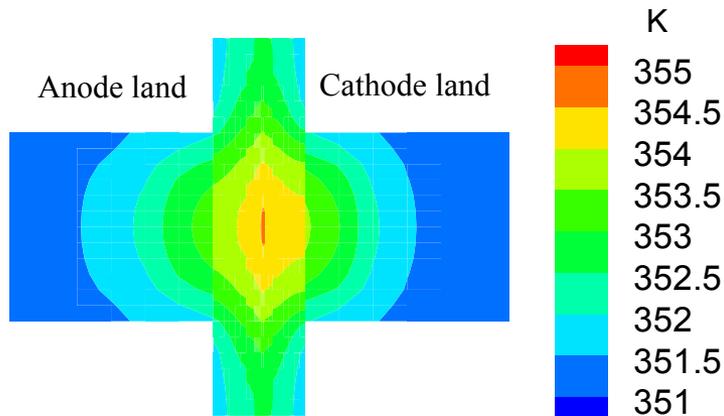
H₂ and O₂ concentration contours at Section A



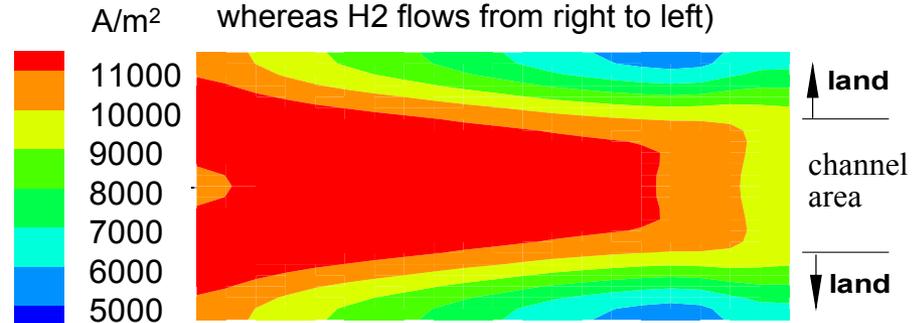
- Quantitative information on H₂ and O₂ concentrations, local current density, flowfield, etc. can be readily obtained from model prediction.

Technical Accomplishments and Progress: Examples of model prediction – 2-D contours plots

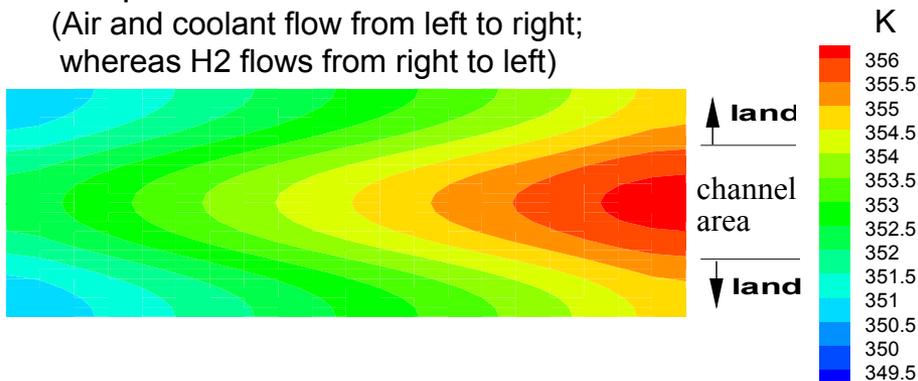
Temperature contours on Section A



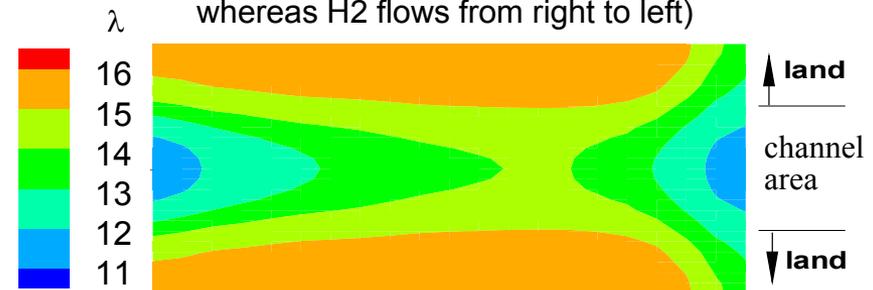
Local current density at Section B
(Air and coolant flow from left to right;
whereas H₂ flows from right to left)



Temperature contours on Section B
(Air and coolant flow from left to right;
whereas H₂ flows from right to left)

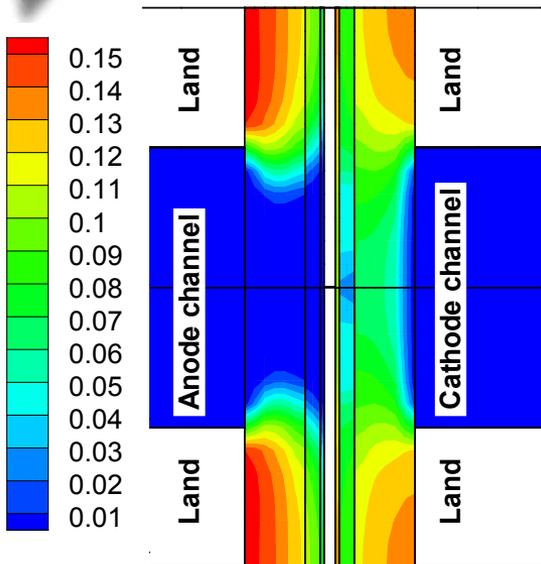


Water content (λ) on Section B
(Air and coolant flow from left to right;
whereas H₂ flows from right to left)

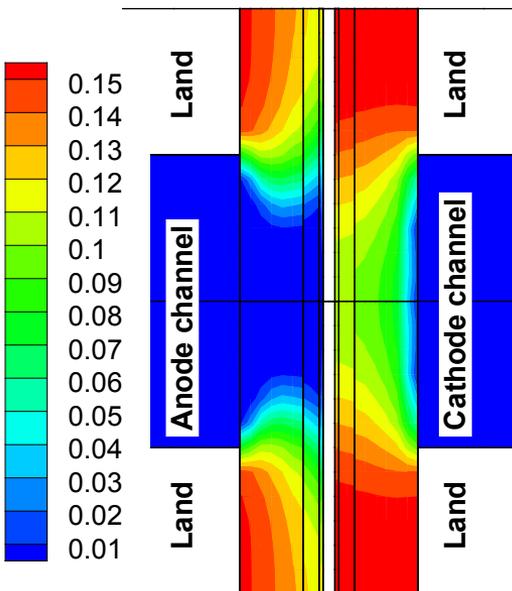


- Quantitative information on temperature, local current density, and water content (λ), etc. can be readily obtained from model prediction.

Technical Accomplishments and Progress: Example of model prediction – MPL effect

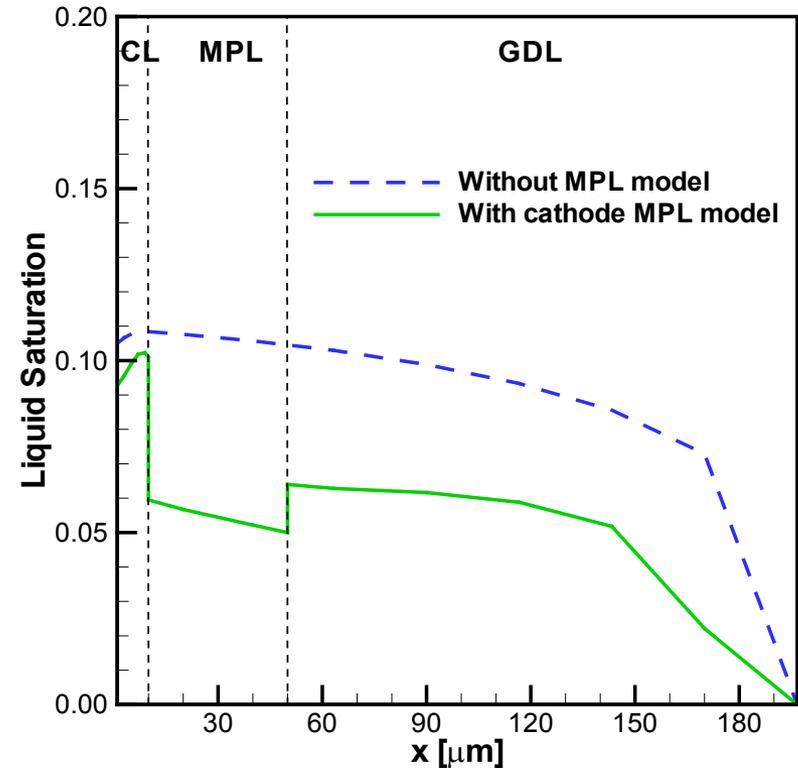


Saturation contours in middle cross section:
With cathode MPL



Saturation contours in middle cross section:
Without MPL

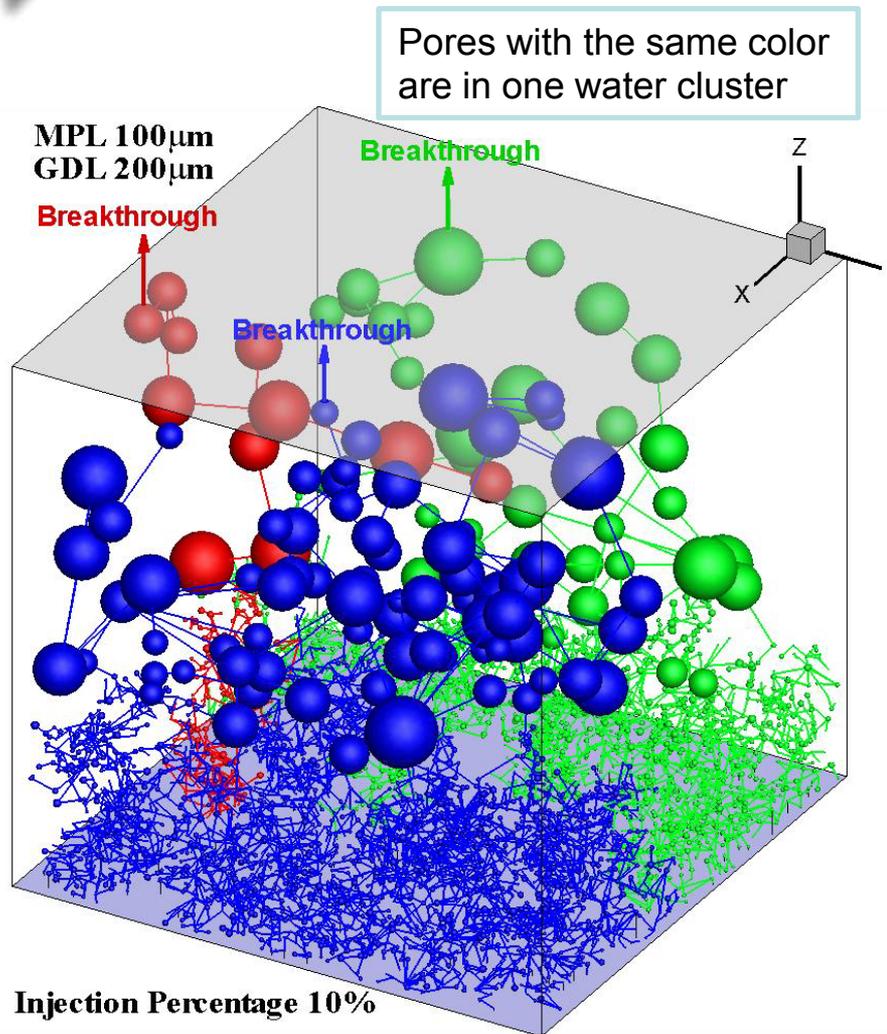
Under-the-Channel Saturation Profile across Cathode CL-MPL-GDL regions



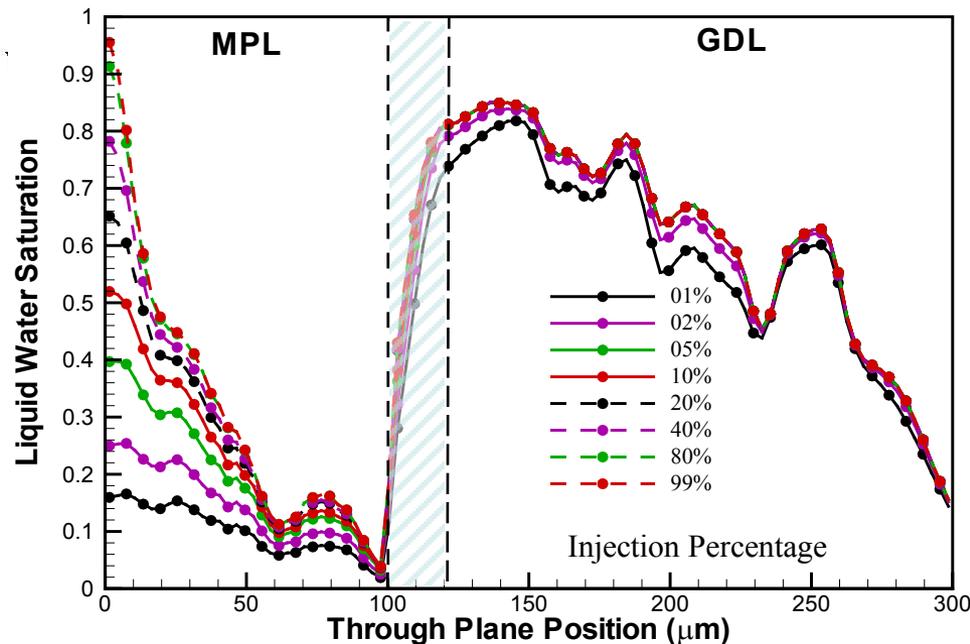
- MPL effectively reduces water saturation in the cathode MPL-GDL regions!

Technical Accomplishments and Progress:

Pore-network modeling of water transport in cathode MPL/GDL



Liquid water saturation profile in the cathode MPL-GDL region computed by pore-network model

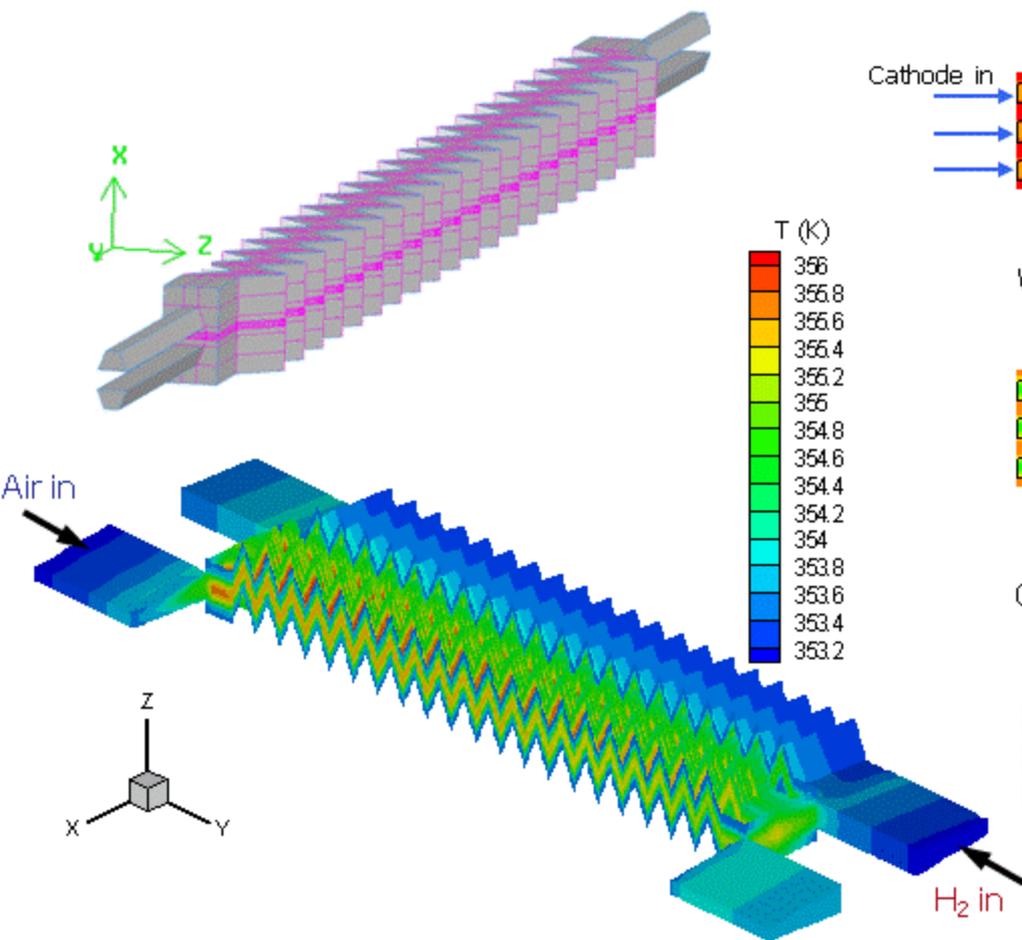


Note: Injection percentages are used to represent current densities.

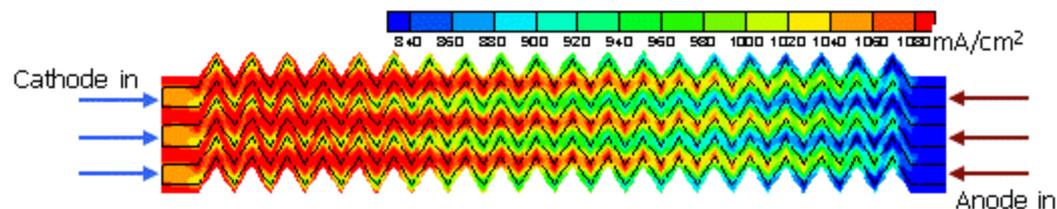
- Pore-network modeling helps elucidate fundamental physics, e.g., saturation discontinuity at MPL/GDL interface!

Technical Accomplishments and Progress:

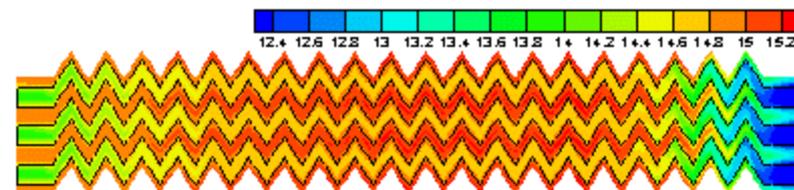
Example of model prediction – PEM fuel cell with *zigzag flow field*



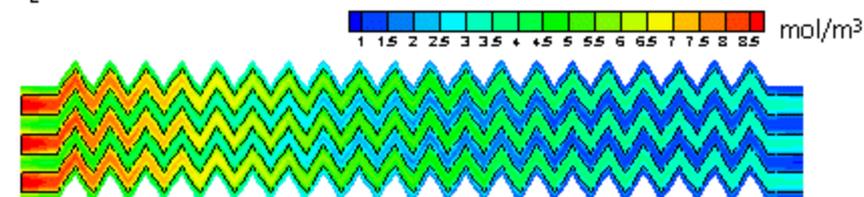
Current density in PEM



Water content in PEM



O₂ concentration in CCL



$I = 1000 \text{ mA/cm}^2$; $St(a/c) = 1.4/1.8$ (h₂/air);
 $T_{\text{cell}} = 80 \text{ }^\circ\text{C}$; Fixed temperature (80 °C) at BP walls; Periodic boundary pair in z direction

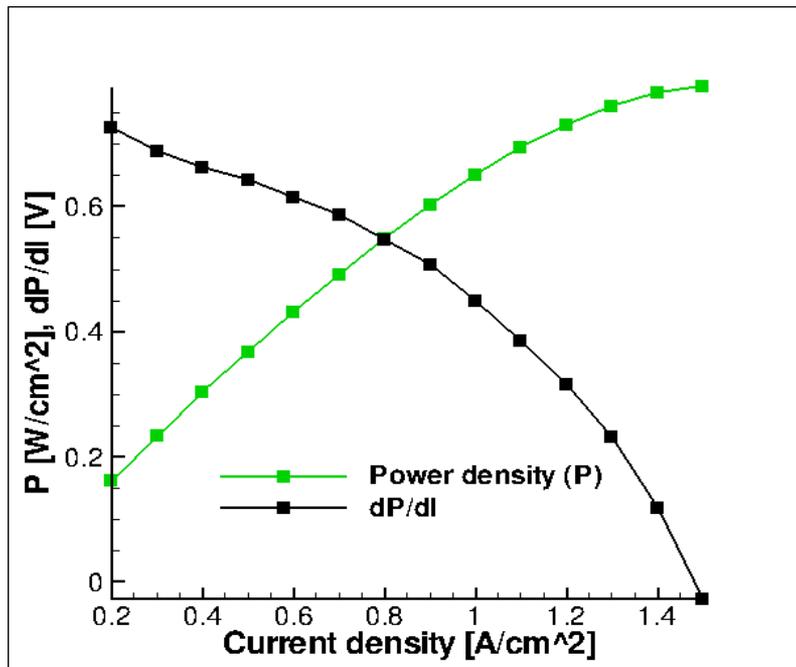
- The present model is capable of simulating practical PEM fuel cells, e.g. a single-channel fuel cell with zigzag and trapezoid-cross-sectional flow field.

Technical Accomplishments and Progress:

Example of Design Studies using the coupled computational capability

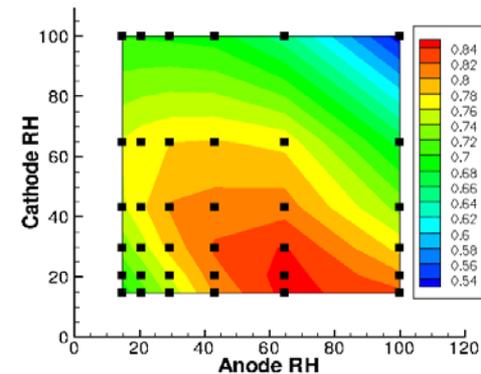
Optimizing RH at Anode/Cathode Inlet

- DAKOTA can also compute sensitivity of outputs (power or voltage) to
 - operating conditions – temperature, RH, stoich, pressure
 - model parameters – for use in calibration with data
- Performance optimization: find RH values for optimal power density

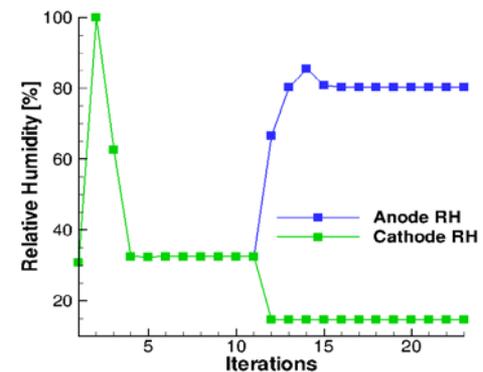


Power density maximum can be detected by computing dP/dI

2D parameter study of a/c RH indicates maximum in power density



Optimizer in DAKOTA can compute optimal values of a/c RH



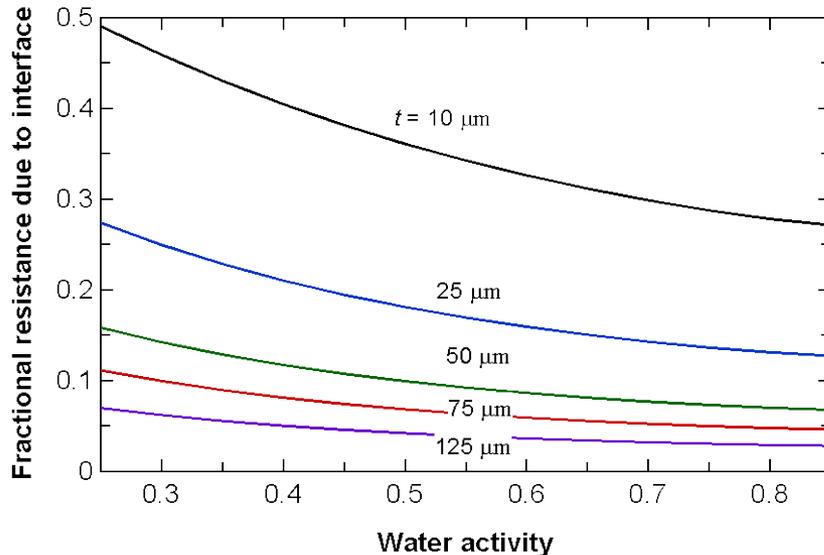
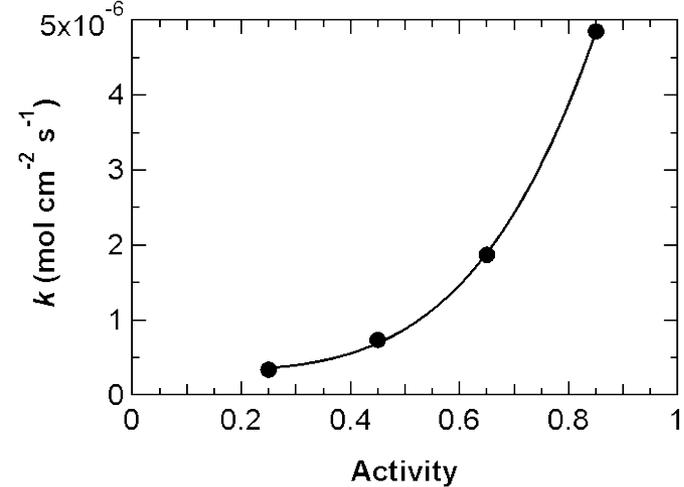
Technical Accomplishments and Progress: Predicting Membrane Interfacial Resistance

- Calculated mass-transfer coefficient for water as a function of RH*

$$N = \frac{\Delta a}{R} = \frac{\Delta a}{\frac{t}{\alpha} + \frac{1}{2k}}$$

- Liquid and membrane interfaces have no resistance
- Resistance believed to be caused by surface reorganization

*B. Kientz et al., *J. Fuel Cell Sci. Tech.*, in press
Work partially funded by Toyota Motor Corporation

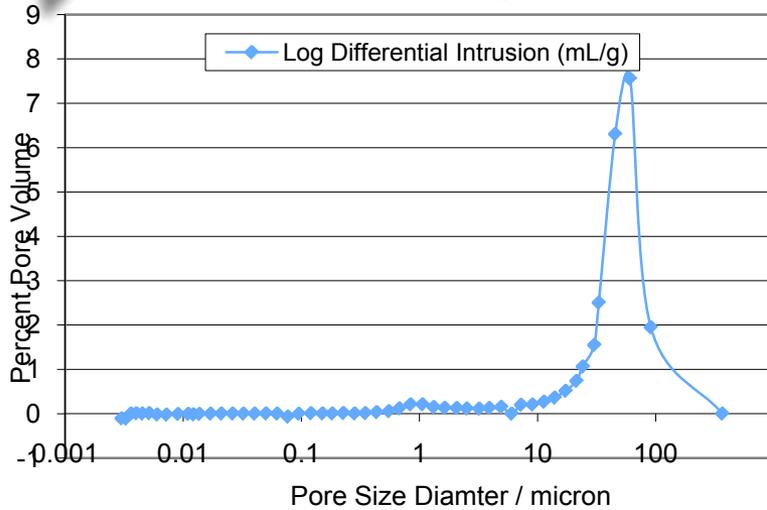


- Impact of resistance
 - Flatten water profiles in the membrane
 - Appreciable for thin, dry conditions
- Future Work
 - Need to implement in CL and cell models
 - Add similar resistances for oxygen and other species

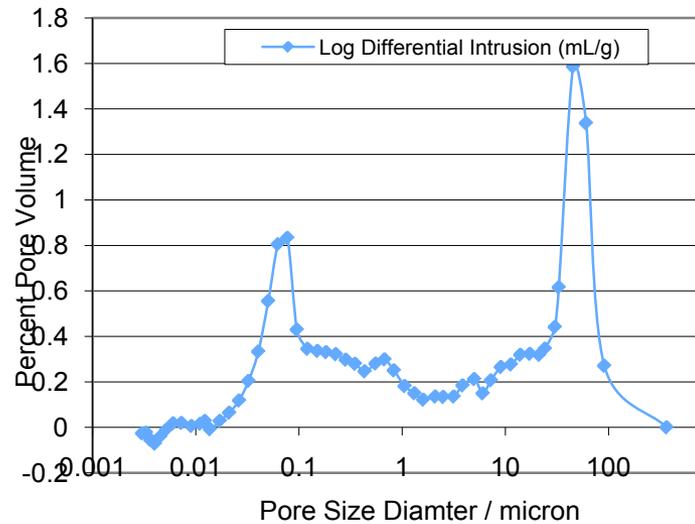
Technical Accomplishments and Progress:

Examples of GDL/MPL pore size measurements via porosimetry

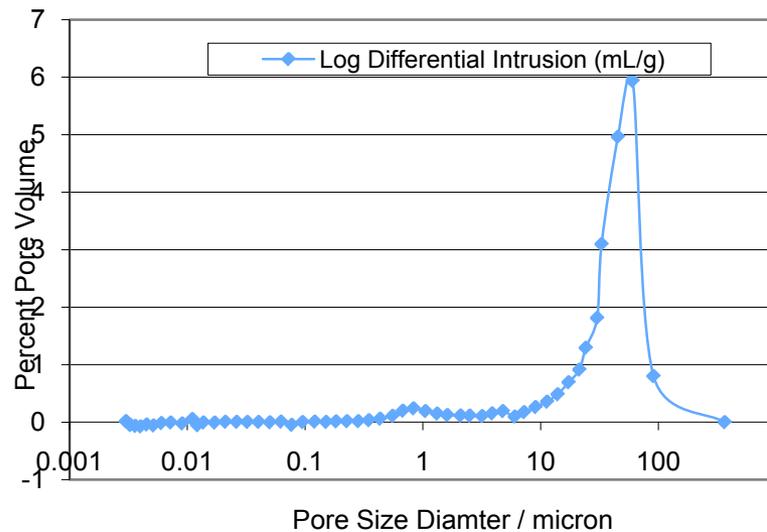
SGL GDL 10AA (plain)



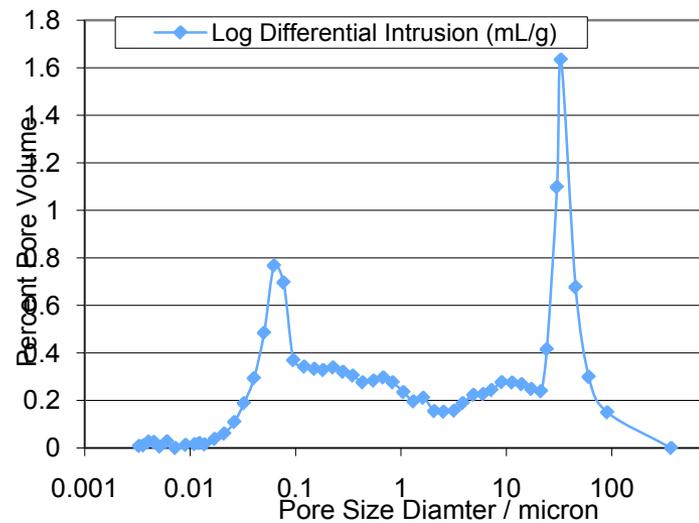
GDL 24D"C", 20 wt% PTFE bulk, 5 wt% PTFE MPL



SGL GDL 10BA, 5 wt% PTFE, no MPL



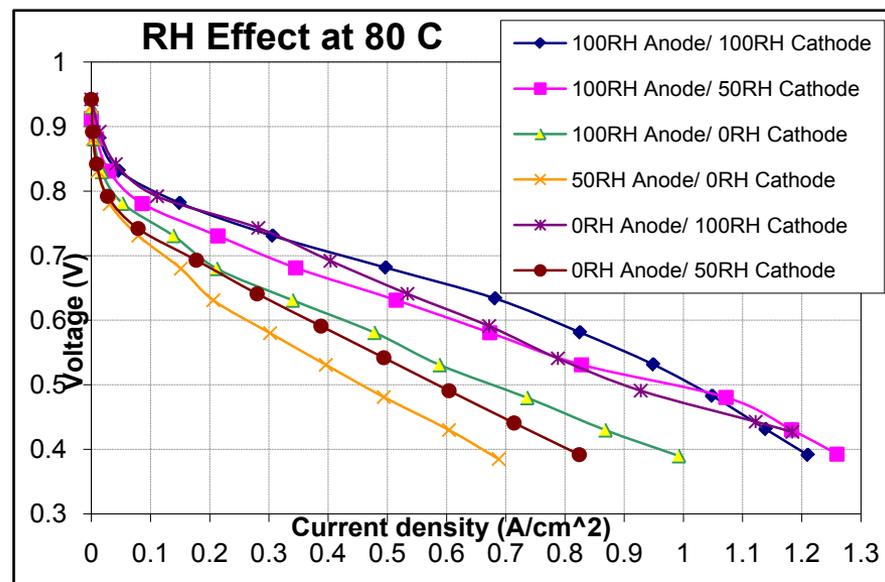
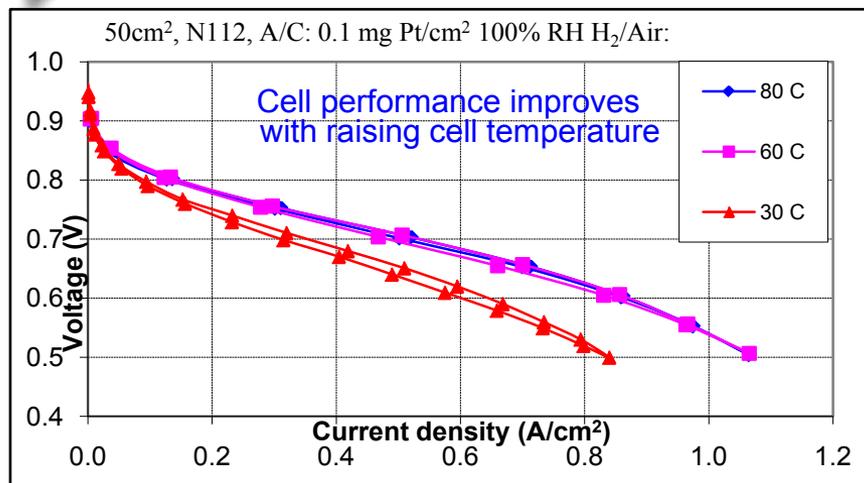
24DC 20% PTFE Bulk / 24% PTFE MPL



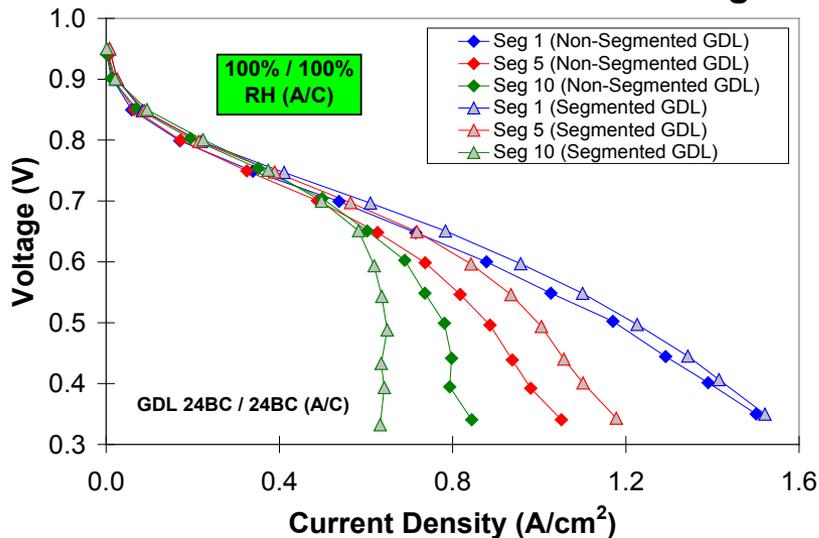
Technical Accomplishments and Progress:

Examples of cell performance measurement – polarization curves

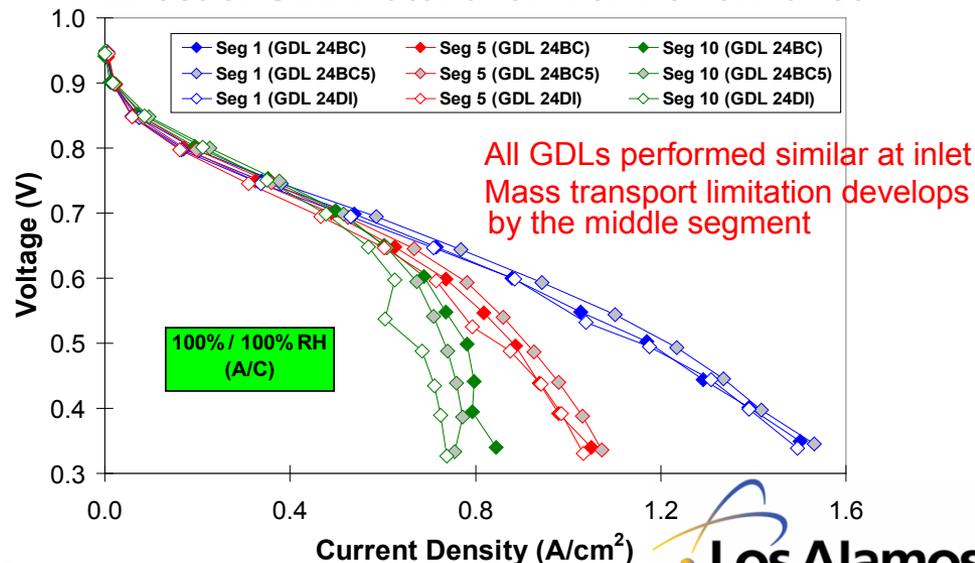
Cell Temperature Effect on Cell Performance



Polarization Curves at Different Cell Segments



Effect of GDL Material on Cell Performance



- Current density spread was less when GDL was not segmented.
- Mass transport limitation worsens at later segments.

Collaborations: Organizations/Partners



Model Development (including numerical implementation), Testing, and Validation:

- Sandia National Laboratories (Ken Chen: kschen@sandia.gov)



Participants: Ken Chen, Brian Carnes, Jay Keller, Marcina Moreno

Roles: project lead; model development, integration, testing, validation, dissemination

- The Pennsylvania State University (Chao-Yang Wang: cxw31@psu.edu)



Participants: Chao-Yang Wang, Fangming Jiang, Gang Luo, Yan Ji, Chris Shaffer

Roles: model development, validation, and dissemination; numerical implementation

- Lawrence Berkeley Lab (Adam Weber: azweber@lbl.gov; directly funded by DOE)



Participants: Adam Weber

Roles: Sub-model development (e.g., membrane, GDL/GFC interface), model dissemination

Model-parameter measurements, Data for Model Validation, Guidance/Applications:

- Los Alamos National Lab (Rod Borup: borup@lanl.gov; directly funded by DOE)



- Ford Motor Company (Atul Kumar: akumar56@ford.com; participate on "in-kind" basis)



- Ballard Power Systems (Patricia Chong: patricia.chong@ballard.com)





Los Alamos Role

(Dusan Spernjak, John Davey, R. Mukundan, Rod Borup)

- Experimental data for model validation
 - Performance, polarization curves, impedance
 - Segmented cell measurements
- Material characterization
 - GDLs
 - Pore-size distributions
 - GDL tomography by imaging (X-ray tomography etc.)
 - Surface energy
 - Dispersive energy vs. specific energy
 - Material hydrophobicity
 - Neutron imaging of water profiles



Project: Development and Validation of a Two-Phase, Three-Dimensional Model for PEM Fuel Cells

- **Ford's Role in this Joint Project**

- Ford is participating in the project as “in-kind” basis
- Ford will provide guidance on recommendations to improve rate limiting steps related to transport of gaseous species, liquid water, H^+ , e^- , and heat
- Ford will support providing range of fuel cell operating conditions pertinent to automotive applications to validate the above recommendations
- In addition, with its expertise in large scale CFD modeling, Ford will advice on as-needed basis towards this project



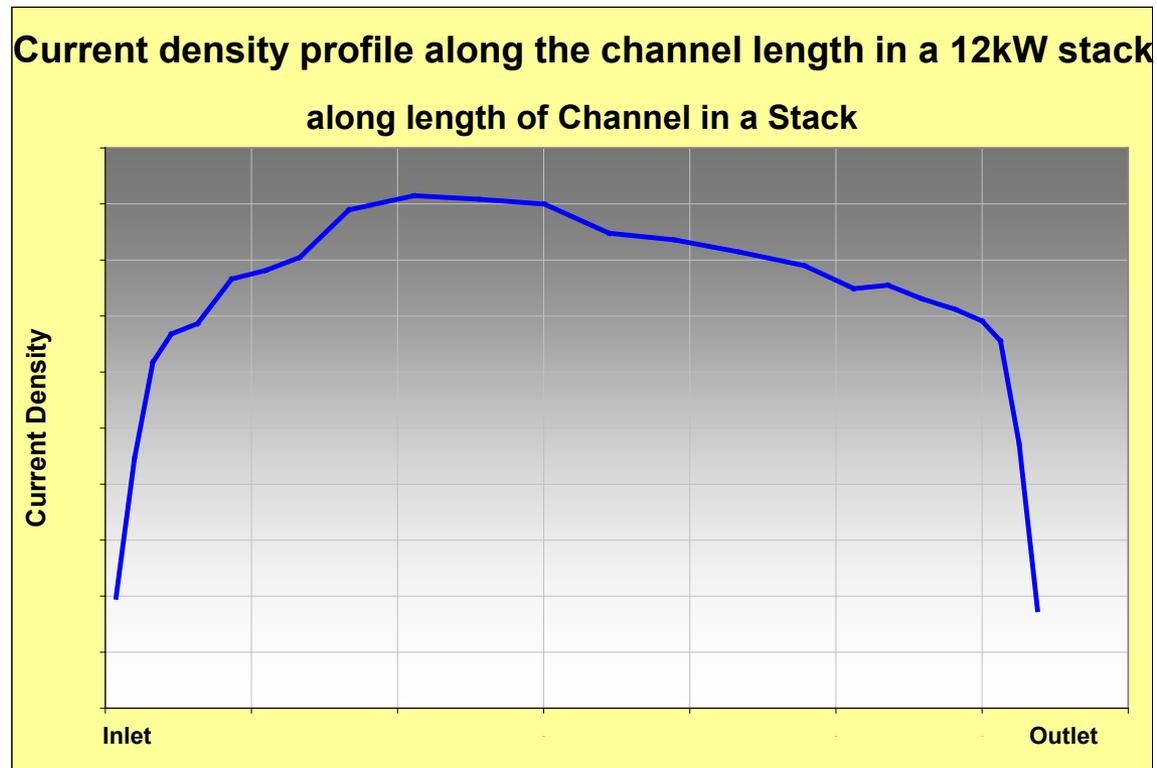
Ballard's Role and Deliverables

(David Harvey, Patricia Chong)

Model Input Parameters	e.g: Material properties, Structural properties, Flow Field Dimensions
Model Validation Data	e.g: Current Map, Temperature Map, Half Cell Potential, Polarization
Operational Parameters	e.g: RH, Temperature, Pressure, Fuel Composition

An Example of Model Validation Data being Provided by Ballard:

Current Density Profile along the channel length in an 12 kW stack



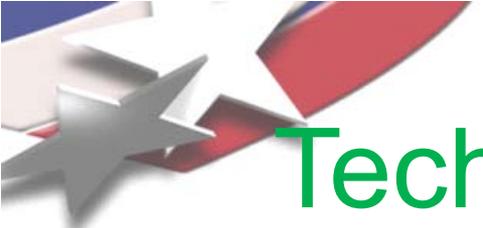
Future Work

PEMFC Model Development, Testing, and Validation

- Complete the development of a 3-D, partially two-phase, single-cell model. (including the incorporation of the microporous layer in anode)
- Complete extensive testing of the 3-D, partially two-phase, single-cell model.
- Perform model validation for single-phase regime, then partially two-phase, ...
- Further develop the coupled PEMFC model/DAKOTA capability for performing parameter estimation and uncertainty quantification.
- Develop a sub-model for specifying physics-based boundary conditions at the GDL/channel interface (to enable the proper account of water flux).

Model-parameter Measurements, Model-validation Data Generation

- Complete measurements of model-input parameters and generation of model-validation data in the single-phase operating regime.
- Initiate and perform measurements of model-input parameters and generation of model-validation data in partially two-phase operating regime.



Summary of Technical Accomplishments

- A single-phase, three-dimensional, single-cell model was developed and significant progress was made toward meeting the year-end milestone (“Develop a three-dimensional, partially two-phase, single-cell model”).
- The present PEMFC model was coupled with DAKOTA* for performing efficient parametric, design and optimization studies.
- Capabilities of the present PEMFC model were demonstrated in case studies by computing effects of operating temperature, cathode RH, back-pressure, and cathode stoichiometric flow ratio on PEM fuel cell performance.
- Utility of the coupled PEMFC model/DAKOTA capability was also demonstrated.
- Capability for simulating cathode MPL effect was implemented and demonstrated.
- The present PEMFC model was used to simulate a PEMFC with zigzag flowfield.
- A sub-model for membrane interfacial resistance was developed and demonstrated.
- Pore size distributions of GDLs w/ and w/o MPL were measured via porosimetry.
- Polarization curves were obtained for different temperature, RH, and cell segments.
- 2 journal publications, 2 proceeding papers, and 3 conference presentations were generated so far. A team member also served as co-editor of a book on PEMFCs.

* DAKOTA refers to a toolkit for design, optimization, and uncertainty quantification developed and being enhanced by Sandia National Laboratories



Thanks to

U. S. DOE EERE Fuel Cell Technologies
Program for financial support of this work
– Program Manager: Jason Marcinkoski

Supplemental Slides



Future work: parameter estimation

- Motivation:
 - Many PEMFC components have variability
 - Different membranes, catalyst loadings, GDL/MPL preparation
 - There is **inherent variability** in model parameters
 - Measurement of some parameters are extremely challenging, if not possible.
 - From previous studies* we learned:
 - Sensitivity analysis (SA) can help match data sets to parameters
 - Bounds are needed on parameters to maintain plausibility
 - Parameter estimation cannot solve model limitations
- Approach:
 - Use the coupled PEMFC model/DAKOTA capability to estimate various parameters using experimental data:
 - IV curves, local data (current, H₂, O₂, Temp)
 - Parameters of interest include:
 - Membrane conductivity, reaction rates, permeability, porosity, two-phase flow parameters, boundary conditions, etc.
 - A nonlinear least squares approach from DAKOTA will be used

* B. Carnes and N. Djilali, "Systematic parameter estimation for PEM fuel cell models," J. Power Sources, 144 (1), 83-93 (2005)

Future work:

Uncertainty Quantification and Validation

- Motivation:

- There is **inherent uncertainty** in many model parameters and data
- Some model parameters are difficult/expensive to estimate accurately
- When model parameter uncertainty can be characterized
 - Bounding intervals
 - Probability distributions

we can propagate the uncertainty through the model into the responses

- Responses may include:

- IV curves, liquid water concentration, localized data

- Approach:

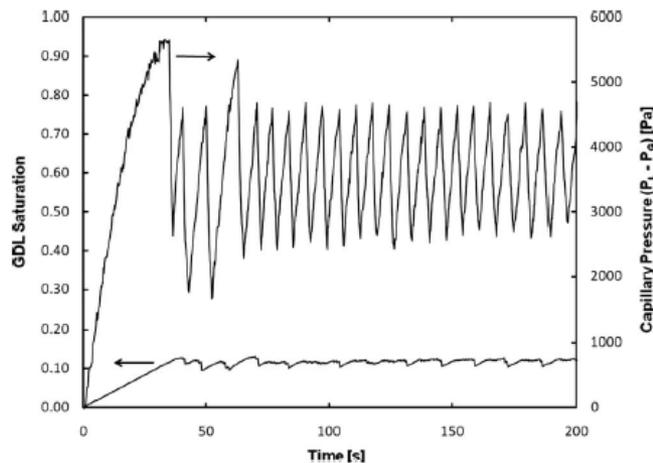
- Use the coupled PEMFC model/DAKOTA capability to quantify various uncertainties
- Methods from DAKOTA for uncertainty propagation:
 - Monte Carlo/Latin Hypercube Sampling
 - Generalized Polynomial Chaos expansions

- Validation will be based on

- Comparing nominal model response to data (point to point comparison)
- Incorporating uncertainty of responses and data (statistical comparison)

Future Work: Gas Channel/GDL Boundary Condition

- The boundary condition at the gas/channel interface can control the liquid saturation in the GDL
 - Impact breakthrough pressure
 - Formation of droplets cause oscillation in the capillary pressure

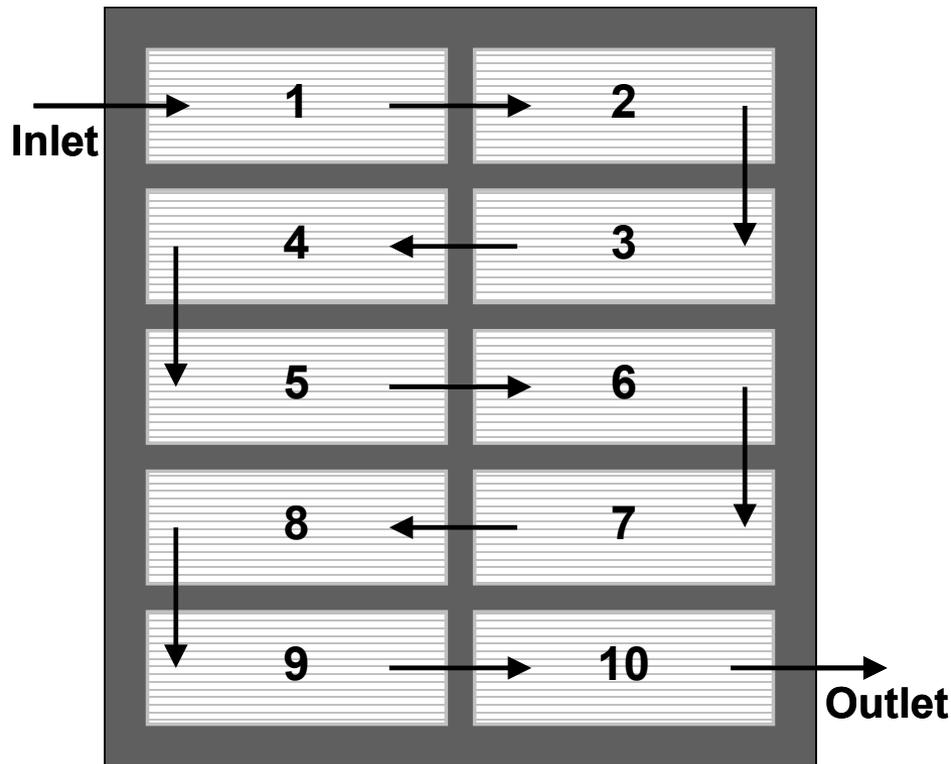


*J.T. Gostick et al., *J. Electrochem. Soc.*, 157, B563 (2010)

- Films may also cause more severe mass-transfer limitations
- Approach
 - Model capillary-pressure oscillations and droplet shear from surface
 - Account for local impact of ribs / channels and the effect of the rib properties
 - Wicking along the rib
 - Do complimentary experimental studies to understand droplet growth and removal mechanisms

Segmented Cell Measurements: Spatial Model Validation

Cathode Flow-Field Segmented Geometry (6-Parallel Serpentine)



- Validate transport models spatially in terms of performance and transport losses (AC Impedance)
- Correlation of polarization and AC impedance data for better understanding of in-plane mass-transport.
- Compare varying cathode GDL performance at different positions within a single cell.
- Investigation of the effects of varying GDL properties.