SAND 2012-XXXX

SAND2012-2037C

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

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Sandia National Laboratories

May 15, 2012

FC027

Overview

Timeline

- Project start date: 10/1/09
- Project end date: 9/30/12
- Percent complete: ~80%

Budget

- Total project funding (over 3 years)
  - DOE share: $2.246M
  - Contractor share: $238K
- Funding received in FY11: $798K
- Funding for FY12: $400K

Barriers

- Barriers addressed
  - **Performance**
  - **Cost**

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

Partners

- Direct collaborations with Industry, University and other National Labs:

  Nissan (no cost), Ballard

  Penn State University

  LANL, LBNL.
- Project lead: Sandia National Labs

* PEM refers to polymer electrolyte membrane

Objective/Relevance

- The project objective is twofold:

  1) to develop and validate a two-phase, three-dimensional transport model for simulating PEM fuel cell performance;

  2) to apply the validated PEM* fuel cell model to improve fundamental understanding of key phenomena involved and to identify performance-limiting phenomena and develop recommendations for improvements so as to address technical barriers and support DOE objectives.

- The coupled DAKOTA/PEMFC model computational capability can be employed to improve and optimize PEM fuel cell design and operation. Consequently, the project helps address the performance and cost technical barriers since improving performance will reduce cost, for example, by using less materials (e.g., catalyst) or minimizing operation cost (e.g., reduce pumping power).

* PEM refers to polymer electrolyte membrane

Approach

Our approach is both computational and experimental with active participation from industrial partners:

- Numerically, develop a two-phase, 3-D, transport model for simulating PEM fuel cell performance.
- Experimentally, measure model-input parameters and generate model-validation data.
- Perform model validation using data available from literature and those generated within the team.
- Apply the validated model to identify performance-limiting phenomena and develop recommendations for improvements.

What distinguishes the present work and previous efforts?

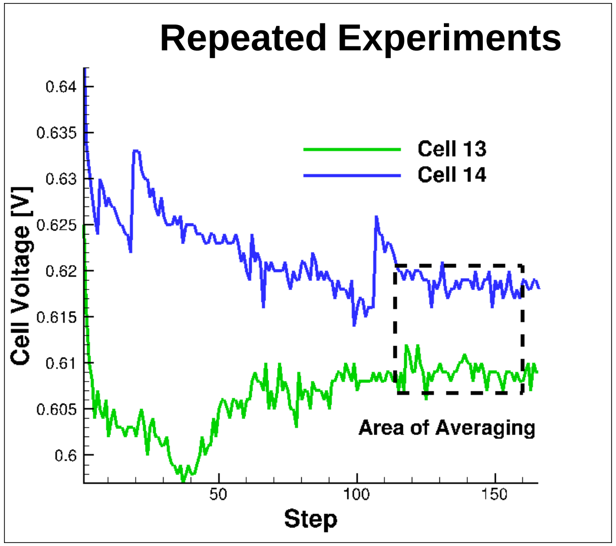
- Couple the PEMFC model with DAKOTA (toolkit for design/optimization) to perform computational DOE (design of experiments) and 3-D detailed probing, sensitivity and variability analyses, and parameter estimation.
- Collaboration with and participation by industry partners, Ballard & Nissan, ensure that the PEMFC model can be used as a practical design tool.

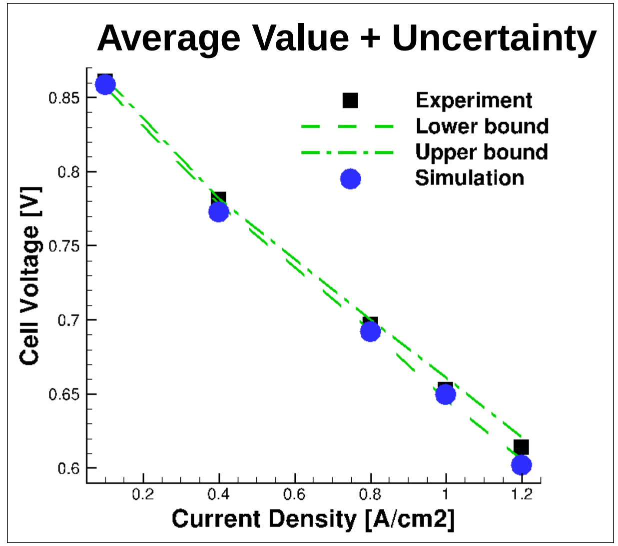
Approach

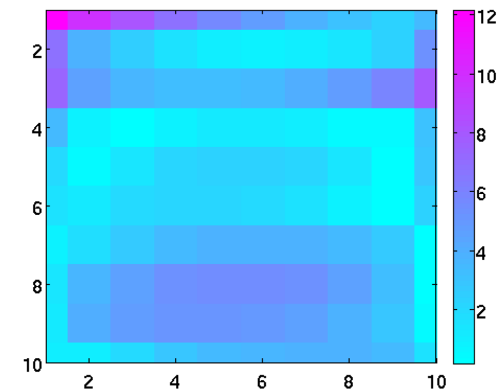
FY12 Milestones, and Current Status

Month/Day/Year	Milestone Descriptions
1/31/2012	Perform the validation of the 3-D, partially two-phase, single-cell PEM fuel cell model. Status: completed.
5/31/12	Validate model under real-world conditions and architectures using data from Ballard and Nissan for non-automotive and automotive applications. Goal is to predict experimental current, temperature and cell voltage within 20% or as defined otherwise by Ballard and Nissan. Status: on track.
7/31/12	Validate fully two-phase, 3-D cell model with microporous layer effect using neutron imaging data. Goal is to match average water thickness in gas diffusion layers within 50% of experimental data. Status: on track.
9/30/12	Generate test suite for PEM fuel cell model and create user manual. Status: on track.

Technical Accomplishment: Uncertainty Quantification of Experiments / Simulations

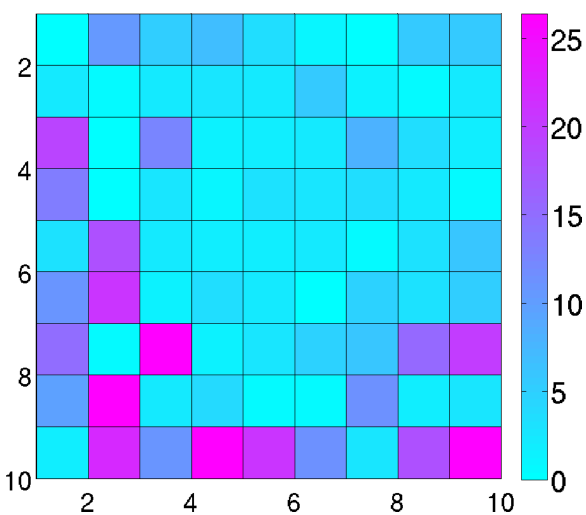






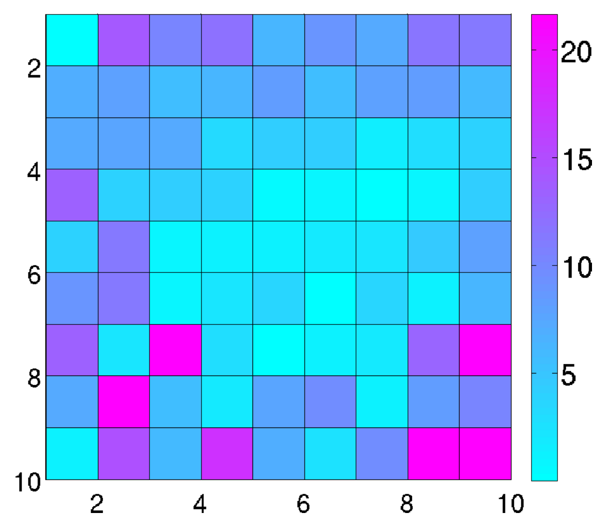
Uncertainty from numerical errors in computations was also quantified using multiple meshes

Repeated experiments enables estimation of uncertainty bounds on cell voltage and current density



Uncertainty at 80C / 50RH / 0.1 CD

Uncertainty in local quantities like **segmented current** (5-25%) can be much higher than for integrated quantities like cell voltage (<5%).



Uncertainty at 80C / 50RH / 1.0 CD

Technical Accomplishment: Validation of Segmented Cells: Cell Voltage

Operating Conditions: (Co-flow)

I= 0.1, 0.4, 0.8, 1.2 A/cm^2

T_{cell} =60, 80 C , P_a=P_c= 25 psig

Inlet %*RH*(a/c)= 25, 50, 75, 100

St(a/c) (H_2/air) =1.2/2.0

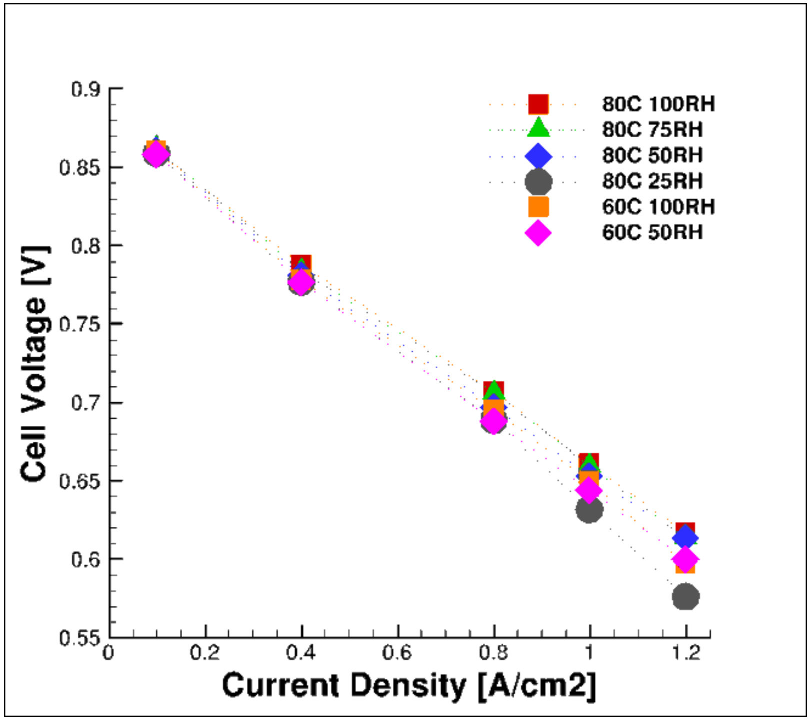
Cell Geometry:

Membrane: 18 um CL(a/c): 7/12 um

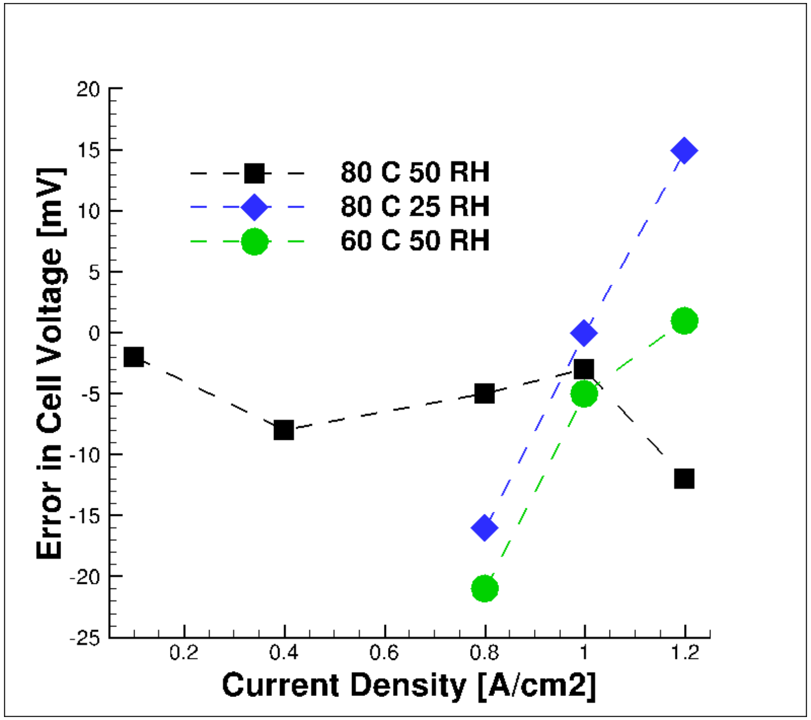
MPL: 40 um GDL: 160 um

GFC: 1×1mm Land: 1.1mm

Cell active area: 50 cm^2

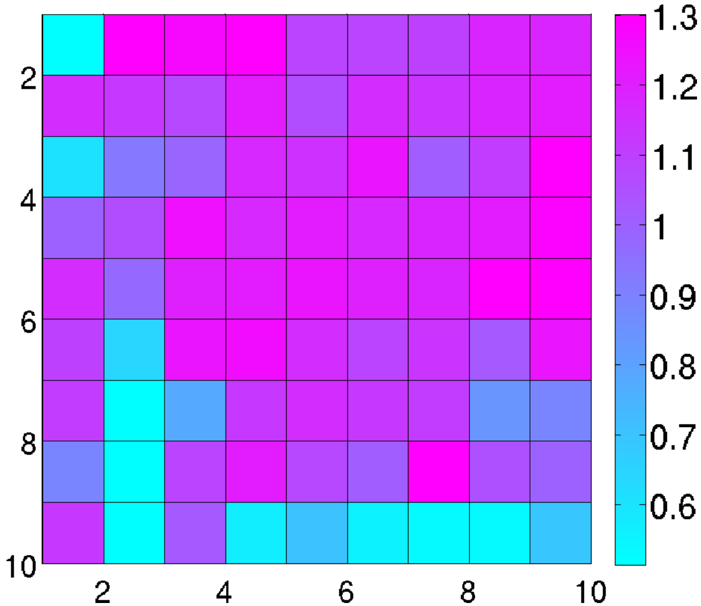


Experimental polarization curves for all 6 operating points

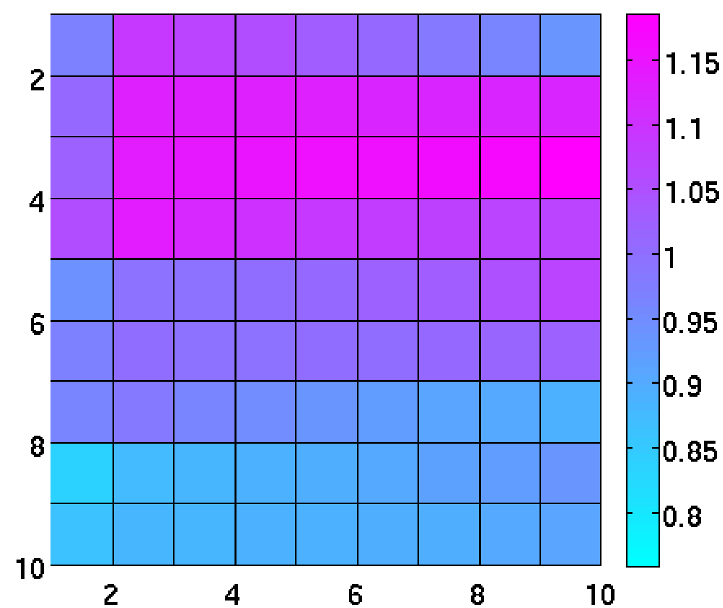


Model validation estimated the cell voltage to within +/-15 mV. Largest errors occurred at high current and at low temperature and relative humidity (RH).

Technical Accomplishment: Validation of Segmented Cells: Current Density (1)



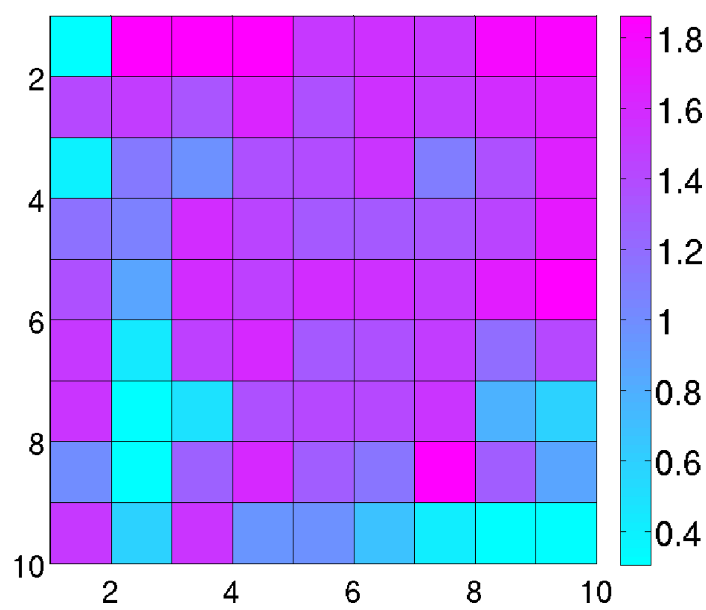
Exp



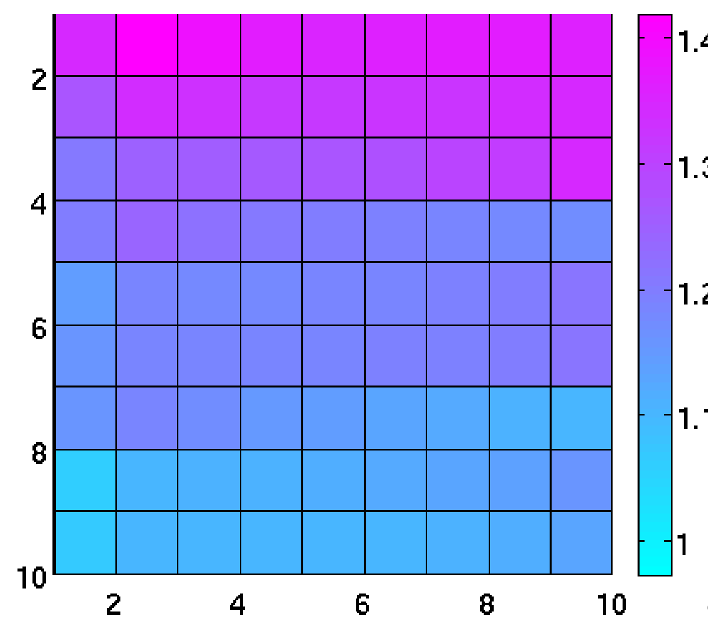
Sim

Comparison of local current density profiles at 80C / 50RH / 1.0 A/cm^2. Note the similar location of the maximum current

Comparison of local current density profiles at 60C / 50RH / 1.2 A/cm^2. Note the similar location of the maximum current



Exp



Sim

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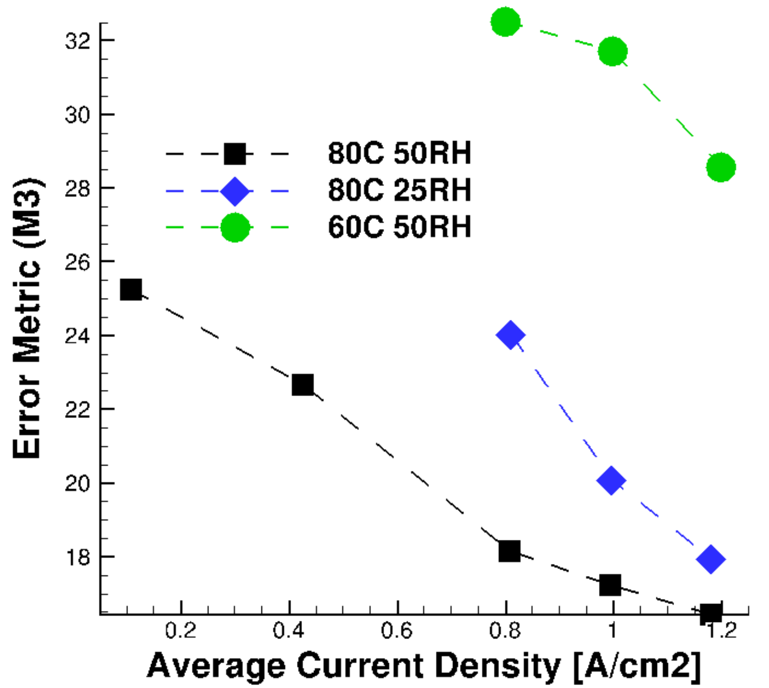
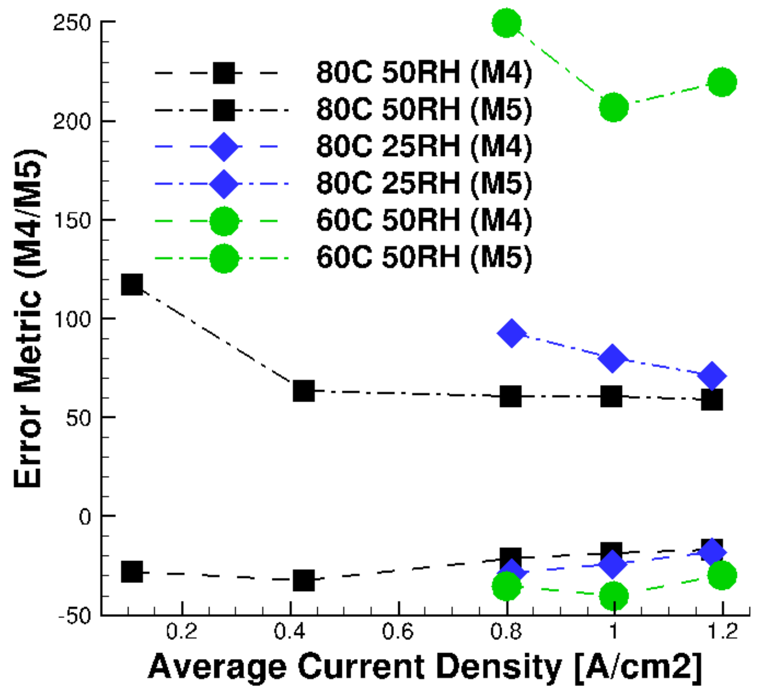
Technical Accomplishment: Validation of Segmented Cells: Current Density (2)



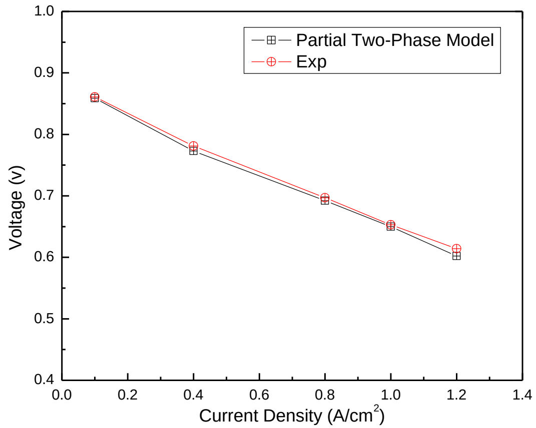


Validation using RMS error in local current density between simulation and experiment at multiple operating conditions.

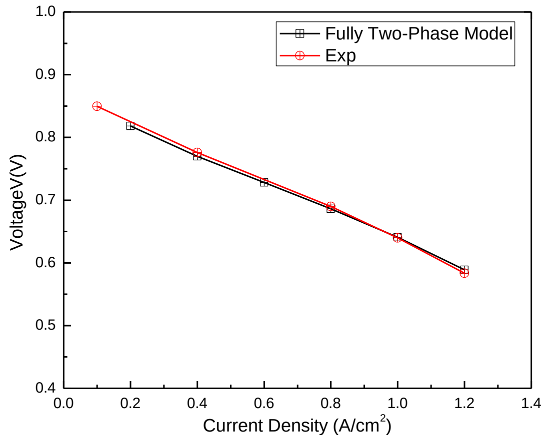
Validation using min/max local error (5/95 percentile) at multiple operating conditions. This shows the largest local error, with over- or under-prediction indicated by a positive or negative sign.

Model validation was acceptable for 80C / 50RH as well as 80C / 25RH. Further improvement is needed for the 60C / 50RH condition.

Technical Accomplishment: Comparison Between Partial and Fully Two-Phase Model



(a) Partial Two-phase Model



(b) Fully Two-phase Model

Polarization comparison between model predictions and measurement

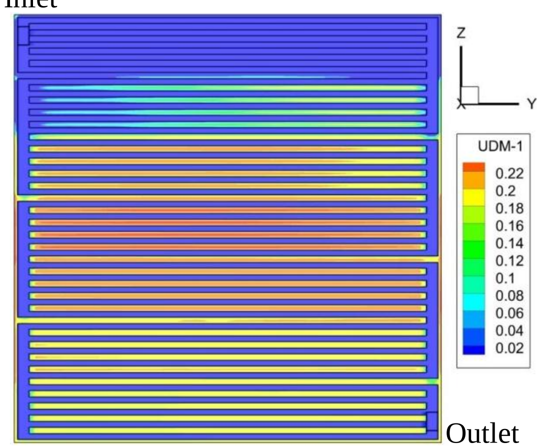
Operating Conditions:

St(a/c) = 1.2/2.0 (H_2/air)

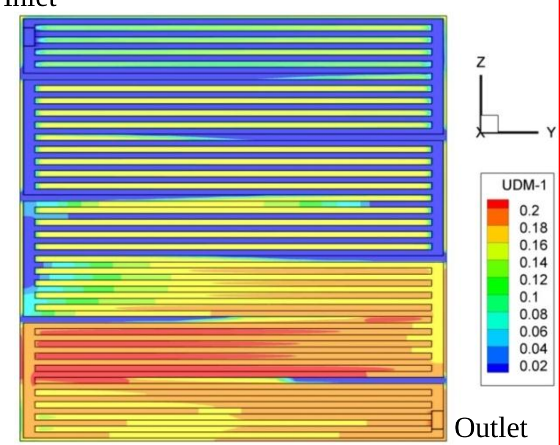
$P_a = P_c$ = 200kPa

T_{cell} = 80 ºC

Inlet %*RH*(a/c)= 50.0/50.0



(a) Partial Two-phase Model



(b) Fully Two-phase Model

Water saturation distribution at cathode gas flow channel/GDL interface

- **Only small difference in polarization prediction between the two models for this large scale cell.**
- **However, the fully two-phase model predicts liquid water in the gas channels comparing to partially two-phase model.**
- **Liquid water predicted by partial two-phase model covers regions only under the bipolar plate.**
- **While liquid water predicted by fully two-phase model appears under both bipolar plate and gas flow channel, especially in the downstream regions near the outlet.**

Technical Accomplishment: Case Study Using Fully Two-Phase Model For Segmented Cell

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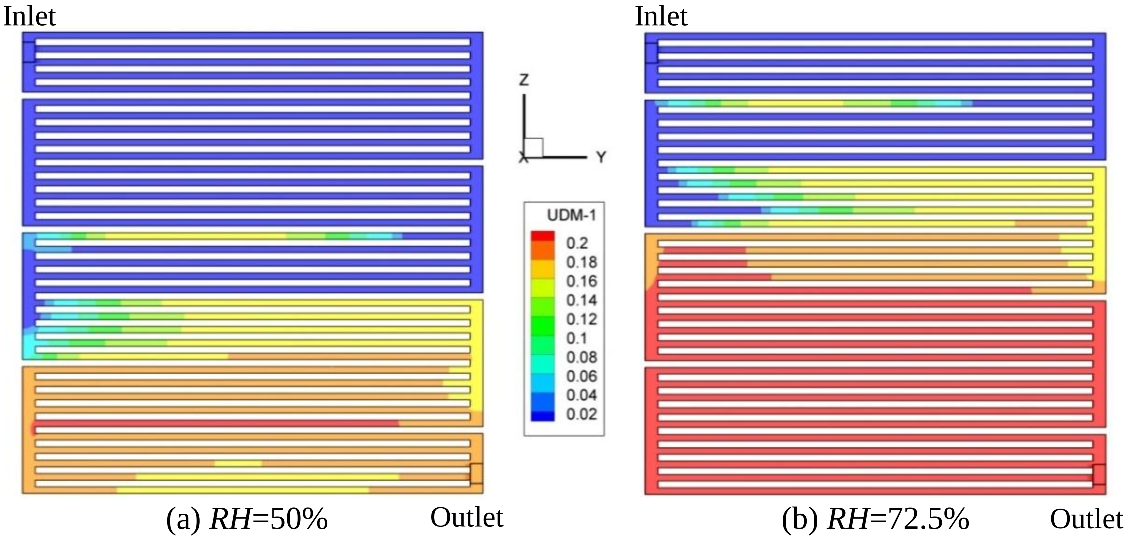
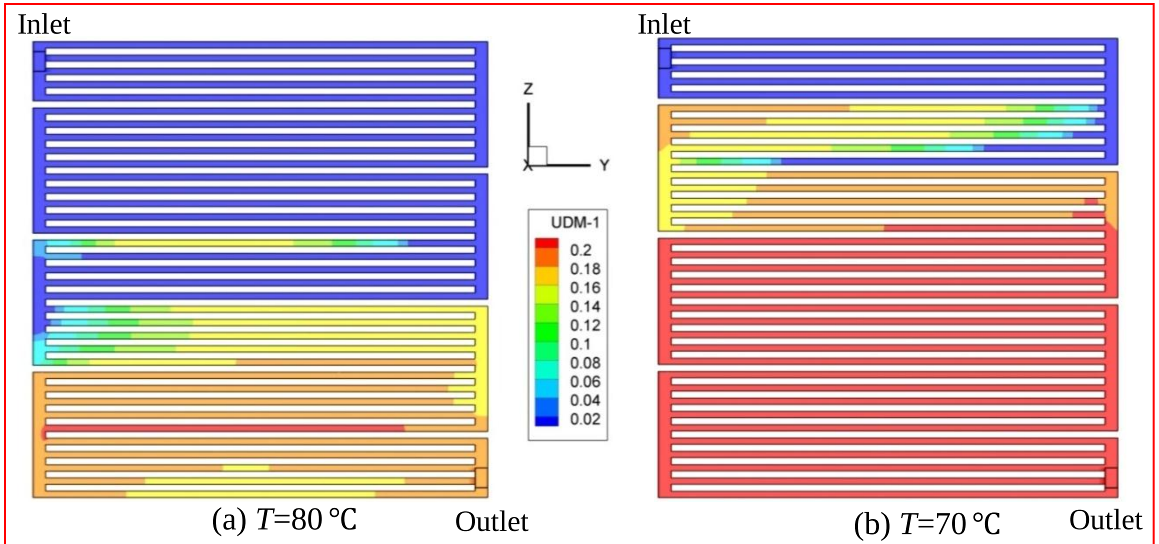


Water saturation distribution in cathode GC for different RH



Water saturation distribution in cathode GC for different cell temperature

Operating Conditions:

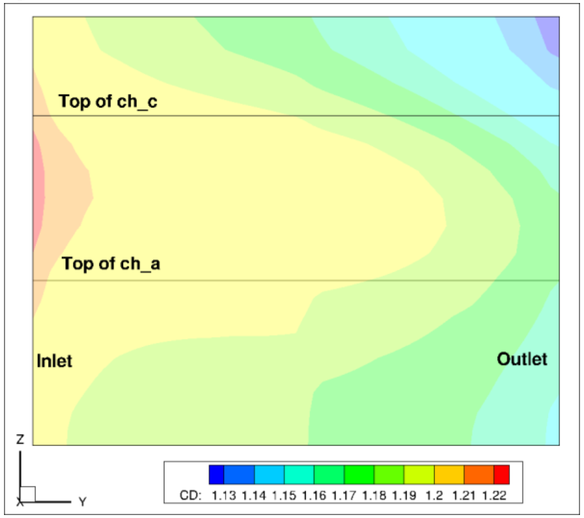
St(a/c) = 1.2/2.0 (H_2/air)

P_a=P_c= 200kPa

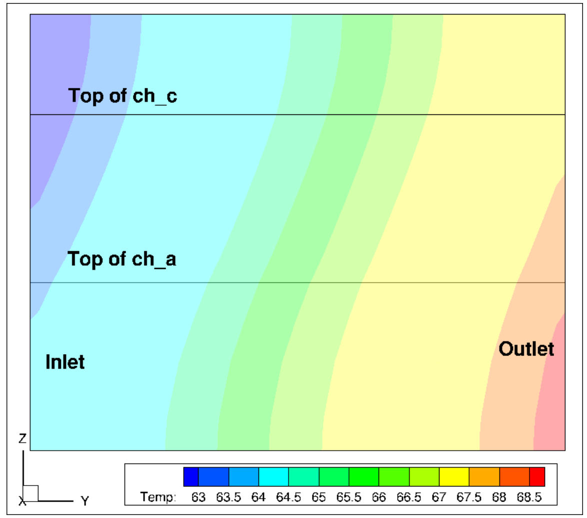
CD=0.8A/cm^2

- **More liquid water appears in the gas flow channel with higher inlet relative humidity.**
- **For lower operating temperature, more liquid water is accumulated inside gas flow channels since low temperature are prone to condensation.**

Technical Accomplishment:
Simulation of Ballard Non-motive Hardware



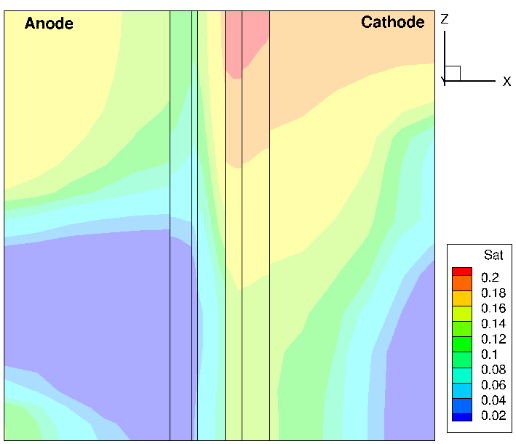
Current at mid-membrane (1.3 A/cm²)



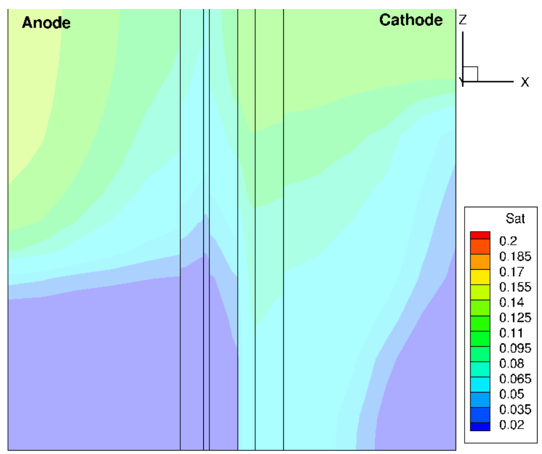
Temp at mid-membrane (1.3 A/cm²)

Operating Conditions:
(Co-flow)

- I= 0.05-1.30 A/cm²
- T_{cell} =70 C
- P_a= 1.15-2.18 atm
- P_c= 1.99-5.10 atm
- Inlet RH(a/c)= 95%
- St(a) (H_2) = 1.6-6.3
- St(c) (air) = 1.8-5.1

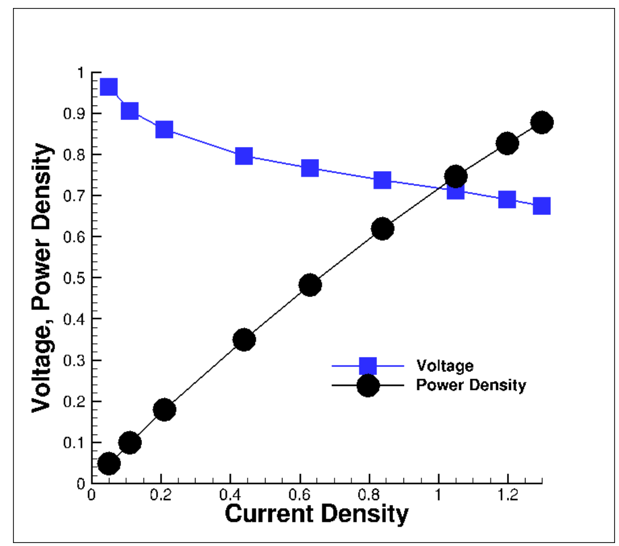


1.3 A/cm²



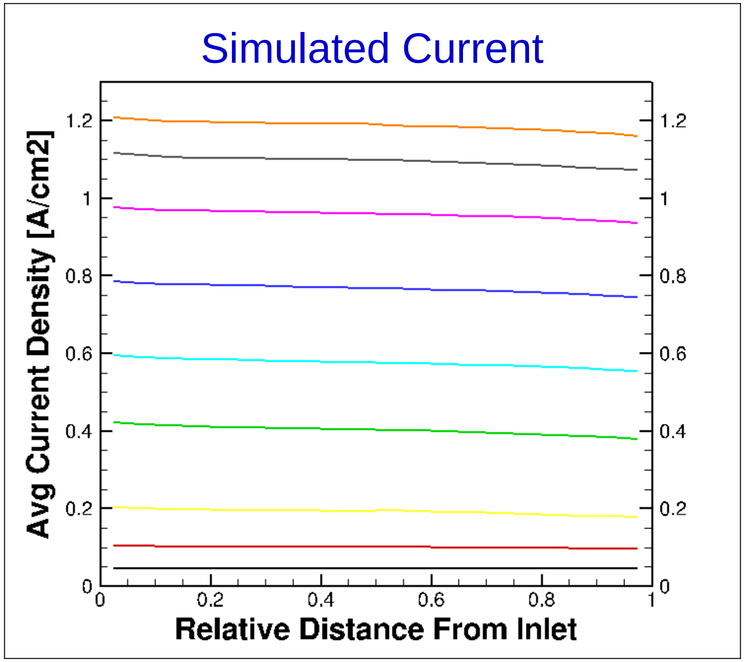
0.44 A/cm²

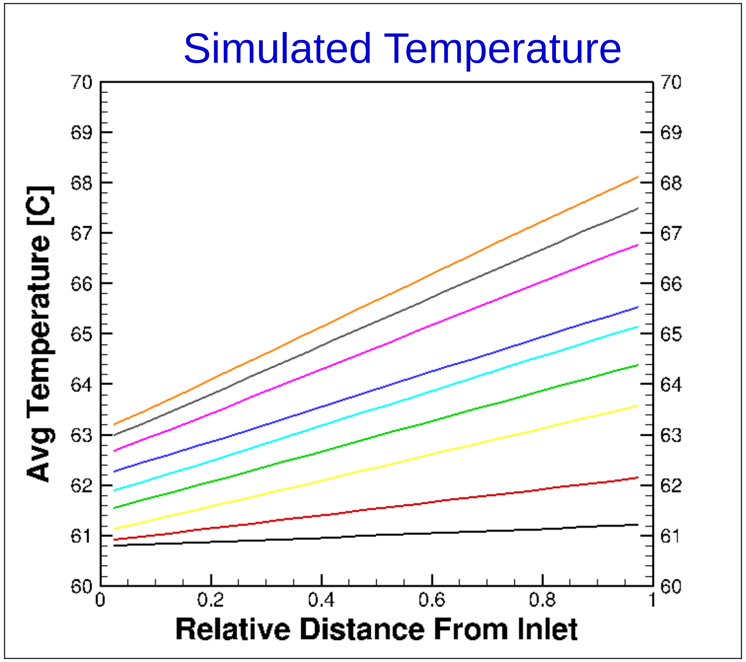
Liquid sat at mid-cell cross section of the membrane-electrode assembly



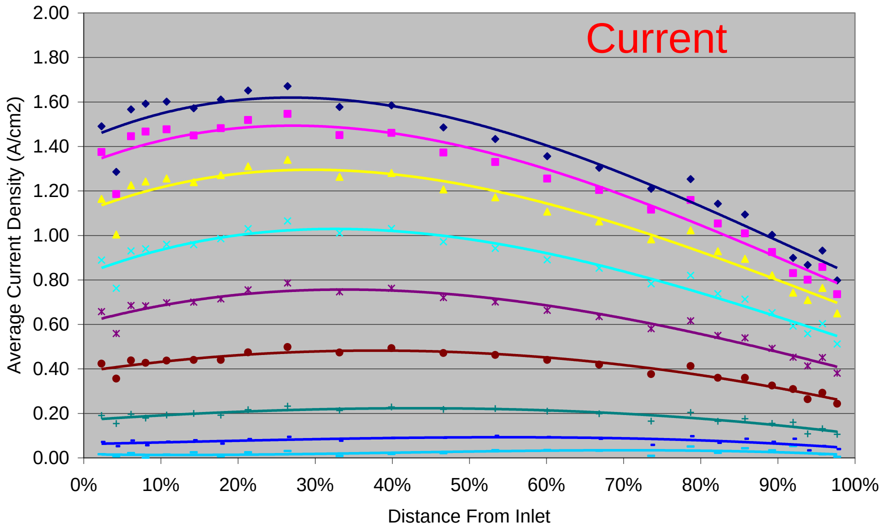
Performance curves

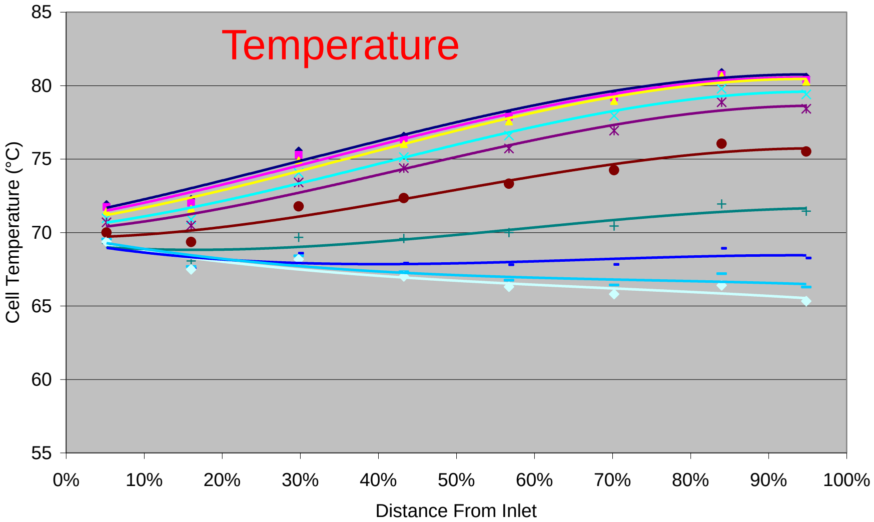
Technical Accomplishment: Validation of Current & Temperature Distribution Measurements





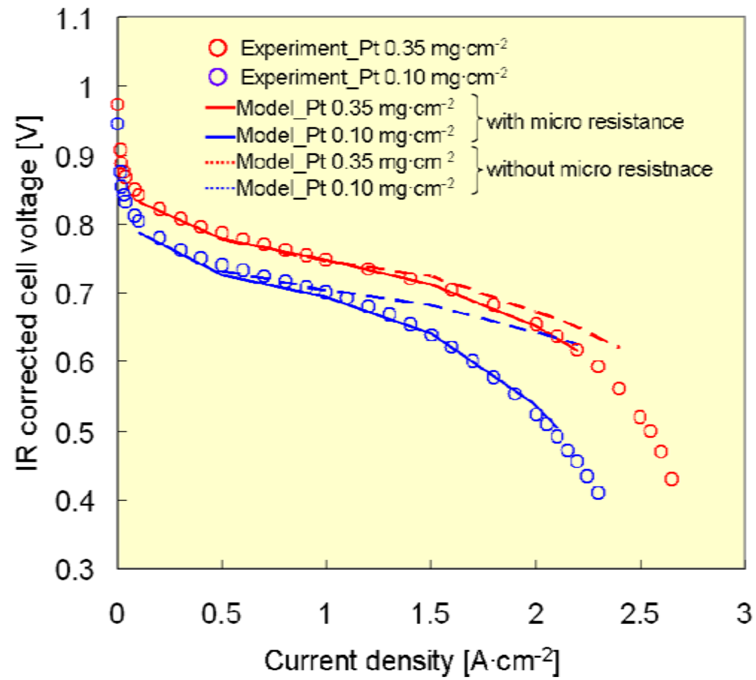
Experimental current/tempèrature distribution obtained by Ballard's mapping tool



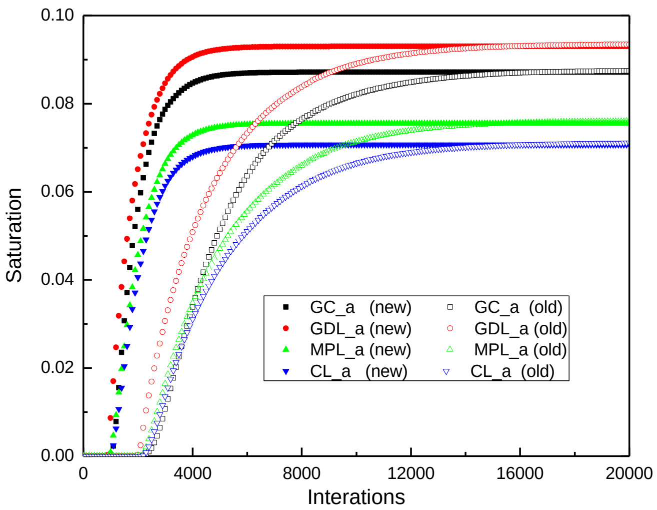


Technical Accomplishment:
Nissan Collaboration and Model Validation

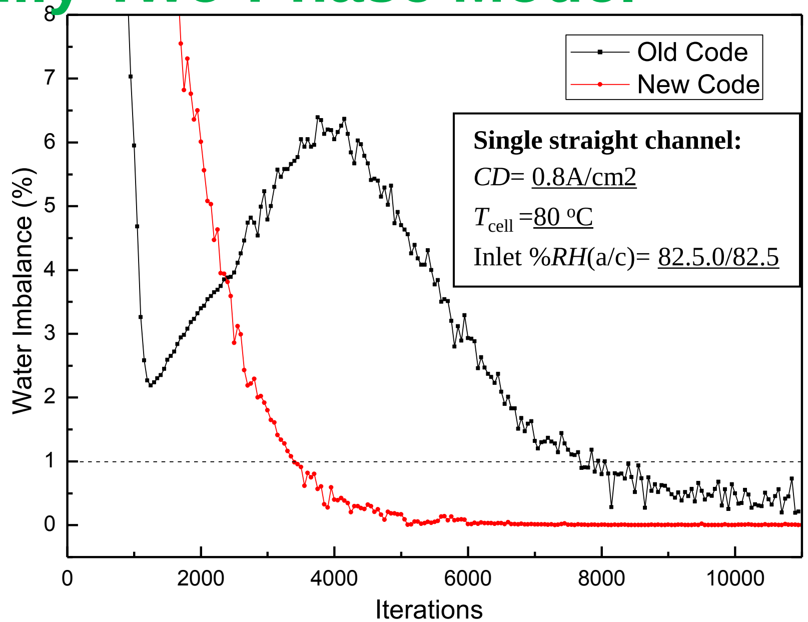
- The team is working closely with Nissan to explore the model application to automobiles.
- Nissan sent a visiting scientist to stay at PSU for one year to collaborate on this project.
- Preliminary success has been achieved by Nissan engineers to modify PSU's two-phase code for predicting fuel cell performance with low-Pt loading catalyst layer, as shown in the figure on the right.



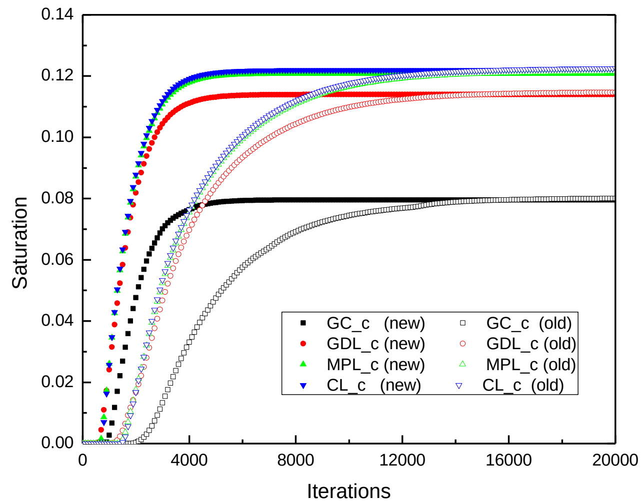
Technical Accomplishment: Improved Convergence for Fully Two-Phase Model



Avg. saturation vs. iteration at **Anode side**



Water Balance for Channel M2 Model

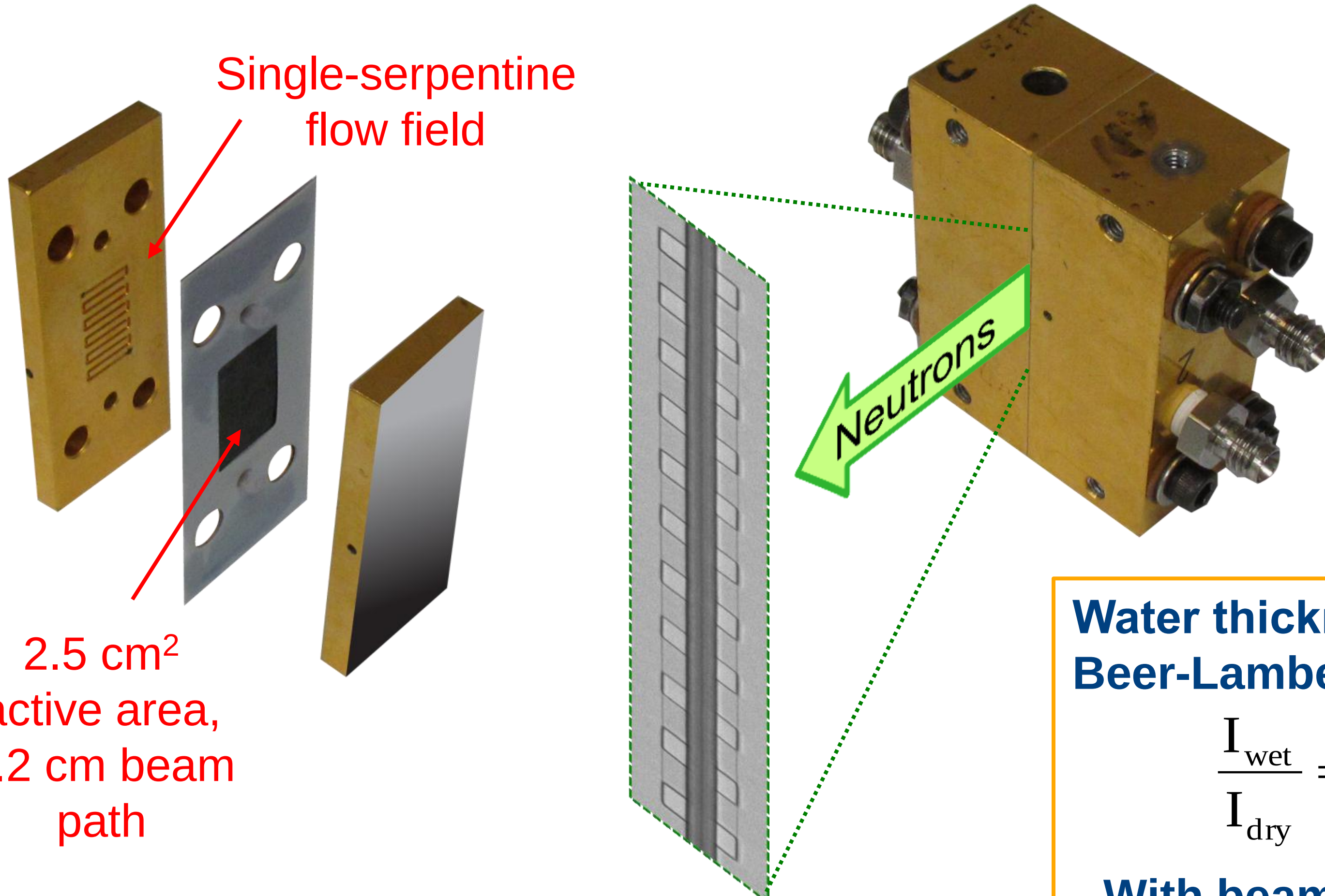


Avg. saturation vs. iteration at **Cathode side**

- **Water saturation convergence at both anode and cathode sides is greatly improved for the latest code.**
- **For a typical case, water saturation converges within about 4,000 iterations for latest code, while it needs about 12,000 iterations for previous version. Thus the simulation time is cut by two thirds.**
- **The water imbalance reaches 1% around 3500 iterations for the latest code, while it needs more than 8,000 iterations for previous code.**

Technical Accomplishment: High-resolution (13 μm) Through-Plane Neutron Imaging

In situ evaluating water content through the thickness.
Varied current density (0.4, 0.8, 1.2 A/cm^2) and RH (50 and 100%)



Water thickness "t" from Beer-Lambert law:

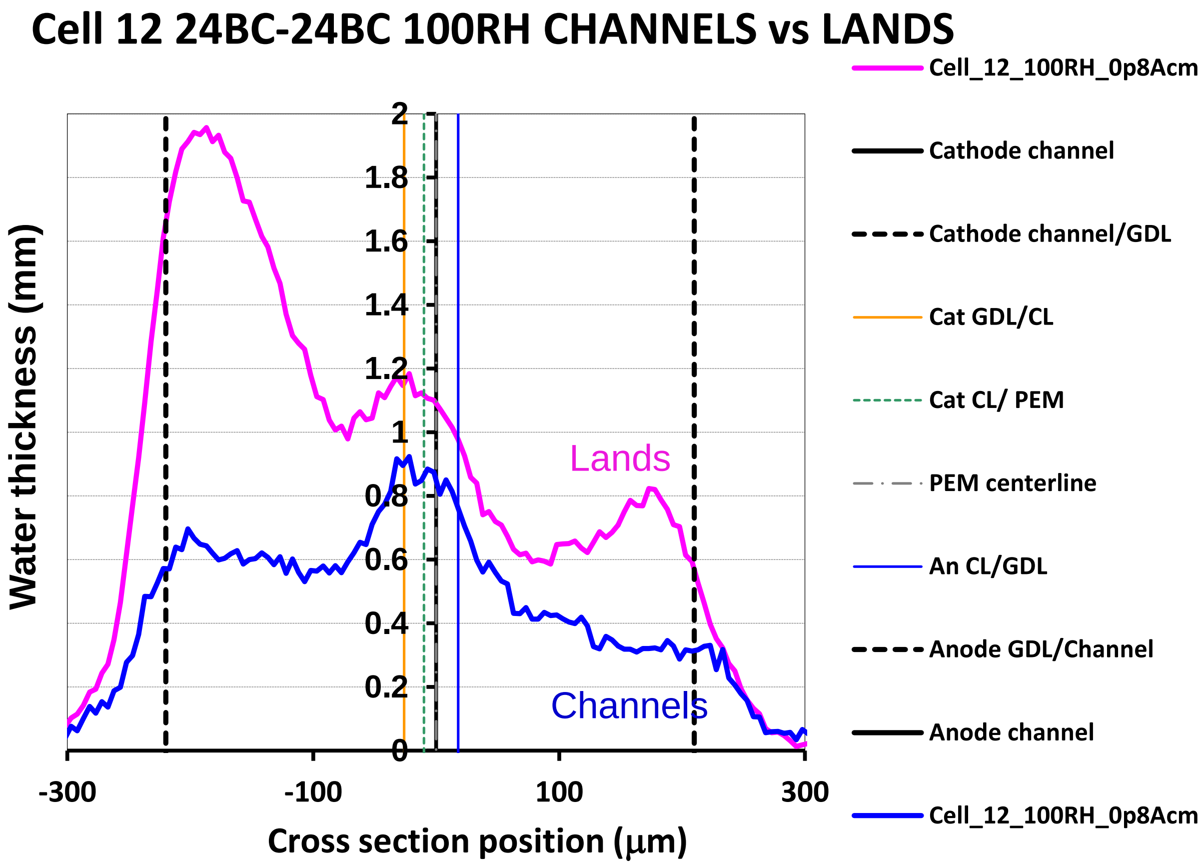
$$\frac{I_{wet}}{I_{dry}} = e^{-\mu t}$$

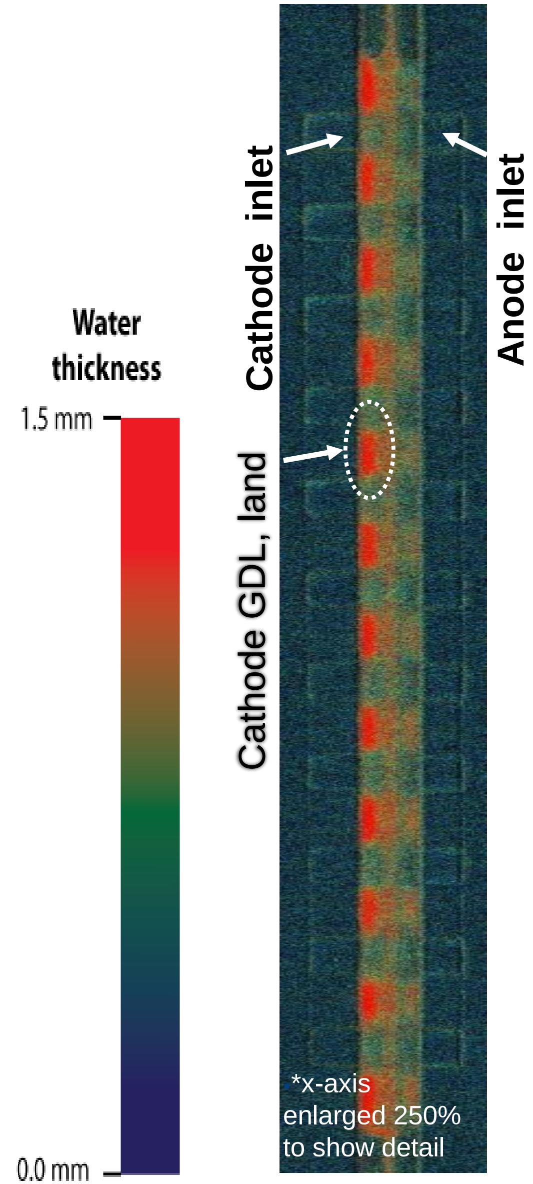
With beam hardening:

$$\frac{I_{wet}}{I_{dry}} = e^{-\mu t - \beta t^2}$$

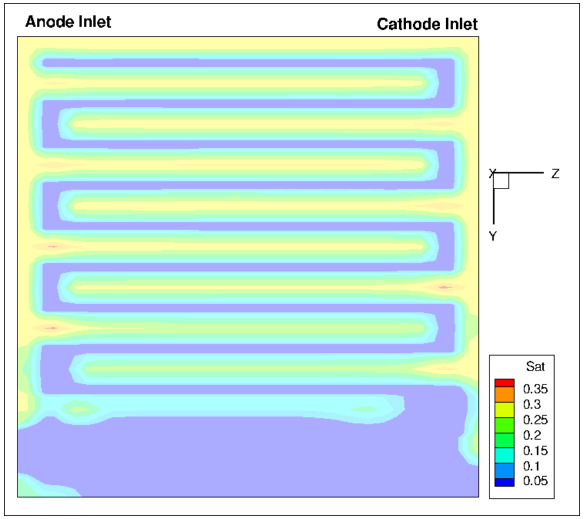
Technical Accomplishment: High-resolution (13 μm) Through-Plane Neutron Imaging

0.8 A/cm², 80°C, 100%RH

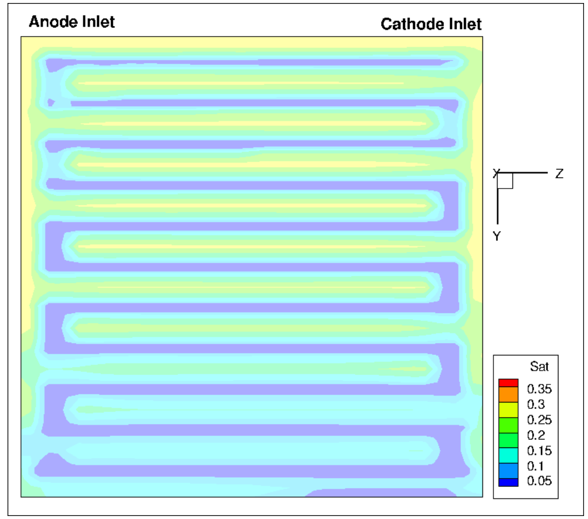




Technical Accomplishment: Prediction of Liquid Water Saturation at Channel/GDL Interface



Anode GDL/Channel Interface



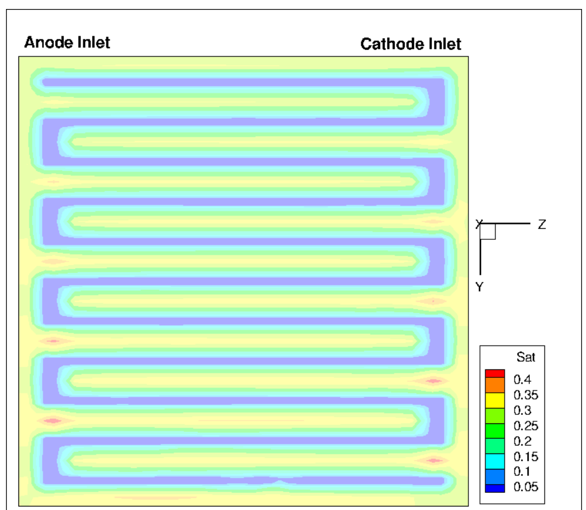
Cathode GDL/Channel Interface

1.2 A/cm^2 **50%RH**

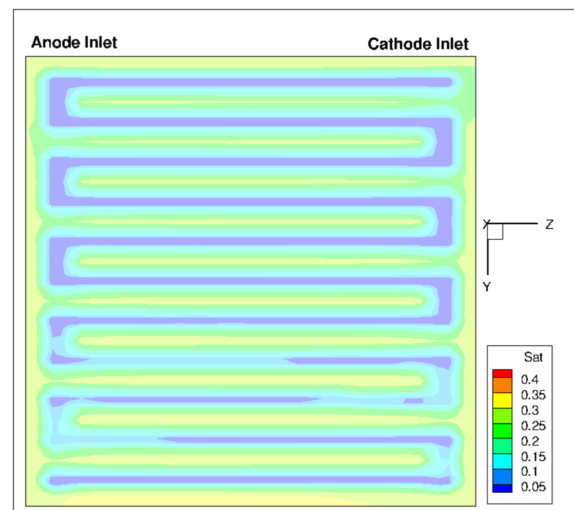
Note the **decreasing** saturation from inlet to outlet and drying of the GDL from the low RH and relatively high flow rates.

1.2 A/cm^2 **100%RH**

Note the increasing saturation from inlet to outlet under fully humidified conditions.



Anode GDL/Channel Interface



Cathode GDL/Channel Interface

Technical Accomplishment: Validation of Liquid Water Predictions Using Neutron Imaging

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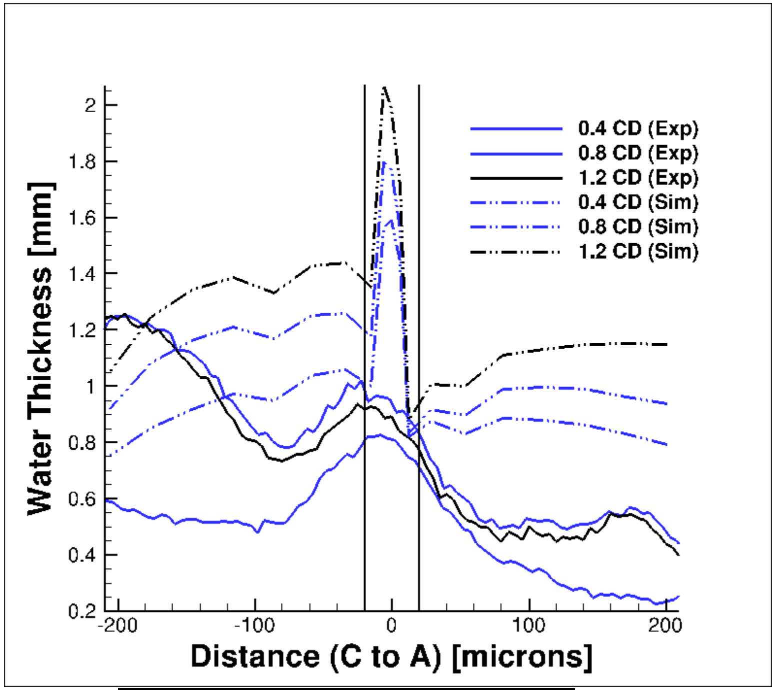
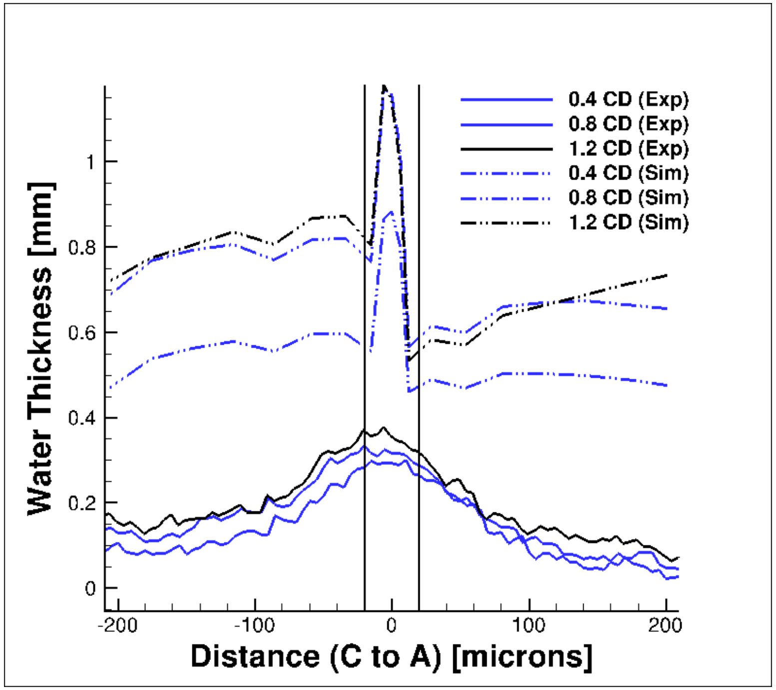


Comparison at 100% RH



Comparison at 50% RH

- **Simulated liquid water saturation was converted to a through-plane water thickness by dividing the cell into small segments (cathode to anode)**
- **The water thickness in each segment was computed by the formula below using saturation (S), area (A), volume (V), porosity (ε):**

$$W = \frac{1}{|A|}\int_V \varepsilon S \, dx$$

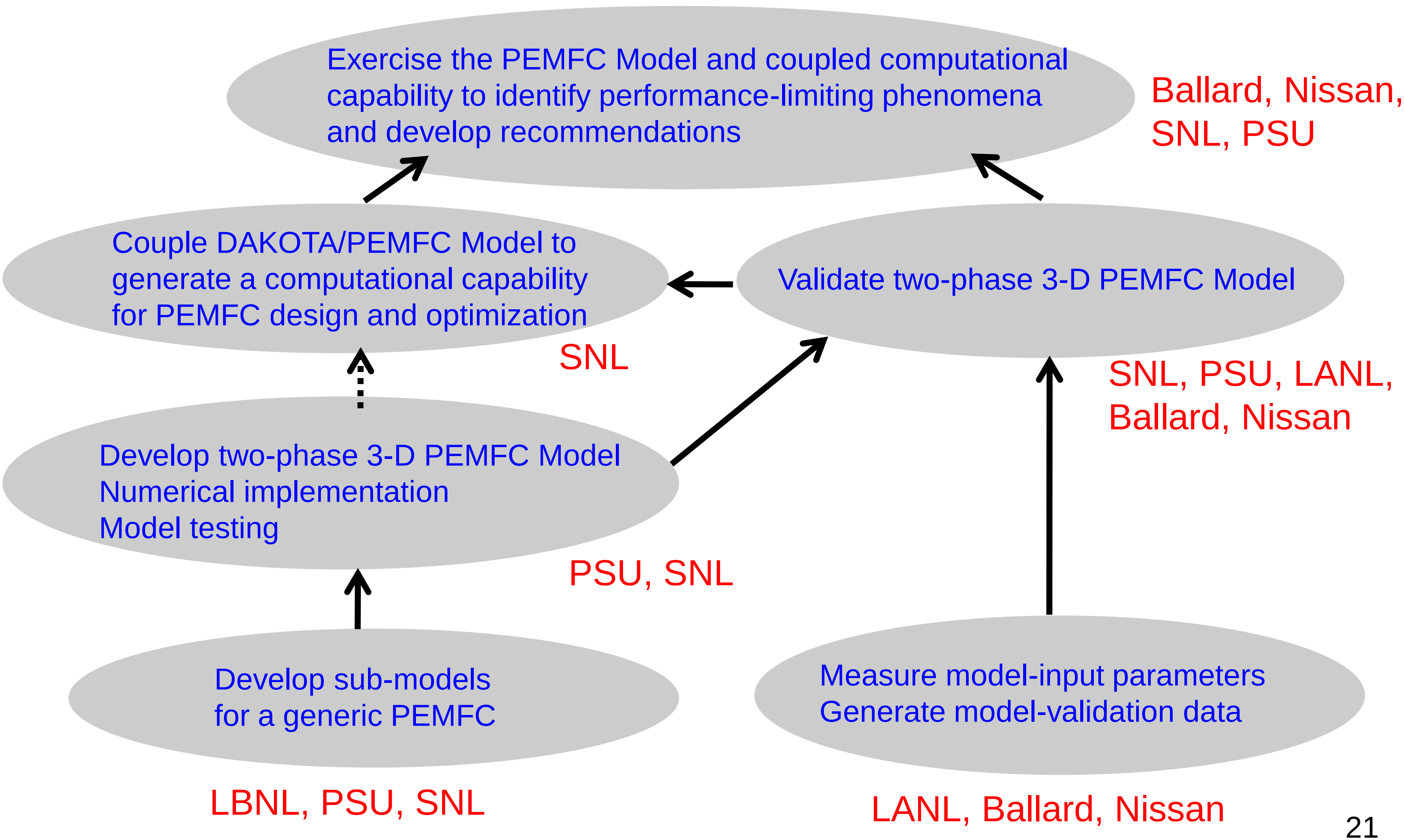
Model validation is not yet acceptable. Further work will be completed to improve the results. However, these initial comparisons indicate that the two-phase model can qualitatively predict the RH effect on liquid water.

Code Dissemination

- A user manual has been documented for the two-phase code we developed over the past decade and further improved in this project.
- The code is currently under testing by project partners, Sandia, Ballard, and Nissan.
- After further development and completion of the project, the software will be made available to the general public under licensing agreements.
- For further information about the two-phase model and computer code, contact Prof. Chao-Yang Wang at cxw31@psu.edu.

Collaborations

Team partners: SNL(prime), PSU(sub), LBNL(sub), LANL(sub), Ballard(sub), Nissan(no cost)



Future Work

Remaining FY12:

1. Complete model validation in the fully two-phase regimes using neutron imaging data obtained by LANL at NIST
2. Complete validation studies using test data from Nissan and Ballard.
3. Complete code manual and test problems.
4. Submit journal articles on model validation.

Summary of Technical Accomplishments

- Year 2 experimental milestone M4 (“Measure *10*x*10* current distribution performance data for model validation for 4 different operating conditions (RH = *25%, 50%, 75%* and *100%*)”) was successfully completed.
- A 3-D, fully two-phase, single-cell model was developed and demonstrated in parametric studies; the Year 2 modeling milestone M3 (“Develop a 3-D, fully two-phase, single-cell model”) is near completion.
- Significant progress has been made in model validation using polarization and current distribution data obtained by LANL using a *10*x*10* segmented cell. Year 2 model-validation milestone M5 is on track.
- Other accomplishments include:
  - Demonstrate the fully two-phase model by simulating a PEMFC with a Chevron flowfield.
  - A nonisothermal pore network model was developed and demonstrated.
  - 3-D CFD simulation was performed to verify the analytical model for droplet detachment.
  - Simplified calculations were performed to estimate water flux at GDL/channel interface.
  - Effect of cell segmenting was investigated and segmentation guidelines were developed.
  - Current/temperature maps and polarization curves with upper/lower bounds were obtained.
- 3 journal publication, 3 proc. papers and 6 conference presentations were generated.

Technical Back-Up Slides

Sensitivity Analysis Using PEMFC/DAKOTA Coupled Model

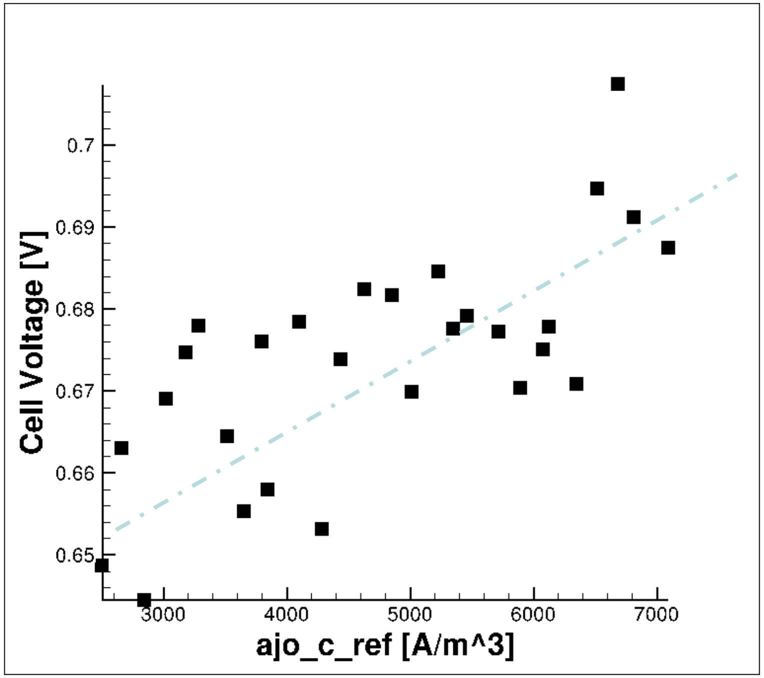
Efficient sensitivity analysis is enabled using the PEMFC/DAKOTA coupled model.

Here we varied 22 parameters to determine the ones with greatest impact on cell voltage.

param	R	m	b
ajo_c_ref	0.71	7.06E-006	0.64
eps_cl_a	-0.57	-0.07	0.71
eps_mpl_c	0.2	0.02	0.66
eps_cl_c	0.16	0.02	0.66
k_p_bl_c	0.13	6.85E+009	0.67
eps_bl_a	-0.13	-0.02	0.68

Linear regression predicts effect of parameter on performance. Positive R value indicates positive correlation.

Cathode exchange current density was most important parameter, followed by anode CL porosity.



Thanks to

U. S. DOE EERE Fuel Cell Technologies Program for financial support of this work

– Program Managers: Jason Marcinkoski
Donna Ho

Reviewer-Only Slides

Addressing reviewers' comments from 2010 AMR

- Collaborative efforts could be a little stronger. Partners seem academia heavy.

  Response: Model development involves one university (PSU) and two national labs (SNL and LBNL). Experimental and validation results come from one national lab and two industrial partners (Ballard and Nissan). We tend to believe that a strong academic presence is a good thing in that it provides a strong theoretical background which is important for such a modeling project. Lastly, the collaboration with industry will become more apparent later this year and in Years 3 and 4, when the model input data and performance data provided by Ballard and Nissan are utilized in model validation, and when Ballard and Nissan start to run the PEMFC model and the coupled DAKOTA/PEMFC model computational capability.

- This project obviously requires a lot of collaboration, since all of the team members must provide substantial input to generate the complex model of the sort envisioned here, especially if the model is also going to be validated (instead of just being used to "predict general trends"). It is also good that the project has an original equipment manufacturer (OEM) like Ford participating with no funding from DOE.

  Response: Nissan has replaced Ford in our project. We are fortunate to have Nissan's participation in this project "with no funding from DOE". We consider Nissan's involvement, and guidance/insights and parameter ranges, etc. provided by Nissan to be very important to the success of this project.

Addressing reviewers' comments from 2010 AMR (Continued)

- The proposed approach of continuing to the partial two-phase model with the validation of the current model seems to be sound. The continued incorporation of the DAKOTA approach to make the model predictive and allow for uncertainty is good. I think it is important to address the water flux as described in the future work.

  Response: Thanks for the positive comments. Yes, it is important to "address the water flux as described in the future work". Specifically, Adam Weber is leading this effort to develop a submodel for properly accounting for water flux at the GDL/channel interface.

- The approach of modeling the behavior and trying to build in the uncertainty is an important step. The focus on generating good data for the model, under a range of conditions, as well as gathering fundamental data on the mass transport and the effect of materials properties, is a definite strength.

  Response: Thanks for the positive and encouraging comments.

- Validation of the modeling to date is weak.

  Response: We've made significant progress in model validation this year. Model validation will be continued in the remaining of this year and also in Years 3 and 4. It should be noted that model validation is being carried out using data obtained by LANL on their segmented cell. In the next stage of model validation, We plan to use cell design and data provided by original equipment manufacturer (OEM) Ballard.

Publications and Presentations

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3. Y. Wang, K. S. Chen, J. Mishler, S. C. Cho, and X. C. Adroher, “A Review of Polymer Electrolyte Membrane Fuel Cells: Technology, Applications, and Needs on Fundamental Research”, *Applied Energy*, **88**, 981(2010).
4. Y. Wang and K. S. Chen, “Elucidating through-plane Liquid Water Profile in a Polymer Electrolyte Fuel Cell”, in *ECS Transaction*, **33** (1) 1605-1614 (2010).
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6. B. Carnes, K. S. Chen, F. Jiang, G. Luo, and C.-Y. Wang, “Systematic parameter estimation and sensitivity analysis using a multidimensional PEMFC model coupled with DAKOTA”, in ASME Proceedings of FuelCell2010, paper #33038.
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11. Y. Ji, G. Luo, and C.-Y. Wang, “Computer Simulation of Liquid Water Transport at Pore Level in MPL and GDL and Their Interface”, presentation at the ASME 8th International Fuel Cell Science, Engineering & Technology Conference, Brooklyn, NY, June 14−16, 2010; paper #33122.

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