

DEVELOPMENT OF A COUPLED 2D-3D FUEL CELL MODEL FOR FLOW FIELD ANALYSIS

George H. Miley*, Glenn Hawkins**, and Jacob Englander**

*Department of Nuclear, Radiological, and Plasma Engineering, University of Illinois at Urbana – Champaign, 61801

**Department of Aerospace Engineering, UIUC, 61801

Abstract

The sodium borohydride and hydrogen peroxide liquid fuel cell developed at the University of Illinois shows promise as a viable energy source for a wide range of applications. To achieve higher powers for a fixed active area, an optimal flow field design is desired. To aid in this venture a coupled 2D-3D model of the fuel cell was developed using the COMSOL Multiphysics software package. At this stage in development, we are comparing the model to experimental data to develop an understanding of its predictive capabilities. The model is governed by the Butler-Volmer equation, Navier-Stokes incompressible flow equations, Darcy's law, and the convection and diffusion equations that express the electrical characteristics, momentum conservation, and mass conservation respectively. Results show modifications in the serpentine flow field design can create a significant improvement in current generation. Calculated voltage versus current plots for the standard design matched experimental results accurately, but deviations entered when other flow fields were used. These inaccuracies are mainly attributed to change in the over potential created by change in the flow field. Despite these difficulties the model correctly predicted the highest performing flow field designs. With these modifications the power can be significantly increased compared to the standard case.

Introduction

The COMSOL model employed in these studies represents the cathode of a working cell. A sketch of the model is shown in fig. 1. The anode is introduced through boundary conditions and is assumed to be working ideally. This is done to reduce the computational requirements of the model so that a solution can be found in a reasonable time, and to allow separate optimization of the cathode and anode in case the best configuration is different for each.

The first and last legs of the continuous channel seen in fig. 1 are the locations of the inlet and outlet. The serpentine design is a one dimensional representation, but the assembly is a three dimensional structure. While the reactant flows along the channel from inlet to outlet, some fluid diffuses into the diffusion layer. Transport through the diffusion layer involves

diffusion and permeation through the porous structure of the layer. The reactant, H_2O_2 , is consumed and water is produced at the bottom surface of the diffusion layer. The current density is computed at this interface.

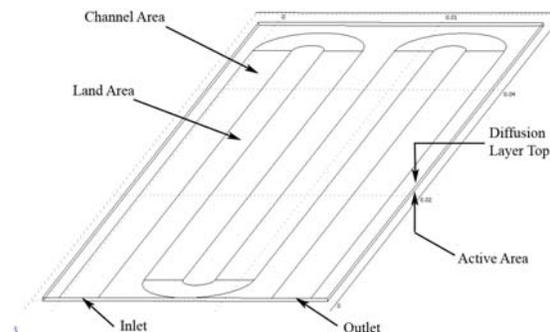


Figure 1: The Model Domain

Governing Equations

The governing equations used in the model are taken from the COMSOL package and include: current generation, momentum conservation, and mass conservation. Each is discussed in the following sections.

Current Generation

Electrical current in this treatment is generated at the interface between the membrane and the diffusion layer, where the catalyst is deposited. This is a first order representation of the actual cell where the catalyst is distributed over the diffusion layer. Within the model this region exists as a boundary on the bottom of the three dimensional diffusion layer. The electrical characteristics are tied to chemical reaction rates through the Tafel type Butler-Volmer equation 1:

$$i_c = -(-S_a \delta_o) \frac{cH_2O_2}{cH_2O_2^{ref}} \exp\left(\frac{F\eta_{op}}{nRT}\right) \quad (1)$$

Here, the current density is a function of the specific surface area of the active layer, S_a , the number of electrons transferred in the reaction, n . Other parameters are Faraday's constant, F , the ideal gas constant R , the cell temperature, T , and the overpotential, η_{op} .

While the reversible potential, which is the theoretical half-cell potential, is a constant for a given temperature, the fuel cell potential varies due to voltage losses due to internal resistance. In order to generate a current versus voltage plot,

Report Documentation Page

*Form Approved
OMB No. 0704-0188*

Public reporting burden for the collection of information is estimated to average 1 hour per response, including the time for reviewing instructions, searching existing data sources, gathering and maintaining the data needed, and completing and reviewing the collection of information. Send comments regarding this burden estimate or any other aspect of this collection of information, including suggestions for reducing this burden, to Washington Headquarters Services, Directorate for Information Operations and Reports, 1215 Jefferson Davis Highway, Suite 1204, Arlington VA 22202-4302. Respondents should be aware that notwithstanding any other provision of law, no person shall be subject to a penalty for failing to comply with a collection of information if it does not display a currently valid OMB control number.

1. REPORT DATE JUN 2006	2. REPORT TYPE	3. DATES COVERED 00-00-2006 to 00-00-2006			
4. TITLE AND SUBTITLE Development of a Coupled 2D-3D Fuel Cell Model for Flow Field Analysis		5a. CONTRACT NUMBER			
		5b. GRANT NUMBER			
		5c. PROGRAM ELEMENT NUMBER			
6. AUTHOR(S)		5d. PROJECT NUMBER			
		5e. TASK NUMBER			
		5f. WORK UNIT NUMBER			
7. PERFORMING ORGANIZATION NAME(S) AND ADDRESS(ES) University of Illinois at Urbana ? Champaign, Department of Nuclear, Radiological, and Plasma Engineering, Urbana, IL, 61801		8. PERFORMING ORGANIZATION REPORT NUMBER			
9. SPONSORING/MONITORING AGENCY NAME(S) AND ADDRESS(ES)		10. SPONSOR/MONITOR'S ACRONYM(S)			
		11. SPONSOR/MONITOR'S REPORT NUMBER(S)			
12. DISTRIBUTION/AVAILABILITY STATEMENT Approved for public release; distribution unlimited					
13. SUPPLEMENTARY NOTES Presented at the 2006 COMSOL Conference Boston, 22-24 Jun, Boston, MA					
14. ABSTRACT The sodium borohydride and hydrogen peroxide liquid fuel cell developed at the University of Illinois shows promise as a viable energy source for a wide range of applications. To achieve higher powers for a fixed active area, an optimal flow field design is desired. To aid in this venture a coupled 2D-3D model of the fuel cell was developed using the COMSOL Multiphysics software package. At this stage in development, we are comparing the model to experimental data to develop an understanding of its predictive capabilities. The model is governed by the Butler-Volmer equation, Navier-Stokes incompressible flow equations, Darcy's law, and the convection and diffusion equations that express the electrical characteristics, momentum conservation, and mass conservation respectively. Results show modifications in the serpentine flow field design can create a significant improvement in current generation. Calculated voltage versus current plots for the standard design matched experimental results accurately, but deviations entered when other flow fields were used. These inaccuracies are mainly attributed to change in the over potential created by change in the flow field. Despite these difficulties the model correctly predicted the highest performing flow field designs. With these modifications the power can be significantly increased compared to the standard case.					
15. SUBJECT TERMS					
16. SECURITY CLASSIFICATION OF:			17. LIMITATION OF ABSTRACT Same as Report (SAR)	18. NUMBER OF PAGES 50	19a. NAME OF RESPONSIBLE PERSON
a. REPORT unclassified	b. ABSTRACT unclassified	c. THIS PAGE unclassified			

one of the main functions of the model, the overpotential must be known for a given cell voltage. The overpotential function is estimated as described later using data from other sources, including other models and experimentation.

Momentum Conservation

In the two dimensional channel system the flow is defined by the incompressible Navier-Stokes equations. Momentum conservation must include both channel flow and porous flow, governed by Darcy's law, in the diffusion layer. The two regions are defined in separate domains connected by a pressure boundary.

i. Flow Field Momentum Conservation

The flow of reactant within the flow field is defined by the Navier-Stokes equations [1, 2].

$$\rho \frac{\partial \mathbf{u}}{\partial t} - \nabla \cdot \mu (\nabla \mathbf{u} + (\nabla \mathbf{u})^T) + \rho (\mathbf{u} \cdot \nabla) \mathbf{u} + \nabla p_{nav} = 0 \quad (2)$$

$$\nabla \cdot \mathbf{u} = 0 \quad (3)$$

This form is called the total stress formulation and is the version used by COMSOL. The first equation represents conservation of momentum, and the second the continuity equation. In this equation, ρ , is the fluid density, μ is the viscosity, and \mathbf{u} is the velocity vector within the channels. Two dimensions are retained in the channels, whereas 3-D flow is used in the diffusion layer. The two are linked through the channel boundary condition. The assumption of incompressible flow is made in order to maintain a constant fluid density. This allows the decoupling of these two equations from the energy equation, assuming that the viscosity is constant as well [3]. It can be seen that the inlet-outlet pressure difference drives the flow in the channel. In the model, an outlet pressure is set as one boundary condition while a velocity at the inlet serves as the second boundary condition to determine the pressure difference over the entire channel. The inlet velocity is perpendicular to the inlet boundary. The inner walls of the channel have a no-slip boundary condition.

The solution gives a pressure distribution and a velocity profile throughout the entire channel system. These distributions are governed by the pressure driven flow along channels combined with flow across the diffusion layer boundaries.

ii. Diffusion Layer Momentum Conservation

The momentum conservation within the diffusion layer is defined by Darcy's law of flow through a porous media [4].

$$\mathbf{u}_{darc} = \left(-\frac{k}{\mu} \nabla p \right) \quad (4)$$

Here the velocity vector within the diffusion layer, \mathbf{u}_{darc} , is proportional to the permeability, k , the inverse of viscosity, μ , and the pressure gradient, Δp , with the dependent variable being pressure. Both the viscosity and permeability are assumed to be constant throughout the entire diffusion layer. The version of Darcy's law used by COMSOL Multiphysics is shown in equation 5.

$$\nabla \cdot \left(\rho \left(-\frac{k}{\mu} \nabla p \right) \right) = F \quad (5)$$

This form sets the net flow in and out of the control volume equal to the source term F. However, no sources occur in the subdomain of the diffusion layer, only on the boundaries, removing the source term from the equation. The ideal conditions for uniform flow through the diffusion layer are a high permeability and a low fluid viscosity, but this is not fully achieved in practice. There are two types of boundary conditions that can be implemented for driving the flow: a pressure condition or a velocity condition on a boundary. These conditions are both utilized on various surfaces. As the Navier-Stokes flow in the channels produces a pressure distribution, these pressures form a top boundary condition for the Darcy's law equation. The land surface area between the channels has an insulation boundary condition. This is also true for any borders along the edges of the diffusion layer that would be contacting the gasket, preventing any external flow. A Flux velocity condition is imposed at the active anode area, attributed to the reactant loss and water creation, as well as water transportation across the membrane.

Mass Conservation

Mass conservation for a species with concentration c is governed by net flow associated with diffusion and convection [5]. The form used by COMSOL is called the non-conservative formulation and is given in equation 6.

$$\delta_{ts} \frac{\partial c}{\partial t} + \nabla \cdot (-D \nabla c) = R_c - \mathbf{u} \nabla c \quad (6)$$

There are two separate sets of these equations, one set for H_2O_2 and another for the water forming the approximately 30% peroxide solution. The time-scaling coefficient, δ_{ts} , is zero for steady-state operation, eliminating time

deviation. R_c is a reaction rate that exists throughout the entire domain. Since current generation was restricted to the active area boundary, R_c is also equal to zero. The diffusion coefficients, D , are inputted by the model user and are specific to the reactant used. For the peroxide solution used, D_{H_2O} is $1e-6$ m²/s and $D_{H_2O_2}$ is $1.37e-9$ m²/s. The velocity vector, \mathbf{u} , is supplied from the solution of Darcy's equation, thus coupling these equations.

Three active boundary conditions are possible. The first is a molar flux, the second a concentration and the third, a convective flux. The first two are used with the flux existing at the active area plus the concentration occurring at the boundary shared with the flow design. Other areas, such as the land area and the diffusion layer edges have an insulation boundary condition, at which no concentration or flux exists.

The fluxes defined as the boundary conditions are dependent on the amount of electrical current that is generated in the cell. For every two moles of electrons produced at the anode, a single mole of H_2O_2 is consumed. Similarly, a single mole of water is produced for every mole of electrons used. With this known, a flux due to the reaction can be found in relation to the current density by converting amps per second into moles of electrons per second by Faraday's constant.

Water is also transported along with protons across the membrane in a phenomenon called electro-osmotic drag. As protons cross through the membrane, they tend to take water molecules with them into the cathode. The drag coefficient, termed *drag* in the COMSOL model, is the number of moles of water entering the cathode per mole of protons. The precise value of *drag* is dependent on many factors including membrane water content, pressure differentials between the anode and cathode, and concentration differentials [6]. However, in the present calculations, the "traditional" value for transport in Nafion of *drag*=3 was assumed.

Note that COMSOL represents molar fluxes as inward, or into the boundary. Therefore a loss of reactant from the domain is actually a positive value. Hence the H_2O_2 flux is positive, representing a loss of reactant, and vice versa for water, representing a gain.

This completes the boundary conditions for mass conservation and the flow model. As witnessed, the model is highly coupled, with information shared in many ways across model domains via boundaries.

Geometry and Extrusion Coupling Variables

The half-cell model consists of two separate geometries that interact with each other: fuel cell flow channels which are two dimensional, and the diffusion layer which is three dimensional.. Both are easily constructed using only a minimal number of geometric functions provided by the COMSOL Multiphysics solid modeling menus.

By default, these two separate geometries do not automatically share information from their respective physics modes. Instead, the information to be shared must be manually selected. This is the case for the pressure coupling between the channels and diffusion layer. To achieve a proper setup, the COMSOL function called "extrusion coupling variables" was implemented. This allowed transfer of data from a subdomain to a boundary. For the present model, the expression transferred is p , and the name it was given was p_{nav} . This name was then used in the boundary conditions to create the pressure boundary condition. The flow between the channels and the diffusion layer is also a boundary condition and is described elsewhere. With this in place, the model was complete and ready to be solved.

Solution Procedure

Initial attempts to solve the model as a single entity from the initial conditions resulted in non-convergence. It was found that the couplings were too complex for the modeling software to evaluate simultaneously without a fairly accurate initial value. Therefore a method to slowly build a better initial value was developed.

There are three steps to the process. The first step is to remove the coupling between the mass conservation and the momentum conservation. The two momentum conservation modes are then solved together using first rough solution.

With the momentum conservation solved for, this partial solution can then be used to solve for the mass balance. By reinserting the coupling, the velocity profile, found without the velocity due to the species flux, is stored in the convection and diffusion mode. Mass conservation for each species is then solved individually. For the velocity profile to exist the previous solution must be used as the initial value in the solver.

Finally, using the decoupled solutions to all of the physics modes, the momentum conservation dependence on the mass flux is re-entered, and a solution for the entire model using the previous solution as the initial value is

carried out. This process initially requires a low overpotential, near $\eta=0.5$, to work effectively. As other overpotentials are required in order to develop the desired voltage versus current plot, the overpotential can be gradually increased, using the old solution at the previous overpotential as the initial value. This also holds true for the variation of other parameters, such as the exchange current density. Following this process provides a well defined and smooth V-I curve.

Model Normalization and Results

With all parameters evaluated and the model constructed, the model is used to generate a current profile within the cell. As an initial test to ascertain that the model was behaving properly, the model was run at a base overpotential $\eta_{op} = 0.5$ [7].

While it was impossible to determine if the average electrical current density was correct until the result was fully benchmarked against experiments, it was still possible to judge the general validity of the predicted current distribution. The expected distribution should follow the concentration profile, with higher currents directly underneath the flow field, and lower current densities underneath the land area. The model did simulate this characteristic, with a typical distribution shown in fig. 2.

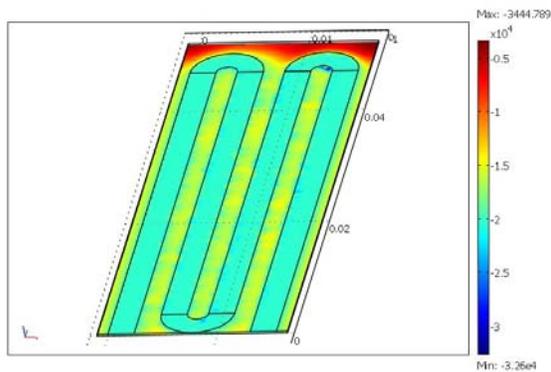


Figure 2: Current density distribution for $\eta_{op} = 0.5$

Another observation is that the regions of lowest electrical current density are those under the channels, where no cross-flow exists. Here, only permeation drives the transport of reactant to the active area. Therefore theoretically it is advantageous to design the cell flow field to achieve a maximum active area. While the current $\text{NaBH}_4/\text{H}_2\text{O}_2$ fuel cell flow field does extend to the active area boundary at the wall, a small artificial gap was necessary in the model

due to meshing constraints. This gap was included in all flow designs studied. This effect should be roughly the same in all configurations and is thought to be negligible.

Finally, a relation between cell voltage and cell overpotential must be found to use in eq. 1. This is described next.

V-I Results Using Overpotentials from Two Dimensional Model

The first attempt at finding correct overpotential values for cell voltages used a 2-D model developed at the University of Illinois [8]. The model, designed for the $\text{NaBH}_4/\text{H}_2\text{O}_2$ fuel cell, requires the user to define the cell voltage, using this information to calculate the overpotential according to equation 7, in which ϕ_m and ϕ_d are the membrane and diffusion layer potentials measured at the active layer boundary.

$$\eta_{op} = \phi_{rev} + \phi_m - \phi_d \quad (7)$$

Using these values for overpotential, a V-I curve was found for several flow field configurations. The land width, l , and the channel width, c , were varied for each design. The starting voltage for each flow model was 1.4 volts as determined in the $\text{NaBH}_4/\text{H}_2\text{O}_2$ characteristics. The final voltage was either 0.4 or 0.3V, with the 0.4V models unable to reach lower voltages due to a build-up of extraneous model data causing convergence errors. The V-I curves found for all the flow field designs are summarized in fig. 3. The designs shown were all serpentine flow but with varying channel widths, c , and land widths, l . The curves exhibit the classic shape attributed to V-I curves, starting with a large drop, typically due to activation losses, with a linear decline in voltage due to resistance losses.

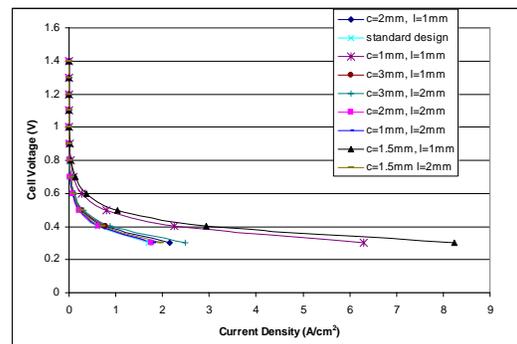


Figure 3: V-I Plots for Varying Flow Designs (c =channel width, l =land width)

For the majority of channel widths c , a small land width l generally gives higher V-I performance. Thus the V-I curves also indicate

that the performance strongly depends on finding the optimal land width. Land width is clearly a crucial parameter to optimize the cell V-I characteristic.

As noted earlier, the major objective of this work was to compare computation with select experimental results using different channel and land widths. The results for current density showed a large amount of error when compared to experimental values of 0.6 to 0.7 A/cm². While some positive difference was expected due to the ideal nature of the half-cell model, the error was excessive in terms of absolute current values. It is planned to correct this in future work by revising the overpotential correlation. Still, the comparative results have provided some valuable information relative to localizing the region of optimal performance.

V-I Results Using Overpotentials from Experimentation

While some important factors in flow field design were observed using the overpotential values found from the two dimensional model, the electrical current and power density values found were significantly different from experimental. Therefore, an attempt was made to normalize the model to past experimental results using the standard “reference” fuel cell flow field design. The currents found experimentally were compared to currents found with the model. By determining the voltage that the model assigned for the experimental current through a linear fit between model points, the corresponding overpotentials were found. The resulting V-I curve proved an excellent match to the experimental results for the reference design, as seen in fig. 4.

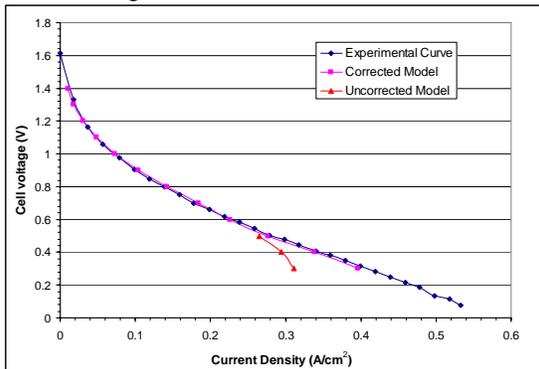


Figure 4: Experimental and Normalized Model V-I Curve

Next, several of the alternate flow field models were modified to use these overpotentials in order to see any differences between the

previous results. The updated V-I curves are seen in fig. 5.

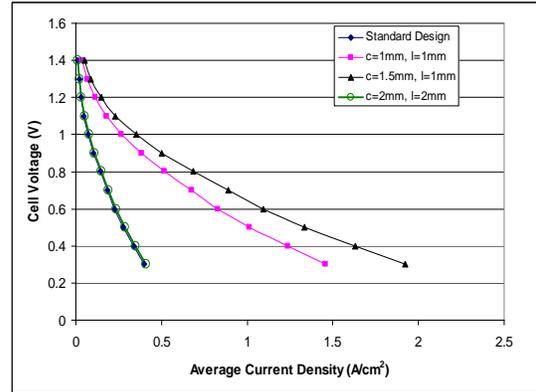


Figure 5: Voltage versus Current using Updated Overpotentials

Observations

To verify the COMSOL mode, the V-I curves found with the model for non-standard flow configurations were compared to experimental results in fig. 6.

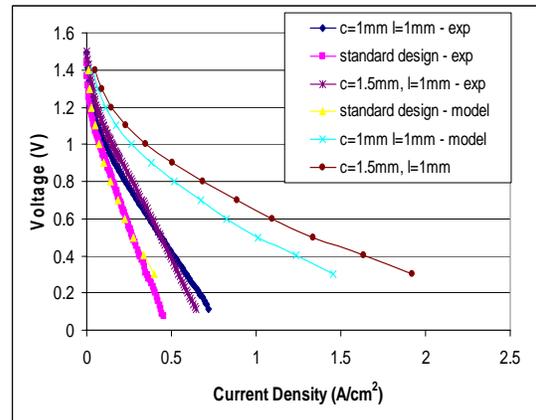


Figure 6: Comparison of normalized model to experimental results for non-standard flow field configurations

As expected, the predicted curve for the standard configuration closely matches the experimental results. However, the modeled results for non-standard configurations show significant deviation from the experimental data. This indicates that normalizing the model to overpotentials found from the standard configuration loses accuracy for significant changes to flow field design. However, as shown from fig. 6, the model successfully predicted the two highest V-I configurations.

Conclusion

An important aspect of the computational technique is the three step procedure described that obtains an appropriate initial condition to allow convergence on a desktop PC. Another key point involves the overpotential correlation which, while a step in the right direction, still needs improvement. This model is still a work in progress and does not yet fully predict the performance of non-standard flow field designs. However, it is already a useful tool for determining trends and the region of optimal performance.

It is vastly preferable to use modeling to localize the region of interest rather than machine new flow plate designs for each data point.

The most important design result found from the modeling process is that the V-I characteristic of the present $\text{NaBH}_4/\text{H}_2\text{O}_2$ cell could be significantly improved by redesigning the flow field to reduce both land and channel width. This change in flow plate design requires the replacement of the present graphite plates with pieces having channel/land dimensions in the region of the optimal space of fig. 6. No other changes in the construction process or the materials used are required.

In conclusion, while the model still contains inaccuracies the desired optimal flow design can be finalized with a few flow-limited experiments located in the region of optimization predicted. Further information can be found in [9].

References

- [1] A. Kumar, R.G. Reddy, "Effect of Channel Dimensions and Shape in the Flow-Field Distributor on the Performance of Polymer Electrolyte Membrane Fuel Cells", *Journal of Power Sources*, vol. 113, pp. 11-18 (2003).
- [2] J.D. Anderson Jr., *Fundamentals of Aerodynamics, Third Edition*, McGraw Hill, Boston, MA (2001).
- [3] M.S. Cramer, *Foundation of Fluid Mechanics*, <http://www.navier-stokes.net>, Blacksburg, VA, (2002).
- [4] P. Nguyen, et al., "Computational model of a PEM fuel cell with serpentine gas flow channels," *Journal of Power Sources*, vol. 130, pp.149-157 (2004).
- [5] F.P. Incropera, D. P. DeWitt, *Fundamentals of Heat and Mass Transfer, Fifth Edition*, John Wiley & Sons, Inc. (2002).
- [6] J.J. Baschuk, X. Li, "A general formulation for a mathematical PEM fuel cell model," *Journal of Power Sources*, vol. 142, pp. 134-153 (2004).
- [7] COMSOL Multiphysics chemical engineering module technical staff, *3D Model of a Serpentine Proton Exchange Membrane Fuel Cell Cathode*, COMSOL Multiphysics (2005).
- [8] E. Byrd, "Modeling a sodium borohydride and hydrogen peroxide fuel cell using COMSOL Multiphysics," *Masters thesis*, Department of Electrical and Computer Engineering, University of Illinois, Urbana Illinois (2006). Also see G. Miley and E. Byrd, these proceedings (COMSOL Annual Meeting 2006).
- [9] G. Hawkins, "Development of a 3D Fuel Cell Model for Flow Field Analysis," *Masters thesis*, Department of Aerospace Engineering, University of Illinois, Urbana Illinois (2006).

Appendix: Constants

S_a	Specific surface area of active layer	$10^8 \text{ m}^2/\text{m}^3$
d	Active layer thickness	10^{-6} m
n	Number of electrons transferred in reaction	2
F	Faraday's constant	96485 J/mol K
R	Ideal gas constant	8.314
T	Cell temperature	303.0 K
η_{op}	Cell overpotential	0.5 V
k	Permeability	$1 \cdot 10^{-11} \text{ m}^2$
$c_{\text{H}_2\text{O}_2}$	Concentration of H_2O_2	$3.029\text{e}+3 \text{ M}$
$D_{\text{H}_2\text{O}_2}$	Diffusion coefficient of H_2O_2	$1.366\text{e}-9 \text{ m}^2/\text{s}$
$c_{\text{H}_2\text{O}}$	Concentration of H_2O	$5.150\text{e}+4 \text{ M}$
$D_{\text{H}_2\text{O}}$	Diffusion coefficient of H_2O	$1\text{e}-6 \text{ m}^2/\text{s}$
R_c	Reaction rate	0
Drag	Electro-osmotic drag coefficient	3.0
μ	Fluid viscosity	$1.056 \cdot 10^{-3} \text{ Pa}\cdot\text{s}$
ρ	Fluid density	$1003 \text{ kg}/\text{m}^3$

Development of a Coupled 2D-3D Fuel Cell Model for Flow Field Analysis

G. H. Miley (Speaker) *Nuclear, Plasma and Radiological Engineering*

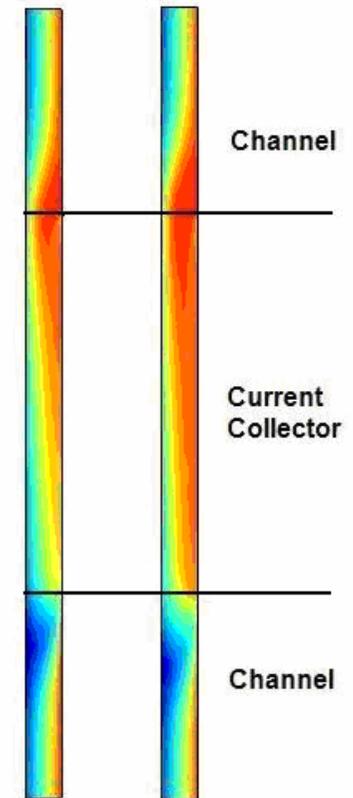
G. G. Hawkins *Aerospace Engineering*

J.A. Englander *Aerospace Engineering*

**University of Illinois at Urbana-Champaign
Urbana, IL 61801**

Outline

- ◆ **NaBH₄/H₂O₂ Fuel Cell**
- ◆ **Description of Model**
- ◆ **COMSOL Application Mode coupling**
- ◆ **Pressure Differential Simulations and Results**
- ◆ **Land Area vs. Permeability and Conductivity Simulation and Results**
- ◆ **Conclusions**



UIUC Fuel Cell Testing Lab



Equipment for Testing of 500W Design includes:

- NI DAQ (Labview)
- Programmable Load
- 2 KW Power Supply
- Water Deionization
- Hot Press
- Pumps / Electronics
- Screen Printer
- Digital Oscilloscope
- Hydrogen Detectors
- Fume Hood
- Safety Shower

NaBH₄/H₂O₂ Fuel Cell

- ◆ Use in fuel cells is a relatively new development
- ◆ H₂/H₂O₂ and NaBH₄/H₂O₂ cells were investigated at NPL Associates, Inc., the University of Illinois (UIUC), and elsewhere
- ◆ Have shown great results, demonstrating the general feasibility of a peroxide based cell
- ◆ Excellent potential for space applications due to high power density and air (oxygen) independence.

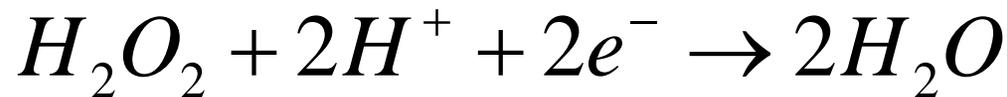
UIUC/NPL Direct Peroxide Fuel Cells

- ◆ **The sodium borohydride/hydrogen peroxide reactions.**

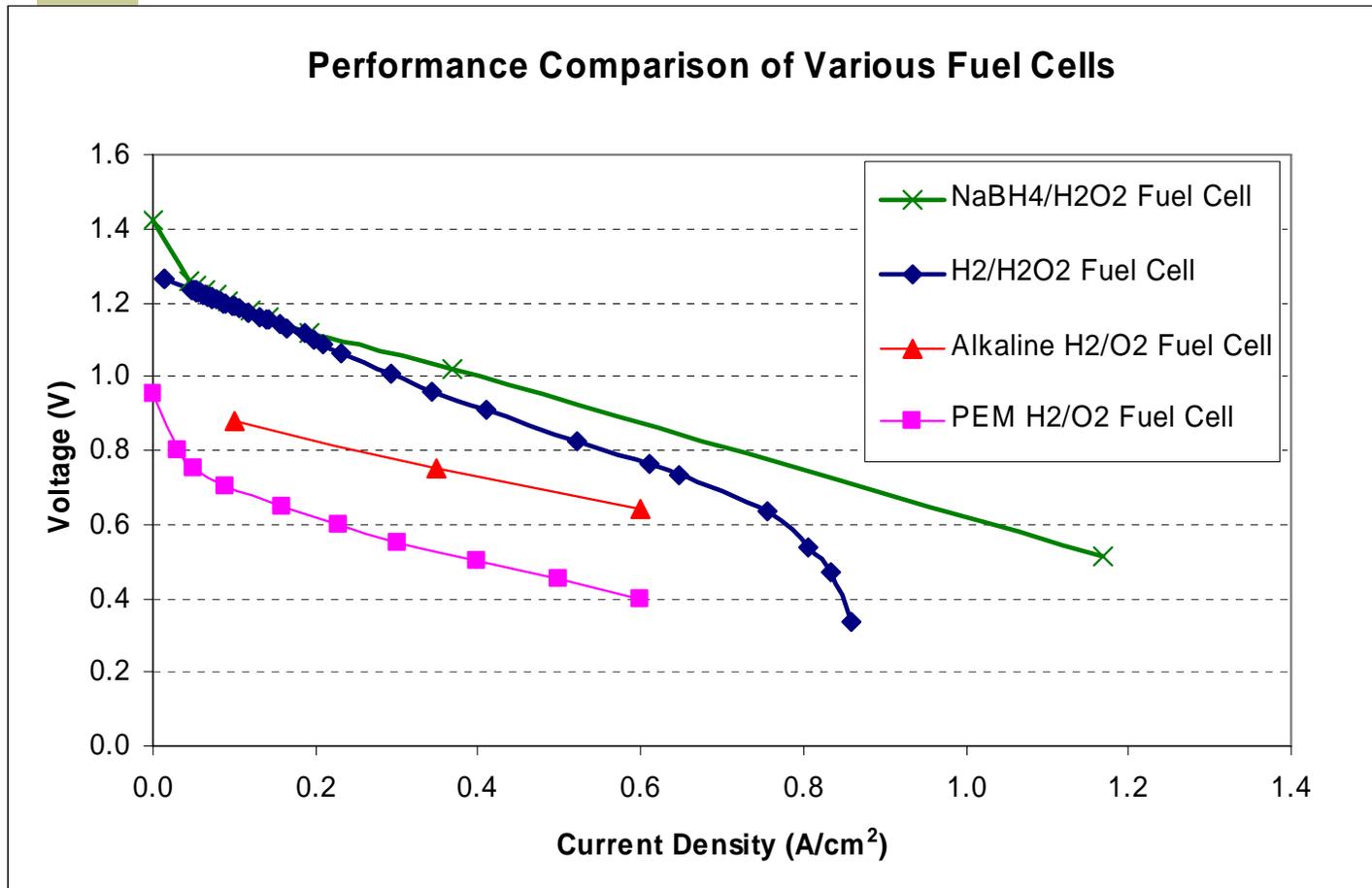
Anode:



Cathode:



Peroxide-based fuel cells (both the H_2/H_2O_2 and $NaBH_4/H_2O_2$ types) offer higher V-I densities than traditional fuel cells under similar conditions



The V-I characteristics of various fuel cells, at room temperature, ambient pressure operation.

COMSOL Users Conf. 2006 Boston,

MA

II – MEA studies at UIUC

- ◆ The UIUC Reference MEA
- ◆ Tests with Various Catalysts (anode and cathode)
- ◆ Results from the 10-40 W-class Test Fuel Cells
- ◆ Implication for the 500-W Stack
- ◆ Initial 500-W Stack testing.

10-40 Watt Class Catalysis Test Cell

- 5 x 5cm flow field/active area
- Nafion 115 untreated w/o catalyst
- Nafion binder for catalyst on diffusion layer
- Graphite felt diffusion layer
- Graphite current collectors
- Aluminum endplates
- Silver epoxy at interface between endplates and graphite plates



10 Watt class fuel cell

Test Cells - Compact 10-40 W Power Units

The 15-W cell shown here uses an integrated cooling channel to dissipate the waste heat generated in the relative small 25-cm² active area.

An optimized version of this small cell generated 36-W at ~ 60°C, representing the highest power density reported to date for a small fuel cell working at sub-100°C.



15-W NaBH₄/H₂O₂ Test Fuel Cell as assembled.

- Flow rate of approximately 200 cm³/min
- Minimal pressure drop even with parallel flow due to low flow rate
- Temperature rise of approximately 15°C
- Heat flux is approximately equal to electrical power (500-W)

Unique Catalyst Issues

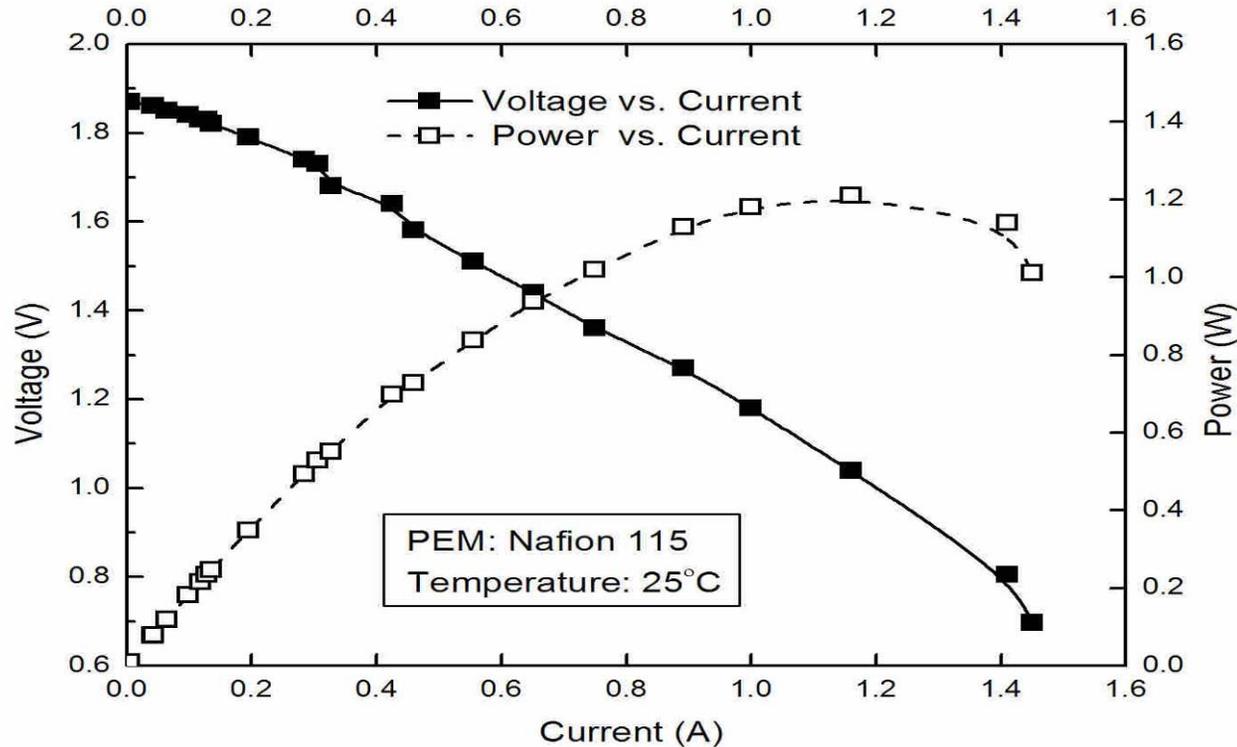
◆ Key issue

- Our reactants decompose violently (gas production) in the presence of conventional fuel cell catalysts such as Pt as well as several other very active catalysts
- This dramatically reduce the cell efficiency

◆ Goals of catalysis studies

- Find catalysts for both anode and cathode reactions that maximize power density without gas production

UIUC/NPL Proprietary Catalysts

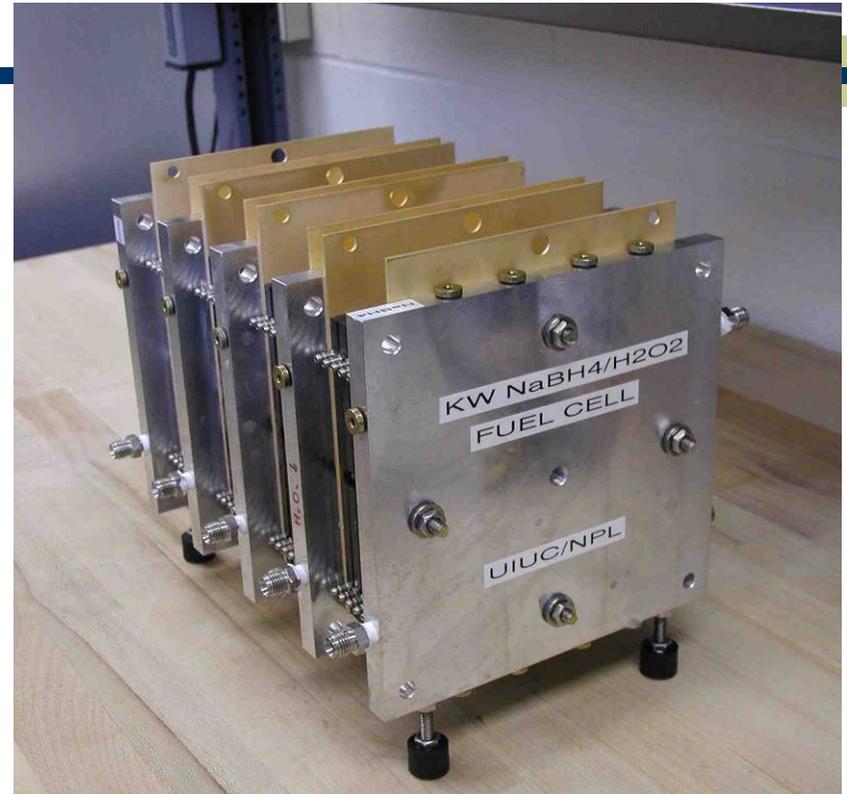
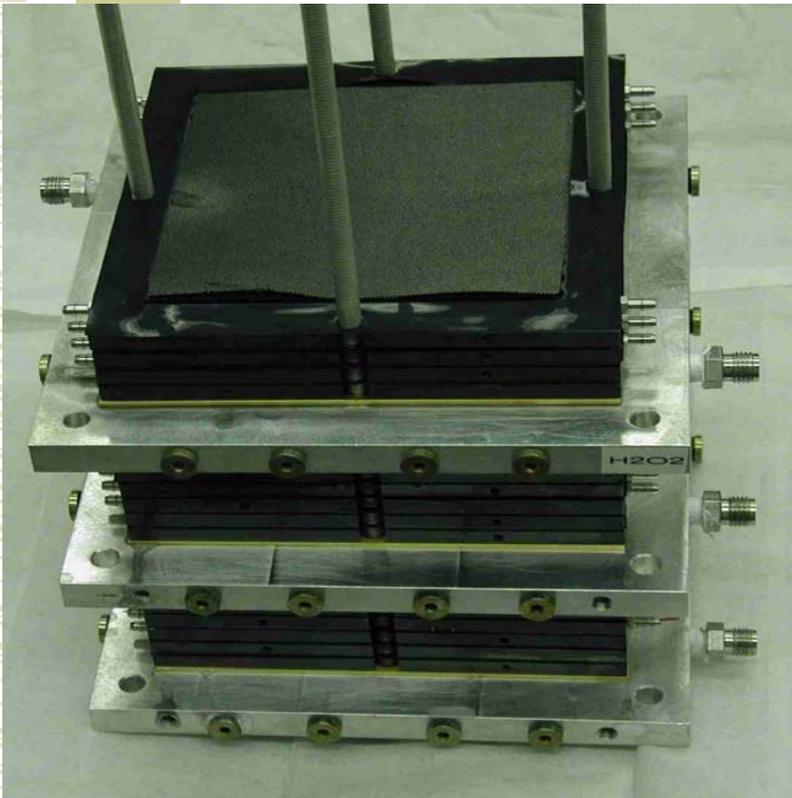


Catalysts optimized for high efficiency

500-W Stack Catalyst Selection

- ◆ For initial tests of the 500-W stack, palladium and gold alloys were finally selected for the anode and cathode catalysts, respectively, for their high performance and relative stability

The 500-W UIUC/NPL $\text{NaBH}_4/\text{H}_2\text{O}_2$ Fuel Cell Stack



The active area per cell was 144 cm^2 and 15 cells were employed to provide a total stack active area of 2160 cm^2 .

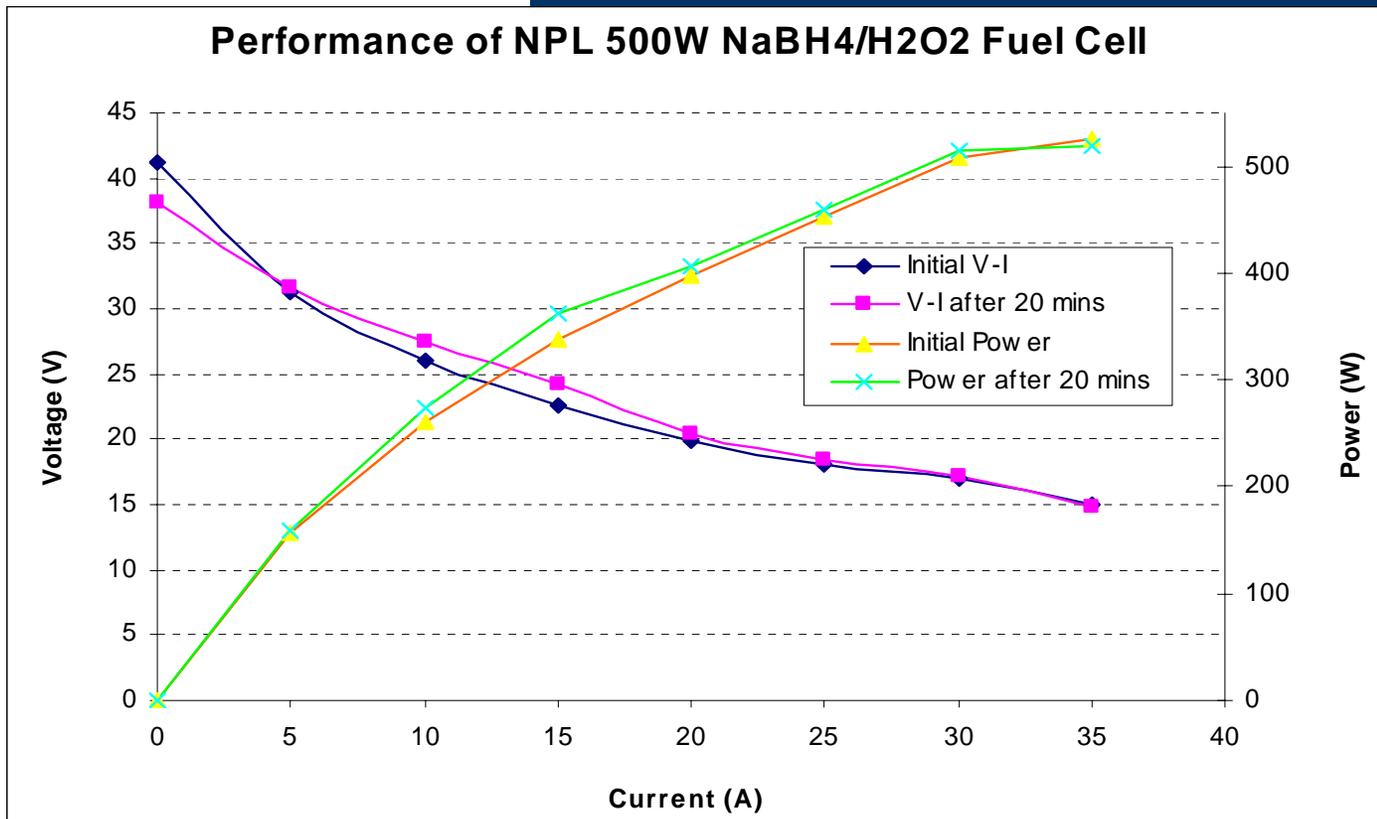
Bipolar Plate Design

- Serpentine design utilized
 - Ensures proper fuel distribution over range of environments
 - Higher pressures caused by serpentine are within design parameters
- External Inlets and Outlets
 - NaBH₄ conductivity causes risk of short circuit when manifolding
 - Non-conductive tubing between cells during series flow prevents short circuit

Stack Parameters

- Flow rate ~ 200 cm³/min
- Pressure drop for series flow is 17 psi for cathode and 8 psi for anode
- Expected temperature rise ~ 15C
- Heat flux ~ 500 W

500-W Stack runs at ~ 29 A, 17 V



V-I and P-I performance of the 500W UIUC/NPL/CUA NaBH₄/H₂O₂ FC.

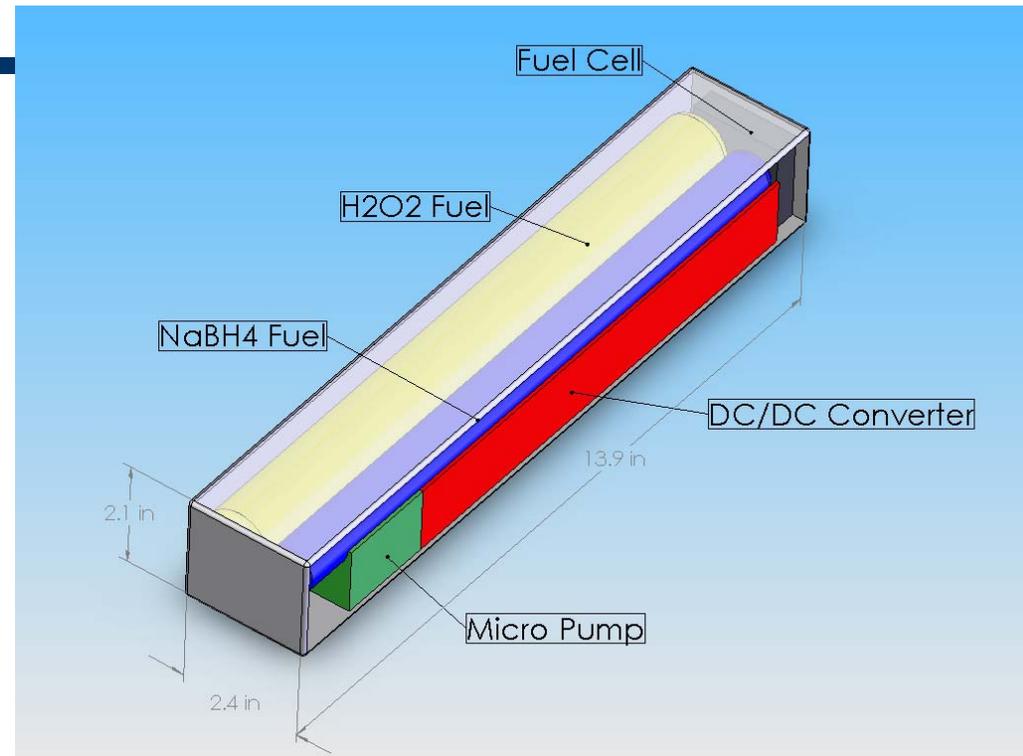
COMSOL Users Conf. 2006 Boston,

MA

6/22/2006-6/24/2006

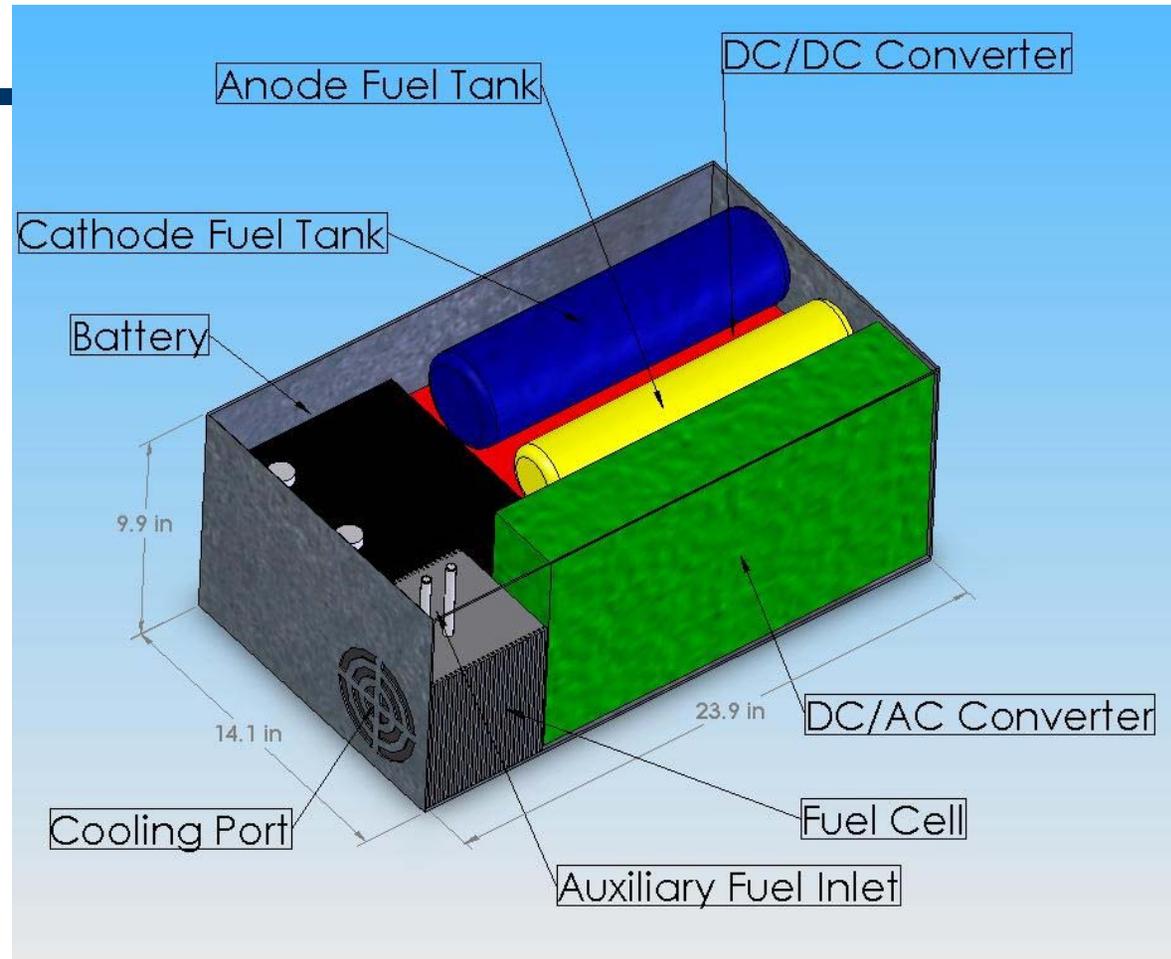
Comments re applications: 20-W I-Charger™ Laptop Power Unit

- ◆ NaBH₄/H₂O₂ I-Charger™ Micro Cell
 - 20 W
 - 500 W-hr
 - ~2.2 lbs
- ◆ SOA Micro DMFC
 - 20 W
 - 500 W-hr
 - 3.5 – 7 lbs



1-kW I-Charger™ UPS System

- ◆ Initial unit designed using retail components.
- ◆ Compact Design providing ~1 hr runtime.
- ◆ Easily expandable with auxiliary tanks for extended operation.
- ◆ Meets or exceeds typical commercial UPS system requirements.
- ◆ ~25% more space efficient than typical UPS



Objectives for COMSOL modeling

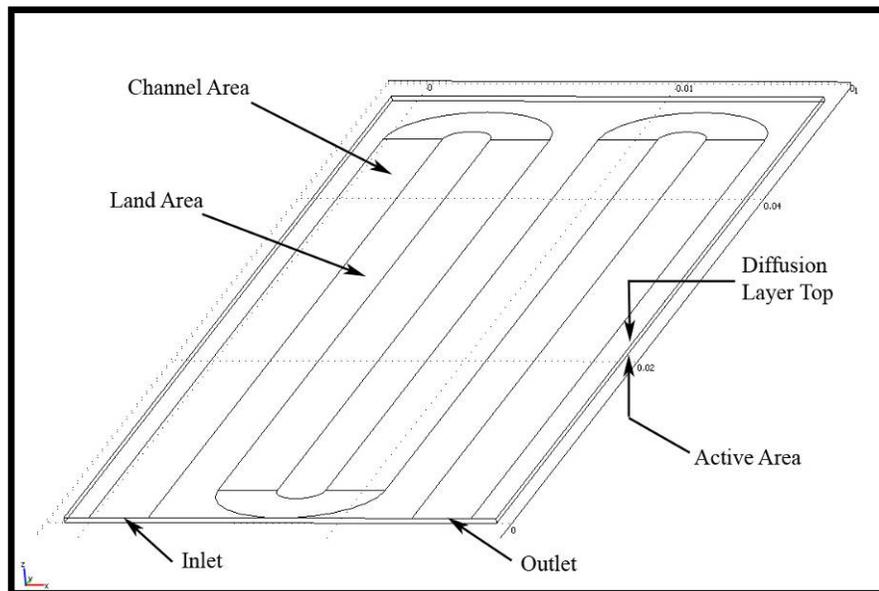
- ◆ Gain insight into behaviors governing flow and current distributions
- ◆ Determine space (diffusion layer parameters, conductivity effects, flow channel and land dimensions) for detailed optimization physics
- ◆ Guide future design improvements

Model developed from COMSOL built-in library

- ◆ Adapted 3D Serpentine Flow model for a cathode in the COMSOL Model Library.
 - The model inputs, such as exchange current density, concentration, and diffusion coefficients were altered.
 - Flow in the channels was condensed from 3D to 2D.
 - Some physics modes were replaced with alternate modes that best represented the cell.

Model Development and Operation

- ◆ End result has a 2D flow field acting as a boundary on the 3D diffusion layer.



The Model Space

COMSOL functions used in NaBH₄/H₂O₂ fuel cell model

◆ Physics Modes

■ Momentum Conservation

- Navier-Stokes Incompressible Flow within the flow field
- Darcy's Law for flow in a porous media within the diffusion layer.

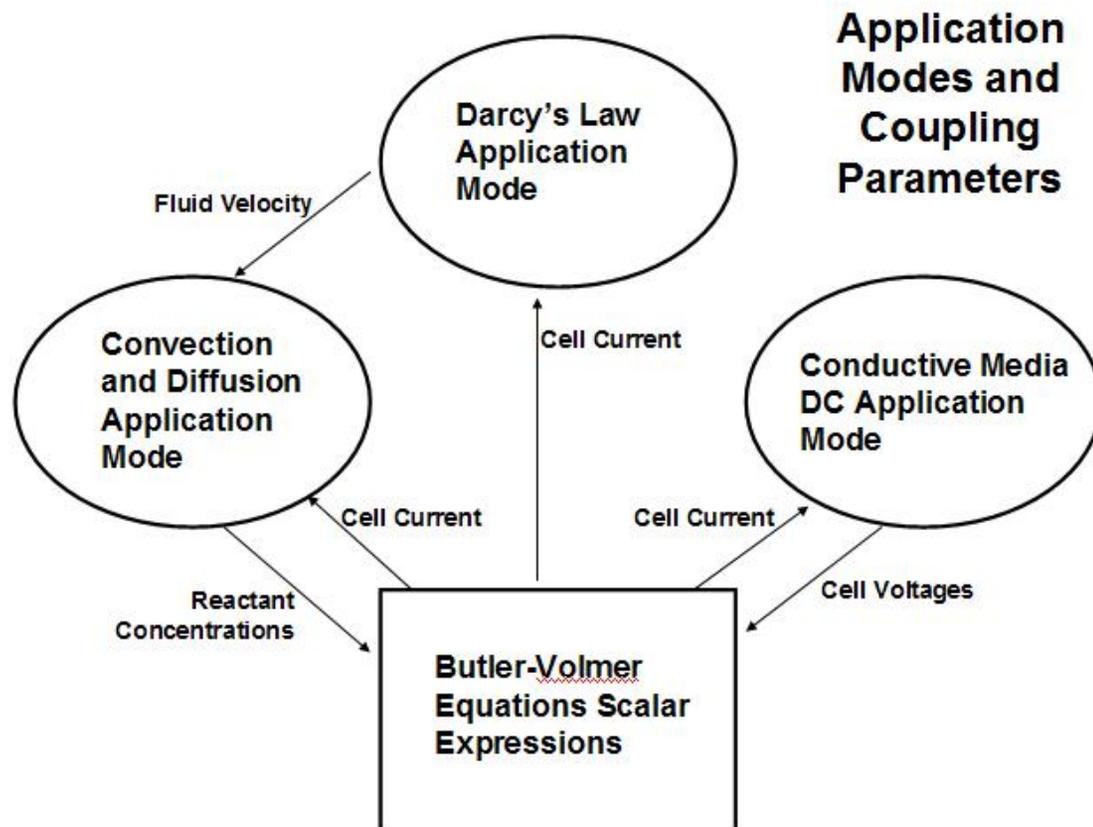
■ Conservation of Mass

- Chemical Engineering Module Convection and Diffusion within the diffusion layer.

COMSOL functions used in NaBH₄/H₂O₂ fuel cell model

- ◆ Current Density and Mass Flux Simulation
 - Tafel type Butler-Volmer Equation used to find current density.
 - Depends on concentration distribution in the diffusion layer and over-potential (directly related to cell voltage).
 - Mass flux at active area proportional to the current generated, with H₂O₂ consumed and H₂O generated

COMSOL Application Mode Coupling



Relationship between COMSOL functions in the model

◆ Couplings

- Pressure coupling between Navier-Stokes in flow field and Darcy's Law in diffusion layer
- Mass flux at the active layer causes small velocity boundary condition for Darcy's Law

Methodology – convergence requires special method to obtain reasonable initial values

◆ Solving

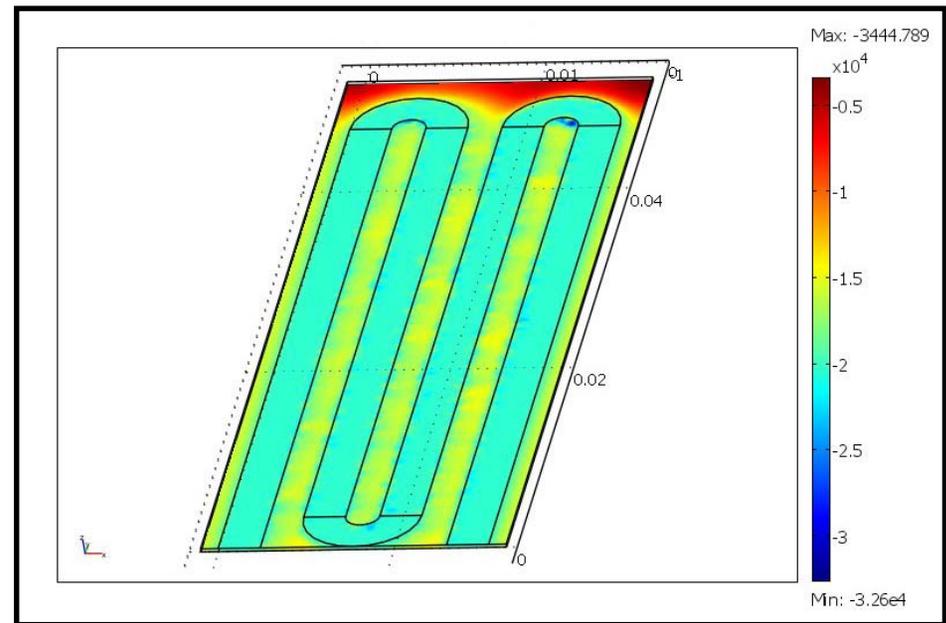
- Initial values found by first decoupling the momentum and mass conservation and solving separately.
- The initial value was used to find the solution with couplings reintroduced
- Over-potential values were varied to generate V-I curves.

Methodology

- ◆ Solutions were found for several different flow fields.
 - Flow field designs varied in channel and land width.
 - All couplings were verified and all boundary conditions found to be satisfied.

Initial Results

- ◆ Current generation greatest under channels where concentration greatest.
- ◆ Current generation underneath the land follows the flow of reactant created by pressure differences between channels

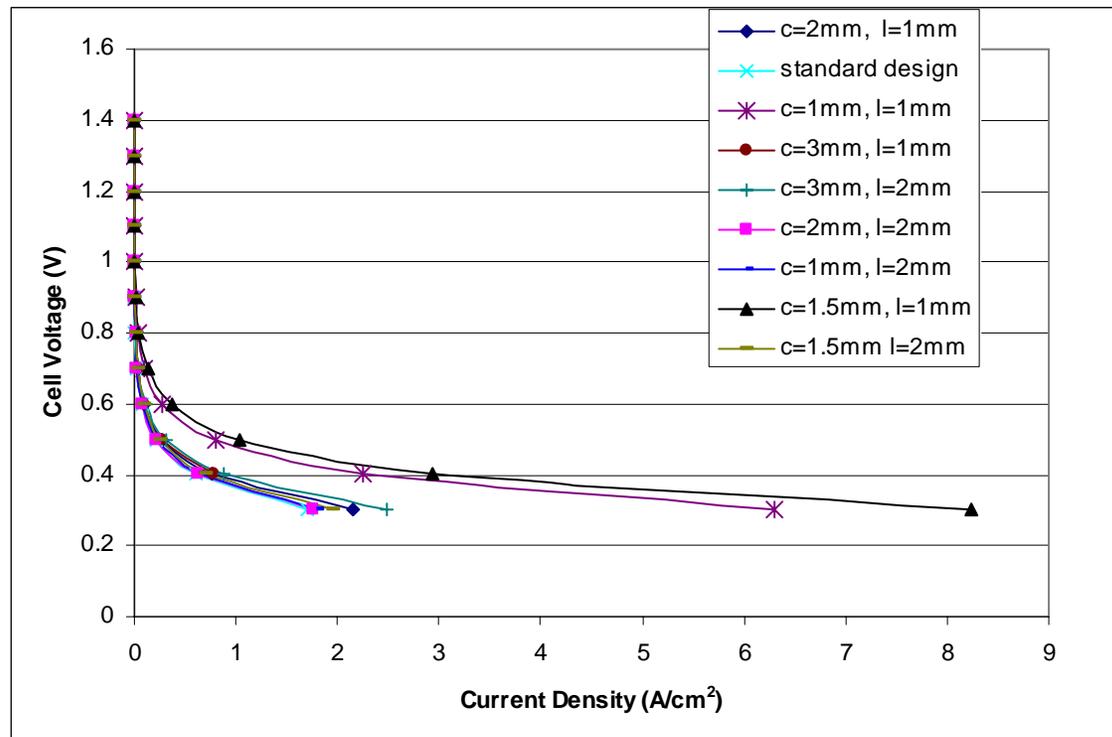


Current Density Distribution

Apparent optimal flow field dimensions found by varying channel and land widths

- ◆ For most channel widths, small land width is desired.
- ◆ V-I curves also indicate that performance greatly depends on finding the optimal land width. Without a properly sized land width, the fuel cell will perform well below potential.

Smaller land width results in greater current density

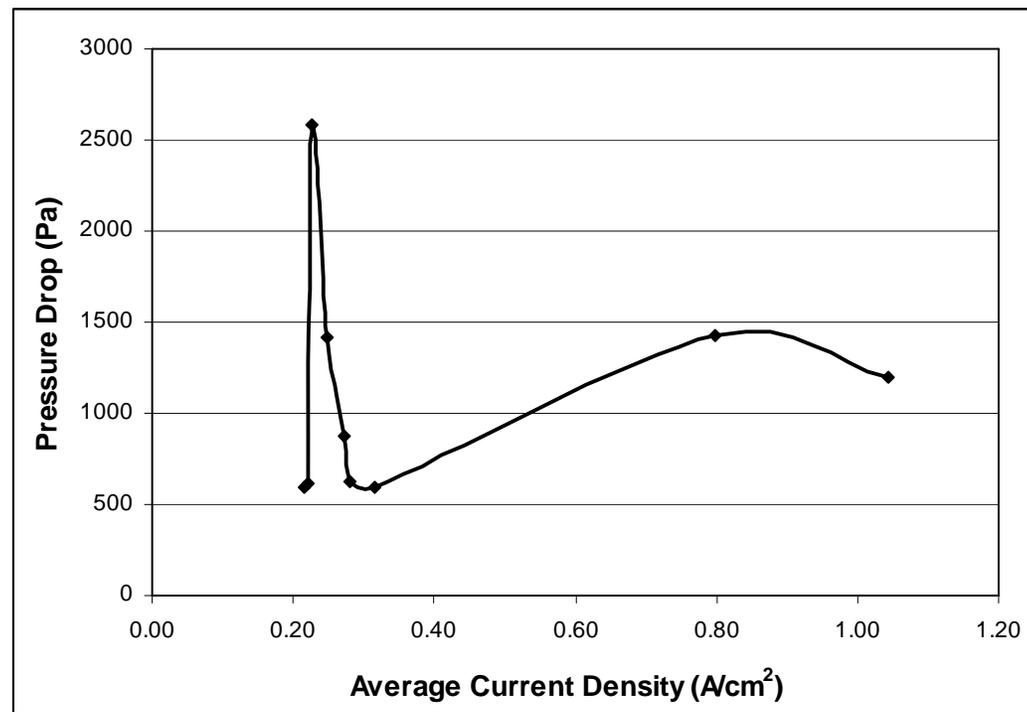


V-I Curve for various flow field dimensions (c=channel width, l=land width)

Model shows variations in pressure not as significant as land width

- ◆ By plotting the current generated at 0.5V as a function of the overall pressure drop, it was determined that there is no strong dependence of current density on pressure drop.
- ◆ Even when the curve is modified to take into account variation in land width, only a weak dependence on pressure is shown.

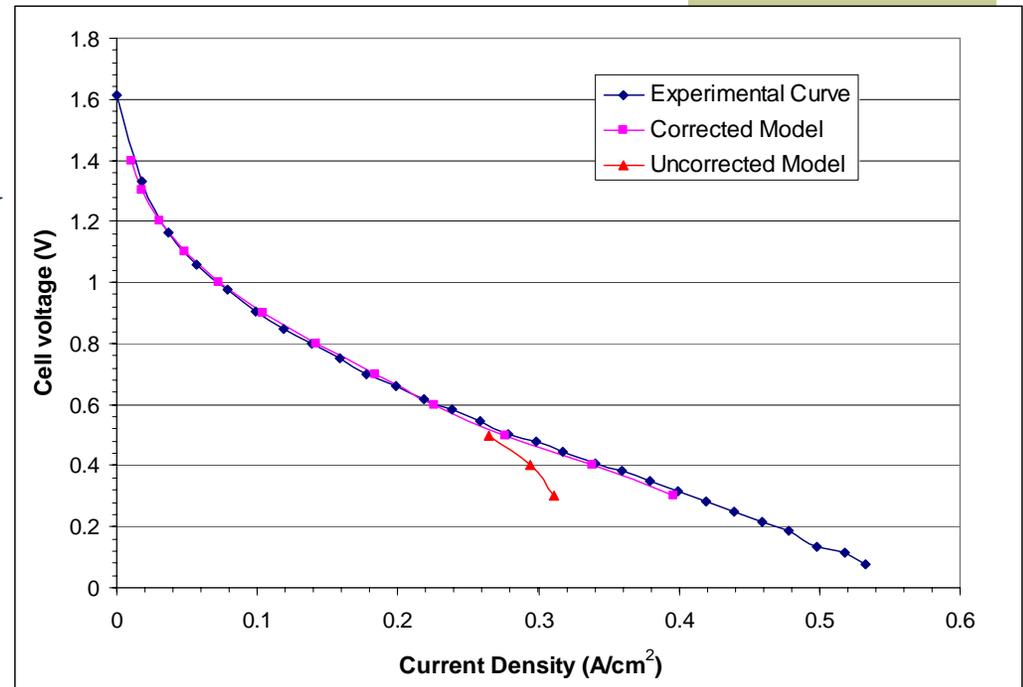
Current density found to be not directly related to pressure



Average Current Density vs. Model Pressure Drop

Normalized model V-I curve closely resembles real data for standard cell

- ◆ The model was normalized to experimentally found over-potentials from the standard cell configuration
- ◆ The resulting normalized curve fit well against the experimental data

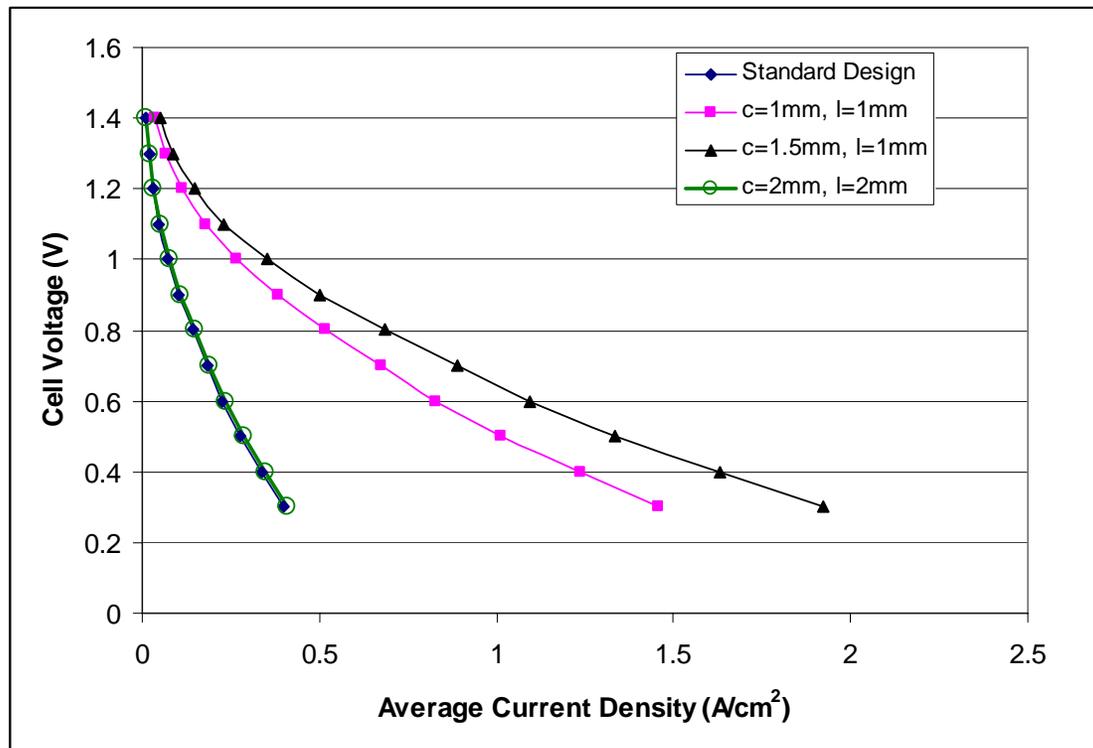


Experimental and normalized model V-I curve

Normalized model used to make new V-I curves for alternate flow fields

- ◆ Normalized model used to generate V-I curves for a variety of alternate flow field designs
- ◆ Use of the normalized model should result in better prediction of experimental results

Model indicates 1.5mm channels, 1mm lands should perform best

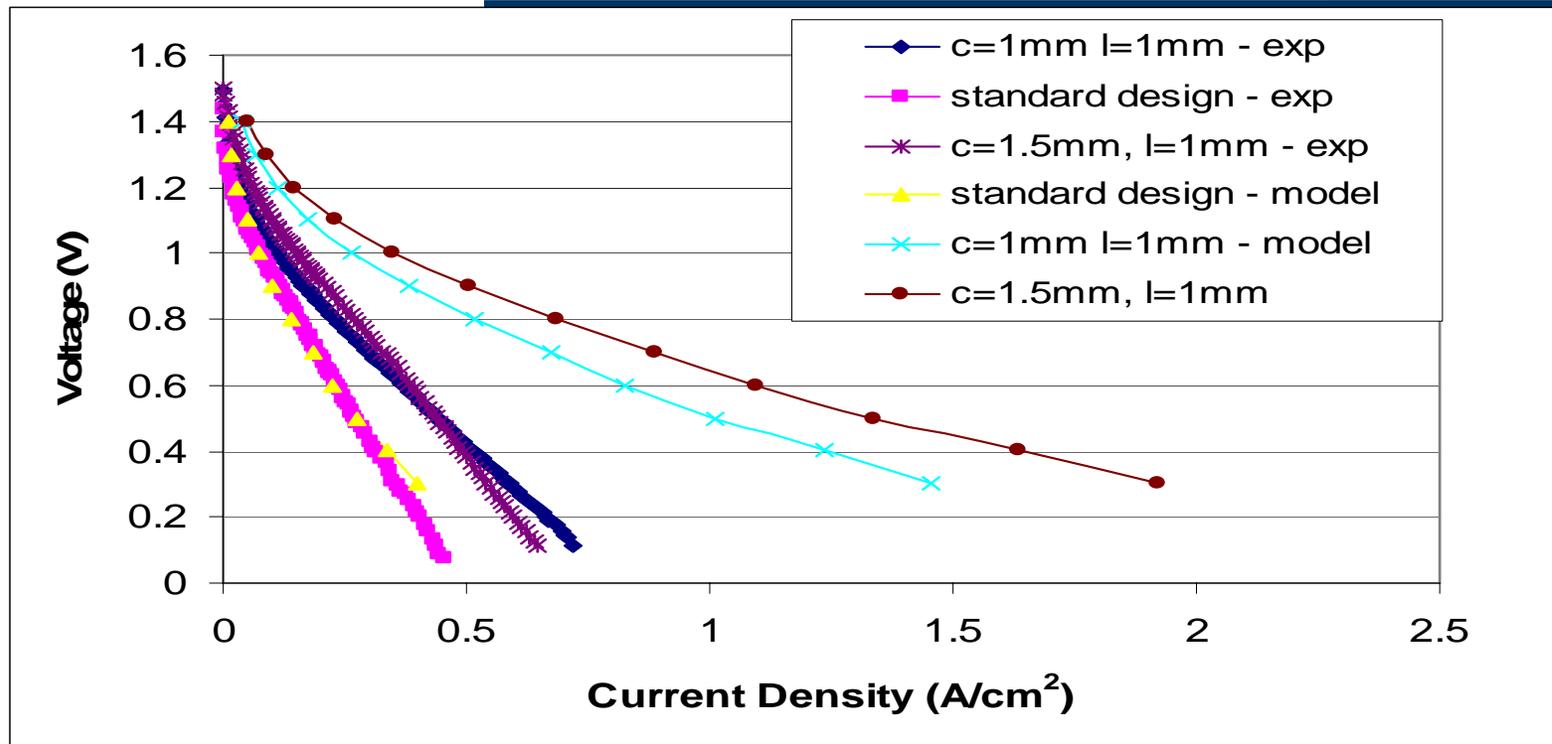


Voltage versus Current using Updated Overpotentials

Verification of model with experiment

- ◆ 6 new flow field plates were constructed matching the configurations explored in the model
- ◆ Cell performance was measured with the new flow plates and compared to the model's predictions

Normalized model does not match experiment well, but predicts optimal performance region in agreement with experiments.



Comparison of normalized model to actual results for alternate flow fields – note that $c=1.5\text{mm}$, $l=1\text{mm}$ performs best, as predicted by the model

COMSOL Users Conf. 2006 Boston,

Normalized model predicts best design region- i.e.
key trends predicted are valid

- ◆ The model results did not accurately match cell performance away from the standard design, but it successfully predicted the design region for best configuration shown in experiments.

Further lessons learned from the model

- ◆ Unlike the model, experimental results indicate that current density is dependent on an ideal combination of both channel and land width, not just land width
- ◆ Normalization from the standard cell did not apply to V-I curves from alternate flow field designs, indicating that the overpotentials change with the flow field

Conclusions

- ◆ COMSOL can be used to model the behavior of an NaBH₄/H₂O₂ fuel cell
- ◆ Model over-potentials must be normalized to experimental values to predict cell performance
- ◆ Normalized model correctly predicted which flow field designs would work best, but did not accurately model their performance
- ◆ Normalization is dependent on flow field parameters, so a more accurate normalization method is desired in the future.

Conclusions - continued

- ◆ The model is requires further work to accuracy predict absolute performance values for case away from the “standard” case used in normalization.
- ◆ Still it provides a very valuable tool for identifying the design space where optimal performance is expected. Thus experiments can be rapidly focused on this space to refine the optimal design. Significant improvements have been achieved in this fashion.

Acknowledgments

We would like to thank:

- **NPL Associates, Inc.** for their support with starting the project.
- Fellow researchers N. Luo, G. Kopec, and E. Byrd
- This research was supported by **DARPA SB04-032**.
- Continuing studies are supported by **DARPA/AFRL**.



Thank You



For more information please contact:

Dr. George. H. Miley

UIUC

Phone: (217) 333-3772

email: ghmiley@uiuc.edu