

Transport in PEMFCs

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Project ID # FC054



Transport in PEMFCs

Timeline

- Project Start Date: 10/5/2009
- Project End Date: 10/31/2013
- Percent Complete: 80%

Budget

- Total Project Funding
 - DOE Share \$2.66M
 - Cost Share \$678K (20%)
- Funding Received in FY10: \$915K
- Funding Received in FY11: \$786K
- Funding Received in FY12: \$400K
- Planned Funding in FY13: \$560K

Barriers Addressed

- Performance
- Water Transport within Stack
- System Thermal and Water Management
- Start-Up and Shut Down

Technical Targets

- Cold Start-up Times
- Specific Power Density
- Stack Power Density
- Stack Efficiency

Partners

- University of South Carolina
- Virginia Tech
- Tech Etch
- AvCarb



Approach: Team and Tasks

Objective: Improve Understanding/Correlation Between Material Properties and Model Equations



- Generate model
- Supply model relevant transport numbers
- Stress the model by developing different materials with different transport properties
- Determine sensitivity of fuel cell performance to different factors
- Guide research

Milestone	Plan Complete	Actual Complete
Baseline PFSA model, with overall results correlating within +-20% of each other. Design the new apparatus for extending the range of electroosmotic drag and diffusivity.	4/15/2011	4/1/2011
Extend Model to a variety of membranes, catalyst content, GDM's, and flow fields. The model should be able demonstrate prediction of the actual data within +-20% of the experimental results.	8/15/2012	90%







Approach & Milestones

	Techniques	Materials	Modeling
Year 1	New technique generation for static and dynamic diffusion, EODC, through plane conductivity confirmation with Baseline materials. Current Distribution Board Demonstration	Baseline hydrocarbon PEM generated and down selected Baseline Gas diffusion Media Delivered First Etched Plates	Set-Up of Model Use of Baseline materials for Testing Model Sensitivity Testing
Year 2	Techniques applied to alternative materials. Diffusivity apparatus used to characterize alternative diffusion media (33%).	Scale-up of Baseline PEM Integration of catalysts Modification of diffusion media Alternative Plates & Design of larger plates.	Performance and water balance modeled and confirmed with baseline materials and hydrocarbon PEM. (50%) Alternative diffusion media tested.
Year 3 (Period 2)	Low Temperature Studies	Delivery of Large PEMs Current Distribution board for larger plate Fabrication of larger plate and current distribution board	Modeling extended to larger cells. Effect of coolant/heat transfer. Model confirmation with current distribution and water balance.

Work on larger cells abandoned in favor of using GM "open source" hardware



Relevance: Use of Modeling in Fuel Cell Development is Widespread. Agreement on Fundamentals *is not*





Average Water Content (λ)

T.A. Zawodzinski, M. Neeman, L.O. Sillerud and S. Gottesfeld, J. Phys. Chem., 95, 6040 (1990)

T.F. Fuller, Ph.D. Thesis, University of California, Berkeley, CA (1992) T.V. Nguyen and R.E. White, *J. Electrochem. Soc.*, **140**, 2178 (1993) Equations of the form of: S. Motupally, A.J. Becker and J.W. Weidner, *J. Electrochem. Soc.*, **147**, 3171 (2000)

- NOTHING EVEN RESEMBLING CONSENSUS ON THESE FUNDAMENTALS
- Systematic approach of generating and developing various materials with better characterization methods is needed

Nafion is a registered trademark of E. I. DuPont de Nemours and Company.

Relevance:

PEM Development

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- Hundreds of PEMs developed for fuel cells
 - Would like to come up with design rules for PEMs
 - How does size/degree of Phase separation affect
 - Conductivity
 - EODC
 - Water Diffusivity
 - Gas Permeability
 - Similar Study done by *Gross et al* for side-chain polymers

Modeling

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- Need to make sure we know how changes in transport numbers effect fuel cell performance
- Transport numbers and model are used to confirm each other
- How sensitive is fuel cell performance to these different parameters?
- What should we be working on?

DOE Hydrogen Program

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Achievements: Copolymer Membrane Design

• Matrix 1: Varied Block Lengths, Annealing Temperature and IEC



	Polymer	Thermal Treatment Temperature (°C)	IEC (meq/g)
1	6FPAEB-BPSH100 7k-7k	110	1.55
2	6FPAEB-BPSH100 15k-15k	110	1.55
3	6FPAEB-BPSH100 10k-18k	110	2.01
4	6FPAEB-BPSH100 7k-7k	220	1.55
5	6FPAEB-BPSH100 15k-15k	220	1.55
6	6FPAEB-BPSH100 10k-18k	220	2.01

• Matrix 2: Varied Oligomer Categories/Properties

Sample	Block Copolymer	Block Length	IEC (meq/g) ^a	Water Uptake (%) ^b	Conductivity (S/cm) ^c
JR-143-2	6FK-BPSH	8K – 8K	1.45	21	0.10
JR-143-3	6FPAEB-BPSH	13K – 13K	1.63	37	0.14
JR-143-4	6FBPS0-BPSH	10K – 10K	1.47	35	0.10







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New Technique: Simultaneous Water Uptake and Diffusivity Extended to Sub-Ambient Conditions





Achievements:

New Technique: Simultaneous Water Uptake and Diffusivity



BPSH-6FPAEB Membranes show nearly identical water uptake with little temperature dependence Water Diffusivity is $\sim \frac{1}{2}$ that of Nafion[®] regardless of temperature.

Achievements: Water Diffusivity of Various Membranes

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- Morphology does not affect water uptake at low RH
- Phase separation on a smaller scale results in lower diffusivity. Annealing increases phase separation and diffusivity



Achievements: New Technique: EODC



- Water/H₂ inlet ratio controlled by controlling saturator temperature and H₂ pressure
- If ratio is too high, not enough water is dragged across and cell floods and fails
- If ratio is too low, membrane dries out and cell fails
- At Water/H₂ = 2*EODC Cell operates in quasistable state

All gas/gas diffusion is eliminated



- All hydrocarbon membranes exhibit lower EODC than Nafion[®]
- Higher thermal annealing, block lengths and IEC seem to increase EODC







Water Balance Measurement Under Different Conditions Experimental vs. CFD (VT membrane)



Water Balance experiment and numerical result of new MEA $(80^{\circ}C, 1.5/2.0 \text{ stoich } H_2/O_2)$

Water balance shows the transport of water inside fuel cell. For different operating conditions and membranes, the water transport inside fuel cell will also be different.

	i	Anode Water Balance (mg/sec) Cathode Water Balance (mg/sec)					e (mg/sec)	Cross		
		RH	Water	Watan aut	Cross to	Water	Can	Water	Cross from	error
	A/cm ²		in	water out	Cathode	in	Gen.	out	Anode	(%)
EXP	1.2	80/80	1.40	0.071	1.32	0.95	2.80	4.82	1.25	5
CFD	1.2	80/80	1.40	0.055	1.34	0.95	2.80	5.10	1.36	1.5
EXP	0.8	100/100	1.27	1.42	-0.15	0.85	1.86	2.44	-0.17	11
CFD	0.8	100/100	1.28	1.44	-0.16	0.85	1.86	2.54	-0.17	6



Water Balance Measurement Under Different Conditions

Water Balance experiment of VT MEA 6FPAEB-BPSH

		i	Anode Water Balance (mg/sec)Cathode Water Balance (mg/sec)					orror			
		1	RH	Water	Watar out	Cross to	Water	Con	Water	Cross from	
		A/Cm ²		in	water out	Cathode	in	Gen.	out	Anode	(%)
ſ	EXP	0.4	75/25	0.75	0.22	0.53	0.59	0.93	2.01	0.49	7.5
	CFD	0.4	75/25	0.75	0.23	0.52	0.59	0.93	2.03	0.51	2.0
4	EXP	0.4	50/50	0.42	0.29	0.13	1.35	0.93	2.42	0.14	7.1
nt	CFD	0.4	50/50	0.42	0.29	0.13	1.35	0.93	2.40	0.12	8.3
	EXP	0.4	95/95	1.10	0.86	0.25	3.51	0.93	4.73	0.29	13.7
	CFD	0.4	95/95	1.10	0.90	0.20	3.51	0.93	4.66	0.22	9.0
	EXP	0.6	75/25	1.12	0.33	0.79	0.88	1.40	3.09	0.81	2.5
	CFD	0.6	75/25	1.12	0.31	0.81	0.88	1.40	3.13	0.85	4.7
ſ	EXP	0.8	50/50	0.85	0.55	0.30	2.70	1.87	4.88	0.31	3.2
	CFD	0.8	50/50	0.85	0.58	0.27	2.70	1.87	4.85	0.28	3.5
nt	EXP	0.8	95/95	2.21	1.68	0.53	7.03	1.87	9.44	0.54	1.8
٦	CFD	0.8	95/95	2.21	1.66	0.55	7.03	1.87	9.45	0.55	0.0
	EXP	1.2	95/95	3.32	1.99	1.34	10.55	2.80	14.60	1.25	7.5
	CFD	1.2	95/95	3.32	2.01	1.31	10.55	2.80	14.72	1.37	4.4

 $(80^{\circ}C, 1.5/2.0 \text{ stoich } H_2/\underline{Air})$

Low current density

High curren density

WIDE AGREEMENT BETWEEN CFD MODEL AND EXP RESULTS

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Local Distributions of Current Density & Water Transport on the Membrane Surface at low inlet RH at 0.4 A/cm²

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Local Distributions of Water Content in Membrane & Liquid Water on Cathode MEA/GDL Interface at <u>high inlet RH</u> at 0.6 A/cm²





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Effect of Inlet Humidity on Current Density at Different Potentials (80 °C, 1.5/2.0 stoich H₂/<u>Air : Memrbrane – NRE212: GDL – EP40T</u>)



Using to probe/characterize efficiency of diffusion media to remove liquid water

Will try to relate to porosity, hydrophobicity McMullin Number



Effect of Inlet RH on CDD (A/cm²) on MEA Surface

at Vcell = 0.6V (80°C, 1.5/2.0 stoich $H_2/Air : Membrane - NRE212: GDL - EP40T$)



Model does a good job of handling flooding by assuming accumulation of water film.

Summary

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□ Membrane design and development (VA Tech) & Characterization (Giner):

- 0 Membranes with similar charge densities but different
 - Chemistries (different hydrophobicity of the non-functional block)
 - Morphologies

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- Block length
- ➤ Annealing
- Increased hydrophobicity of the non-functional group, longer block lengths and annealing all lead to a more distinct separation of phases, on a larger length scale
 - Increases conductivity at low RH
 - Increases diffusivity
 - Increases EODC

Transport Modeling, GDL & Current Distribution Board Characterization (USC & Giner):

- Model successfully predicts:
 - Dry, Wet Conditions. Hydrocarbon and PFSA membranes
 - Performance and Water Balance
 - Increased flooding for PFSA membranes compared to hydrocarbon membranes

DOE Hydrogen



Future Work

- Emphasis on Saturated Conditions
 - Diffusion Media
 - Membrane Transport Properties
- Electrode Effects
 - High Current Density at Low Catalyst Loading
 - Flooding of NSTF layers



Technical Back Up Slides

- Not Discussed:
 - Current Distribution Boards
 - Verification of Model with GM Open Data
 - Diffusion Media



Achievement: Model Verification:

Distributions of current density and temperature of 50-cm² GM-Down-the-Channel flow-field compared with

www.pemfcdata.prg

(*I_{avg}* = 1.5 A/cm², <u>counter-current flow</u>: 50/50%RH, 150/150kPa, 80°C, 1.5/2.0 stoich)



www.pemfcdata.org

USC data matches published data very well, both with performance and model results

CFD comparison with pemfcdata.org at different operating conditions

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Local distributions of current density and temperature on membrane surface at $I_{avg} = 0.4 \text{ A/cm}^2$ $T_{cell} = 60C, \text{ RH} = 95\%/95\%, \text{ P} = 150\text{kPa}/150\text{kPa}.$ Stoich = 1.5/2.0



Temperature Distribution







USC CFD prediction

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Achievements: New Materials: Diffusion Media

- AvCarb added to the program recently
- Started with Toray Materials
 - Variable Wet-Proofing
 - Microporous Layer
- AvCarb will provide more custom materials
- Want to generate differences in:
 - MacMullin Number
 - Porosity
 - Tortuosity
 - Hydrophobicity

- Tortuosity
 - Ratio of the actual path length through the pores to the shortest linear distance between two points.

•Porosity

Ratio of void volume (volume of pores) to the total volume.

MacMullin Number

Function of tortuosity and pososity.









Achievements: Design of Gas Diffusion Media



MPL 1 MPL 2

Carbon Substrate

Baseline Material at start of program was Toray H060

The new design of GDLs have been modified from standard AvCarb GDLs by adding two micro porous layers. Each set has been treated with two different methods in order to provide two different values of diffusivity.

Substrate	Diffusivity Modification	MPL 1/MPL2 (carbon particle size)
P50	Low	Small/Large
EP40		
P75	High	Large/Small

Total of 12 new papers generated 5 characterized ex-situ to date



AvCarb GDLs Status

5 sets of 12 assigned new design of GDLs with two different micro porous layers

Substrate	Diffusivity	MPL 1	MPL 2	# of Samples	MacMullin No.
P50	low (<0.15)	Small	Large	5	
P50	low (<0.15)	Large	Small	2	2.63
P50	high (>0.25)	Small	Large	5	
P50	high (>0.25)	Large	Small	2	2.16
EP40	low (<0.25)	Small	Large	5	
EP40	low (<0.25)	Large	Small	5	
EP40	high (>0.35)	Small	Large	5	
EP40	high (>0.35)	Large	Small	2	2.34
P75	low (<0.20)	Small	Large	5	
P75	low (<0.20)	Large	Small	2	2.26
P75	high (<0.3)	Small	Large	5	
P75	high (<0.3)	Large	Small	2	1.88

The new design of GDLs have been modified from standard Ballard GDLs by adding two micro porous layers. Each set has been treated with two different methods in order to provide two different values of diffusivity.



Achievements: Design of Gas Diffusion Media Comparison of Mercury pore size distributions of new design GDLs

Baseline Substrates



Modified Substrates

EP40T has largest pore volume, concentrated at 50 μm

Modification greatly reduces volume of large pores

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AvCarb GDLs Status

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Baseline perf	ormance at Icell	$\ell = 80^{\circ}C$	MEA =	= Gore ^{1M} 5/; S	erpentine
Substrate	Diffusivity	MPL1	MPL2	Macmullin#	Status
P50	Low (<0.15)	Large	Small	2.63	Done
P50	High (>0.25)	Large	Small	2.18	Done
EP40	High (>0.25)	Large	Small	2.34	Done
P75	Low (<0.20)	Large	Small	2.14	Done
P75	High (<0.30)	Large	Small	1.92	Done
$ \begin{array}{cccccccccccccccccccccccccccccccccccc$	Condition: 100 with 5 PSI	D/50%RH	← P75,I ← P75,I ← P50,I ← P50,I ← EP40	Diff: High Diff: Low Diff: Low Diff: Low Diff: High Diff: Low Diff: High Diff: High Diff: High Diff: High Diff: High Diff: High Diff: A bar Diff: High Diff: Low Diff: High Diff: High Diff: Low Diff: High Diff: High Diff: High Diff: High Diff: Low Diff: High Diff: High Diff: High Diff: High Diff: High Diff: High Diff: Diff: High Diff: Diff: High Diff: High Diff: Diff: High Diff: Diff: High Diff: Diff: High Diff: Diff: Diff: High Diff: Diff: Dif	

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- EP40,Diff: high shows the highest • performance for all three cases
- P75, Diff: low shows the lowest performance for all three cases
- Both P50, Diff: high and Diff: low • show similar performance





Using data of pore size distribution information and McMullin Number from GDL characterization, CFD predictions are able to compare well with experimental data.

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0.20

0.10

0.00

0

200

Condition: 25/25 %RH

600

Current density (mA/cm2)

800

1000

1200

400

2

0

1400





Water balance measurement under different conditions Experimental vs. CFD (<u>Nafion[®] membrane</u>)



Water Balance experiment and numerical result of the parallel Channel flow-field (80°C, 1.5/2.0 stoich H₂/Air)

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Water balance shows the transport of water inside fuel cell. For different operating condition the water transport inside fuel cell will be different.

	i		Anode V	Vater Balanc	e (mg/sec)	Catho	ode Wat	er Balance	e (mg/sec)	orror
		RH	Water	Water out	Cross to	Water	Gan	Water	Cross from	
	A/cm ²		in	water out	Cathode	in	Gen.	out	Anode	(70)
EXP	0.4	25/25	0.37	0.42	-0.20	1.15	1.87	2.81	-0.21	4.6
CFD	0.4	25/25	0.37	0.52	-0.15	1.15	1.87	2.85	-0.17	10
EXP	0.4	75/25	1.50	0.55	0.94	1.15	1.87	3.79	0.91	3.8
CFD	0.4	75/25	1.50	0.53	0.97	1.15	1.87	4.00	0.98	1
EXP	0.6	75/25	2.25	0.87	1.38	1.70	2.80	5.80	1.30	5.9
CFD	0.6	75/25	2.25	0.81	1.44	1.70	2.80	6.00	1.50	4





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Polarization curves of <u>VT MEA</u> (update 01/14/2013) 6FPAEB-BPSH

(<u>co-current flow</u>: 80°C, 1.5/2.0 stoich, H_2 /<u>Air</u>)



95/95 %RH 150 kPa (Air)

100/50 %RH 5 PSI (Air)



Polarization curves of <u>VT MEA</u> (update 01/14/2013) 6FPAEB-BPSH

(<u>co-current flow</u>: 80°C, 1.5/2.0 stoich, H_2/\underline{Air})



50/50 %RH 150 kPa (Air)

i-V polarization curve



Backup Slides for Model's Input Parameters and Output Variables



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Material Properties							
Material 1: Anode Side Fluid							
Density (rho)	ldeal gas	28.96	g/gmole				
Viscosity (mu)	multicomp	1.81e-05	kg/m-s				
Conductivity (k)	multicomp	0.02637	W/m-K				
Spec Heat (Cp)	constant	1006	J/m-K				
Material 2: Cath	node Side F	luid					
Density (rho)	ldeal gas	28.96	g/gmole				
Viscosity (mu)	multicomp	1.81e-05	kg/m-s				
Conductivity (k)	multicomp	0.02637	W/m-K				
Spec Heat (Cp)	constant	1006	J/m-K				
Material 3: ME	A Solid						
Density (rho)	constant	200	kg/m^3				
Conductivity (k)	constant	0.16	W/m-K				
Spec Heat (Cp)	constant	500	J/kg-K				
Material 4: Bipo	plar Solid Pla	ates					
Density (rho)	constant	200	kg/m^3				
Conductivity (k)	constant	15.7	W/m-K				
Spec Heat (Cp)	constant	500	J/kg-K				
Apply		Reset to Det	faults				

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Porous Resistance Coefficients

Porous Media 1: Anode Side GDM					
	alpha	beta			
X- Direction	0	2.0e+07			
V- Direction	0	2.0e+07			
Z- Direction	0	2.0e+07			
Porosity Fac	tor (-)	0.7			
Effective Co	onductivity (W/m-K) 0.25			
🔟 User C	oding for Poros	ity			
Porous Media	2: Cathode Sid	e GDM			
	alpha	beta			
X- Direction	0	2.0e+07			
V- Direction	0	2.0e+07			
Z- Direction	0	2.0e+07			
Porosity Fac	:tor (-)	0.7			
Effective Conductivity (W/m-K) 0.25					
User Coding for Porosity					
Apply Reset to Defaults					

Write Operating Conditions Input File

- Operating Parameters				
Initial Cell Voltage for all Cells (V)	0.72			
Membrane Thickness (mm)	0.018			
Anode GDM Thickness (mm)	0.260			
Cathode GDM Thickness (mm)	0.260			
Cell Temperature (C)	80			
Dry Membrane Density (g/cm^3)	2			
Equiv Wt of Dry Membrane (g/gmol)	1100			
Equil Int. of Dify monitorialis (giginoly	1100			
Electrochemical and Kinetic Para	meters —			
Open Circuit Voltage (V)	0.92			
Oxygen Exch Current Density (A/m^2)	500			
Hydrogen Exch Current Density (A/m^2)	5000			
Anode Transfer Coefficient (-)	2			
Cathode Transfer Coefficient (-)	0.717			
Hydrogen Inlet Mole Fraction (-)	0.763			
Oxygen Inlet Mole Fraction (-)	0.160			
Other Parameters				
Evaporation / Condensation Rate (/s)	1.0			
Starting Iteration for Reacting Flow	50			
No. of Cells in Z-Dir in the Anode GDM	5			
No. of Cells in Z-Dir in the Cathode GDM	5			
No. of Fuel Cells in the Model	1			
Average Current Density (A/m^2)	0.4			
Auto-adjust Cell Voltage (VCEL)				
Write Operating Conditions Input File	Reset			

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DOE Hydrogen Program

Modeling Input Parameters (Cont.)

Constitutive Equation Panel (Advanced)						
Input of Coefficients of Parametric Constitutive Equations						
Main Equation:	Wate Equ	r Diff. ation	Overpot. Electrical Equation Properties			
Membrane Properties are a function of: Anode water activity Cathode water activity Average between anode and cathode water activities						
Water Content in the Membrane (lambda)						
B1 =	0.043	(-)	B5 =	0.0	(-)	
B2 =	17.81	(-)	B6 =	14.0	(-)	
B3 =	-39.85	(-)	B7 =	1.4	(-)	
B4 =	36.0	(-)				
Electro-osmotic Drag Coefficient (n)						
E1 =	0.05	(-)	E3 =	0.0	(-)	
E2 = 🔲	0.0029	(-)	E4 =	-3.4e-19	(-)	
Local Membrane Conductivity (sigma)						
H0 =	0.0	(S/m)	H3 =	1268.0	(K)	
H1 =	0.514	(S/m)	H4 =	303.0	(K)	
H2 =	0.326	(S/m)			()	
Store C	Coefficient	ts	Re	set to Defa	ults	

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Constitutive Equation Panel (Advanced)				
Input of Coefficients of Parametric Constitutive Equations				
Main Equations Water Diff. Overpot. Electrical Equation Properties				
Water Diffusion Coefficient (D)				
$G0 = 2416.0 (K) G1 = 1e-10 (m^2/s)$ $G2 = 1e-10 (m^2/s)$ $G3 = 1.25e-10 (m^2/s)$				
G4 = 303.0 (K)				
Membrane Water Content Range (lambda) L1 = 1.67 L2 = 2.0 L3 = 3.0 L4 = 4.5				
Store Coefficients Reset to Defaults				

Constitutive Equation Panel (Advanced)					
Input of Coefficients of Parametric Constitutive Equations					
Main Water Diff. Overpot. Electrical Equations Equation					
Overpotential Equation Term Selection					
Term 1					
Term 2					
Term 3					
Term 4					
🗖 Term 5					
🔟 Term 6					
Term 1 and Term 5 cannot be selected at					
Term 2 and Term 6 cannot be selected at the same time.					
Please refer to the Methodology section of the Tutorial Manual for a description of the six terms involved.					
Store Coefficients Reset to Defaults					

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DOE Hydrogen Program



Modeling Outputs

Scalars	Scalars	Scalars
Active Passive Generic	Active Passive Generic	Active Passive Gene
Active Scalars Description No. Name Nitrogen Gas 1 N2 Hydrogen Gas 2 H2 Oxygen Gas 3 O2 Water Vapor - Anode 4 WVA Water Vapor - Cathode 5 WVC Liquid Water - Anode 6 LWA Liquid Water - Cathode 7 LWC	Passive Scalars Description No. Name Current Density 8 CD Net Water Flux per Proton 9 ALPHA Kinetic Overpotential 10 KOP Anode Overpotential 11 AOP Cathode Overpotential 12 COP Membrane Conductivity 13 MC Water Diffusivity 14 WDC Water Content inside MEA 15 LAMBDA Anode Activity 16 AA Cathode Activity 17 CA MEA Liquid Film Thickness 18 LFT Local MEA Voltage* 20 MEA_POTENT. * Only used when Electron Transport is On No	Generic Scalars Description No. Name Potential [#] 19 POTENTI * Only used when Electron Transport is On
Apply	Apply	Apply

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Generic

Name POTENTIAL