

High Performance PEFC Electrode Structures

PI: Robert Darling

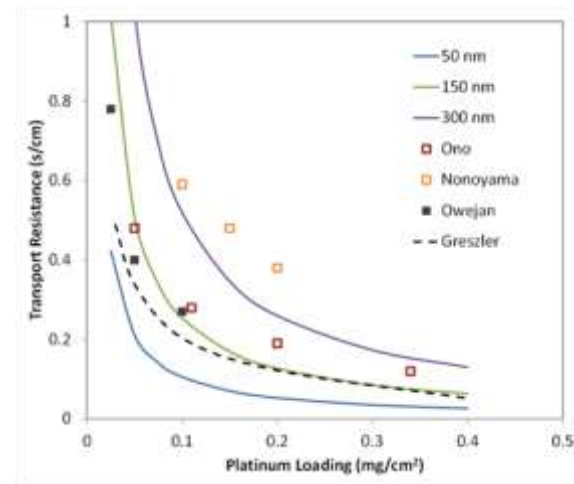
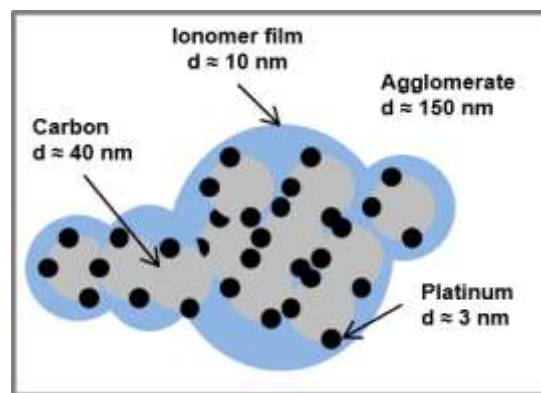


FC PAD Consortia
Project

Project ID: FC157

DE-EE0007652

May 30, 2020



Acknowledgment: This material is based upon work supported by the U.S. Department of Energy's Office of Energy Efficiency and Renewable Energy (EERE) under FCTO's FC PAD Program, Award Number DE-EE0007652.

Overview

Timeline

Project Start: Oct 1, 2016
Actual Start: Jan 1, 2017
Project Duration: 48 months (with
no-cost extension)
Project End Date: Sep 30, 2020

Budget

Total Project Budget: \$3,019K
▪ Federal Share: \$2,415K
▪ Cost Share: \$604K (20%)
Total Funds Spent*: \$2,327K
* as of 4/30/2020

Key Barriers

Achieve DOE's 2020 Targets for MEAs

Characteristic	Units	2015 Status	2020 Targets
Platinum-group metal (PGM) total loading (both electrodes)	mg PGM /cm ²	0.13	≤ 0.125
Performance @ 0.8 V	mA/cm ²	240	300
Power @ rated conditions (150 kPa _{abs})	mW/cm ²	810	1,000

Partners



Relevance

Objective: develop quantitative fundamental understanding of transport limitations in SOA MEAs and use this knowledge to develop and demonstrate high-performing MEAs with ultra-low platinum-group metal loadings

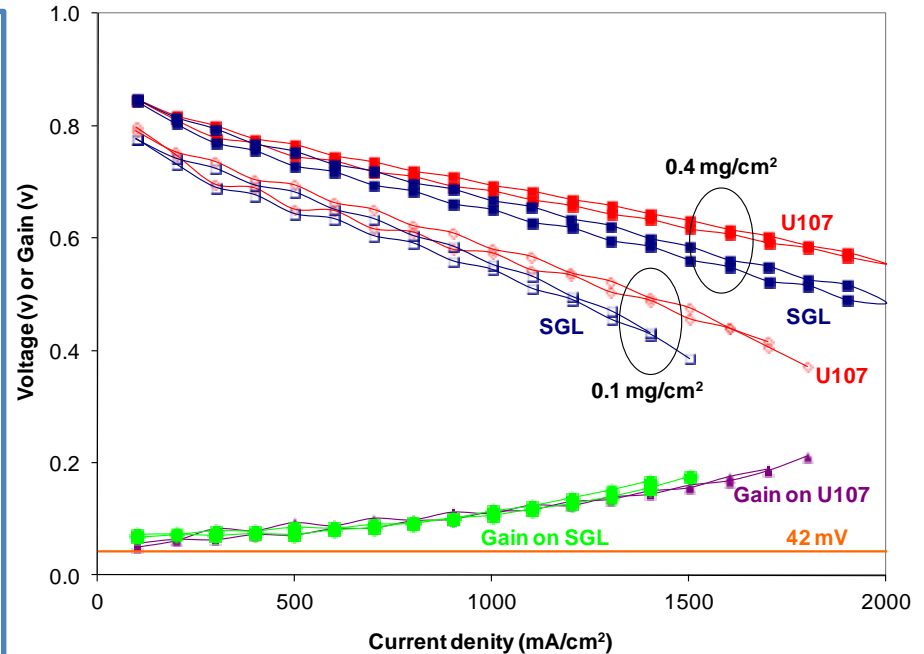
Very active alloy catalysts for electrochemical oxygen reduction have been demonstrated
 MEAs with ultra-low catalyst loadings meet activity targets
 Good performance at high current elusive

Transport losses are a major barrier

Flux per Pt site increases as Pt load drops
 Transport worsens in catalyst layer
 MEAs with low loadings cannot meet power density targets

Traditional agglomerate and thin-film models do not explain observations

Impact: developed models that explain ohmic and oxygen transport losses in catalyst layers with Pt/V and Pt/KB



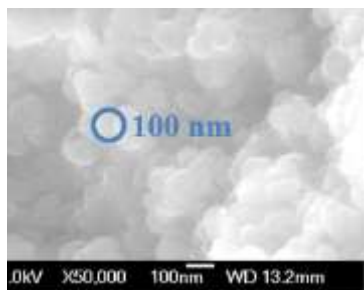
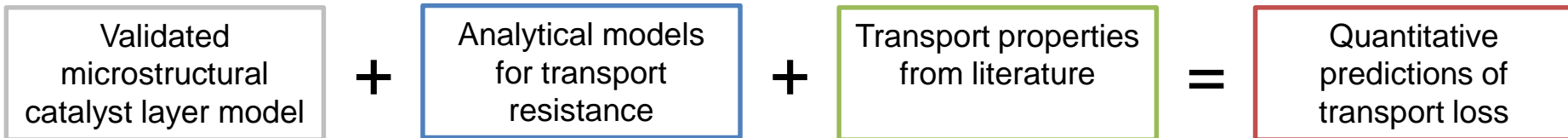
Characteristic	Units	2015 Status	2020 Targets
Total PGM loading	mg/cm ²	0.13	≤ 0.125
Power at 0.8 V	mA/cm ²	240	300
Rated power at 150 kPa	mW/cm ²	810	1,000

Approach: Milestones

Milestone	Task Title	Milestone Description	Status
Q8 Go/No-Go	Model validation and C-supported MEA performance	Validated microstructural model and MEA with significantly improved transport-limited performance	100%
Q9	Carbon-supported model development and validation	Extend Pt/C hierarchal CCL model to incorporate HSA carbons and validate with HSA MEA performance	100%
Q10	Alternative catalyst MEA Fab and MEA performance	Validated microstructural model for MEA with alternative catalysts	75%
Q11	Carbon-supported model and performance degradation	Validate Pt/C hierarchal model to include both BOL and EOL performance after selected AST Protocols	75%
Q12	Alternative catalyst MEA Fab and MEA performance	Complete validated model of at least two different alternate catalyst structures, including recommending changes to catalyst layer morphology to improve the performance	25%
Q12	Model validation and MEA Performance	Recommend optimal catalyst layer structures based on model learnings	0%

- **Major goals for final year of project (beyond BP-2 G/NG):**
- Continue to develop improved understanding of transport losses in CCLs
- Work to resolve results from different CCL characterization methods
 - Focus on porosity and agglomerate size
- Collaborate with groups working on alternative catalyst layer architectures

Approach: Transport Resistance Calculated from Feature Dimensions and Bulk Transport Properties



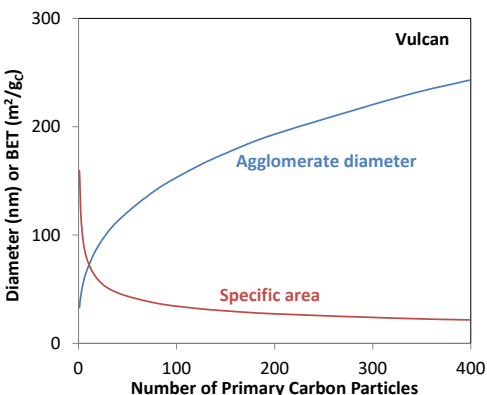
Model incorporates agglomerate and nano scale diffusion effects

$$R_{tx} \approx \frac{\sqrt{a_a d_a^2 l_{Pt}}}{6RT\psi_i} = \sqrt{R_a R_n}$$

$$R_{n,LSAC} = \frac{d_{Pt}}{RT\psi_i}$$

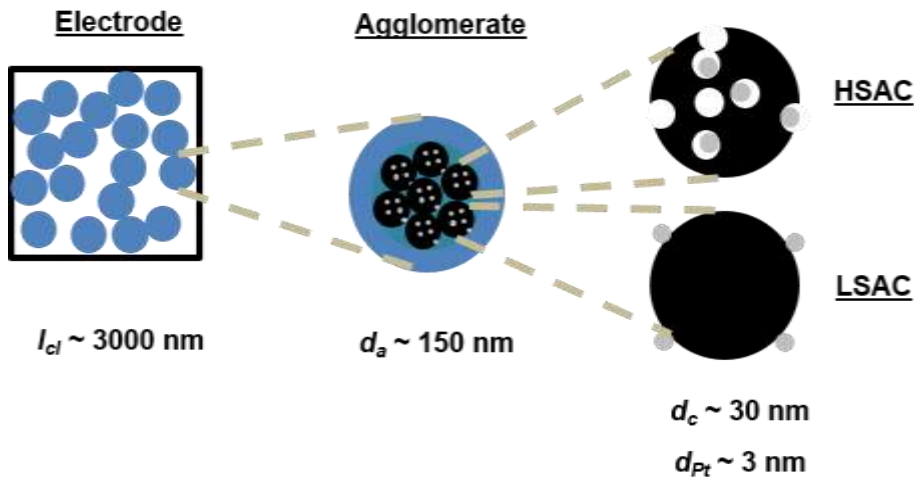
$$R_{n,HSAC} = \frac{\pi d_{Pt}}{2RT\psi_i} + \frac{4l_\mu}{RT\psi_w}$$

	Kongkanand et al. (12)		Harzer et al. (7)	
Carbon black	V	KB	KB(NH _x) _{PO}	KB _{IW}
Percent Pt on surface	95%	38%	100%	0%
Pt load (mg/cm ²)	60	60	63	64
Pt wt%	20	20	20	20
I/C ratio	0.95	1.33 ^(a)	0.65	0.65
Equivalent weight	1100	1100	700	700
H ₂ O/SO ₃ ⁻ , λ	3.9	3.9	3.9	3.9
f _v (%) ^(b) (41)	12	12	18	18
ψ _{i,b} (x10 ¹⁴ mol·m ⁻¹ ·Pa ⁻¹ ·s ⁻¹)	1.2	1.2	1.5	1.5
ψ _{w,b} (x10 ¹⁴ mol·m ⁻¹ ·Pa ⁻¹ ·s ⁻¹)	2.6	2.6	2.6	2.6
ψ _i /ψ _w ^(d)	2.97	0.77	1.02	1.02
d _a (nm)	150	150	300	300
η(φ)	0.49	0.59	0.23	0.44
R _{meas} (s/cm)	13	17	18	39
R _{pred} (s/cm)	10.3	15.0	15.6	35.5

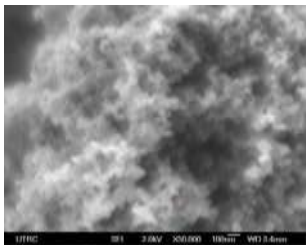


Approach: Modeling Catalyst Layer Structure

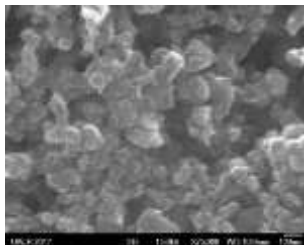
Develop and validate a model for transport resistance that uses measured microstructural details and transport properties – no free parameters.



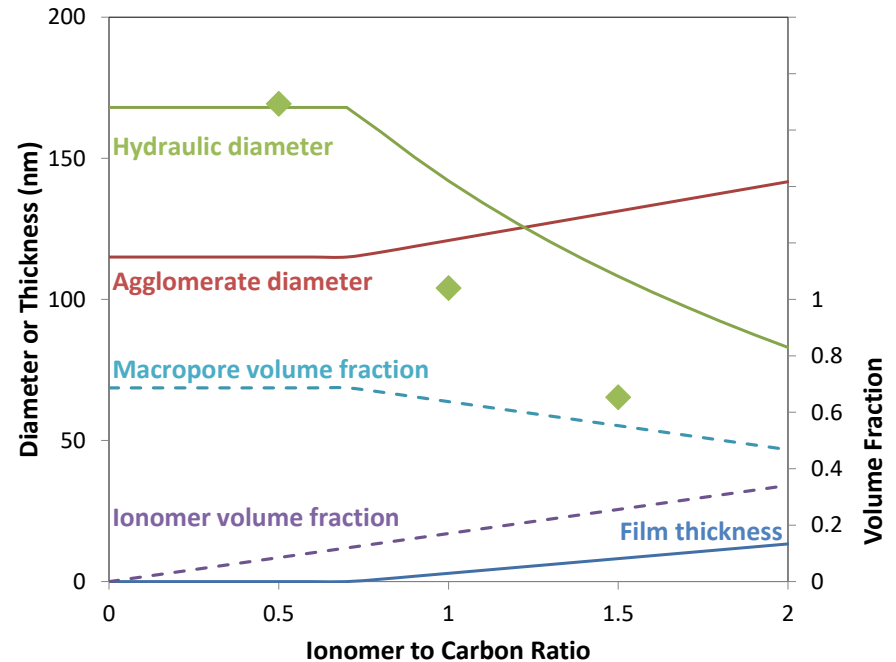
Pt/C Catalyst



Catalyst Layer



Catalyst Layer Dimensions



Data \blacklozenge from Hg porosimetry: Yu and Carter, *ECS Trans.*, **19**(17), 1 (2009).

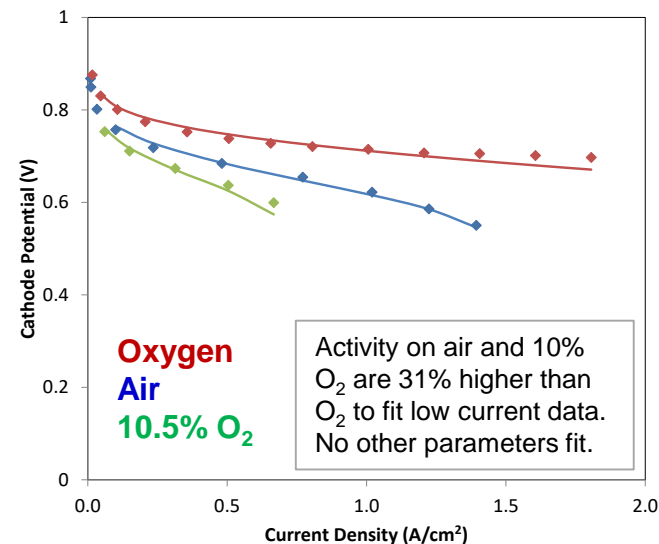
Accomplishment: BP-2 Go/No Go Milestone Performance, Part A

Part A: Validate microstructural model, including performance predictions, for MEAs with carbon-supported catalysts. The model will be able to reproduce measured pore-volume distributions to within 25% and predict key polarization metrics to within 25%.

Summary of *ex-situ* measurement on Ion Power MEAs

Property	Value	Measurement Technique
Platinum loading	0.046 mg/cm ²	Mass measured after decal transfer, 0.065 mg/cm ²
		Inferred from ECA measurements on cell, 0.046 mg/cm ²
		XRF, 0.08 mg/cm ²
Platinum weight %	20%	Measured by TKK
Ionomer to carbon ratio	0.94	Ion Power estimated from ink formula, 0.81 TGA, 0.94
Electrode Thickness	5 μm	Cross-section of MEA (I.P. Web 874)
Electrode porosity	63%	Calculated from electrode thickness and material amounts
		TEM of secondary pores, 36%
Agglomerate diameter	130 nm	2D HAADF STEM C-diluted CCLs (Web 928, 26% cat C)
Meso-porosity of carbon	0.34 cm ³ /g _C	Published for TKK catalysts
Ionomer Film thickness	2.6 nm	Calculated from agglomerate diameter and loadings
Platinum diameter	3.6 nm	Published ECA in flooded RDE (77.6 m ² /g)
		TEM of unprocessed catalyst, 2.4 nm
Platinum utilization	80%	ECA in liquid versus cell

Comparison of model to MEA



IR-free voltage	0.8 V	0.6 V
Measured	35 mA/cm ²	1.145 A/cm ²
Predicted	30 mA/cm ²	1.130 A/cm ²

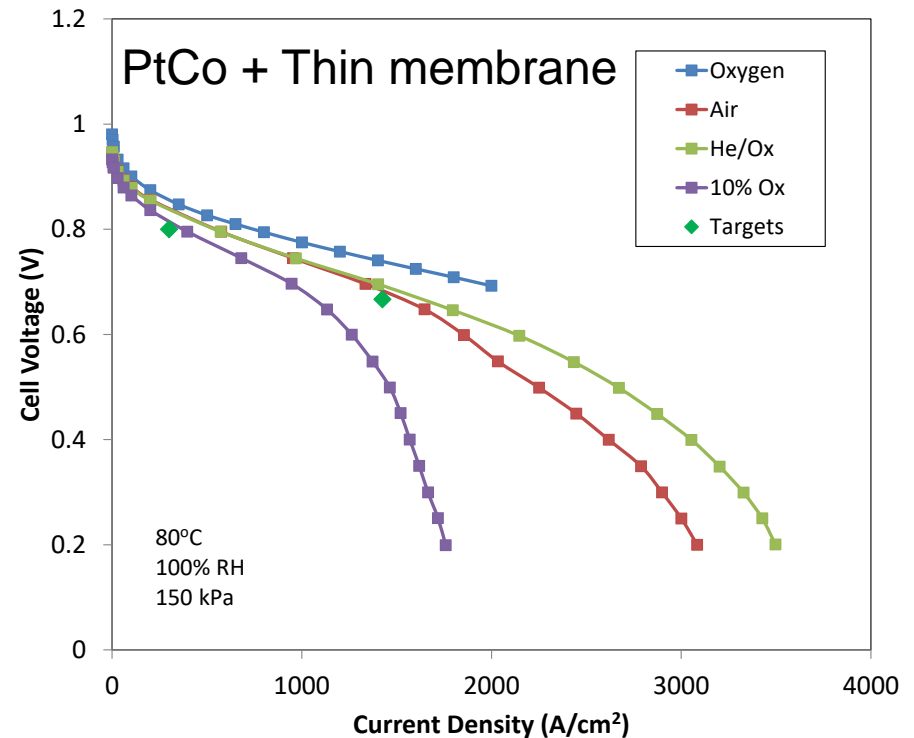
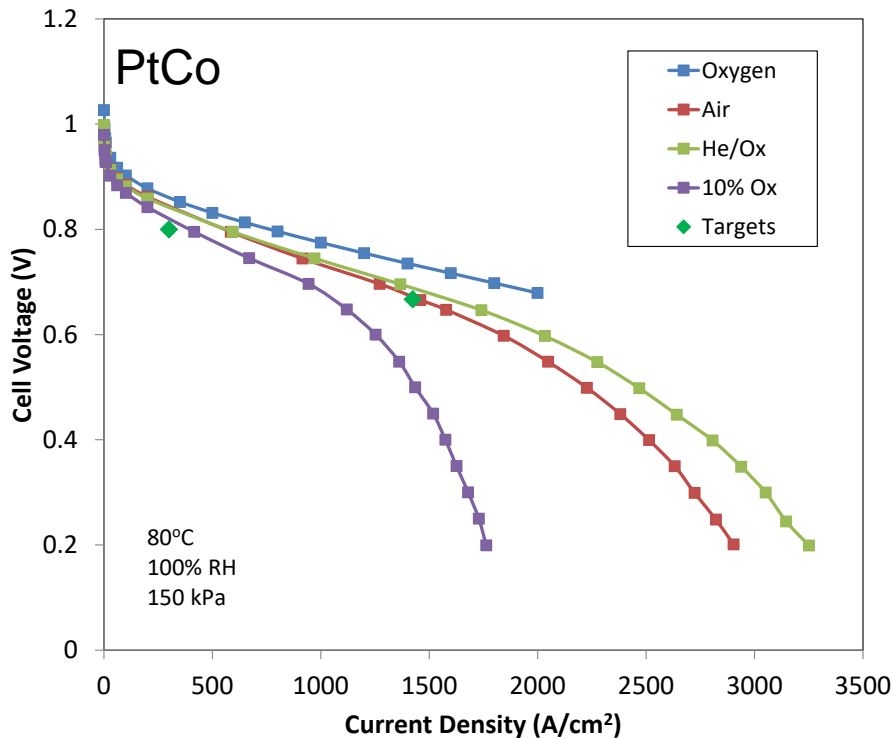
Ion Power

Los Alamos
NATIONAL LABORATORY

OAK
RIDGE
National Laboratory

Accomplishment: BP-2 Go/No Go Milestone Performance, Part B

Part B: Demonstrate progress towards meeting 2020 MEA performance targets with carbon supported catalyst and a total catalyst loading of $\leq 0.125 \text{ mg}_{\text{PGM}}/\text{cm}^2$; specifically: $\geq 240 \text{ mA}/\text{cm}^2$ at 0.8 V and $\geq 905 \text{ mW}/\text{cm}^2$ at rated power measured using the specified polarization curve protocol in FCTO's MYRDD (Table P.6).

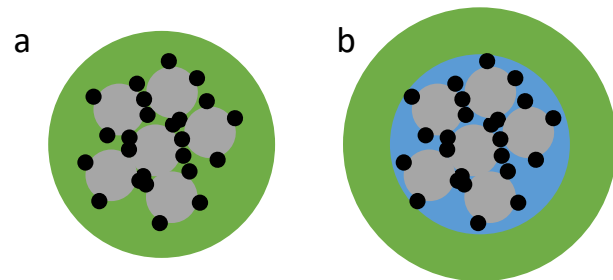


Accomplishments: Modeling Intra-Agglomerate Ohmic Losses (Q9)

Two extreme models for agglomerates interiors in the literature

- (a) ionomer filled
- (b) water or gas filled

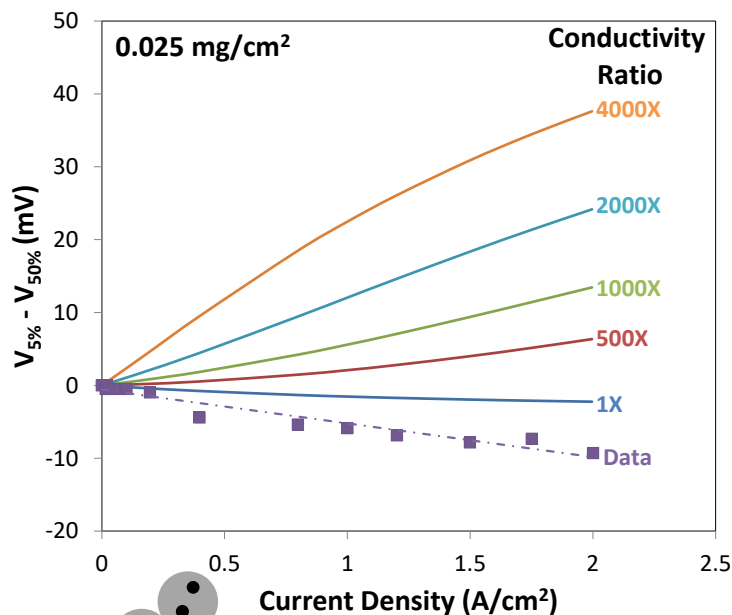
Water or gas filled agglomerate interiors have low conductivity



Compare polarization on oxygen to minimize transport losses

Look at different wt% Pt catalysts to give different agglomerate scale losses

Diluted to same thickness to give same electrode scale losses

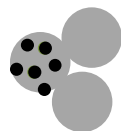


Low wt% catalyst predicted to be better than high wt%

Benefit increases with ratio of ionomer to intra-agglomerate conductivity

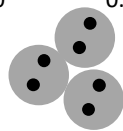
Experimental data does not behave as model predicts

No evidence for extremely low intra-agglomerate conductivity



50 wt% Pt/C + C

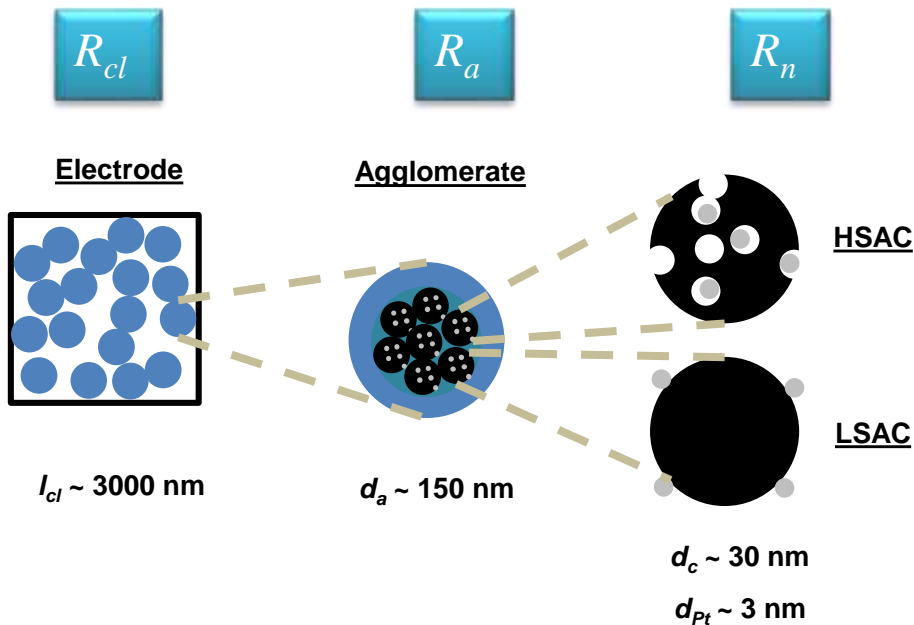
vs.



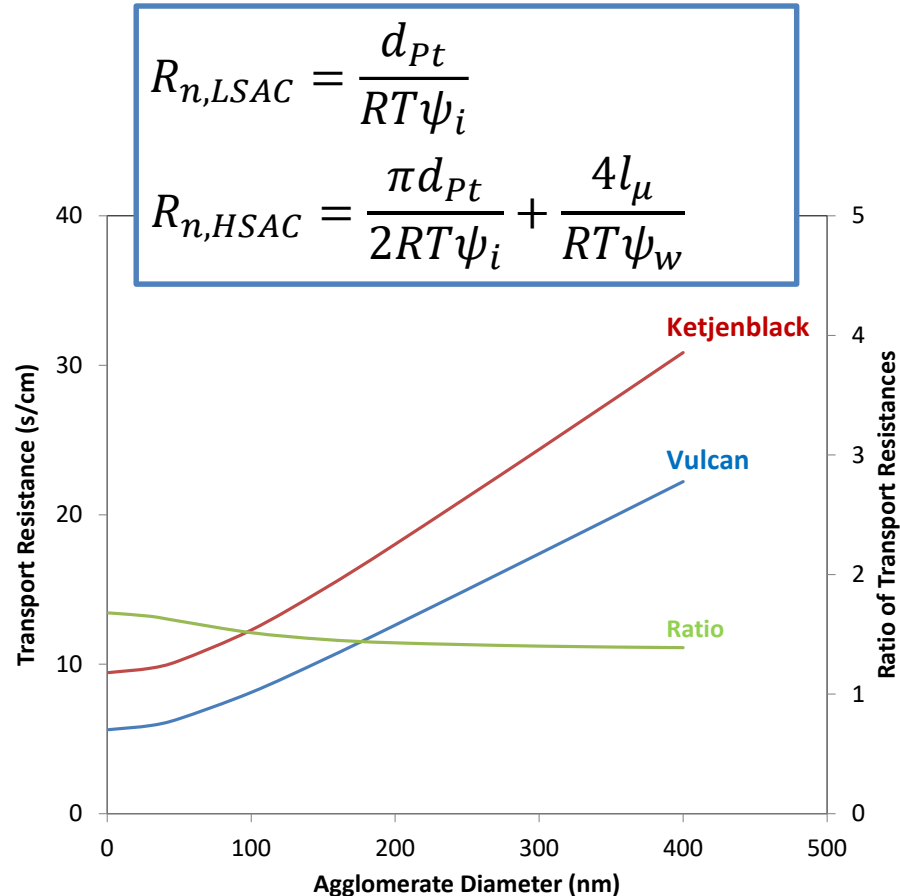
20 wt% Pt/C

Accomplishment: Transport Resistance in Pt/HSAC (Q9)

Two carbon black supports are common in fuel cells: high and low surface area (HSAC, LSAC)
 HSAC – carbon contains micropores that exclude ionomer and host platinum
 LSAC – carbon has few micropores, most platinum is on surface touching ionomer

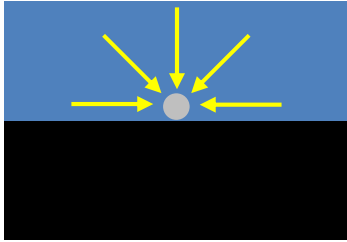


We modified LSAC model to treat HSAC by assuming that platinum sits in water filled pits with diameter equal to platinum nanoparticles

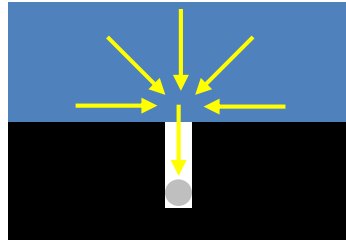


Accomplishment: Ohmic Resistance in Micropores in Pt/HSAC Catalysts (Q9)

LSAC



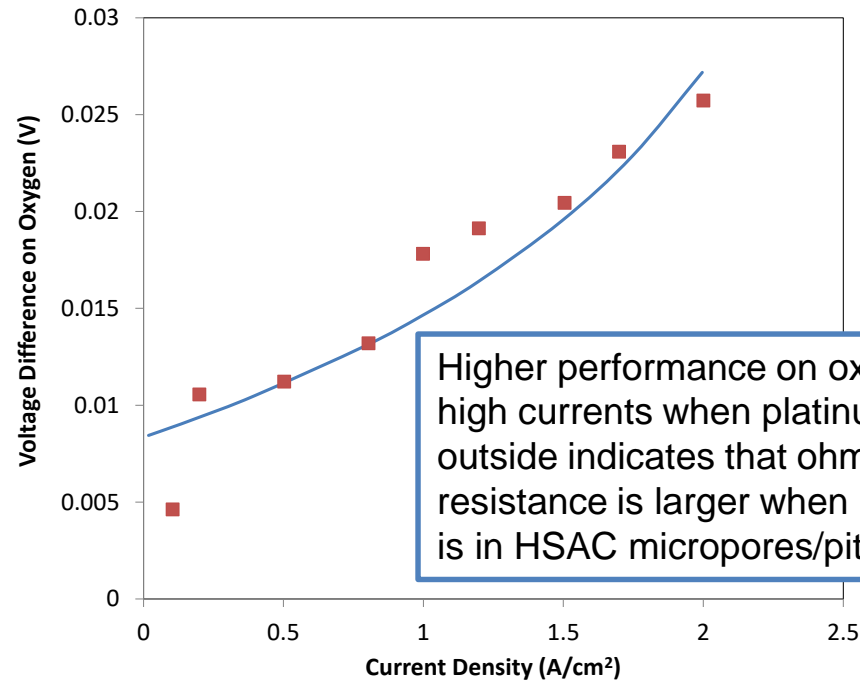
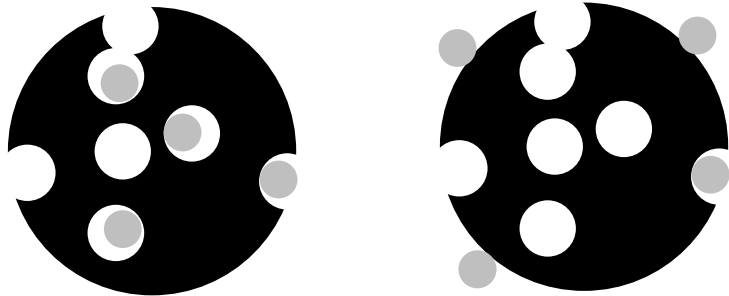
HSAC



Nanoscale O₂ transport losses are 6x higher in HSAC

Ohmic losses in pits can be large even though the pits are small if the conductivity of the pit solution is very small.

Harzer *et al.* (*J. Electrochem. Soc.*, **165**, F770 (2018)) preferentially deposited platinum on the outside and inside of Ketjenblack. Performance at high current much better when platinum on outside.

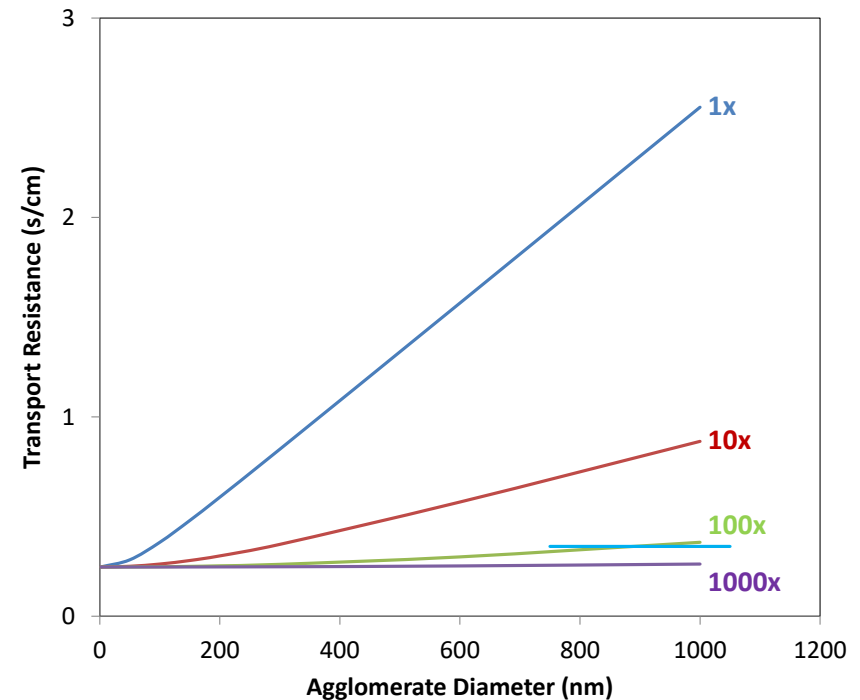
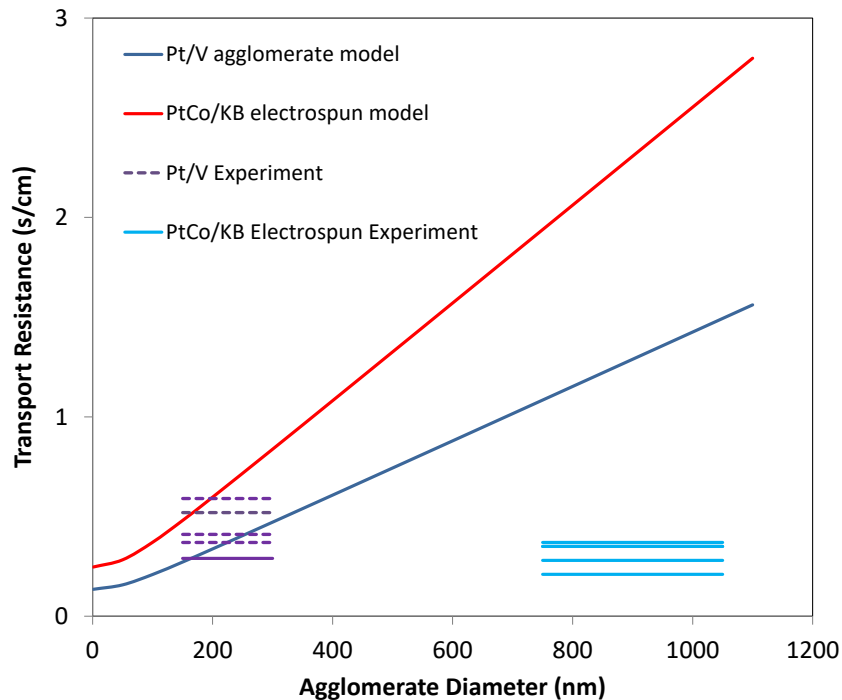


Higher performance on oxygen at high currents when platinum on outside indicates that ohmic resistance is larger when platinum is in HSAC micropores/pits.

Accomplishment: Transport Resistance in Electrospun Catalyst Layers (Q10)

Pintauro (*J. Electrochem. Soc.*, **167**, 054517 (2020)) developed high performance electrospun electrodes. The fibers can be regarded as long cylinders. Modeling them as ionomer filled spheres with 1.5x cylindrical radius (same volume / area ratio) significantly overestimates transport resistance.

Model can be matched to experiments by dramatically increasing oxygen permeability. Consistent with 31% porosity measured in these fibers. According to model nanoscale transport resistance is dominant, and predicted magnitude aligns with measurements.

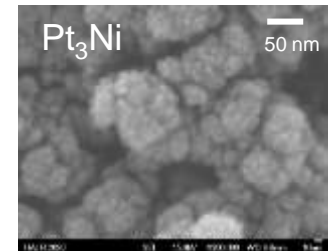
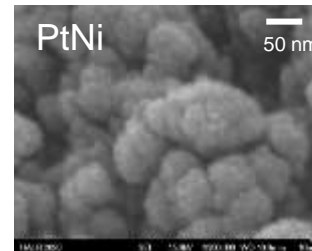
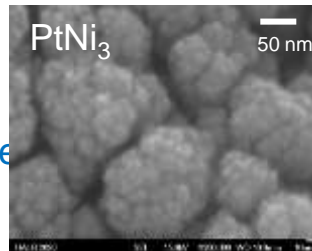


Accomplishment: Nanocolumnar Pt-Ni Thin Film Electrocatalyst (Q10)

Tansel Karabacak's Group, University of Arkansas at Little Rock, collaboration with LANL and ORNL

- Self-supported nanocolumnar Pt-Ni alloy thin films with different Pt:Ni ratios and Pt weight loadings were deposited by high pressure sputtering on an MPL-like surface composed of carbon particles in order to mimic the catalyst-coated gas diffusion layer (gas diffusion electrode, GDE) in an MEA.
- Cauliflower-like microstructure was observed (See the SEM images below. TEM imaging by ORNL is under progress).
- Benchtop CV and RDE measurements show that Pt:Ni (1:3) gives the highest electrochemical performance compared to other ratios.
- MEA tests were performed at UTRC. Due to non-sufficient catalyst porosity, there was oxygen transport issues. In-cell test results did not differ significantly for different Pt:Ni ratios. Actual activity is believed to be shadowed by the mass transport issues.
- Surface diffusion rate of Ni is high. This leads to large particle/grain/cluster sizes with small particle-particle gaps during thin film deposition, which is expected result in poor mass transport in MEA tests.
- Currently working on depositing Pt and Pt:Ni nano-cauliflowers onto carbon powder to increase the surface area and accessibility, and therefore improve mass transport..

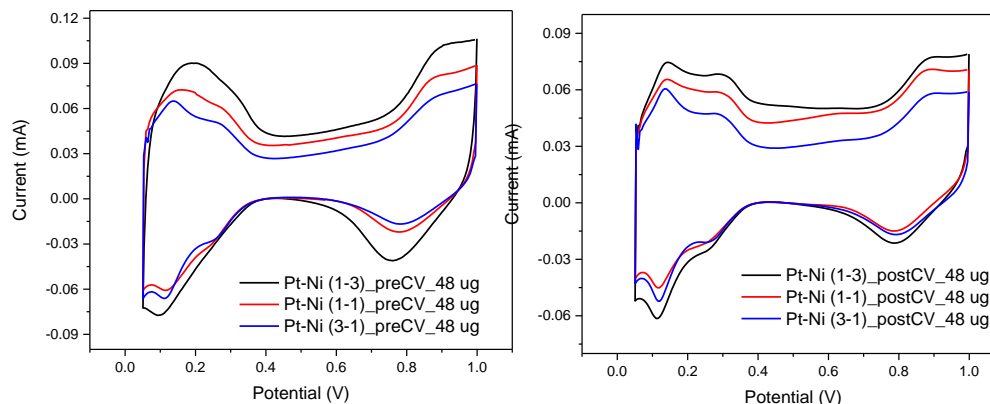
- High pressure sputtering
- Deposited on MPL-like surface
- Cauliflower-like columnar structure



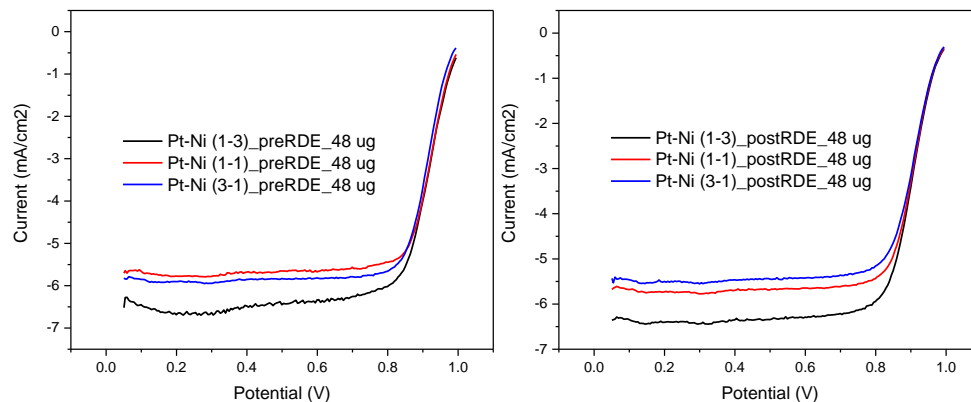
*pre = initial
*post = after 3000 cycles of stability test

Pt loading ($\mu\text{g}_{\text{Pt}}/\text{cm}^2$)	Pt:Ni (1:3)						Pt:Ni (1:1)						Pt:Ni (3:1)					
	ECSA (m^2/g)		SA ($\mu\text{A}/\text{cm}^2$)		MA (A/mg)		ECSA (m^2/g)		SA ($\mu\text{A}/\text{cm}^2$)		MA (A/mg)		ECSA (m^2/g)		SA ($\mu\text{A}/\text{cm}^2$)		MA (A/mg)	
	pre	post	pre	post	pre	post	pre	post	pre	post	pre	post	pre	post	pre	post	pre	post
~94	34	21	1842	921	0.64	0.19	26	20	1654	890	0.43	0.17	22	15	955	799	0.21	0.12
~48	38	26	1781	855	0.66	0.22	27	21	1183	793	0.33	0.16	28	22	710	706	0.20	0.16
~20	42	33	1196	809	0.51	0.27	34	25	1095	794	0.37	0.20	35	26	586	583	0.20	0.15

Accomplishment: Nanocolumnar Pt-Ni Alloy Thin Film Electrocatalyst (Q10)



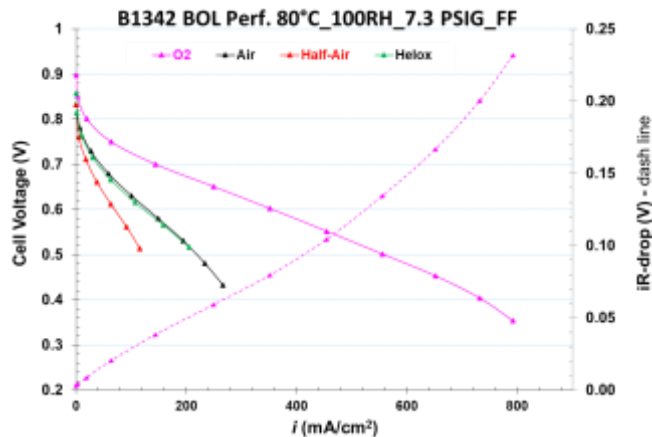
Cyclic voltammograms of PT-TFs deposited on MPL-like surface before and after 3,000 cycles of durability test



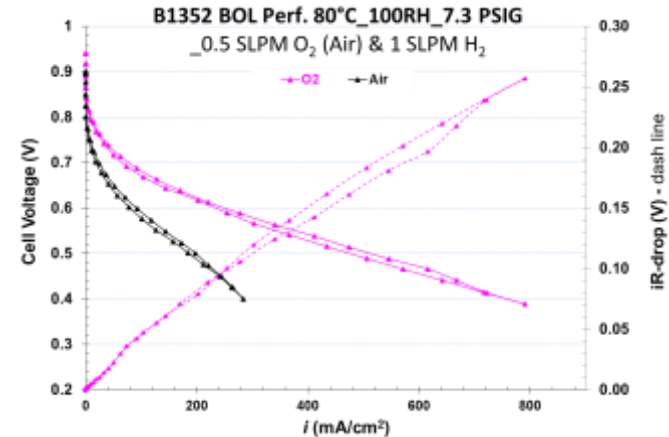
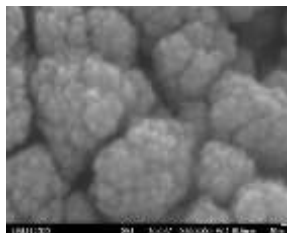
ORR polarization of Pt-Ni alloy TFs deposited on MPL-like surface before and after 3,000 durability cycles

Accomplishment: Nanocolumnar Pt-Ni Alloy Thin Film Electrocatalyst

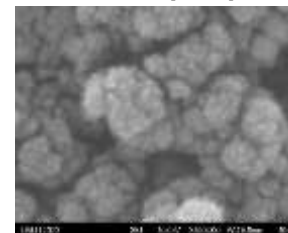
- MEA tests were performed at UTRC. The mass (O_2) transport losses dominate the cell performance due to non-sufficient catalyst porosity. In-cell test results do not differ for different Pt:Ni alloy ratios. Actual activity is believed to be shadowed by the mass transport issues.
- The Helox performance overlaps with Air, indicating the O_2 transport losses is occurring in solid phase(s) and/or nano-pores in the electrode, rather than in meso-pores/micro-pores in the GDL.



Pt:Ni (1:3)



Pt:Ni (3:1)



Accomplishment: Models of Thin-Film Catalyst Layers (Q10)



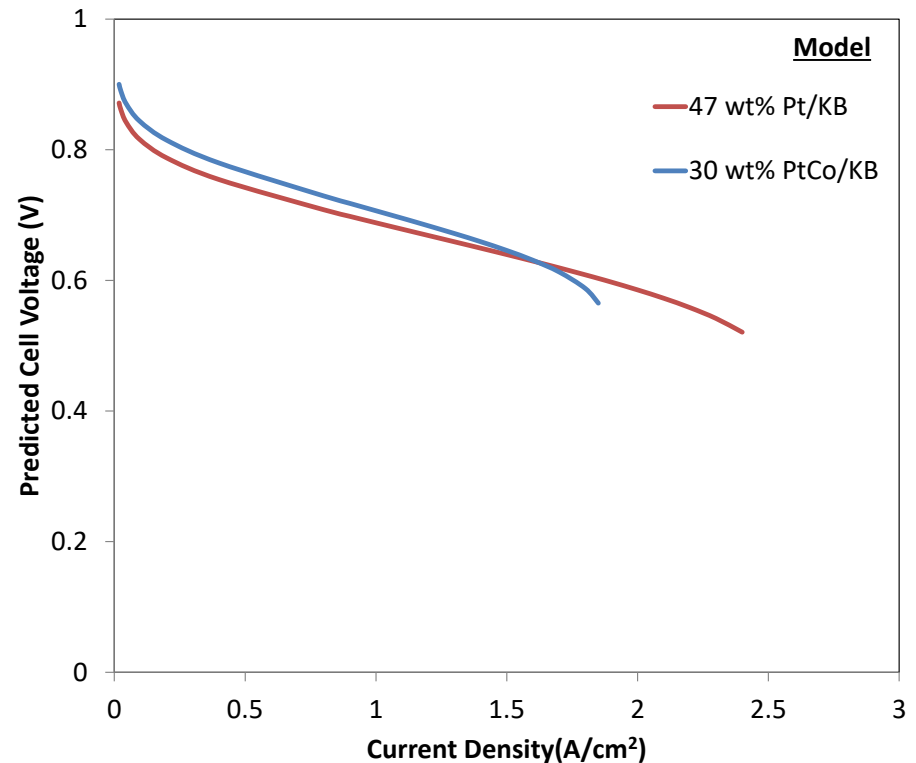
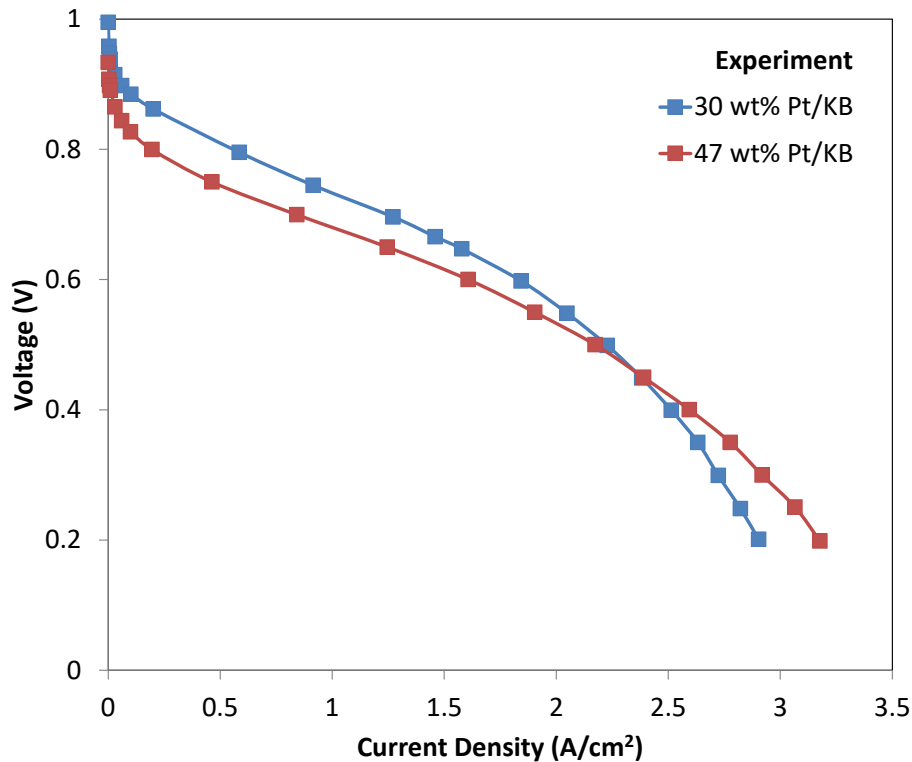
Description	Water film	Water filled	Ionomer filled	Ionomer film	Ionomer, flooded
Effectiveness at 0.65 V	0.98	0.073	0.091	0.98	0.05
Thiele Modulus	0.02	13.7	11	0.02	17.7
Resistance ($\Omega\text{-cm}^2$)	17.8	17.8	0.1	0.55	0.55

$$\text{Effectiveness Factor} = \frac{\text{Current density limited by transport \& kinetics}}{\text{Current density limited by kinetics only}}$$

Key model inputs:

- Pt loading = 0.1 mg/cm²
- $k_{ORR} = 0.053$ cm/s (at 0.65V)
- $D_{O_2} = 5.7 \times 10^{-7}$ cm²/s
- $\kappa_{ionomer} = 89$ mS/cm

Accomplishment: Qualitative Predictions for PtCo vs. Pt Catalyst Layers (Q10)

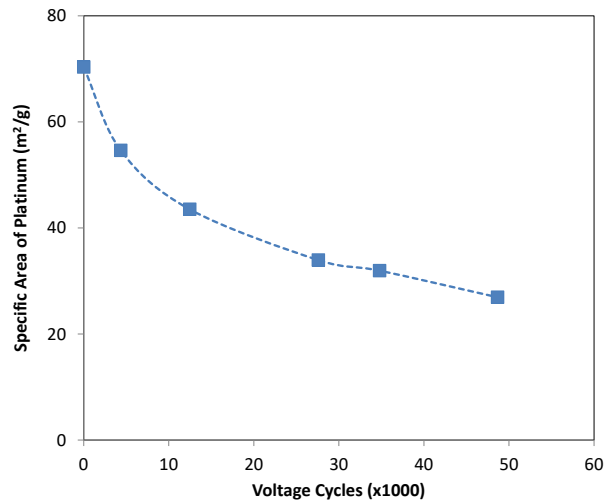


Simulations of PtCo/HSAC show a declining benefit versus Pt/HSAC at high current density caused by the smaller surface area, larger particles.

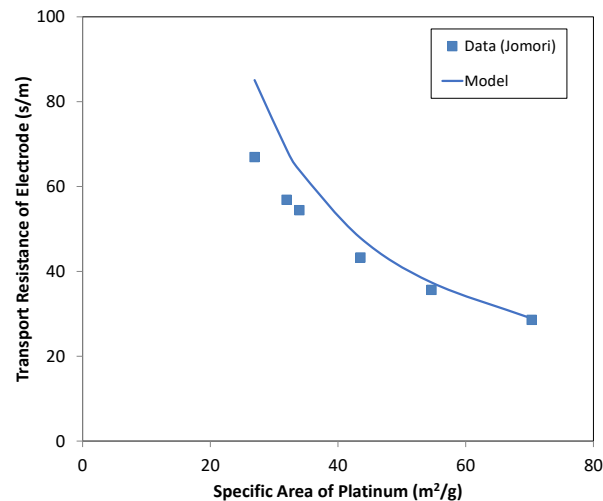
Accomplishment: Transport Losses Predicted from Surface Area Loss (Q11)

Increase in transport resistance in Pt/HSAC caused by voltage cycling predicted from measured loss in surface area

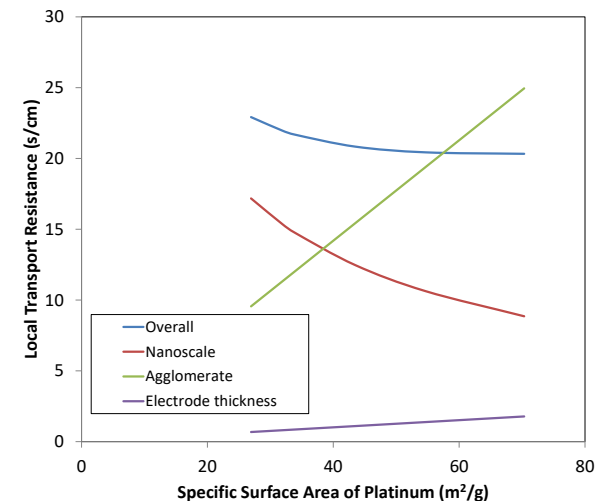
Experimental ECA loss, square wave 0.7 to 0.9 V cycling



Comparison of measured and predicted transport resistance



Predicted components of local transport resistance



Data from: Jomori *et al.*, *J. Power Sources*, **215**, 18 (2012)

0.1 mg/cm², 30% Pt/HSAC,
0.75 l/C, 150 nm
agglomerates, 5 nm micropits

Nanoscale transport length
approaches particle diameter

Responses to Reviewers' Comments (1/2)

- The most critical Comments & Scores were on:
 - Relevance and Future Work
- Also scored low on:
 - Approach and Accomplishments

Critical Comments on Approach and Accomplishments:

- Many comments on UTRC's model that were fundamentally incorrect

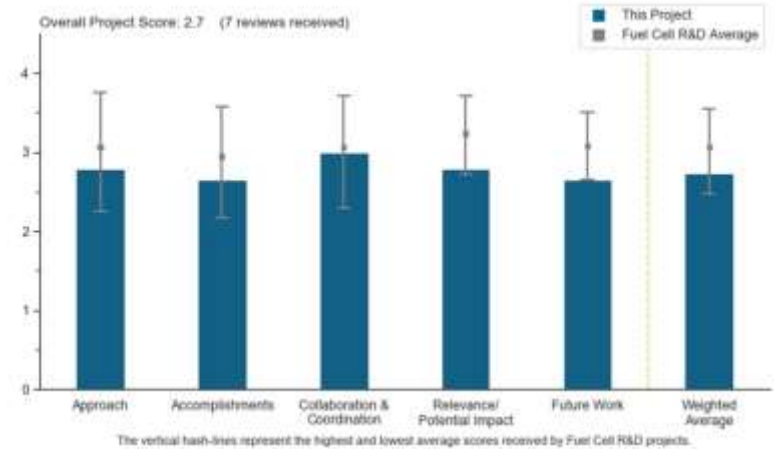
The Hierarchal Model focuses on transport losses with low catalyst loadings by treating oxygen transport in ionomer in novel way; it is NOT limited to “gas-phase transport losses” and it does not neglect ohmic losses; nor does the model need to include complexities that are obviously not well correlated with the phenomena that have been observed (e.g., pore-size distributions or GDL properties, which will primarily impact gas-phase transport) and are not consistent with how transport resistance varies with RH, E_A , etc.

- Concerns about the model being validated or enabling improved MEA performance

The model has been validated, with multiple data sets. The team continues to reduce transport losses by utilizing predictions from the model.

- Claims that the team is repeating what has already been done or is not innovative

Prior to this project, no published model could effectively explain the data (e.g., how transport resistance varies with loading, catalyst wt. %, RH, T, etc.). We have shown that the Hierarchal Model does fit the data of interest here, and compared 10 different models to show this was the case, both qualitatively and quantitatively (3 peer-reviewed publications, to date). Cause of large transport resistances was unexplained for the previous decade.

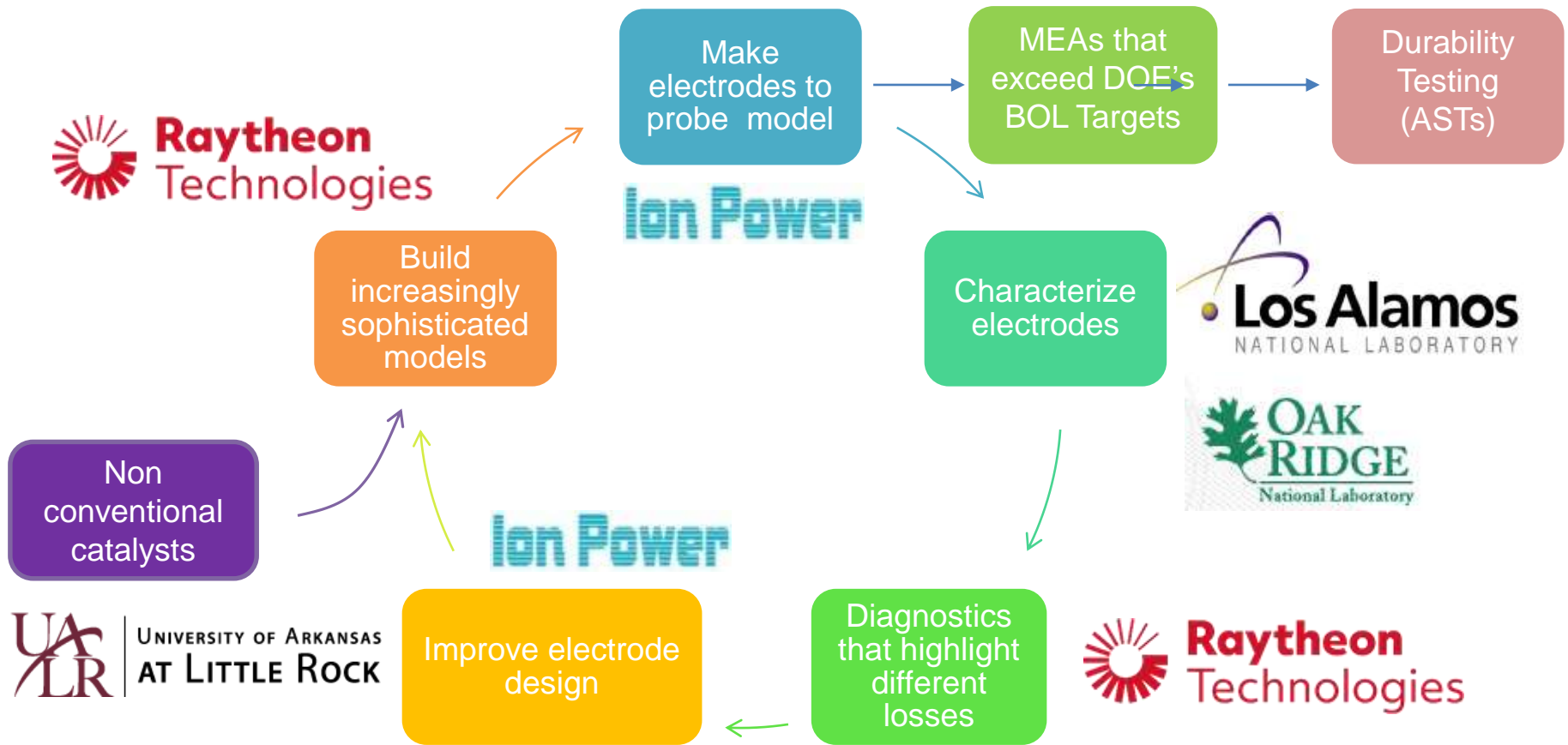


Responses to Reviewers Comments (2/2)

Critical comments on Relevance and Future Work:

- “Nearly all FC developers have already found some way to treat O₂ transport resistances”
Not at DOE’s target catalyst loadings (e.g., TMC stack has higher loadings).
- A couple of comments to the effect of: “modeling alone cannot fix the problem.”
The model does not address how to make the required structures, but it does provide guidance on what types of CCL architectures should enable improved performance.
- More misunderstandings about the model (e.g., model does not include ohmic losses)
We have published models that incorporate kinetics, ohmics, and multiple kinds of transport, and our third paper explicitly shows the relative importance of oxygen transport and ohmic resistance.
- Doubts raised about the team being able to substantially vary agglomerate size
Agreed, this is challenging, and the team does not plan to explore a variety catalyst ink solvents and mixing methods (utilizing FC PAD’s experience on ink formulations)
- There was a lot of skepticism about the thin-film (TF) catalyst work included here
The team has successfully fabricated and tested some MEAs using UALR’s TF catalysts. UALR can provide the team with variations of TF catalysts, which has been their primary focus to date. These materials can then be used to study and develop new CCL architectures that utilize TF catalysts. Currently, there are very few sources for TF catalysts.
- Multiple suggestions to use other available catalysts and/or alternative structures
Agreed, the team has been trying to collaborate more with others working on alternative CCLs and plans to make this a major focus of the last year of the project (if funded).

Collaboration and Coordination



Prime:  **Raytheon Technologies**
Industry

Subs:  **Ion Power**
Industry

 **UNIVERSITY OF ARKANSAS AT LITTLE ROCK**
University

Remaining Barriers and Challenges

- Verification of model predictions for alternative catalyst layers rely on published data because laboratories are closed
- Verification of hierarchical model pre- and post durability cycling rely on published data because laboratories are closed
- Need to acquire sufficient data for electrospun catalyst layers to complete model verification

Proposed Future Work

Milestone	Task Title	Milestone Description	Status
Q10	Alternative catalyst MEA fab and performance	Validated microstructural model for MEA with alternative catalysts	75%
Q11	Carbon-supported model and performance degradation	Validate Pt/C hierarchal model to include both BOL and EOL performance after selected AST protocols	75%
Q12	Alternative catalyst MEA fab and MEA performance	Complete validated model of at least two different alternate catalyst structures, including providing recommend changes to catalyst layer morphology to improve the performance	25%
Q12	Model validation and MEA Performance	Recommend optimal catalyst layer structures based on model learnings	0%

- Improve both Pt/C and thin-film catalyst layer models
- Resolve agglomerate diameters from different characterization methods
- Study the impact of surface and buried platinum on the decay of Pt/HSAC
- More work on making, testing, and modeling alternative catalyst layer structures

Summary

- Mass transport losses in catalyst layers are an impediment to high efficiency at high power density
- A hierarchical catalyst layer model that incorporates transport at multiple length scales is required to describe experimental trends at low platinum loadings
- RTRC's hierarchical model was extended to describe oxygen transport and ohmic losses in high surface area carbons
- Predictions from the model have assisted in developing catalyst layers with lower oxygen transport losses
- The Pt/HSAC model quantitatively predicts the increase in transport resistance caused by ripening of catalyst particles
- Thin-film catalyst layers without carbon supports show promising catalytic activity and performance

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Nancy Kariuki
Debbie J. Myers

Technical Back-Up

Publications

- R. M. Darling, “A Hierarchical Model for Oxygen Transport in Agglomerates in the Cathode Catalyst Layer of a Polymer Electrolyte Fuel Cell”, *J. Electrochem. Soc.*, **165**, F571 (2018).
- R. M. Darling, “A Comparison of Models for Transport Resistance in Fuel-Cell Catalyst Layers”, *J. Electrochem. Soc.*, **165**, F1331 (2018).
- L. M. Pant, Z. Yang, M. L. Perry, and A. Z. Weber, “Development of a Simple and Rapid Diagnostic Method for Polymer-Electrolyte Fuel Cells”, *J. Electrochem. Soc.*, **165**, F3007 (2018).
- R. M. Darling, “Modeling Air Electrodes with Low Platinum Loading”, *J. Electrochem. Soc.*, **166** F3058 (2018).
- A. Chowdhury, R. M. Darling, C. J. Radke, A. Z. Weber, “Modeling Water Uptake and Pt Utilization in High Surface Area Carbon”, *ECS Trans.*, **92**, 247 (2019).
- R. M. Darling, “Examining Ohmic Losses in Fuel-Cell Catalyst Layers with Different Pt/C Ratios”, *J. Electrochem.*, **167**, 084505 (2020).
- R. M. Darling and S. F. Burlatsky, “Modeling Oxygen Transport in High-Surface Area Carbon Supports for Polymer-Electrolyte Fuel Cells”, *J. Electrochem. Soc.*, Accepted.

Presentations

August 2018: “Use of Modeling for Diagnostics of Polymer Electrolyte Fuel Cells”

- Presented by L. M. Pant (**LBNL**) at ISE Meeting (Providence, R.I.)
- Co-authors: M. L. Perry (**UTRC**) and Adam Z. Weber (**LBNL**)

October 2018: “Platinum Nanorod Arrays as ORR Electrocatalyst for Polymer Electrolyte Membrane Fuel Cells”

- Presented by M. Begum (**UALR**) at Fall ECS and AiMES Meeting (Cancun, Mexico)
- Co-authors: B. Ergul & T. Karabacak (**UALR**), Nancy Kariuki & Debbie J. Myers (**ANL**), & M. L. Perry (**UTRC**)

October 2018: “A Model-Based Approach to Improved Understanding & Mitigation of Transport Losses in PEFCs”

- Presented by M. L. Perry (**UTRC**) at Fall ECS and AiMES Meeting (Cancun, Mexico)
- Co-authors: Z. Yang & R. M. Darling (**UTRC**)

September 2018: “High Performance PEFC Electrode Structures”

- Presented by R. M. Darling (**UTRC**) at FCTT Meeting (Detroit, MI)
- Co-authors: Z. Yang & M. L. Perry (**UTRC**)

November 2018: Platinum Nanorod Arrays as ORR Electrocatalyst for Polymer Electrolyte Membrane Fuel Cells

- Two talks presented by B. Ergul and M. Begum at 2018 American Chemical Society (ACS) Southwest Regional Meeting (Little Rock, AR)
- Co-authors: T. Karabacak (**UALR**)

January 2019: “High Performance PEFC Electrode Structures”

- Presented by M. L. Perry (**UTRC**) during breakout session at FC PAD Consortia Meeting (Santa Fe, NM)
- Co-authors: Z. Yang & R. M. Darling (**UTRC**)

August 2019: “High Performance PEFC Electrode Structures”

- Presented by M. L. Perry and R. M. Darling (**UTRC**) at FCTT Meeting (Detroit, MI)

January 2020: “High Performance PEFC Electrode Structures”

- Presented by R. M. Darling (**UTRC**) during breakout session at FC PAD Consortia Meeting (Santa Fe, NM)

Modeling of Catalyst Layers with Platinum Alloys on Carbon

Previously reported and published

Oxygen transport resistance with nanoscale diffusion for low surface area carbon (LSAC)

Comparison of newly developed transport resistance model to literature approaches

Modeling cell performance with LSAC showing importance of Pt/C ratio at different RH

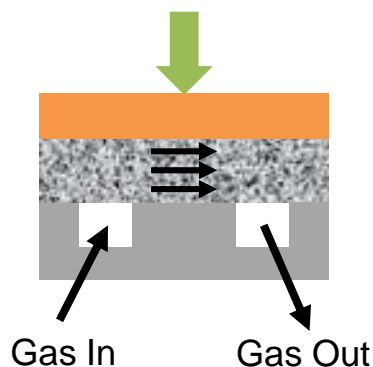
Modeling ohmic resistance at agglomerate scale showing importance of Pt/C

Transport resistance in electrodes with high-surface area carbon supports

Comparison of model and experiment for electrodes subjected to AST cycling

Oxygen transport and ohmic resistances in electrodes with HSAC

Agglomerate Size from Gas Permeability



Measure gas permeability of electrode and use Carman-Kozeny equation to calculate grain size.

Force (psi)	Gas Press (psi)	Porosity	Electrode thickness (μm)	Permeability (cm ²)	Grain size (nm)
100	7	65%	12.0	3.31E-12	163
100	15	65%	12.0	5.52E-12	211
200	7	40%	7.0	3.75E-13	195
200	15	40%	7.0	3.17E-13	179
200	25	40%	7.0	2.58E-13	162
300	7	36%	6.6	1.56E-13	157
300	15	36%	6.6	1.16E-13	135
300	25	36%	6.6	2.76E-13	209

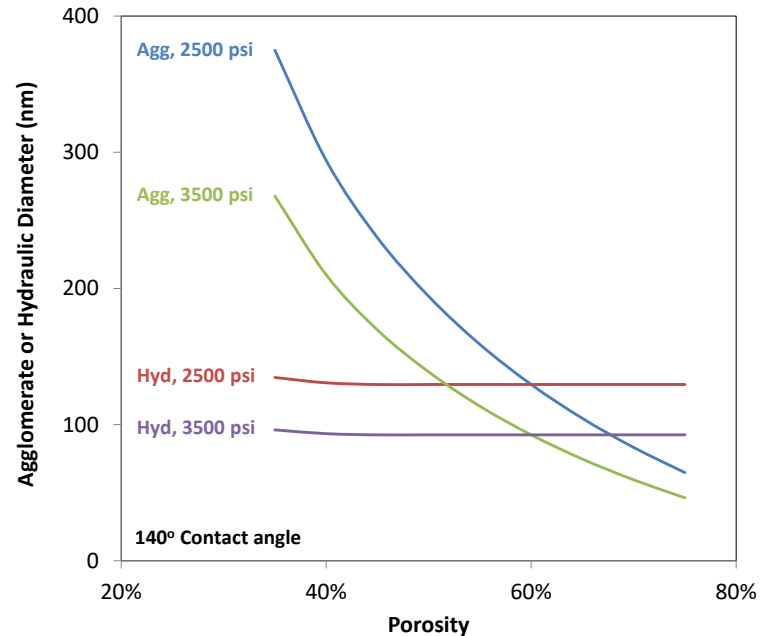
Agglomerate Size from Porosimetry



Mayer-Stowe model for intrusion

$$P^*(\varepsilon, \sigma) = \frac{P_c d_a}{2\sigma}$$

$$P^*(\varepsilon, \sigma) \approx - \frac{m \cdot 6(1 - \varepsilon) \cos \theta}{\varepsilon}$$



Yu and Carter (*ECS Trans.*, **19**(17) 1 (2009)) gave 3000 psi for an electrode with $I/C = 1$. Porosity was 42% from MIP or 65% from density calculations. Consistent with $100 \text{ nm} < d_a < 200 \text{ nm}$