



Northeastern University
Center for Renewable Energy Technology



Vapor Deposition Process for Engineering of Dispersed PEMFC ORR Pt/NbO_x/C Catalysts

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Ford Motor Company

May 1, 2019

FC162

Overview

Timeline

- Project Start Date: 1/1/2017
- Project End Date: 3/31/2020
- Percent complete: 60%

Budget

- Total project budget: \$2,594,412
 - Total recipient share: \$518,883
 - Total federal share: \$2,075,529
 - Total DOE funds spent*: \$1,073,311

* As of 1/31/2019

Barriers

- A. Durability
- B. Cost
- C. Performance

Partners

- Oak Ridge National Laboratory
 - Small batch catalyst production
- Exothermics, Inc.
 - Large batch catalyst production
- University of Michigan
 - TEM, XPS, other characterization
- Northeastern University
 - X-ray absorption spectroscopy
- EWii (IRD) Fuel Cells LLC
 - CCM fabrication, fuel cell testing
- Other contributions: NREL (mass transport resistances)

Relevance

OBJECTIVE: Develop, integrate, and validate a new cathode catalyst material by developing and optimizing a vacuum powder coating physical vapor deposition (PVD) process

- Develop a New Cathode Catalyst Powder
 - Pt/NbOx/C for high durability, power density, mass activity
- Improve the Catalyst Powder Manufacturing Process
 - PVD with superior reproducibility over solution based methods
- Demonstrate the PVD Process is Scalable in a Cost Effective Manner
 - Scale up from small batch (~ 1-2 g) to large batch (20-40 g)
- Show Ease of Integration
 - Powders amenable to already established CCM/MEA manufacturing processes (ink processes)

This project focuses not just on a higher performing and more durable novel catalyst, but also on making the catalyst with a reproducible, scalable process.

Relevance: Targets and Status

The targets below are specified as part of quarterly milestones or go/no-go decisions:

MYRDD Table 3.4.5 Technical Targets: MEAs for Transportation Applications

Characteristic	Units	2020 Target	Project Status
Performance at rated power	mW/cm ² at 150 kPa (abs)	1000	410^{1a}

MYRDD Table 3.4.7 Technical Targets: Electrocatalysts for Transportation Applications

Characteristic	Units	2020 Target	Project Status
PGM content at rated power	g _{PGM} /kW _{gross} at 150 kPa (abs)	0.125	0.366^{1a}
PGM loading	mg _{PGM} /cm ² total	0.125	0.150 ¹⁻⁵
Mass activity	A/mg _{PGM} at 900 mV _{ir-free}	0.44	0.389¹, 0.335²
Electrocatalyst stability (0.6 ↔ 0.95 V)	% mass activity loss after 30K cycles	<40	14%¹, 9%³, 10%⁴, 1%⁵
Loss at 0.8 A/cm ² (0.6 ↔ 0.95 V)	mV loss after 30K cycles	<30	70 mV¹, 71 mV³, 40 mV⁴
Support stability (1.0 ↔ 1.5 V)	% mass activity loss after 5K cycles	<40	TBD
Loss at 1.5 A/cm ² (1.0 ↔ 1.5 V)	mV after 5K cycles	<30	TBD

¹ Measured using Exothermics 180920 (20/2.5% Pt/NbOx on acetylene black) ^a At 250 kPa. ² Measured using ORNL-L-013 (36/0.6% Pt/NbOx on acetylene black). High current measurements at 0.6 V, 80°C (Q/ΔT=2.44), fully humidified.

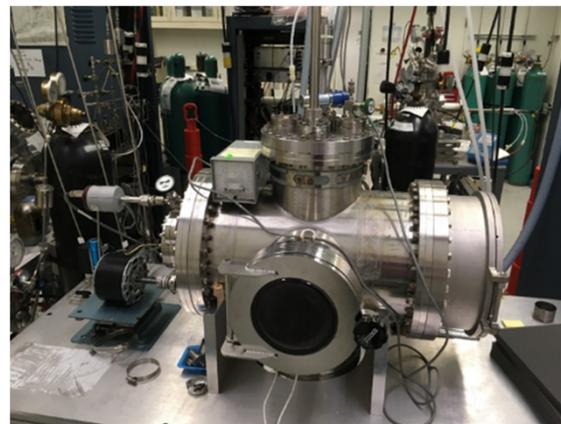
^{3,4,5} Measured using ORNL-L-034 (18/8.0% Pt/NbOx), ORNL-L-030 (18/6.3% Pt/NbOx), ORNL-L-032 (31/3.8% Pt/NbOx) respectively. All supported on acetylene black; anode loading 0.05 mg_{PGM}/cm²

Budget Period 2 focus was on greater durability with increased NbOx

Approach: Tasks and Schedule

Quarter	2017			2018				2019					
	0	1	2	3	4	5	6	7	8	9	10	11	12
TASK 1: Development, Characterization, and Validation of Catalyst Material and Development and Implementation of PVD process parameters													
1.1	Develop the Catalyst Matrix												
1.2	Small Batch Catalyst Synthesis and Carbon Fluidization												
1.3	Catalyst Powders Materials Characterization												
1.4	Synthesis of Catalytic Material with Solution Based Method												
1.5	Target Processing, Chamber Conversion, and Large Batch Carbon Nanoparticle Vacuum Fluidization												
1.6	XAS Evaluation of BOL, Cycled Catalyst Powders												
1.7	Processing of Large Batch Sputter Deposition on Powder												
TASK 2: Catalyst Layer Development and MEA Validation													
2.1	Baseline Materials Specification and Testing Protocol Development for DOE MEA Validation Cycling												
2.2	Catalyst Layer and MEA Development with Process Optimization												
2.3	MEA BOL Validation												
2.4	MEA Cycling Validation												
TASK 3: Project Management and Reporting													
3.1	Project Management												
3.2	Documentation and Reporting												

The approach has been to use small batch sampling at ORNL (Task 1.2) to direct large batch production at Exothermics (Tasks 1.5, 1.7). Material characterization (Tasks 1.3, 1.6) and fuel cell testing (Task 2) provide feedback data on understanding activity and durability.



ORNL

Small batch: 0.5-2.5 gram batches from ORNL
Large batch: 35-40 gram batches at Exothermics



EXOTHERMICS

Small catalyst batches at ORNL, along with characterization at UM, Northeastern, and EWII, have influenced desired production parameters at Exothermics

Approach: Budget Period 1 Showed High, Repeatable Activity

Budget Period 1 Milestones: 1/1/2017 – 3/31/2018

Milestone #1 (Q1): Demonstrate that small batch PVD catalyst synthesis has a narrow metal particle size distribution (2-10nm) on the carbon powder via TEM



Line-of-sight sputtering yields wide Pt particle size distributions. Narrow distribution may not be necessary for performance, durability. Future particle size distribution milestones removed.

Milestone #2 (Q2): Demonstrate that small batch PVD catalyst synthesis is able to reliably reproduce Pt and Nb loadings (with <10% variation) on the carbon powder by XRF measurement.



Repeatability shown for Pt within +/- 10% of average. Nb within +/- 40% at very low average weight percent (0.9%).

Milestone #3 (Q3): RDE testing of PVD Pt/NbO_x/HSAC catalysts demonstrates a BOL mass activity (at 0.9 V) above 0.40 A/mg_{Pt}



Project has shown 23 samples to have met this milestone: 10 from ORNL, 8 from Ford APD (small batch), and 5 from Exothermics (large batch). 14 non-alloyed, 9 alloyed. Pt wt% from 13-71% (22 with 17% or higher).

Go/No-Go Decision Point for Budget Period 1 (end of Year 1): Validation of PVD deposited catalyst powders via MEA BOL Testing with 40-50 cm² single cell having a cathode loading of ≤ 0.150 mg_{Pt}/cm² giving a **BOL mass activity of ≥ 0.30 A/mg_{Pt}** at 900 mV_{iR-free} following the protocols given in Table 3.4.7 of FCTO's MYRDD Plan.



Project has shown 4 samples to have met this milestone:

- ORNL-L-013 (Pt/NbO_x/C): 335 A/g_{Pt}
- Exothermics 180109 (Pt/NbO_x/C): 328 A/g_{Pt}
- Exothermics 180302 (PtCo/NbO_x/C): 309 A/g_{Pt}
- Exothermics 180308 (PtCo/NbO_x/C): 352 A/g_{Pt}

Project has passed budget period 1 milestones.

Approach: Budget Period 2 Milestones Revised to Focus on Material Characterization and Durability

Budget Period 2 Milestones: 4/1/2018 – 3/31/2019

Revised: Milestone #4 (Q5): Determine Pt, NbOx, C interactions in PVD Pt/NbOx/C catalysts using elemental mapping, TEM, XAS, XPS; provide comparisons between PVD Pt/NbOx/C and Pt/C



Pt adjacent to or on top of deposited NbOx (amorphous, electronically conducting), enhancing ORR activity and durability.

Revised: Milestone #5 (Q6): Durability comparison between PVD Pt/NbOx/C, PVD Pt/C, and Pt/C



Partially crystallized nano-carbon particle with proper amount of NbOx and Pt necessary

Milestone #6 (Q7): Large batch PVD catalyst - $> 500 \text{ mW/cm}^2$ at $Q/\Delta T_i < 1.45 \text{ kW/}^\circ\text{C}$, $0.125 \text{ mg}_{\text{PGM}}/\text{cm}^2$, $P_{\text{air,in}} \leq 150 \text{ kPa}$



608 mW/cm^2 for Pt/C; 410 mW/cm^2 for Pt/NbOx/C (0.15 mg/cm^2 , 250 kPa)

Milestones #7-8 (Q8): $< 40\%$ mass activity loss and $< 100 \text{ mV}$ loss at 0.8 A/cm^2 in electrocatalyst cycle ($0.6\text{-}0.95 \text{ V}$); $< 40\%$ mass activity loss and $< 200 \text{ mV}$ loss at 1.5 A/cm^2 in support corrosion cycle ($1\text{-}1.5 \text{ V}$)



Mass activity % loss goal and $< 100 \text{ mV}$ loss @ 0.8 A/cm^2 in electrocatalyst cycle have been shown. (Q7-8)

Support corrosion test on going. (Q7-8)

Go/No-go Decision Point for Budget Period 2 (end of Year 2):

Large batch PVD catalyst, total loading of $\leq 0.125 \text{ mg}_{\text{PGM}}/\text{cm}^2$: **BOL mass activity of $\geq 0.30 \text{ A/mg}_{\text{PGM}}$. Mass activity following electrocatalyst and support cycling $> 0.21 \text{ A/mg}_{\text{PGM}}$ (70% of $0.30 \text{ A/mg}_{\text{PGM}}$).**

Go/no-go decision point met for total loading $0.15 \text{ mg}_{\text{PGM}}/\text{cm}^2$; testing at lower loading is on-going.

Milestones 4 met with line-of-sight to meeting other milestones and GNG

Approach: Budget Period 3 Milestones Conform to Target Tables

Budget Period 3 Milestones: 4/1/2019 – 3/31/2020	
Milestone #10 (Q9): Large batch PVD catalyst - 1) mass activity > 0.40 A/mg _{PGM} and 2) electrocatalyst AST (30K cycles) with < 40% loss in mass activity (RDE)	
Milestone #11 (Q10): Large batch PVD catalyst, ≤ 0.125 mg _{PGM} /cm ² – BOL mass activity of ≥ 0.44 A/mg _{PGM}	
Milestone #12-14 (Q11): Large batch PVD catalyst - > 1,000 mW/cm ² at $Q/\Delta T_i < 1.45$ kW/°C, for 0.125 mg _{PGM} /cm ² , $P_{air,in} \leq 150$ kPa; < 40% mass activity loss and < 30 mV loss at 0.8 A/cm ² in electrocatalyst cycle (0.6-0.95 V); < 40% mass activity loss and < 30 mV loss at 1.5 A/cm ² in support corrosion cycle (1-1.5 V)	
Milestone #15 (Q12): A set of MEAs (6 or more, each with active area ≥ 50 cm ²) is made available for independent testing at a DOE-approved location.	

Any proposed future work is subject to change based on funding levels.

Collaborations

Partner	Project Roles
	<p>Prime, Industry. Responsible for project management (Task 3), XRF measurements for Pt, Nb wt%, RDE testing (Task 1). Support role for MEA fabrication and fuel cell testing (Task 2).</p>
	<p>FFRDC partner. Responsible for production of small batch Pt/NbO_x/C and for transferring lessons learned small batch catalyst optimization to Exothermics for large batch production. Support role for XRF measurements. (Task 1)</p>
	<p>Sub-contractor, Industry. Responsible for production of large batch Pt/NbO_x/C. Can support with BET, PSD characterization. (Task 1)</p>
	<p>Sub-contractor, University. Responsible for TEM and particle size measurements. Can also support with SEM, XPS, and other characterization techniques. (Task 1)</p>
	<p>Sub-contractor, University. Role is to help understand by XAS whether coordination numbers, interatomic distances, and the presence of adsorbates (e.g. –OH) influence performance and durability (Task 1).</p>
	<p>Sub-contractor, Industry. Responsible for MEA fabrication using Pt/NbO_x/C catalysts on the cathode, and for fuel testing (Task 2).</p>

All collaborations are within the DOE Hydrogen and Fuel Cells Program

Accomplishments and Progress: Experimental Design

Strategy: optimize the concentration and morphology of NbOx and Pt on acetylene black (AB) carbon nano-particles:



- ORNL made 6 samples with 1.5, 3 and 6 wt.% NbOx and 20, 30 wt.% Pt.
- Exothermics: two large batch samples with and without NbOx.
- Umicore wet-chemical synthesized sample with comparable PVD Pt.

EXPERIMENTAL DESIGN WITH NOMINAL WT%

	1.5% NbOx	3% NbOx	6% NbOx
20% Pt	ORNL-L-037	ORNL-L-035	ORNL-L-034
30% Pt	ORNL-L-038	ORNL-L-032	ORNL-L-030

Sample Name	Temperature	Nb Deposition: Ar/O2 Ratio	Nb Deposition: Time (hours)	Nb Deposition: Power (W)	Pt Deposition: Time (hours)	Pt Deposition: Power (W)	Pt Loading from XRF (wt.%)	NbOx Loading from XRF (wt.%)
ORNL-L-030	Room (off-gassed to 400C for 12 hours)	10	18.7	150	0.97	140	27.7	6.3
ORNL-L-032	Room (off-gassed to 400C for 12 hours)	10	9.3	150	1	140	30.6	3.82
ORNL-L-034	Room (off-gassed to 400C for 12 hours)	10	18.7	150	0.67	140	18.1	8.0
ORNL-L-035	Room (off-gassed to 400C for 12 hours)	10	5.6	150	0.58	140	21.5	2.72
ORNL-L-037	Room (off-gassed to 400C for 12 hours)	10	3.9	150	0.63	140	21.3	1.77
ORNL-L-038	Room (off-gassed to 400C for 12 hours)	10	3.7	150	1	140	28.2	1.34
EXO 180109*	off-gassed to >200C; sputter at >100C	32.3	8	100	5	300	19.9	1.75
EXO 180920	off-gassed to >200C; sputter at >100C	32.3	8	100	4.5	300	20.0	2.5
EXO 181114	off-gassed to >200C; sputter at >100C	NA	NA	NA	4.5	300	30.2	NA
Umicore [§]	wet chemical commercial	NA	NA	NA	NA	NA	28.3	NA

* Ketjen black carbon

§ high surface area carbon

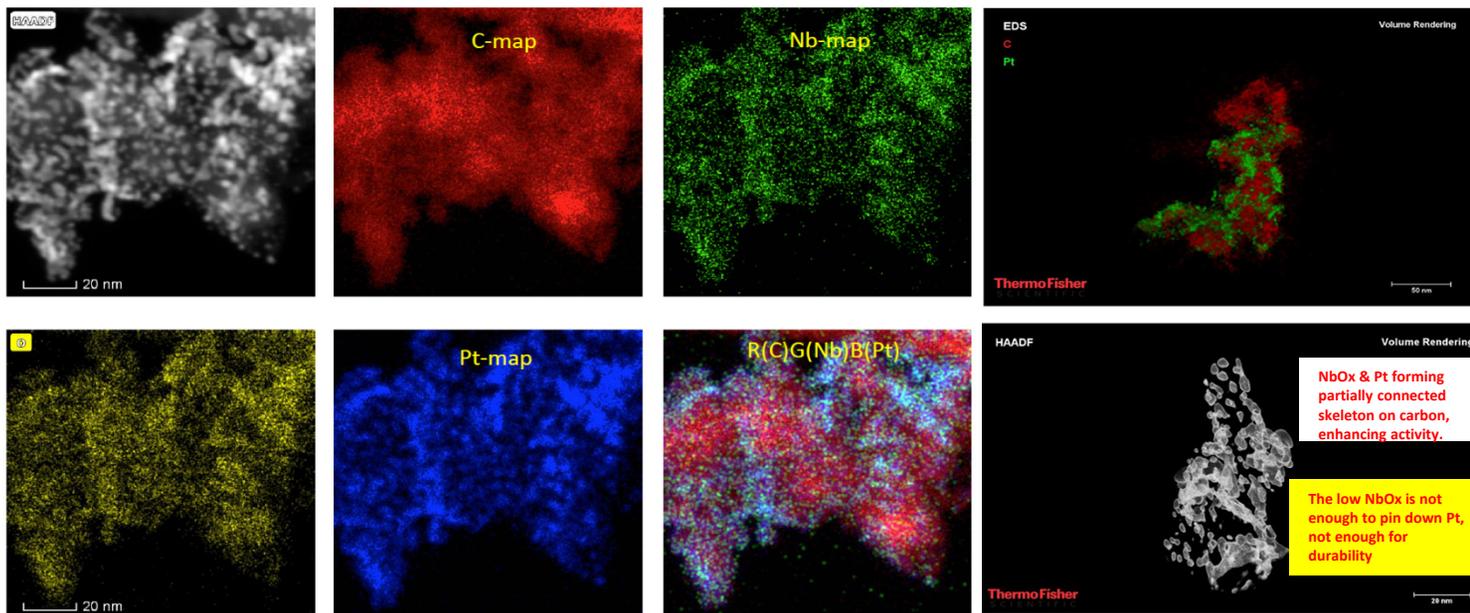
All other samples are supported on acetylene black

Small batch experimental design was used to point toward better Pt, NbOx composition for large batch samples

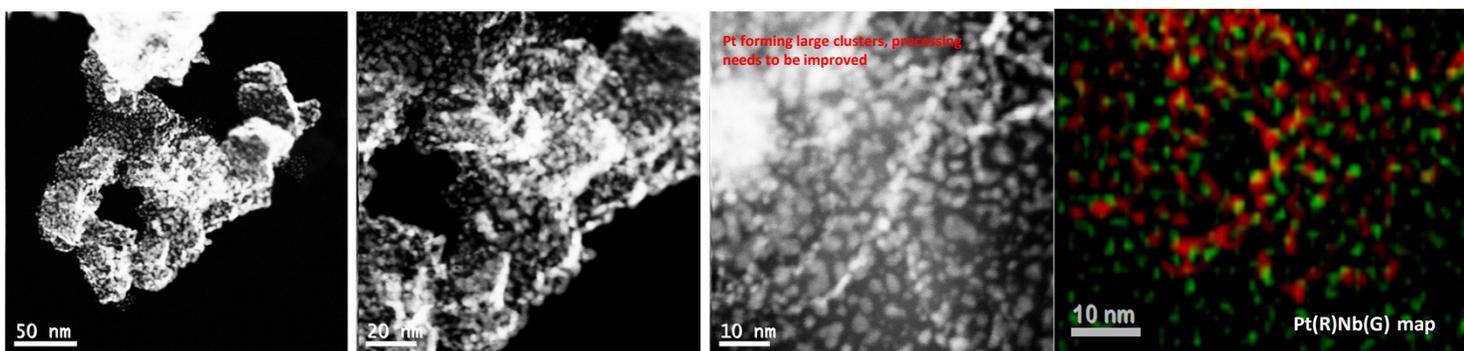
Milestone #4: Microstructural Analysis with TEM & XPS - understanding the morphology of NbOx and Pt



ORNL-L-013 (35.8% Pt, 0.61% NbOx, AB, MA=335 A/cm², MA drop =43%, 0.8 A/cm² voltage drop = 120 mV)



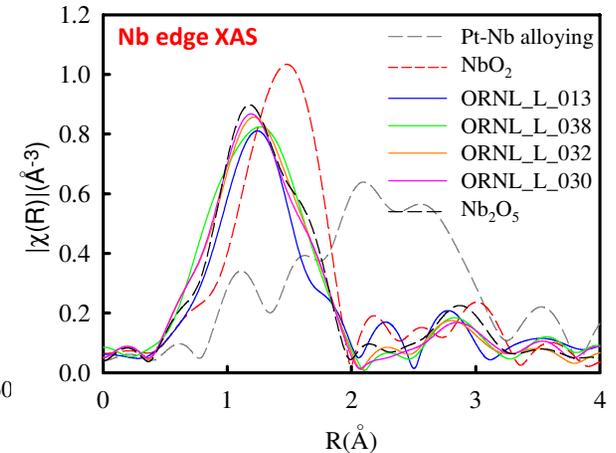
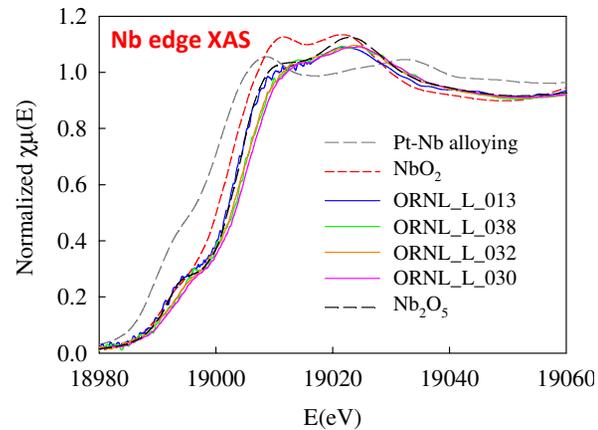
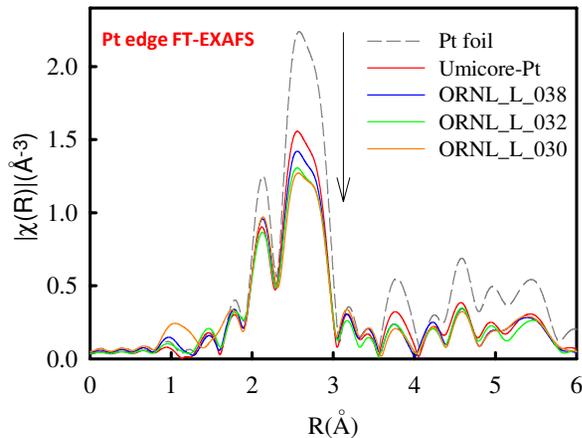
Exothermics 180920 (20.0% Pt, 2.5% NbOx, AB, MA=389 A/cm², MA drop =25%, 0.8 A/cm² voltage drop = 70 mV)



Greater NbOx → uniform distribution of NbOx. Pt forms partially connected network on top & adjacent to NbOx, enhancing ORR activity & durability.

Milestone #4: Electronic Interaction through XAS

NbOx wt%: ORNL-L-013 (0.61%) < ORNL-L-038 (1.34%) < ORNL-L-032 (3.82%) < ORNL-L-030 (6.3%)



- With increasing NbOx content, the intensity of the FT-EXAFS peak decreases.
- The shape of the FT-EXAFS peak does not change much with increasing Nb content, indicating the lack of significant Pt-Nb interactions.

- The Nb XAS signal does not change with Nb content.
- No Nb-Pt interactions are detectable.
- The bulk oxidation state of Nb is nearly saturated close to +5 ($x=4.6$)

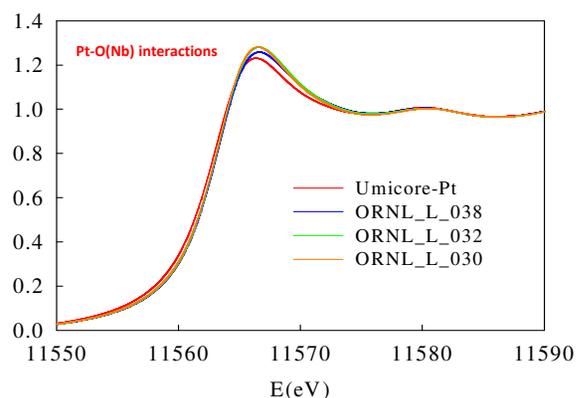
There are no Pt-NbOx interactions



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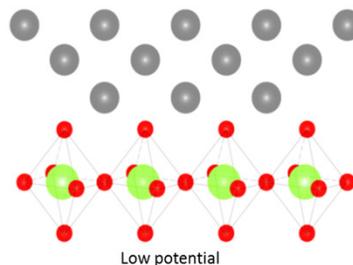
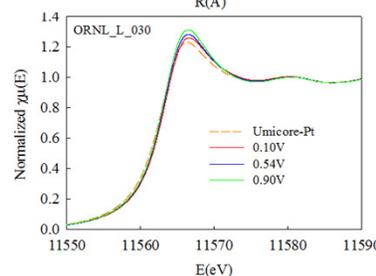
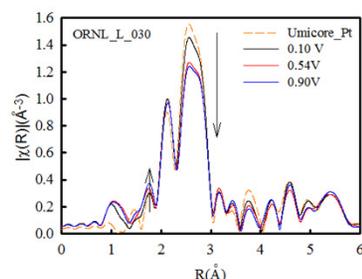
Milestone #4: Electronic Interaction through XAS

NbO_x wt%: ORNL-L-013 (0.61%) < ORNL-L-038 (1.34%) < ORNL-L-032 (3.82%) <

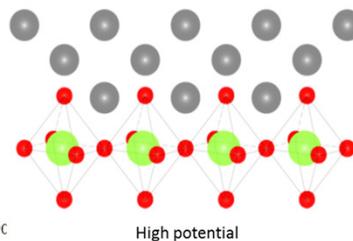


With increasing NbO_x content, the white line intensity of the Pt XANES increases, indicating less number of electrons in the Pt 5d orbitals. This suggests the interaction between Pt and O that comes from NbO_x wherein charge transfers from Pt to O.

ORNL-L-030 (6.3%)



Low potential



High potential

The *in-situ* data suggest the Pt-O interaction throughout the whole potential region, which indicates that Pt is tightly anchored onto NbO_x in contact with O.

The increase in the FT-EXAFS intensity or the coordination number at low potentials indicates that the Pt clusters move away from the substrate at low potentials, which may account partly for degradation.



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NbO_x-induced durability improvement may come from interaction between Pt and O from NbO_x

Milestones #5, 7: Durability Comparison



Electrocatalyst AST (MYRDD P.1) run at EWii with 100 ugPt/cm² on cathode using 50 cm² FCT single cell. Fuel cell mass activities at 0.9 V_{IR-free} and corrected for H₂ x-over.

Sample	Carbon	% Pt	% NbOx	I/C	Anode Loading (ug/cm ²)	RDE BOL Mass Activity (A/gPt)*	FC Mass Activity Loss (%)	FC ECA Loss (%)	Voltage Loss at 0.8 A/cm ² (mV)
Commercial reference	HSA	28.3	0	0.5	50	560	35	40	59
				0.8	50	574	46	70	70
ORNL-L-013	AB	35.8	0.61	0.5	50	409	43	62	120
ORNL-L-019	AB	37.5	0	0.5	50	385	52	55	180
ORNL-L-035	AB	21.5	2.72	0.5	50	287	4	10	160
ORNL-L-032	AB	30.6	3.82	0.5	50	355	1	48	130
ORNL-L-030	AB	27.7	6.3	0.5	50	231	10	3	40
ORNL-L-034	AB	18.1	8.0	0.5	50	144	9	50	71
Exothermics 180109	Ketjen	19.9	1.75	0.5	50	455	39	67	224
Exothermics 180209	Ketjen	26.8	0	0.8	50	440	35	65	180
Exothermics 180920	AB	20.0	2.5	0.5	50	525	14	60	190
				0.8	50	476	33	78	70
				0.8	25	490	15	27	65

* same ink as FC test
AB = acetylene black

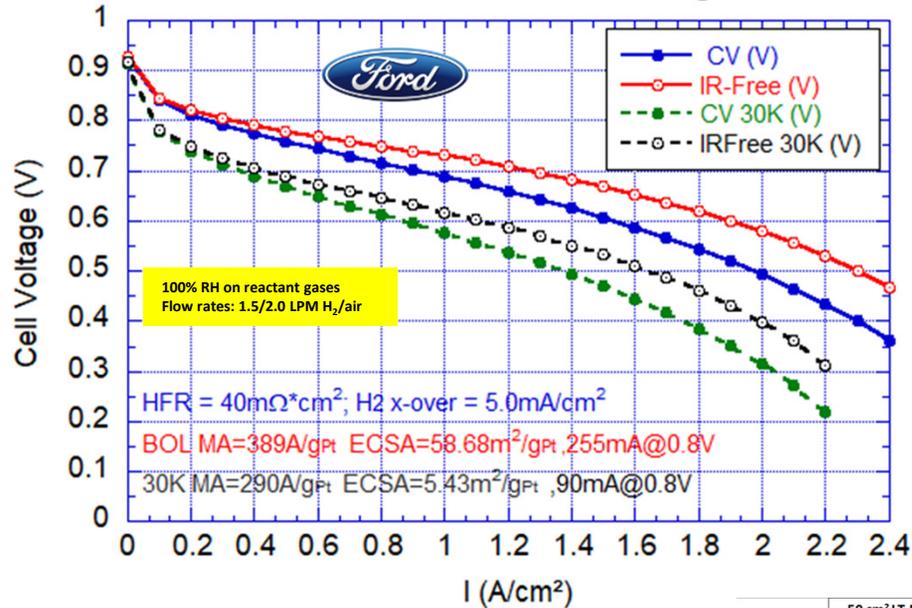
Aligned with final project target

Aligned with Milestone #7

Most Pt/NbOx/C show <40% mass activity loss. Lower V loss shown with ↑ NbOx.

G/NG for Budget Period 2: Electrocatalyst AST

180920 I/C=0.6 Pt/Fcarbon/NbOx with 0.10mg/cm² 80C 150Kpa

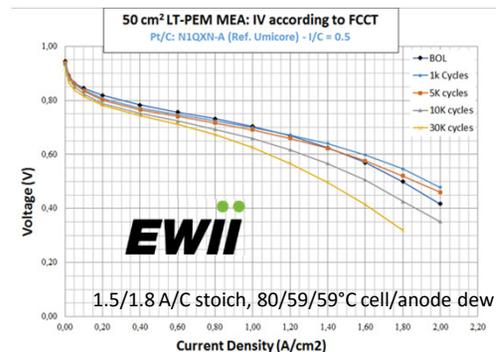


Ford testing: 5 cm² single cell test with Exothermics 180920 at anode / cathode Pt loadings of 0.05/0.1 mg_{Pt}/cm² show samples can meet 2nd go/no-go mass activity target

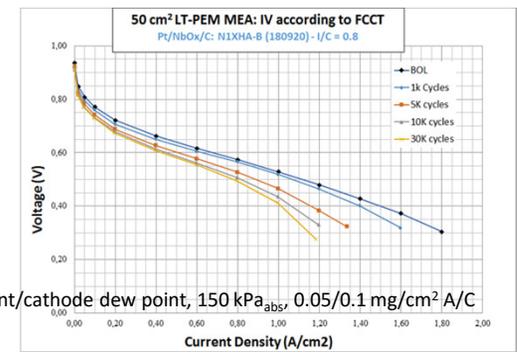
- **BOL mass activity of 389 A/g_{Pt} and EOT mass activity of 290 A/g_{Pt}**
- Need to repeat with 0.025 mg_{Pt}/cm² anode loading

EWii testing:

- During ASTs, mass activity measurements were not performed with proper electrode conditioning – ASTs to be redone
- Lower performance also observed for Pt/NbOx/C polarizations using MYRDD Table P.6 (a.k.a. FCTT method)



number of cycles	Voltage at 0.8 A/cm2	Voltage loss (mV)
0	0,73	0
1000	0,72	8,46
5000	0,72	15,56
10000	0,69	38,66
30000	0,67	58,66



number of cycles	Voltage at 0.8 A/cm2	Voltage loss (mV)
0	0,570	0
1000	0,570	0,00
5000	0,530	40,00
10000	0,510	60,00
30000	0,500	70,00

Ford fuel cell tests show mass activities that align with Budget Period 2 Go/No-Go

Milestone #6: Power Density



Exothermics 180920 (Pt/NbOx/C):

From Ford test (prior slide, differential, 100% RH):

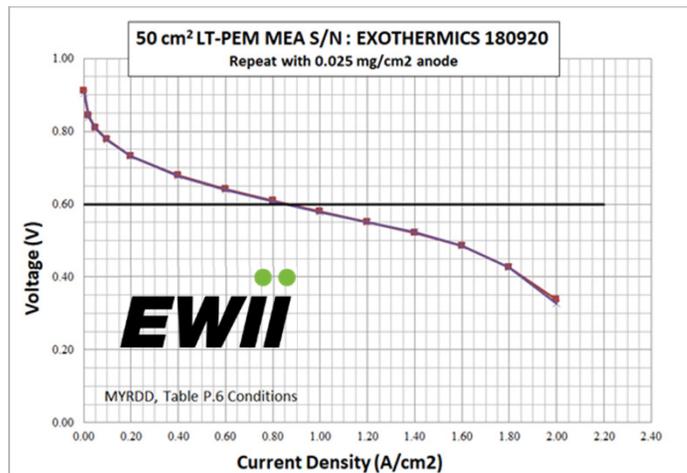
930 mW/cm² at 0.6 V, 80°C ($Q/\Delta T=2.44$ kW/K)

From EWii test (below, MYRDD Table P.6):

538 mW/cm² at 0.6 V, 80°C ($Q/\Delta T=2.44$ kW/K)

From NREL test (MYRDD Table P.6 with 90°C, 250 kPa_{abs}):

410 mW/cm² at 0.692 V ($Q/\Delta T=1.45$ kW/K)



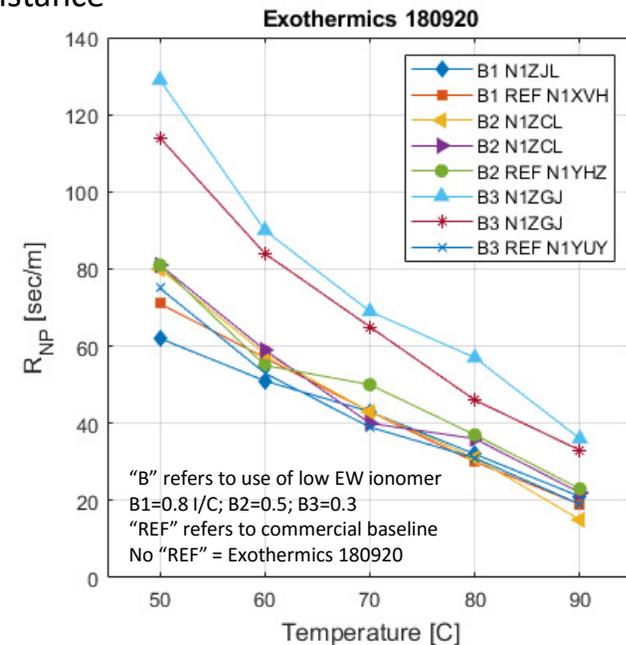
EWii has been focusing on the durability of the catalyst under electrocatalyst AST conditions, did not optimize the initial cell performance.

Exothermics 180209 (Pt/C):

From EWii test (MYRDD Table P.6 with 94°C, 250 kPa_{abs}):

608 mW/cm² at 0.668 V ($Q/\Delta T=1.45$ kW/K)

NREL performed limiting current testing on 3 cm² cell. R_{NP} = non-pressure dependent mass transport resistance



- At higher I/C (0.5 and 0.8), non-pressure dependent mass transport resistance (R_{NP}) is roughly the same for Exothermics 180920 and the commercial standard
- Low I/C catalyst layers: increase in R_{NP} for Exothermics 180920 versus commercial baseline

PVD Pt/C sample tested at $Q/\Delta T=1.45$ kW/K showed >500 mW/cm². Limiting current measurements do not show added resistances from PVD catalyst.

Select Reviewer Comments

- “The effect of Nb on the catalysts is not very clearly illustrated, nor is the parameter space. It is unclear what the catalyst parameters to be varied are.” “More science is necessary; it is important to understand the NbOx coverage and bonding on carbon and how this affects the carbon porosity. The team needs to understand the Pt bonding and how much is on NbOx and on carbon, respectively. The Pt bonding interactions should be measured, and durability measurements should be taken.”
 - Response: The effects of NbOx and Pt concentration are being systematically studied through an experimental design with the small batch preparation system at ORNL. Microstructural analysis by TEM indicates larger Pt sitting adjacent to or on top of pre-deposited NbOx. XPS and XAS confirmed that the Nb valence state is around 4.6 in the NbOx phase, and XAS also confirmed there is a Pt-O interaction between Pt and the O in NbOx, which contributes to the enhanced durability. Electrocatalyst cycling shows enhancement in durability with increased NbOx. Exothermics is making a larger batch of samples following EWii test results on activity and durability.
- “The following additions will help the project understand the quality of PVD-made catalysts better:
 - Performance stability under load cycling and start/stop cycling
 - Structural stability of the catalyst under the ink-making process
 - Understanding of the shelf life of PVD catalysts”
 - Response: Performance stability under load cycling has been performed for electrocatalyst cycling at EWii. Other issues will be addressed in budget period 3 or may be outside the scope of this project.

Future Work

- **Go/No-Go Decision Point for Budget Period 2 (end of Year 2):**

Validation of PVD deposited catalyst powders via MEA BOL Testing with 40-50 cm² single cell having a cathode loading of $\leq 0.125 \text{ mg}_{\text{PGM}}/\text{cm}^2$ giving a **BOL mass activity of $\geq 0.30 \text{ A/mg}_{\text{PGM}}$** at 900 mV_{iR-free} and MA drop of less than 30% after electrocatalyst cycling of 30K cycles, following the protocols given in Table 3.4.7 of FCTO's MYRDD Plan.

- **Actions related to Go/No-Go:**

- EWii: Ink processing & fuel cell testing on candidate samples to obtain mass activity values and AST durability (both support corrosion and electrocatalyst) for both the catalyst & substrate.
 - Mass activity measurements to be done with conditioning same as for measurements prior to GNG1 in 2018.
 - Full examination of voltage losses in kinetic region, and at 0.8 A/cm², including proton conductivity.
- ORNL: Produce samples at higher batch size in Big Chamber so that fuel cell testing can be done.
- Exothermics: Improving deposition conditions to make large batch of catalysts based on ORNL experimental design results.
- Northeastern U: Help test MEAs made by EWii for ORR activity and durability.

- **Beyond Go/No-Go**

- Continue XAS studies to understand source of activity for newer samples
 - Use XAS as function of potential to understand Pt-NbOx interactions
 - Use XAS to observe Nb oxidation state after fuel cell testing
- Work with Strategic Analysis to refine cost estimates on a scaled up Exothermics PVD process
- Work to characterize performance stability for PVD processed catalysts under start/stop cycling, as well as structural stability of the catalyst under ink-making
- Modifying and refining synthesis procedures to achieve homogeneous Pt distribution in Exothermics processed large batch samples.
- Durability: Run support corrosion AST, SU/SD AST, and OCV AST for high performing materials after go/no-go.
- Robustness tests
- Continued measurements on all samples using TEM, XRF, XPS, XAS, RDE

Any proposed future work is subject to change based on funding levels.

Summary

- Go/No-Go still to be confirmed as of early March 2019
 - Best mass activity and durability after electrocatalyst AST has been achieved in cell testing at Ford with anode Pt loading of $0.05 \text{ mg}_{\text{Pt}}/\text{cm}^2$. EWII is working to confirm on 50 cm^2 cell at anode loading of $0.025 \text{ mg}_{\text{Pt}}/\text{cm}^2$.
- Milestone #1-4 has been met, #5, 7-8 mostly met, #6 is being tested.
- Northeastern has shown almost no Pt-Nb interactions (no alloying), but a Pt-O interaction between the Pt and the O in NbOx, and the interaction increases as NbOx concentration increase from 1.5 wt.% to 6 wt.%, contributing to the enhanced durability of the PVD Pt/NbOx/C catalysts. TEM microstructural analysis shows physical evidence that NbOx is homogeneously deposited on top of the carbon powers, and Pt is adjacent to or on top of the pre-deposited NbOx. However, the Pt is prone to clustering during large batch deposition and modification is being made to change the catalyst processing.
- Actions are being taken to address cost analyses (Strategic Analysis), as well as performance stability under start/stop cycling, and structural stability of the catalyst under the ink-making process.

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TECHNICAL BACKUP SLIDES

FC-162

Accomplishments and Progress: GNG 1



- Go/No-Go Decision Point for Budget Period 1 has been passed
 - Four catalysts showed $> 300 \text{ A/g}_{\text{PGM}}$ mass activity

Sample	Composition (AB = acetylene black) (KB = Ketjen black)	Mass Activity (A/g_{Pt})	Test at:	Cell Size (cm^2)	I/C	Cathode / Anode Loadings (mg/cm^2)
ORNL-L-013	35.8% Pt, 0.61% NbOx, AB	335	Ford	5	0.6	0.1/0.05
Exothermics 180109	19.9% Pt, 1.75% NbOx, KB	328	EWII	50	0.8	0.1/0.05
Exothermics 180302	21.5% Pt, 0.23% NbOx, 1.10% Co, KB	309	EWII	50	0.5	0.1/0.05
Exothermics 180308	28.8% Pt, 0.50% NbOx, 1.54% Co, KB	352	Ford	5	0.6	0.1/0.05

Fabrication conditions:

- ORNL-L-013: AB carbon off-gassed at 400°C for 12 hours, followed by Nb sputtering in 10:1 Ar: O_2 environment for 1.5 hours at 150 W. Pt sputtering followed in Ar for 1.5 hours at 140 W.
- Exothermics samples were off-gassed at $>200^\circ\text{C}$, and sputtered at $>100^\circ\text{C}$. Nb sputtering was done in 3% O_2/Ar for 180109 and 180302 (2% for 180308) for 8 hours at 100 W. Pt or PtCo sputtering followed in Ar for 5 hours at 300 W.

Both small and large batch samples have passed the go/no-go criteria

Fuel Cell Testing Results: AST



All anodes comprising 0.05 or 0.025 mg Pt/cm² and all cathodes comprising 0.10 mg Pt/cm²

- MA % loss (A/g Pt) are at 0.9 V iR free and corrected for H₂ x-over
- All AST measured in 50 cm² single-cell hardware (FCT hardware)

Sample	Carbon Support	% Pt	% NbOx	I/C	FC TEST			RDE BOL Mass Activity (A/gPt)	FC TEST			MA (% Loss)	Voltage Loss at 0.8 A/cm ² after 30 K Cycles (pre-ECA, mV)
					BOL ECA _{Avg} (m ² /g _{pt})	EoL ECA _{Avg} (m ² /g _{pt})	BoL ECA @ RT		BOL Mass Activity (A/gPt)	EOL Mass Activity (A/gPt)	ECA (% Loss)		
Exothermics 180109	Ketjen	19.9	1.75	0.5	20.4	6.7	455	107	65	67	39	224	
Exothermics 180209	Ketjen	26.8	0	0.8	23.2	8.1	440	93	60	65	35	180	
Exothermics 180920	Acetylene black	20.0	2.5	0.5	23.6	9.4	525	109	94	60	14	190	
Exothermics 180920	Acetylene black	20.0	2.5	0.8	33.2	7.3	476	73	49	78	33	70	
Exothermics 180920 repeat low PGM	Acetylene black			0.8	27.0	19.7	490	95	81	27	15	65	
Exothermics 181114	Acetylene black	20.0	0	0.5	12.5	-	430	111	-				
Exothermics 181114	Acetylene black	20.0	0	0.8			422						
Commercial reference	High surface area	28.3	0	0.5	25.1	15.0	560	300	196	40	35	59	
Commercial reference	High surface area	28.3	0	0.8	33.0	9.9	574	375	202	70	46	70	
ORNL-L-013	Acetylene black	35.8	0.61	0.5	26.8	10.2	409	113	64	62	43	120	
ORNL-L-019	Acetylene black	37.5	0	0.5	25.1	11.4	385	260	125	55	52	180	
ORNL-L-034	Acetylene black	18.1	8.0	0.5	30.9	15.4	144	58	53	50	9	71	
ORNL-L-030	Acetylene black	27.7	6.3	0.5	12.9	12.5	231	92	83	3	10	40	
ORNL-L-032	Acetylene black	30.6	3.82	0.5	19.1	10.0	355	74	73	48	1	130	
ORNL-L-035	Acetylene black	21.5	2.72	0.5	19.5	17.5	287	57	55	10	4	160	

