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Novel Bifunctional Electrocatalysts, Supports and Membranes for High Performing and Durable Unitized Regenerative Fuel Cells

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Proton/NEL: Chris Capuano, Kathy Ayers

Ballard Power Systems: Dustin Binny, Shanna Knights

Pajarito Powder: Alexy Serov, Barr Zulevi

04/29/20

Overview



Timeline

- Project Start Date: 07/01/2019
- Project End Date: 06/30/2021
- Percent complete: 40%

Budget

- Total Project Budget: \$ 1,250K
 - Total Recipient Share: \$ 250K
 - Total Federal Share: \$ 1,000K
 - Total DOE Funds Spent*: \$ 183,951

* As of 04/28/20

Partners

- Project lead: Danilovic (LBNL)
- Subs:
 - NEL
 - WUSTL
 - Ballard Power Systems
 - Pajarito Powder

Barriers

- Barriers addressed
 - No *regenerative* fuel cell specific barriers, optimization between fuel cell and electrolyzer barriers:
 - Fuel cells
 - Catalyst, Catalyst support and Membrane electrode assembly:
A: Durability; B: Cost; C: Performance
 - Hydrogen Production
 - Catalyst, Catalyst support and Membrane electrode assembly:
F: Capital cost; G: System efficiency and electricity cost

Relevance - Objectives

- The main focus of this project is to demonstrate a highly efficient and stable unitized regenerative fuel (URFC) achieved *through novel membrane, advanced bi-functional electrode and critical hardware optimization*

Project Targets

Specification	Baseline FC	Baseline Electrolyzer	Baseline URFC	Proposed URFC
Membrane thickness (microns)	25	90	125-175	15-30
Total cell Pt loading (mg/cm²)	0.3	1	6	0.55
Ir catalyst loading (mg/cm²)	n/a	2	4	0.6
Fuel cell stack efficiency (%)	>40% (@2 A/cm ²)	n/a	<40%	>63% (@1 A/cm²)
Electrolysis stack efficiency (%)	n/a	>60% (@2 A/cm ²)	<55%	>80% (@1 A/cm²)
Round trip efficiency (%)	n/a	n/a	<25%	>50% (@1 A/cm²)
Durability	5,000 auto 28,000 bus	50,000 steady state	n/a	100 hrs cycling URFC 1000 hrs discrete < 100 ∞V/hr

Approach

WUSTL

Develop membrane/ionomer and catalyst supports

- Develop and characterize membrane and ionomer for URFC operation
- Develop and characterize engineered catalyst supports

LBNL/Pajarito Powder

Integrate WUSTL membrane into MEA, integrate bifunctional catalyst onto WUSTL supports

- Deposit bifunctional catalyst onto supports and evaluate properties
- Fabricate and test MEAs at up to 25cm² under discrete and URFC conditions
- Determine if support interactions stabilize bifunctional catalyst

Approach

NEL

Demonstrate MEA performance and durability in electrolysis testing at $>50 \text{ cm}^2$

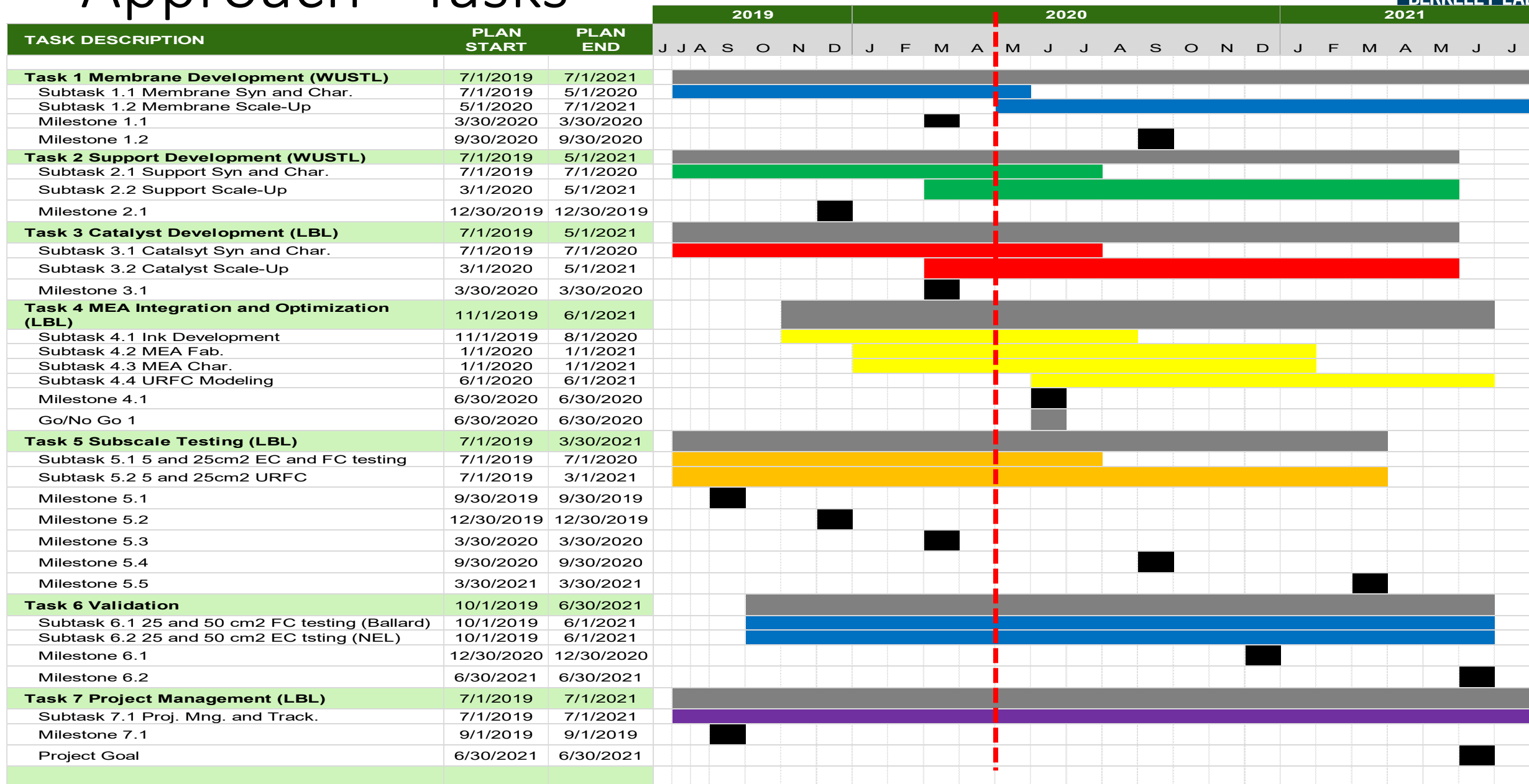
- Baseline performance of SOA membrane and catalysts
- Evaluate WUSTL membrane properties
- Fabricate MEAs from WUSTL materials
- Demonstrate $> 100\text{hrs}$ durability at 50 cm^2

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Demonstrate MEA performance and durability in fuel cell testing at $>50 \text{ cm}^2$

- Baseline performance of SOA membrane and catalysts
- Evaluate WUSTL catalyst support durability
- Fabricate MEAs from WUSTL materials
- Demonstrate $> 100\text{hrs}$ durability at 50 cm^2

Approach - Tasks



Approach - Milestones

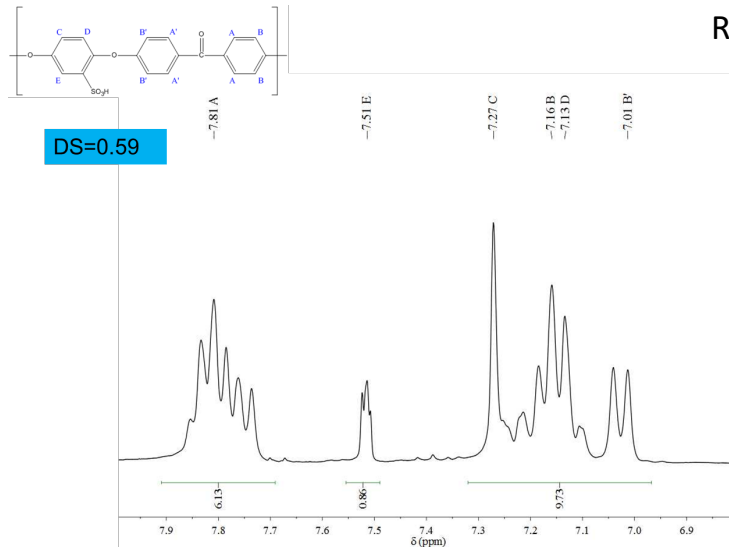


Task Number	Task	Milestone Or Go/No Go Decision Type	Milestone Description (Go/No-Go Criteria)	Anticipated Dates	Responsible	Status
5.1	5 and 25cm ² EC and FC testing	Milestone 5.1	Advanced baseline supported bifunctional catalyst from Pajarito Powder characterized via physical and electrochemical techniques	9/30/2019	LBL	Complete
2.1	Support Synthesis	Milestone 2.1	Synthesize and characterize 3 proposed supports and deliver to Pajarito Powder for catalyst integration	12/31/2019	WUSTL	Complete
5.1	5 and 25cm ² EC and FC testing	Milestone 5.2	Advanced Baseline MEA baselined under MEA experimental conditions in discrete fuel cell (OER/ORR) and electrolysis (HER/HOR) modes at LBL, Proton and Ballard in order to standardize performance	12/31/2019	LBL	Complete
1.1	Membrane Syn and Char	Milestone 1.1	Synthesize, cast and characterize 3 proposed membrane concepts and down-select for MEA integration	3/31/2020	WUSTL	Complete
3.1	Catalyst Syn. and Char.	Milestone 3.1	Synthesize and integrate Pt-Ir catalyst onto WUSTL supports, validated with MEA or RDE testing	3/31/2020	Pajarito	Complete
5.1	5 and 25cm ² EC and FC testing	Milestone 5.3	Pt, Ir and Pt-Ir alloy catalysts baselined under RDE and MEA experimental conditions in discrete fuel cell (ORR) and electrolysis (OER) modes at LBL	3/31/2020	LBL	Complete
4.2	MEA Fabrication	Milestone 4.1	Down-select membrane and ionomer, and supported catalyst based on MEA performance at the 25 cm ² level, showing potential to meet 1A/cm ² currents at >43% round trip efficiency	6/30/2020	LBL	On track
	MEA Fabrication	Go/No Go 1	Demonstrate URFC MEA performance at the 25 cm ² level, of 1A/cm ² current at > 43% round trip efficiency at 80 oC and 20 PSI balanced pressure in PEMFC mode and 20 PSI differential pressure in PEMWE mode	6/30/2020	LBL	On track

Accomplishments and Progress – Milestone 1.1

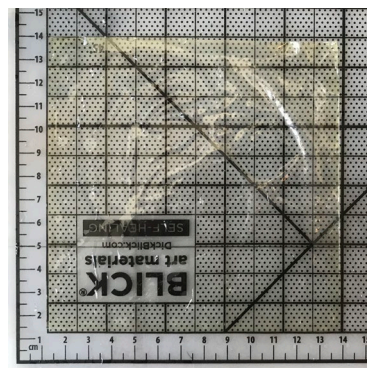
- Two membranes were synthesized, SPEEK and PTFE reinforced SPEEK

Membrane	Proton conductivity, mS/cm @ 60°C	Thickness, μm	Uniformity, %	DF value	Theoretical IEC, mmol/g
SPEEK	124.9 ± 2.7	26.1 ± 2.61	90	0.59	1.75
Reinforced SPEEK	100.6 ± 10.6	24.7 ± 2.42	90	N/A	N/A

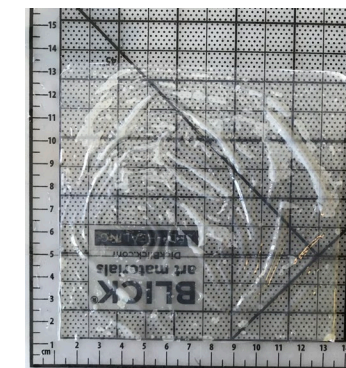


Reinforced SPEEK: using poly(tetrafluoroethylene) (PTFE) as the porous polymeric support

SPEEK



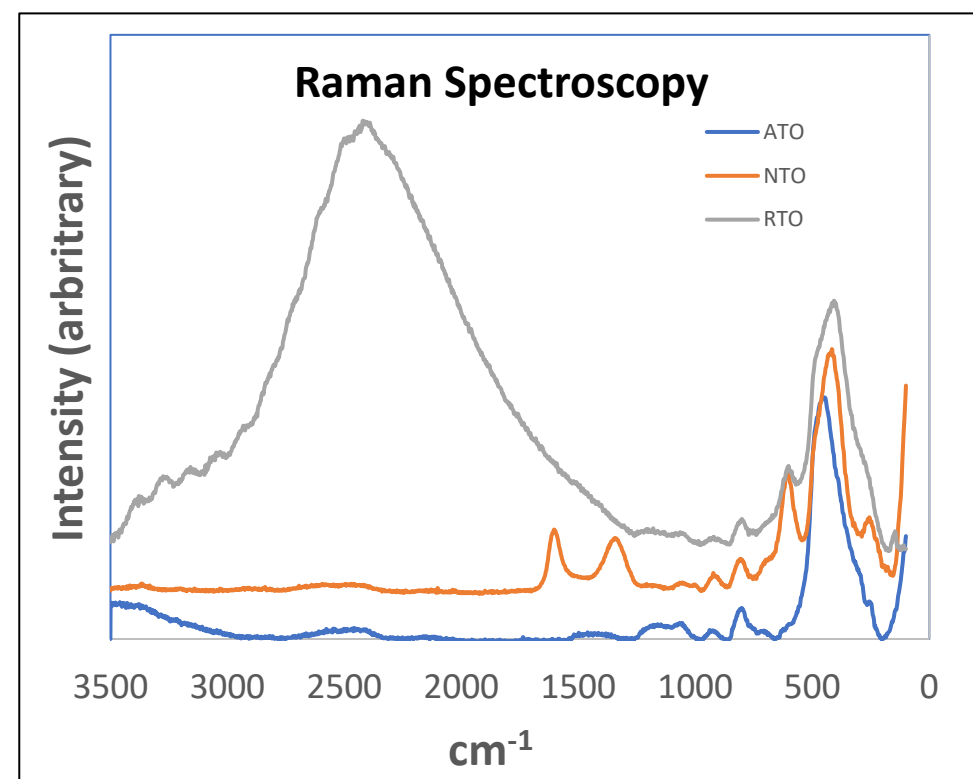
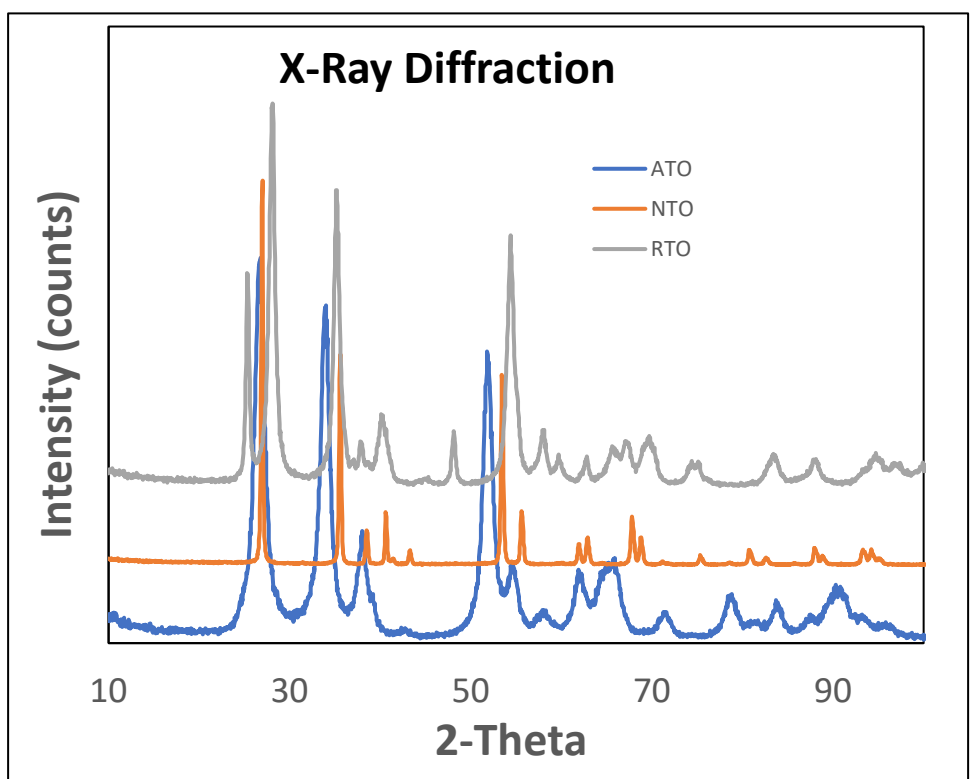
Reinforced SPEEK



NMR spectra of SPEEK

Accomplishments and Progress – Milestones 2.1 and 5.3

- WUSTL supports: Ru-doped TiO₂, Nb-doped TiO₂, Sb-doped SnO₂
- Delivered to PP and started integration of Pt and Ir onto supports

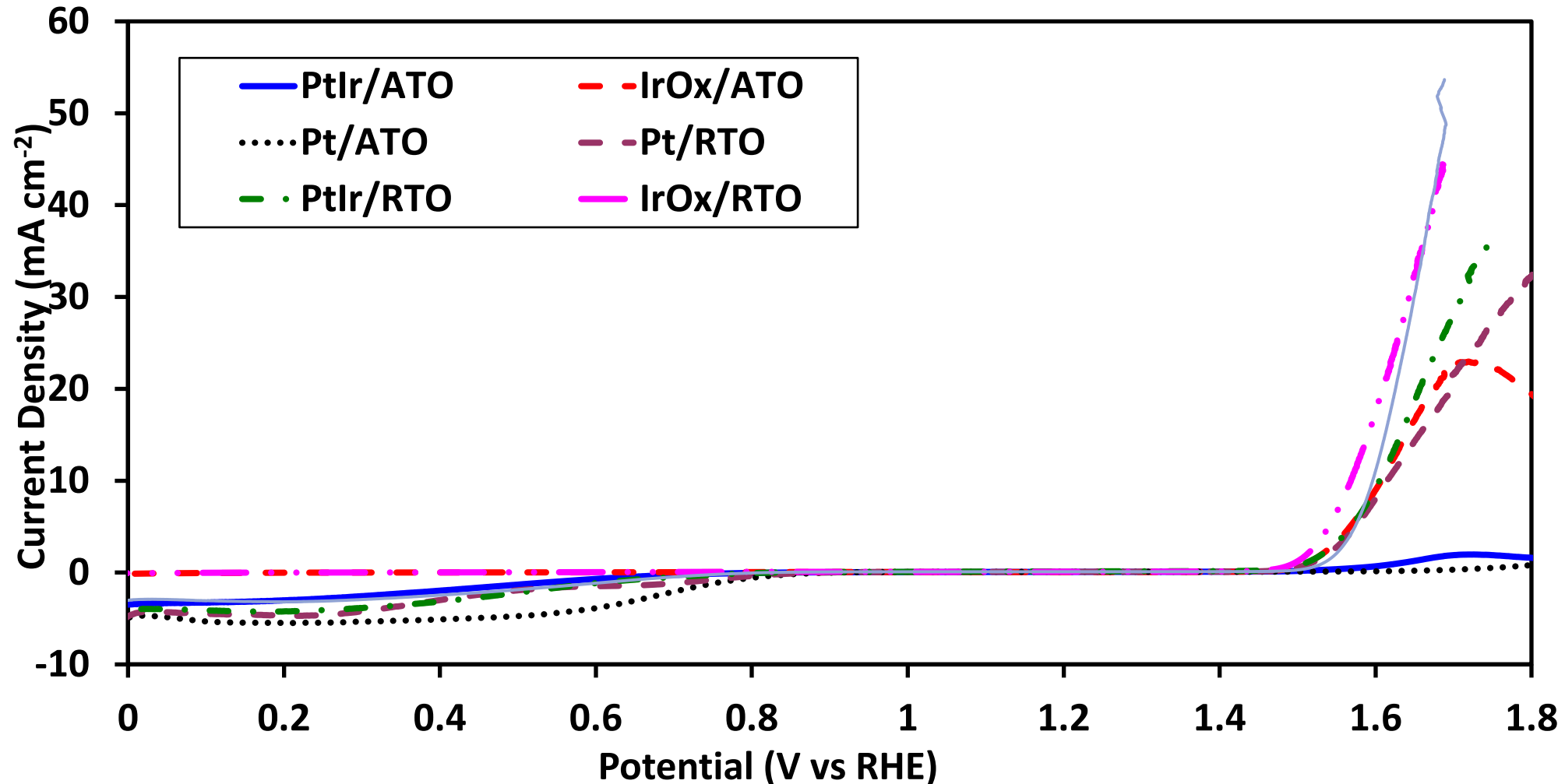


	Zeta Potential, IPA	BET
ATO	-0.56	56m ² /g
NTO	-17.53	13m ² /g
RTO	-0.72	36m ² /g

WUSTL supports:
 Ru doped TiO₂ (RTO)
 Nb doped TiO₂ (NTO)
 Sb doped SnO₂ (ATO)

Accomplishments and Progress-Milestones 2.1, 3.1 and 5.3

- WUSTL supports: Ru-doped TiO_2 , Nb-doped TiO_2 , Sb-doped SnO_2
- Pt, Ir and Pt/Ir coated supports evaluated for ORR and OER activity
- Hard to obtain good ORR performance > focus on IrOx-Pt to elucidate



Accomplishments and Progress-Milestones 2.1, 3.1 and 5.3



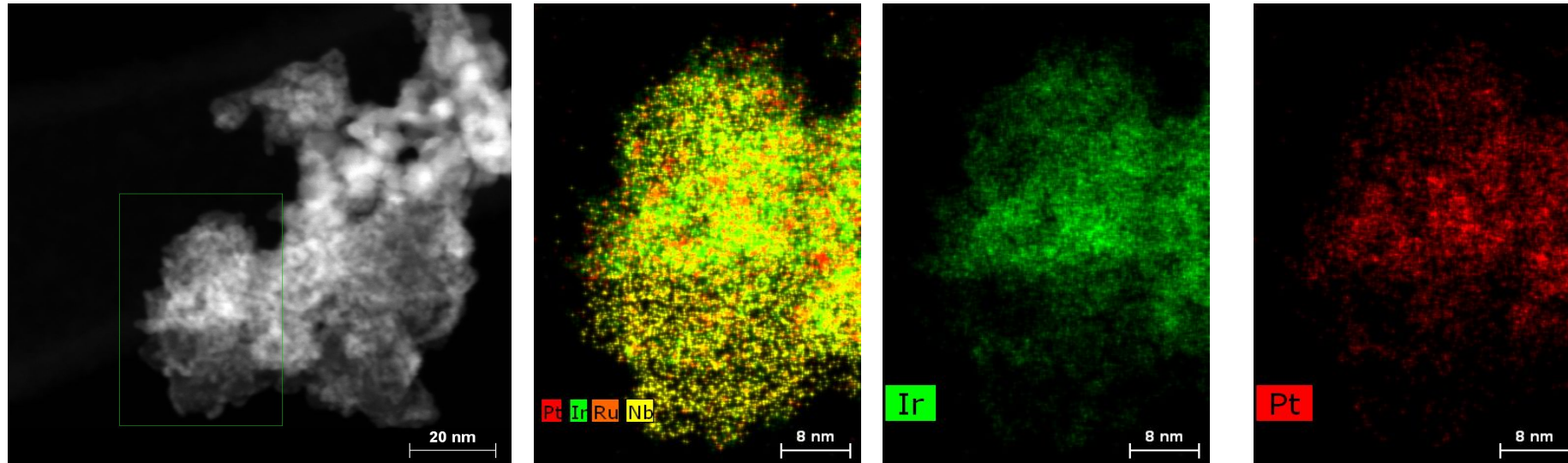
- Summary of RDE tests of WUSTL supported Pt, Ir and Pt/Ir



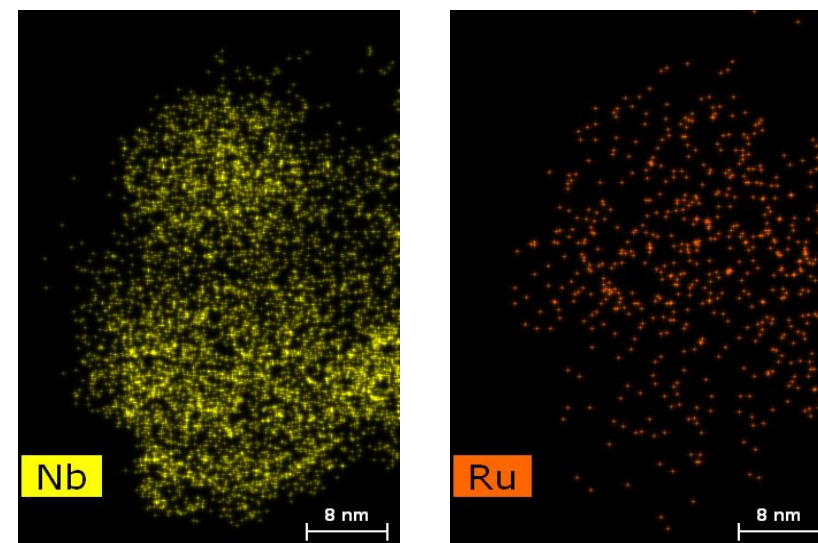
Materials	Pt/ATO	Pt/RTO	PtIr/ATO	PtIr/RTO	IrOx/ATO	IrOx/RTO
ECSA (m ² /g)	4.9	20	7.8	8.3	-	-
OER onset (V vs RHE) before, after	1.9, 2.99	1.5, 1.5	-, -	1.5, 1.5	No OER	1.47, 1.47
η (V@10 mA cm ⁻²) before, after	-, -	1.62, 1.65	-, -	1.61, 1.65	No OER	1.57, 1.62
ORR onset (V vs RHE) before, after	0.86, 0.79	0.77, 0.72	0.73, 0.68	0.75, 0.60	-0.21, -0.05	NA
η (V@-3 mA cm ⁻²) before, after	0.65, 0.46	0.39, 0.02	0.19, 0.07	0.41, 0.22	-0.23, -0.23	NA

Accomplishments and Progress – Milestone 5.1

- PP produced PtIr supported by Ru doped NbO_x (PtIr/RuNbO_x) as an alternative supported catalyst

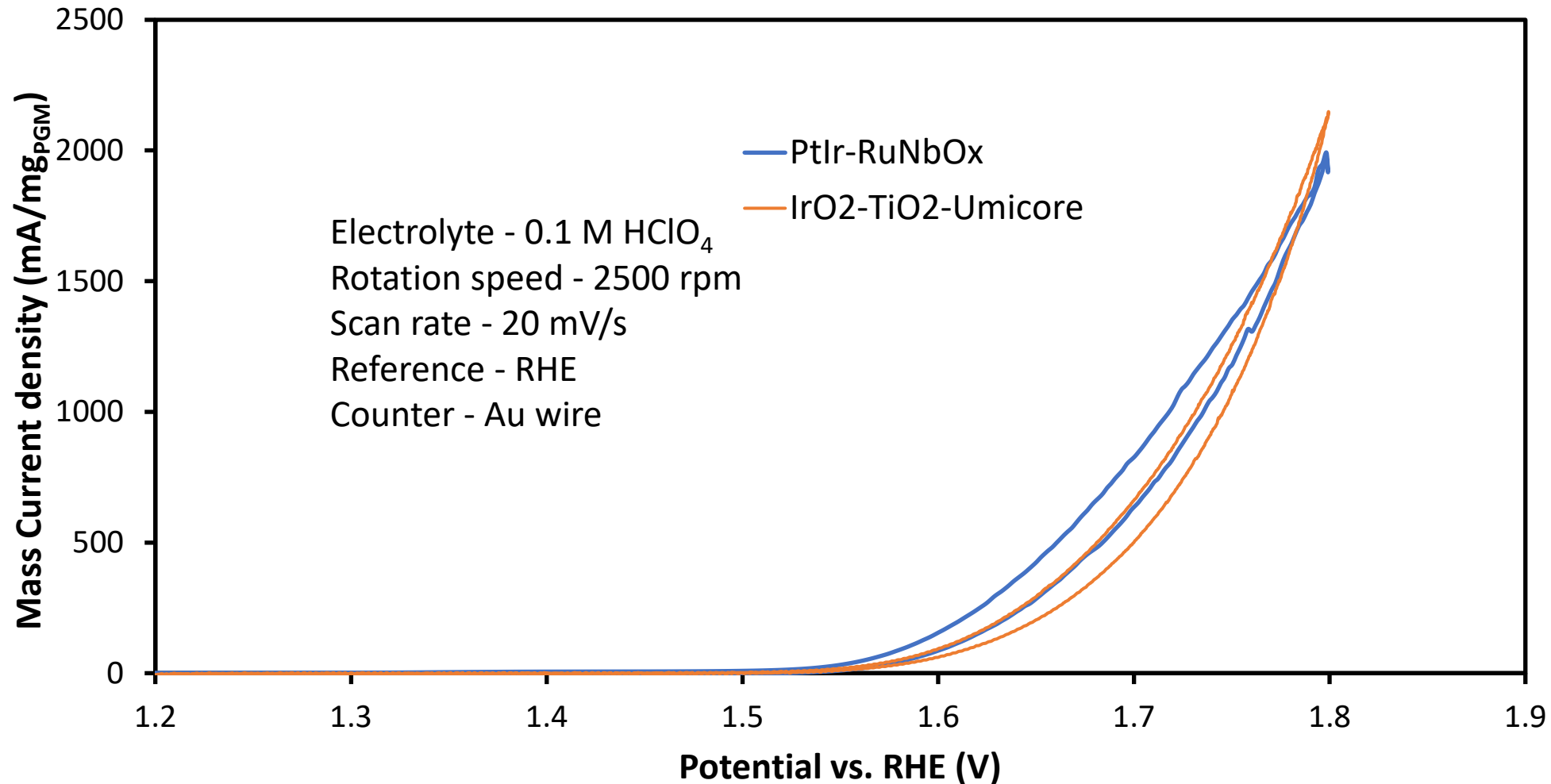


Element	series	Net	[wt.%]	[norm. wt.%]	[norm. at.%]
Platinum	L-series	6490	13.2	13.2	10.6
Iridium	L-series	32783	65.2	65.1	53.0
Ruthenium	L-series	1109	1.4	1.4	2.2
Niobium	K-series	3926	20.2	20.3	34.2
		Sum:	100	100	100



Accomplishments and Progress – Milestone 5.1

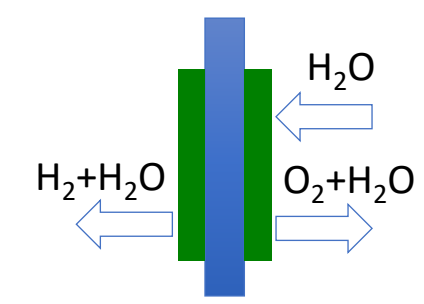
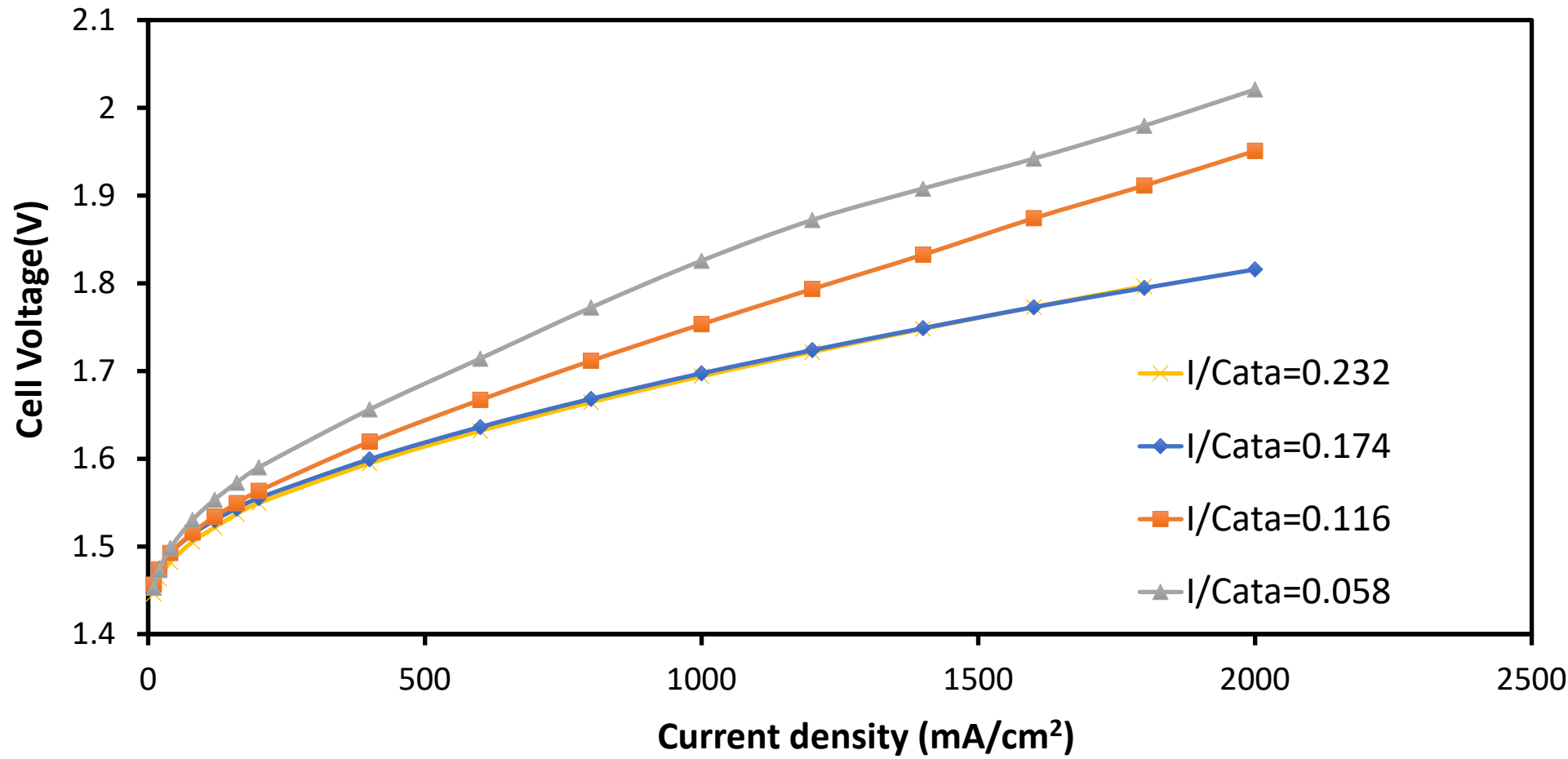
- PtIr/RuNbO_x RDE Testing for OER
- Once again poor ORR performance was found



Accomplishments and Progress – Milestone 5.1



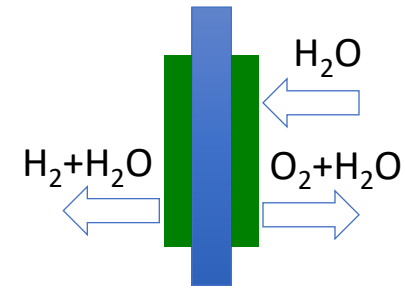
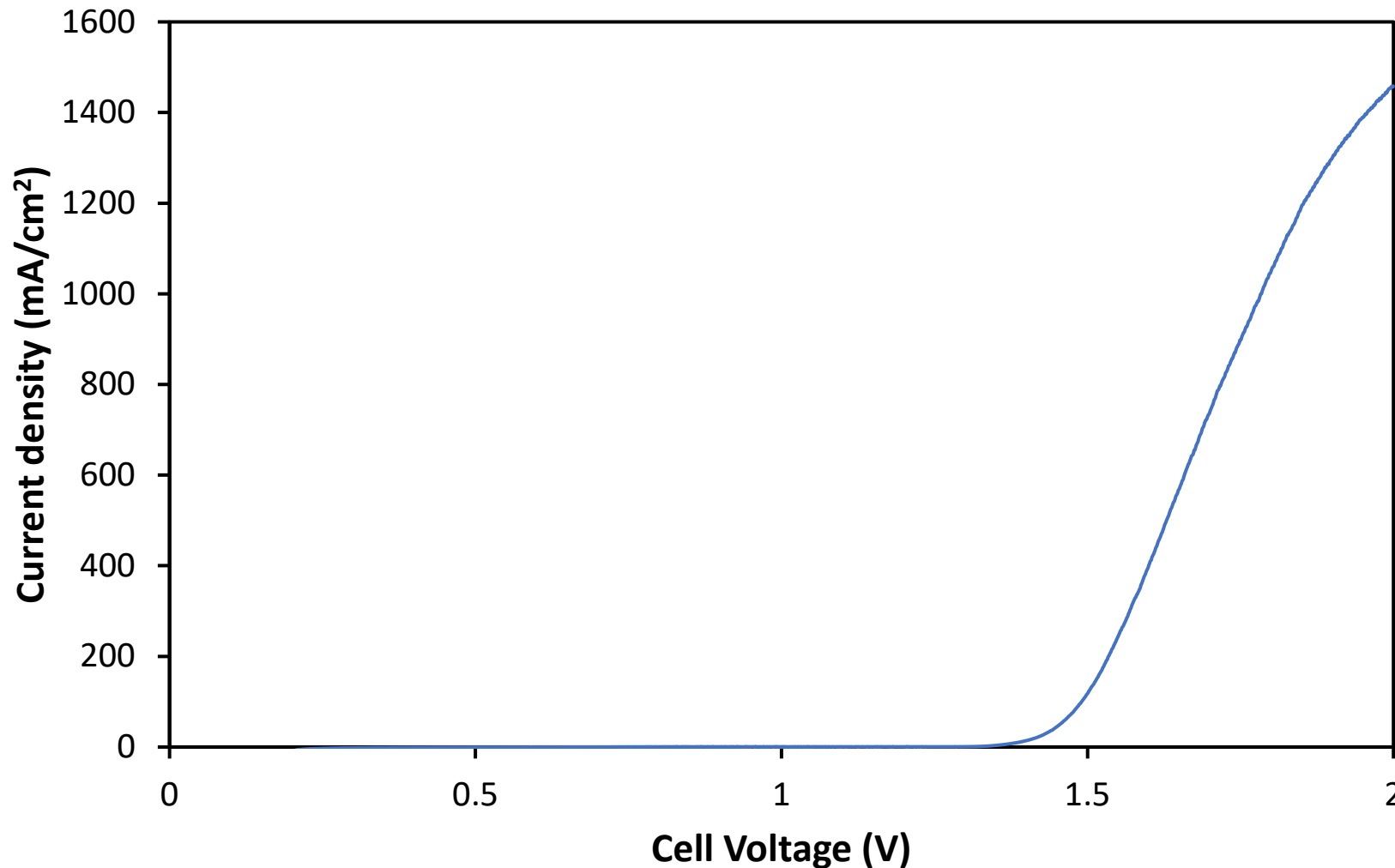
- PtIr/RuNbO_x MEA Testing for PEMEC



- While performance translation from RDE to MEA is encouraging Pt-Ir/MO_x based supported electrocatalysts underperform in URFC relative to Pt and Ir black

Accomplishments and Progress – Milestone 3.1

- Alternative approach to use IrO_x as support for Pt
- To get a better handle on improving ORR activity
- Use as model system to help move forward on down-selected supports

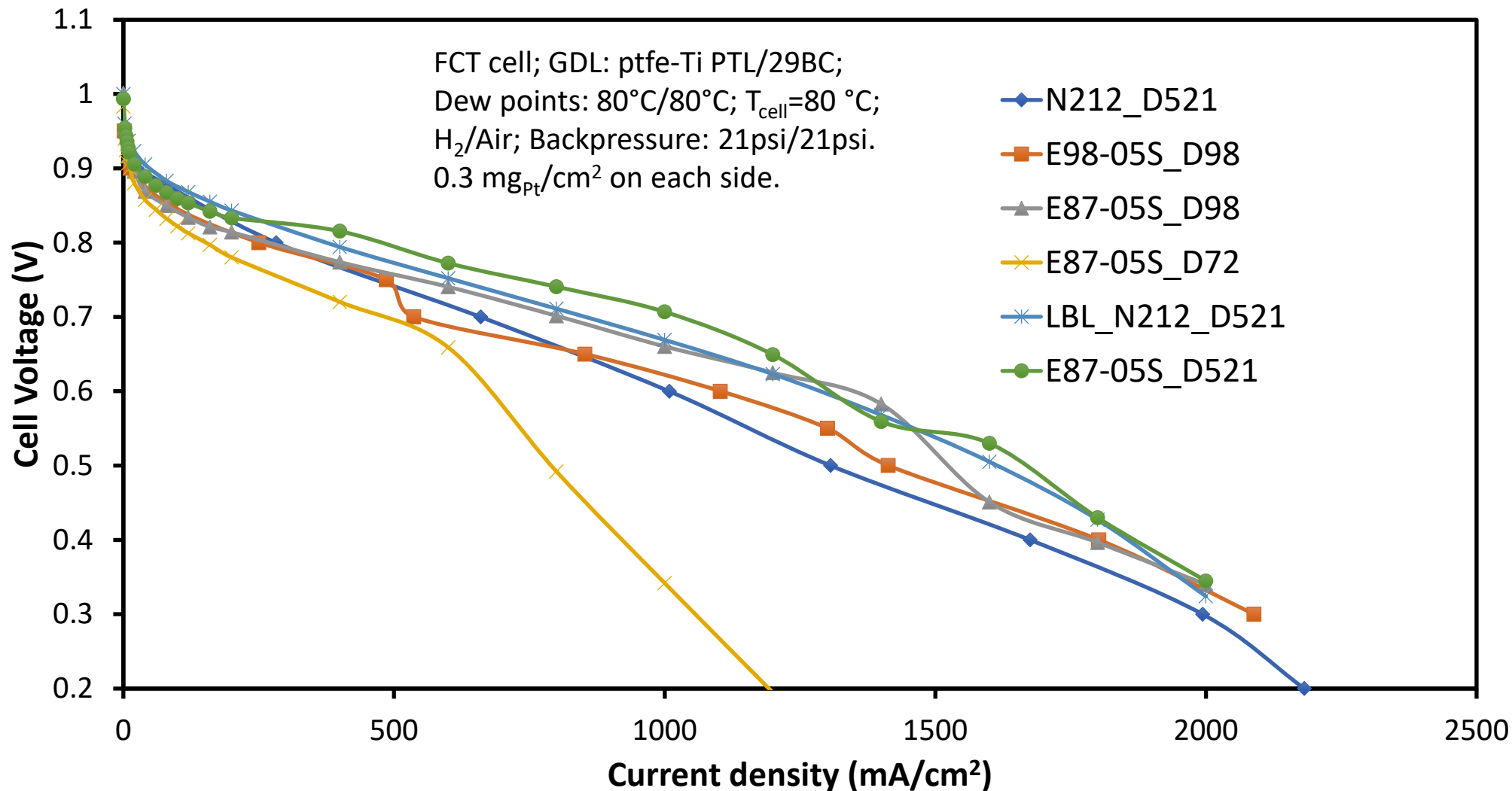
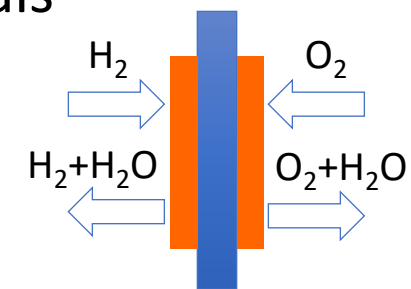


MEA performance of PtIrO_x:

1. Decent PEMEC performance
2. Low PEMFC performance

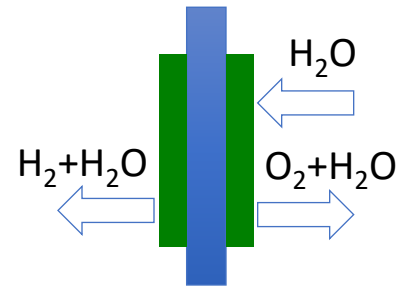
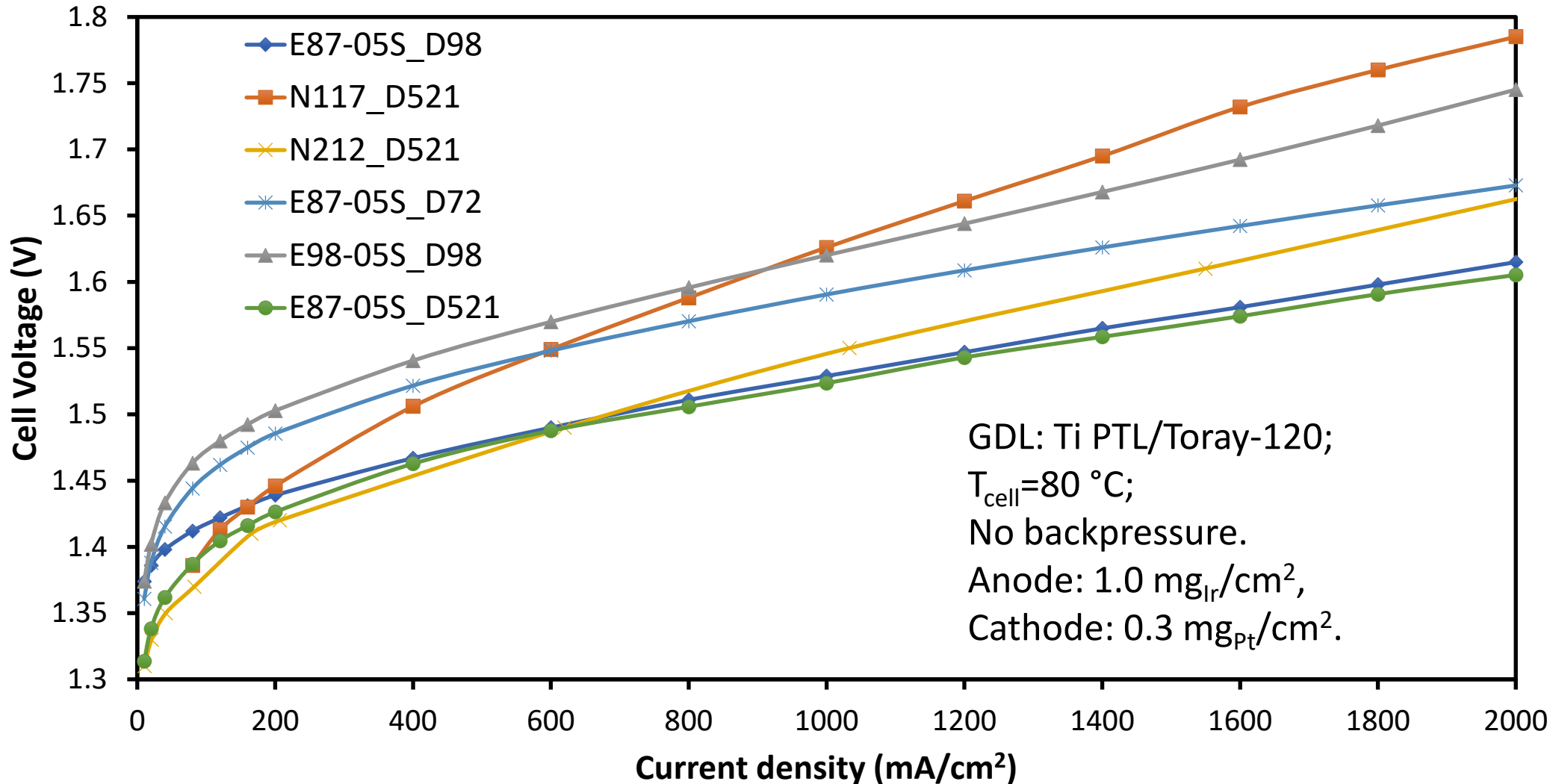
Accomplishments and Progress – Milestone 4.1

- System optimization to achieve better performance using commercial materials
- Membrane & Ionomer down selection at discrete mode PEMFC



Accomplishments and Progress – Milestone 4.1

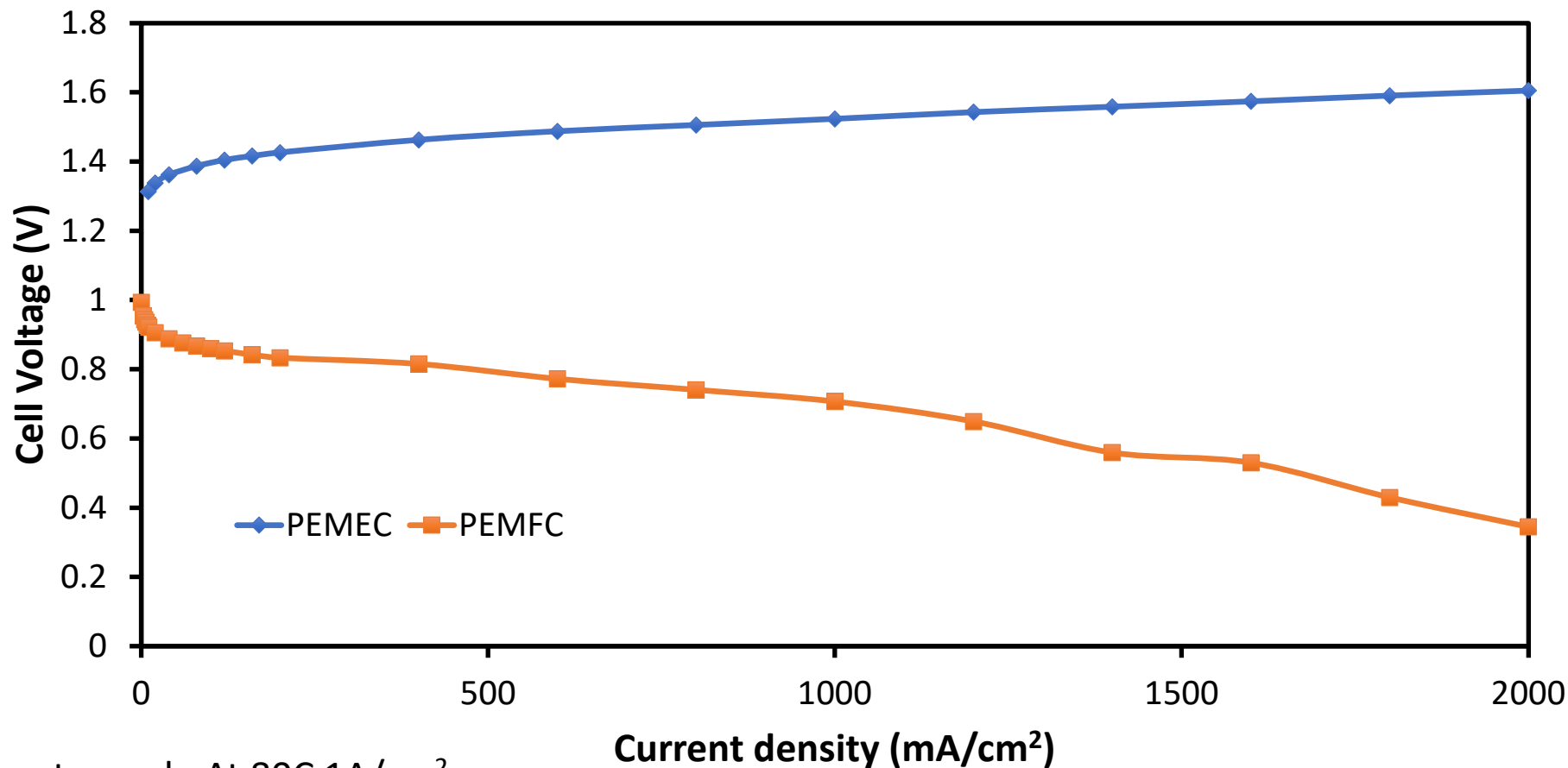
- Membrane & Ionomer down selection at discrete mode PEMEC



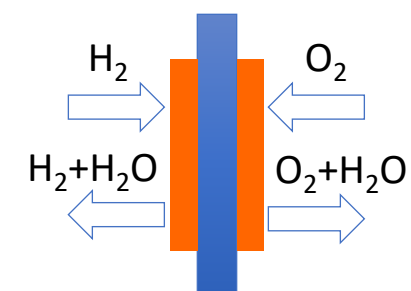
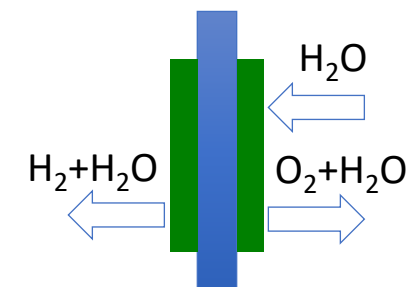
Accomplishments and Progress – Milestone 4.1



- Discrete mode URFC round-trip efficiency
- Materials set from slide 14 used going forward



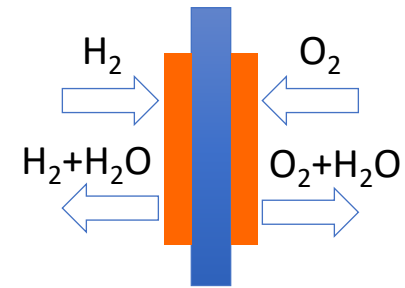
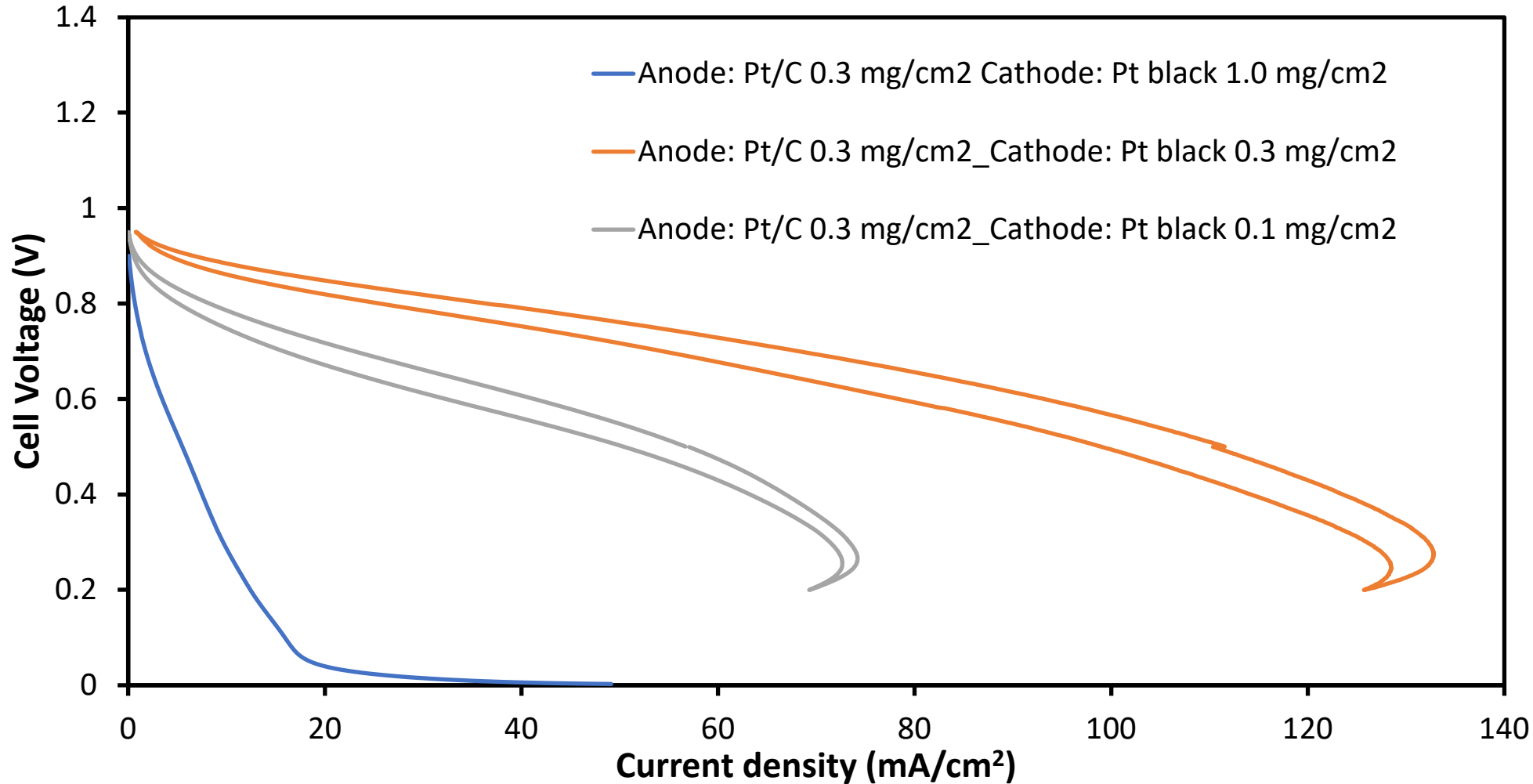
Discrete mode At 80C 1A/cm²:
PEMFC Efficiency: 57.5%;
PEMEC Efficiency: 97.2%
URFC: ~56%



Unitized Mode!!!
Year 1 target:
43%

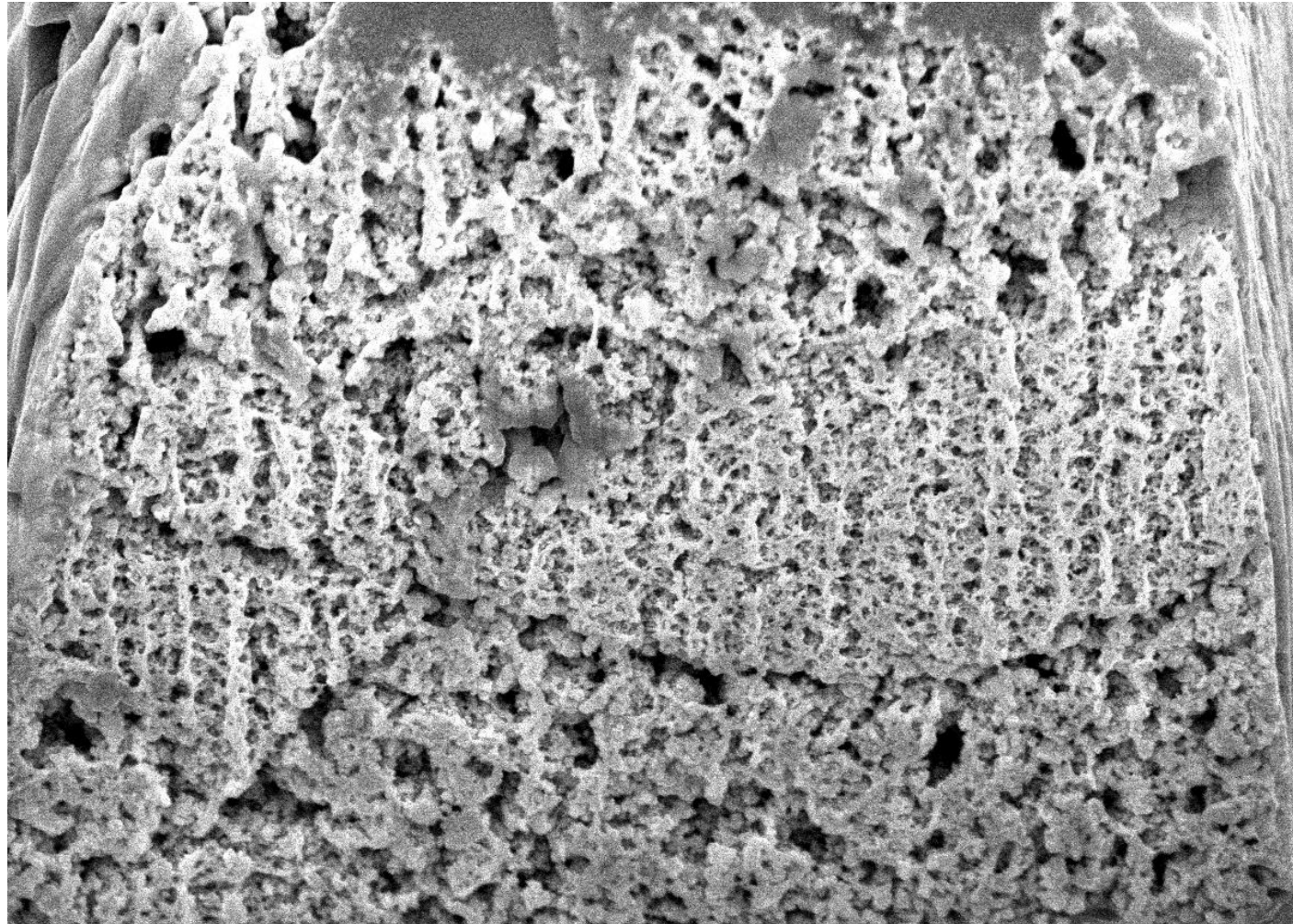
Accomplishments and Progress - MEA Integration

- Performance limitation using Pt black as cathode for PEMFC

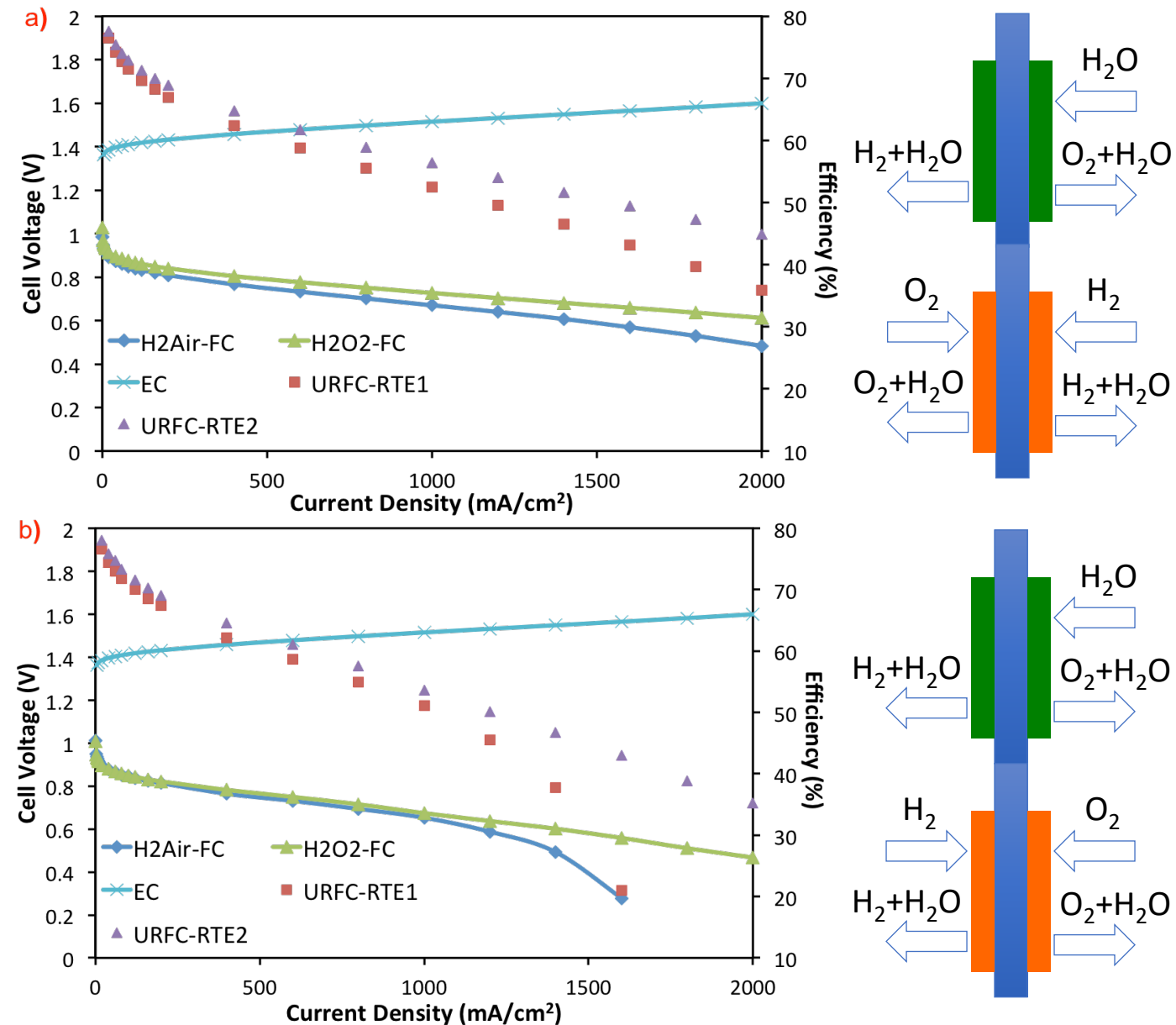


Accomplishments and Progress - Electrode Characterization

- Creating more porous catalyst layer just using un-supported catalyst



Accomplishments and Progress - MEA Integration in URFCs

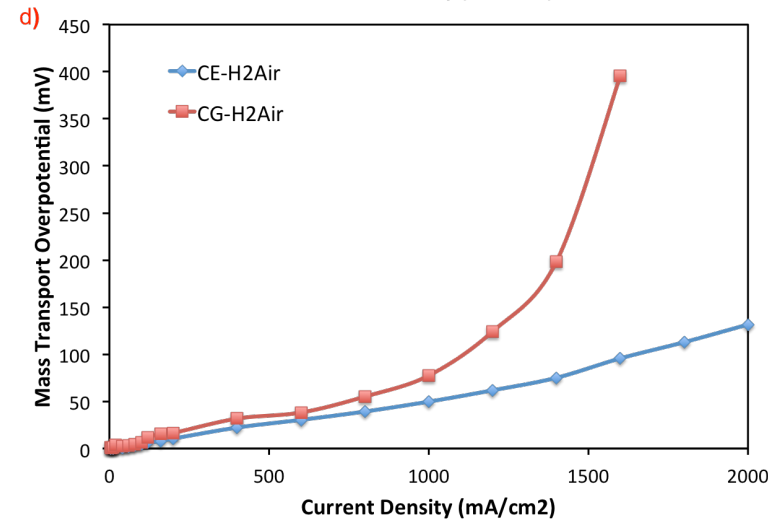
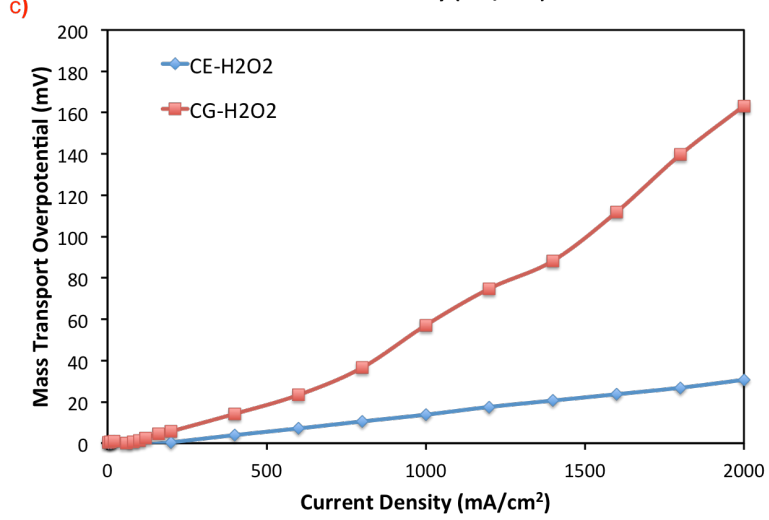
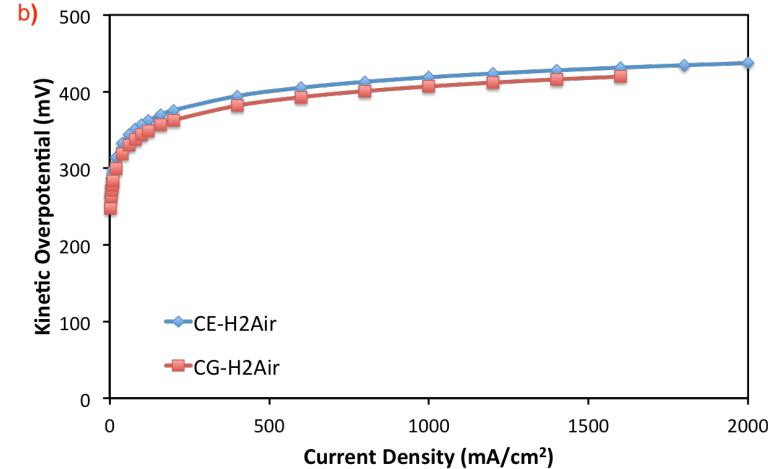
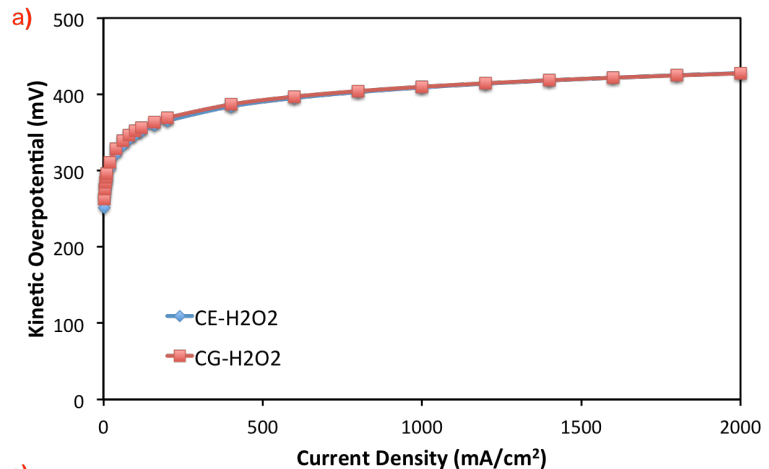


a), b) URFC polarization curves and RTEs evaluation using Pt&Ir black CCM under CE and CG mode, respectively; URFC-RTE1 and URFC-RTE2 are calculated when air and oxygen are used as oxidant at fuel cell mode, respectively. Nafion 212 was selected as membrane for all tests. Cells were operated at 80 ° C. Data was presented without *iR* correction.

5 cm² MEA @ 1A cm⁻²

Mode	H ₂ /O ₂	H ₂ /Air
CE-URFC	56.4%	52.3%
CG-URFC	53.6%	51.1%

Accomplishments and Progress - Performance Analysis



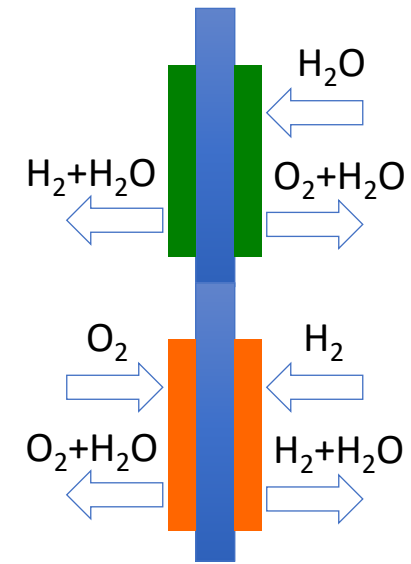
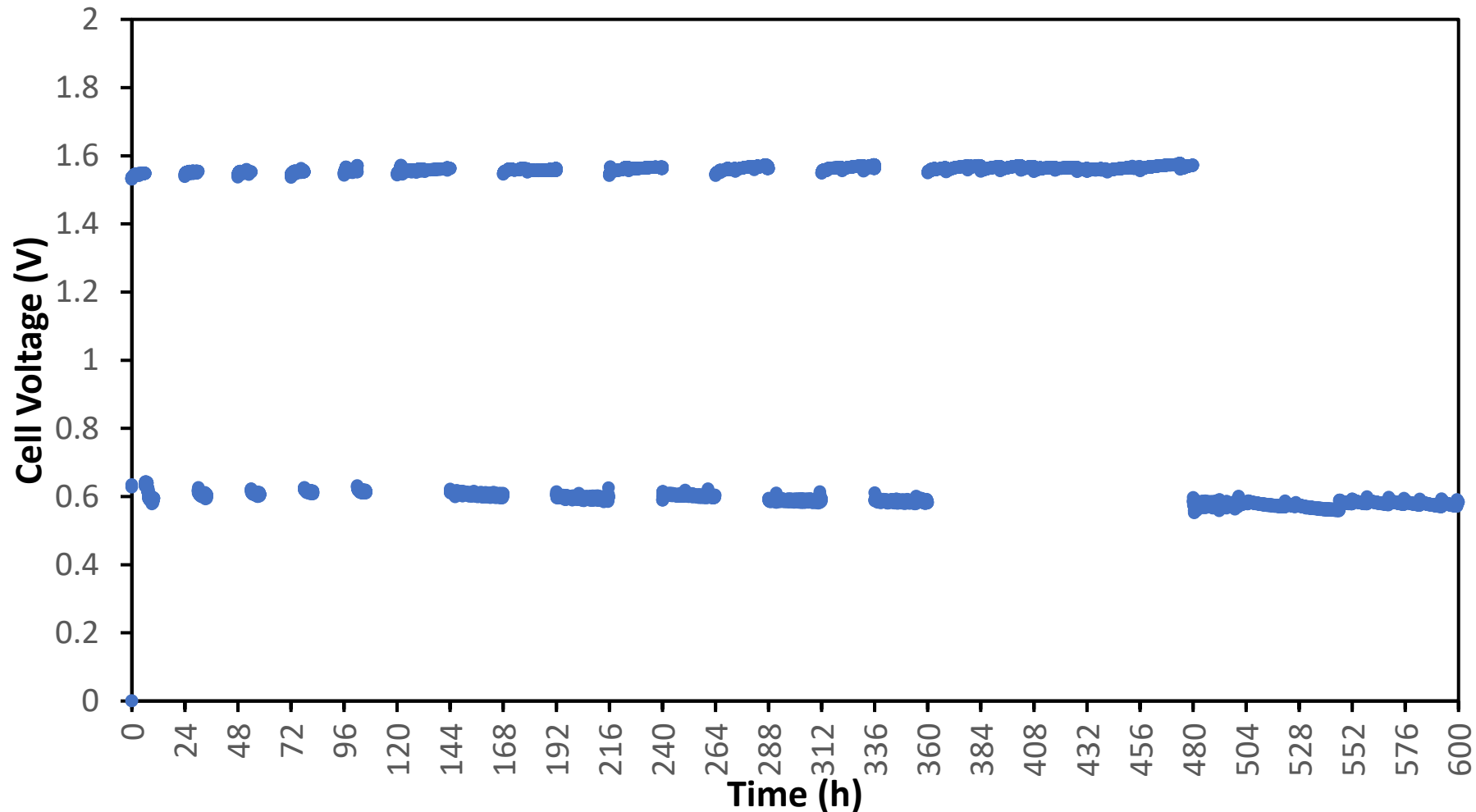
- Kinetic overpotential is almost the same between CG and CE mode

- Mass transport limits CG mode URFC performance

a), b) Kinetic overpotential of CCM1 under CE and CG mode when air and oxygen are used as oxidant at fuel cell mode, respectively; c), d) Mass transport overpotential of CCM1 under CE and CG mode when air and oxygen are used as oxidant at fuel cell mode, respectively.

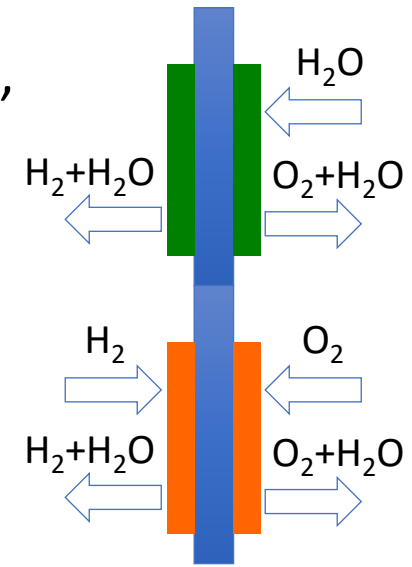
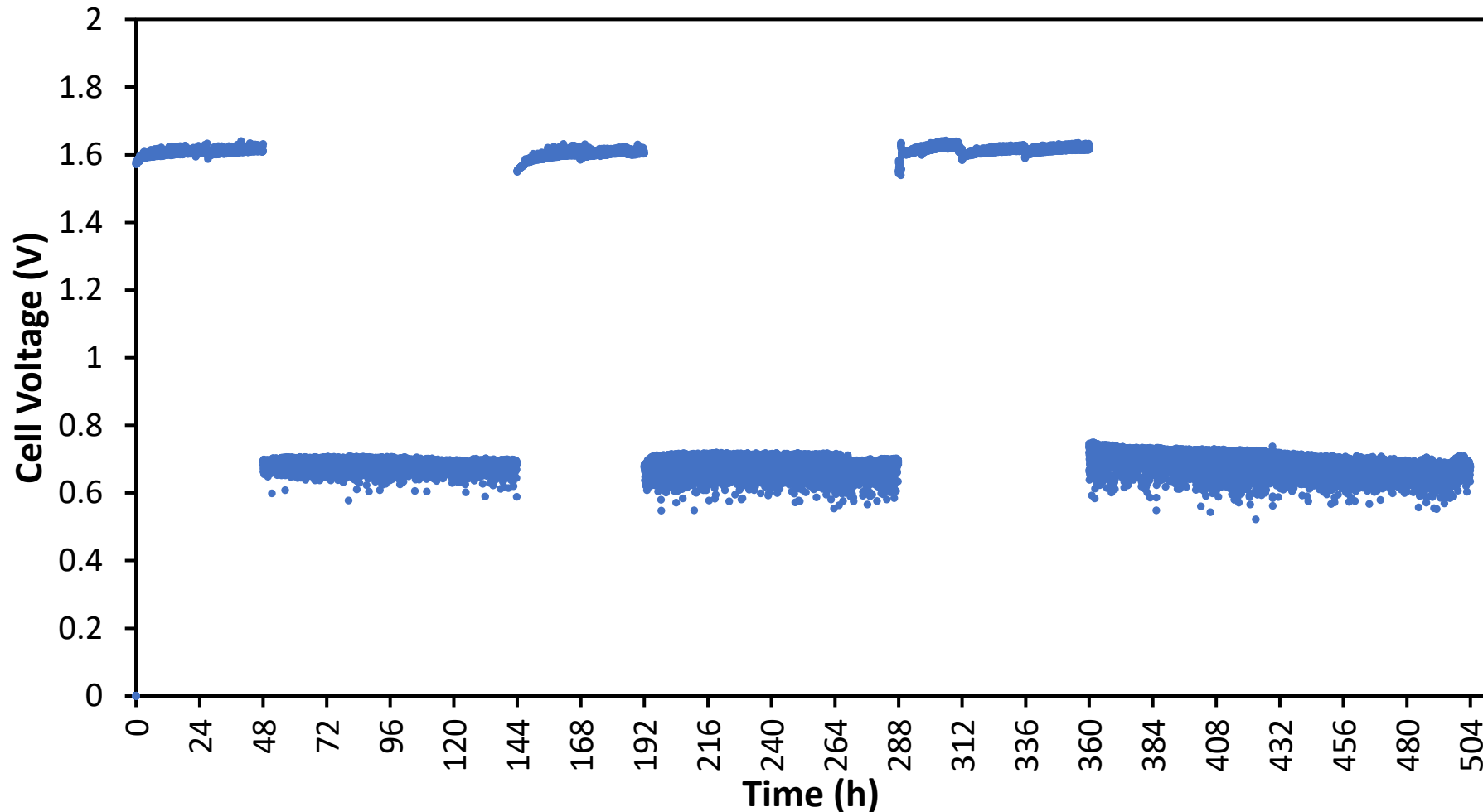
Accomplishments and Progress - CE Stability Tests

- Stability test of Ir-Pt black catalyst layer in CE-URFC mode
 - Charge and discharge @ 1 A/cm² for 600h, negligible degradation



Accomplishments and Progress - CG Stability Tests

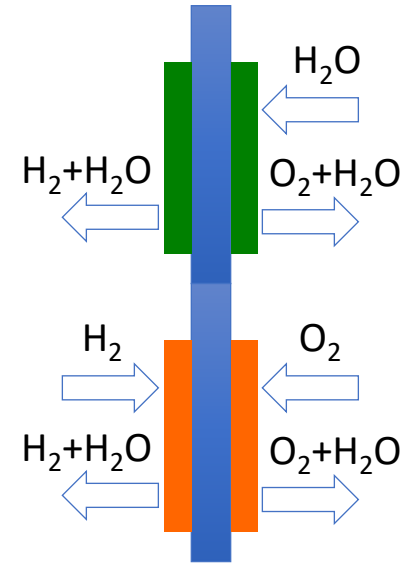
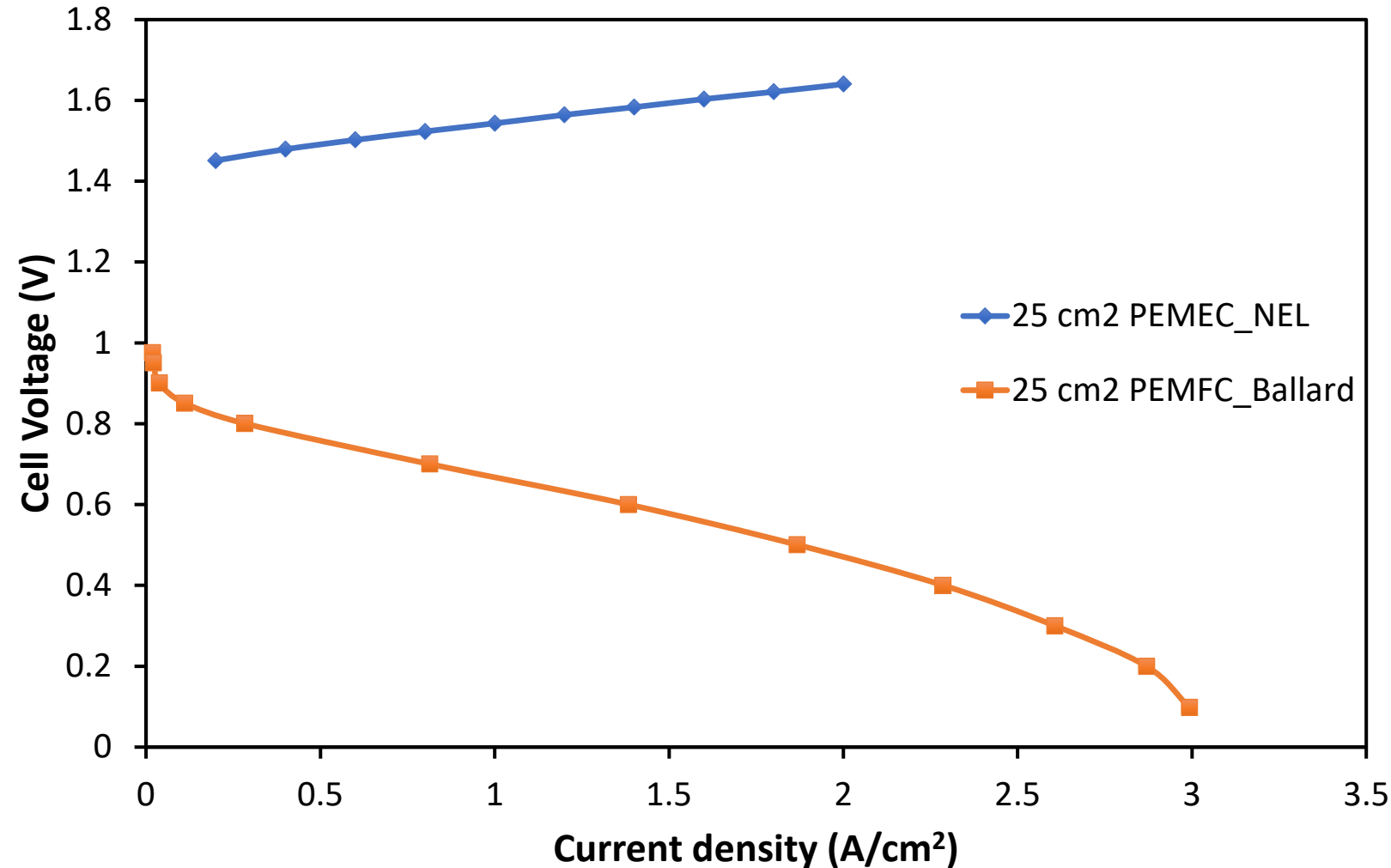
- Stability test of Ir-Pt black catalyst layer in CG-URFC mode
 - Charge @ 1 A/cm² and discharge at 0.5 A/cm² , negligible degradation in EC mode, voltage oscillation in FC mode.



Accomplishments and Progress

- Go/No Go 1

- 25 cm² Ir-Pt black catalyst layer in CG-URFC mode



25 cm² CCMs were prepared and tested at Ballard and Nel, respectively.

@ 1 A cm⁻² the RTE is 51.1%
Year 1 Target Met !

Accomplishments

Membrane Screening

- Screening for H₂ permeation under relevant conditions
- Pressurized operation may be necessary for storage but comes at a penalty of loss of storage efficiency



Test	Membrane	Operating Pressure (psi)	LFL% (1.86A/cm ²)	LFL% (1.16 A/cm ²)	LFL% (0.58 A/cm ²)	Diffusion, mL/min
1	Solvay 90	15psi	18.1%	15.6%	13.9%	0.20
		100psi	21.5%	20.7%	20.6%	2.04
		200psi	27.4%	29.5%	32.2%	3.50
		400psi	36.1%	41.3%	47.9%	5.52
2	Solvay 50 (980EW)	15psi	45.8%	39.5%	34.9%	6.00
		100psi	50.0%	50.0%	50.0%	16.00
		200psi	50.0%	50.0%	50.0%	21.14
		400psi	50.0%	50.0%	50.0%	29.33
3	Solvay 50 (870 EW)	15psi	35.0%	32.0%	31.0%	-
		100psi	42.0%	46.0%	50.0%	-
		200psi	44.0%	50.0%	50.0%	-
		400psi	46.0%	50.0%	50.0%	-
4	N212	15psi	41.2%	36.5%	32.0%	19.00
		100psi	50.0%	50.0%	50.0%	126.67
		200psi	50.0%	50.0%	50.0%	316.67
		400psi	50.0%	50.0%	50.0%	-
5	N211	15psi	50.0%	50.0%	50.0%	11.67
		100psi	50.0%	50.0%	50.0%	27.60
		200psi	50.0%	50.0%	50.0%	46.50
		400psi	50.0%	50.0%	50.0%	76.92

Accomplishments and Progress

- Accelerated Stress Test Development

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- Accelerated Stress Tests (ASTs) allow for failure analysis characterization to an end-of-test (EOT) fingerprint similarly matched to a standard durability test, but in a shortened timeframe
- Current AST does not have a failure fingerprint that matches the standard durability lifecycle
 - Current AST does not degrade the sample in the same way as the standard durability lifecycle, so is not suitable for screening materials
- **Objective**
 - Develop a new AST that has a failure analysis fingerprint that matches the standard durability lifecycle, with an acceleration factor $> 2x$
- **Path Forward**
 - Execute a design of experiment (DOE) to establish optimal square wave AST upper and lower potential limits and dwell times to match the EOT failure analysis fingerprint to the standard durability test

Collaboration and Coordination



- Collaborations

- Pajarito Powder

- Added to project post FOA award
 - Collaboration turned into subcontract on project
 - Leverages Pajarito catalyst experience and worldwide supply base for materials

- Solvay

- Aquivion chosen as baseline membrane/ionomer material
 - Direct supply by lots

- Coordination

- Benchmarking and AST development

- Crosscuts with HydroGEN and FCPAD

- URFC Technoeconomic Analysis

- Max Wei at LBL

Summary

- Baseline material set and testing protocols established
 - Unsupported bifunctional catalyst and mechanically robust thin membrane
 - URFC initial performance and stability testing protocols established
- Key findings in terms of CG-URFC performance limitation
 - Electrode structure plays very important role in determining CG-URFC performance
 - Unsupported electrode could achieve high URFC performance in both CE and CG modes
 - Mass transport limits CG-URFC performance due to PTL thickness and hydrophobicity
- Year 1 go/no-go target met
 - 25 cm² CCM achieved RTEs of 51% even using air during discharging

Remaining Challenges and Barriers

- Non-technical
 - Complete subcontracting and adjust affected milestones
 - Export control and sharing transport layers internationally
- Technical
 - Gas Diffusion Layer/Porous Transport Layer
 - Bifunctional OER/ORR electrode requires GDL/PTL that can manage liquid water and water vapor
 - Advanced Baseline Bifunctional Supported Catalysts
 - The current developed material showed excellent OER activity with relatively low ORR activity
 - Materials need to be tuned at single particle level to expose more ORR sites
 - Accelerated Stress Tests
 - What is the main degradation mechanism, how to accelerate/characterize
 - Large scale ($\geq 50 \text{ cm}^2$)
 - Large scale CCMs need to be fabricated and tested without performance compromise

Proposed Future Work

- YR2:
 - Advanced membrane
 - Develop highly conductive and mechanically stable PEMs
 - PEMs need to have low H₂ permeability at high pressure gradient
 - Advanced transport layer
 - Current porous transport layer will be thinned while maintaining high porosity
 - PTL Hydrophobicity treatment will be optimized to balance H₂O transport and O₂/Air transport
 - Membrane Electrode Assembly
 - Incorporate WUSTL supported catalysts into MEA
 - Develop AST for durability and degradation mechanism identification for supported catalysts

Technology Transfer Activities

- IP Management Plan
 - Being developed to protect existing IP
 - WUSTL: V. Ramani patents on supports
 - Pajarito
 - LBNL
 - To protect IP generated within project between partners

Acknowledgments



- Department of Energy for support

Response to Previous Year's Reviewer Comments

- **Question 1: Approach to performing the work**

- There are issues in this project that need to be addressed.
- (1) This project is confusing. It is regenerative, but in barriers it states "no regenerative-fuel-cell-specific barriers."
 - *Response:* We were referencing barriers in the MYRD&D. There are none for URFCs or RFCs.
- (2) The proposal is quite broad, with membranes and supported catalysts all in one project.
 - *Response:* Very good point by the reviewer. The current URFC research has been stagnant for years. To successfully achieve high-performing URFCs, it needs research on all aspects, even more than what we propose to do in this project. Therefore, we team with experts in catalysis, polymer and electrochemical systems to hopefully set a benchmark study for the URFCs and open different fields to future research
- (3) It would be nice to see a flowchart of how the contributors are interfacing.
 - *Response:* Very good suggestion by the reviewer. We will include flowchart in future reports.
- (4) It was stated that key personnel may have changed, which has impacts on the approach.
 - *Response:* Good point by the reviewer. We are conducting a dynamic research on URFC. At different stage of the research, we may need new input from different field. Therefore, we remain open in teaming with other experts. However, the key members including LBL, WUSTL, Ballard and Nel will stay through the project.
- (5) The challenges list the gas diffusion layer (GDL) and porous transport layer (PTL), which seem to be out of scope.
 - *Response:* We agree with reviewer that the PTL studies seem to be out of the scope. However, from our research in the past year, we actually found that the PTL properties played very important role in determining URFCs' performance by governing mass transport. Detailed data will be included in the year 1 AMR report.
- (6) There is no mention of what will be unique about the membranes; they seem to be just commercially bought.
 - *Response:* We expect to synthesize a membrane with high conductivity, low thickness and high mechanical stability to ensure stable URFC operation under charging mode. The reason we started with commercial membranes is that we'd like to learn how different membranes behave under URFC operation to better strategize our synthesis path and methodology for membrane development.

- **Question 4: Relevance/potential impact**

- (1) Using existing commercial membranes does not create much new impact. Pt-Ir systems have been around for a long time. The main impact, it seems, is the Washington University in St. Louis's supports for stability.
 - *Response:* It is good point by the reviewer. On one hand, the idea of starting with commercial membranes is to understand how different membrane properties (ionic conductivity, ion exchange capacity, water uptake, thickness et.al) impact URFC operation. It will provide guidance on how a good membrane should be designed for URFC operation. On the other hand, since new membrane development is expected to take time and using commercial won't delay the research of electrode, transport layers and other URFC critical component development. From a catalysis point, Pt-Ir systems won't create new impacts, however, it is very important to understand how a Pt-Ir catalyst layer would impact mass transport for URFC systems. The criterion we hold to access a good URFC catalyst is the device performance not half-cell activity.

- **Question 5: Proposed future work**

- (1) In year 1, the next developments are planned for the catalyst's support and integration, but nothing is mentioned for the membranes; this should be justified, considering the title of the project, including the membrane aspect. MEA optimization should focus on the reversible operation; performance for discrete operation could be second in the order. This is the same for the accelerated stress test (AST); it would be better justified to focus on defining the right duty cycles for URFCs or a specific AST for URFCs, if needed. In year 2, scale-up and discrete testing are planned; it should be that scale-up and reversible testing are planned for validation.
 - *Response:* Very good catch by the reviewer. We would include all membrane development work by WUSTL in the AMR presentation.
- (2) The GDL and PTL seem to be out of scope. It seems there is flux in the project.
 - *Response:* The GDL and PTL properties significantly impact the mass transport of URFCs, which is proved to be the major limiting factor to both the initial performance and stability from our year 1 work. Details are included in the AMR presentation.

Response to Previous Year's Reviewer Comments

- **Project weaknesses:**
- (1) The complementarity between partners is not always clear, particularly for MEA manufacturing and testing; the choice to double some actions should be justified. More efforts seem to be put on the separate assessment in fuel cell or electrolyzer modes instead of pushing URFC tests, which is the core scope. There is a lack of technical information about developments concerning the membrane in particular. Concerning catalysts, results could have been shown (electrochemical analyses or microstructure observations could have been mentioned, if available). The differences in the level of information expected from 25 cm² and 50 cm² is not obvious; the choice of considering these two areas for evaluating up-scaling should be clarified. For the validation task, the reason that Proton OnSite and Ballard are performing fuel cell or electrolyzer testing only is not justified. Validation should be done at larger scale for actual URFC cases. Duty cycles are defined as a challenge; this should be considered more of a required objective for final validation.
- *Response:* Very good suggestions by the reviewer. LBNL is mainly responsible for small scale (5 cm²) MEA manufacturing and testing. LBNL will scale up MEA fabrication to 25 cm² and 50 cm² and transfer the testing knowledge to NEL and Ballard for large scale MEA testing. NEL and Ballard will use their expertise in stack level testing and access the performance and stability of large scale MEAs, which is crucial to commercialization of URFCs. In year 1 work, we mainly focused on URFCs research instead of assess separate fuel cell or electrolyzer modes. There is a decent portion of work done in the catalysis research in year 1, which is shown in AMR presentations. The choice of considering 25 cm² and 50 cm² is based on the hardware size of Nel and Ballard R&D lab. We are pushing both Nel and Ballard to perform URFC testing instead of separate electrolyzer and fuel cell testing. However, due to some NDA reasons, part of the work has been delayed. We expect to overcome the hurdles in year 2 and performance URFC testing at both Nel and Ballard. We have achieved significant progress in duty cycles. Related work is included in AMR presentation.
- (2). The project has a broad, unfocused scope. Out-of-scope challenges are listed. The project has confusing targets and seemingly arbitrary baselines. Using a 175 mm membrane as a baseline needs explanation as to why it is so thick. Also, the team is in flux.
- *Response:* We have ambitious goal in this project. The reason the focus seems to be broad is because the URFC research has been stagnant over the years. To achieve URFCs at high round trip efficiency and high stability, it requires an interdisciplinary work among catalysis, membrane, transport layers, testing and technoeconomic analysis. The target and timeline are well established in initial proposal. 175 mm membrane is industrial baseline for electrolyzer operation. We believe that membrane mechanical stability will have huge impact on device stability especially under charging mode.
- **Recommendations for additions/deletions to project scope:**
- (1). It is recommended that the project clarify whether the membrane development aspects are still needed for the project outcomes and, in this case, what the routes explored and next steps are and what the possible role of Solvay might be. Focusing on the assessment and validation of materials and devices in URFC conditions with the definition of proper duty cycles would add value to the project (instead of focusing on discrete validation), since other projects are already considering the specific developments for fuel cell or electrolyzer materials and improvements. For the next review, more details should be given and more results shown.
- *Response:* Thanks for reviewer's recommendation. We have clarified the membrane and catalyst reasoning. We have largely focused on electrode and transport material validation in URFC devices and lots of progress has been made (see AMR presentation).
- (2). The project should eliminate or clarify the membrane part and flesh out the catalyst development paths and reasoning. Out-of-project components should be eliminated.
- *Response:* Thanks for reviewer's recommendation. We have clarified the membrane and catalyst reasoning.