Durable MEAs for Heavy-Duty Fuel Cell Electric Trucks

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Nikola Motor Company

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Carnegie Mellon University

Northeastern University
Center for Renewable Energy Technology

Georgia Tech

FCPAD
FUEL CELL PERFORMANCE AND DURABILITY

Project ID: fc326

Project Overview

Timeline

- Project Start: Q3 2020
- Project End: Q3 2023

Budget

- Total project budget: $2,125,000
  - Total Recipient Share: $425,000
  - Total Federal Share: $1,700,000
  - Total DOE funds spent*: $0

  * As of 05/30/2020

Barriers

- Durability
  - Improve stability of MEA for HD truck relevant operating conditions
- Performance
  - Increase catalyst while reducing ionomer poisoning effects to achieve high power density and higher efficiency
- Cost
  - Enable reduction in PGM catalyst loading and improve ionomer utilization

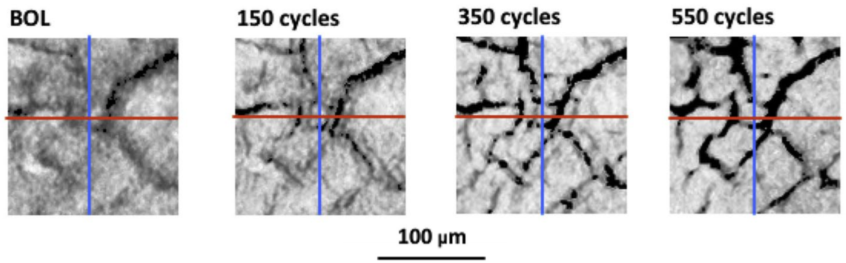
Partners

- Carnegie Mellon University
  - Modeling and characterization for MEA optimization
- Northeastern University
  - Durable IBAD catalysts
- Georgia Tech.
  - Durable alloy catalysts
- FC – PAD
  - Conventional electrode fabrication, MEA component ASTs, post-mortem characterization

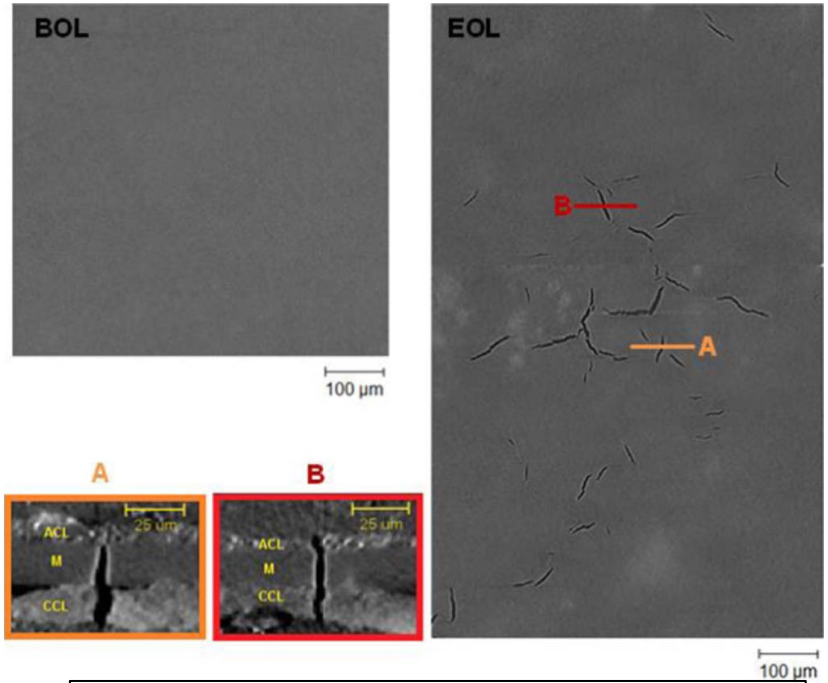
Relevance: Overcoming Durability and Cost Targets

OBJECTIVE: Develop novel MEA architecture to eliminate the formation of desiccation structures during MEA fabrication and fuel cell operation

- **Lifetime Target: 25,000 hours**
- **SOA MEA Durability Issues – addressed in this project**
  - Pt catalyst dissolution and stability
  - Catalyst and ionomer utilization
  - Electrode Corrosion & Cracking during Cycling



Microstructure of cathode catalyst layer after voltage cycling. Cracks are present even at BOL due to uneven drying of solvent and are exacerbated after operation. [1]



Top-down and cross-sectional views of of cathode catalyst layer and MEA after cycling. Cracks cause **CL degradation** and catastrophic **membrane failure**. [2]

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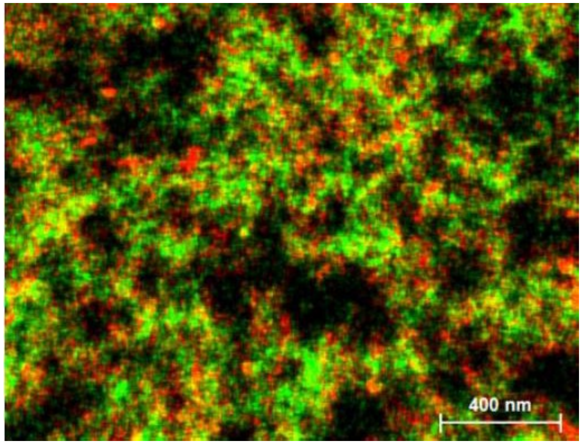
1. R.T. White et al. *Journal of Power Sources,* 350, 94-102 **2017** ***(**0.6 V for 30 s, to an upper potential of 1.4 V for 60 s in a square wave pattern).
2. Y. Singh et al. *Journal of The Electrochemical Society*, 164 (13) F1331-F1341, **2017**

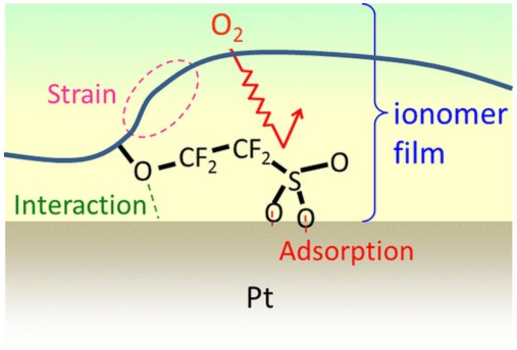
Relevance: Overcoming Durability and Cost Targets

Motivation & Current Shortcomings

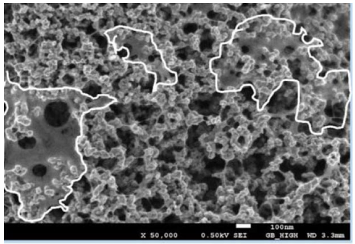
- Current catalyst layer (CL) fabrication techniques have several shortcomings including
  - The formation of cracks during fabrication
  - The formation of cracks during operation
  - Overuse of ionomer
    a) This reduces ORR activity
    b) This increases **O_2 gas transport resistance**
    c) Increases CL hydrophilicity, leading to **flooding** and greater **catalyst metal dissolution**



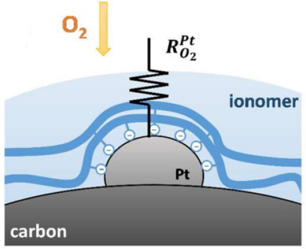
Current state-of-the-art catalyst layer catalyst/ionomer dispersion[1]



Proposed mechanism for catalyst poisoning via ionomer adsorption.[2]



Excessive ionomer can lead to agglomerates and poor durability.[4]



Adsorbed ionomer can increase oxygen gas transport resistance.[3]

1. Kumaraguru S. *General Motors FC-PAD Tech Team*, **2018**
2. K. Kodama et al. ACS Catalysis, 8, 694-700, **2018**
3. A. Kongkanand and Mark Mathias, *J. Phys. Chem. Lett.*, 7, 1127−1137 **2016**
4. Journal of The Electrochemical Society, 165 (14) F1254-F1263, **2018**

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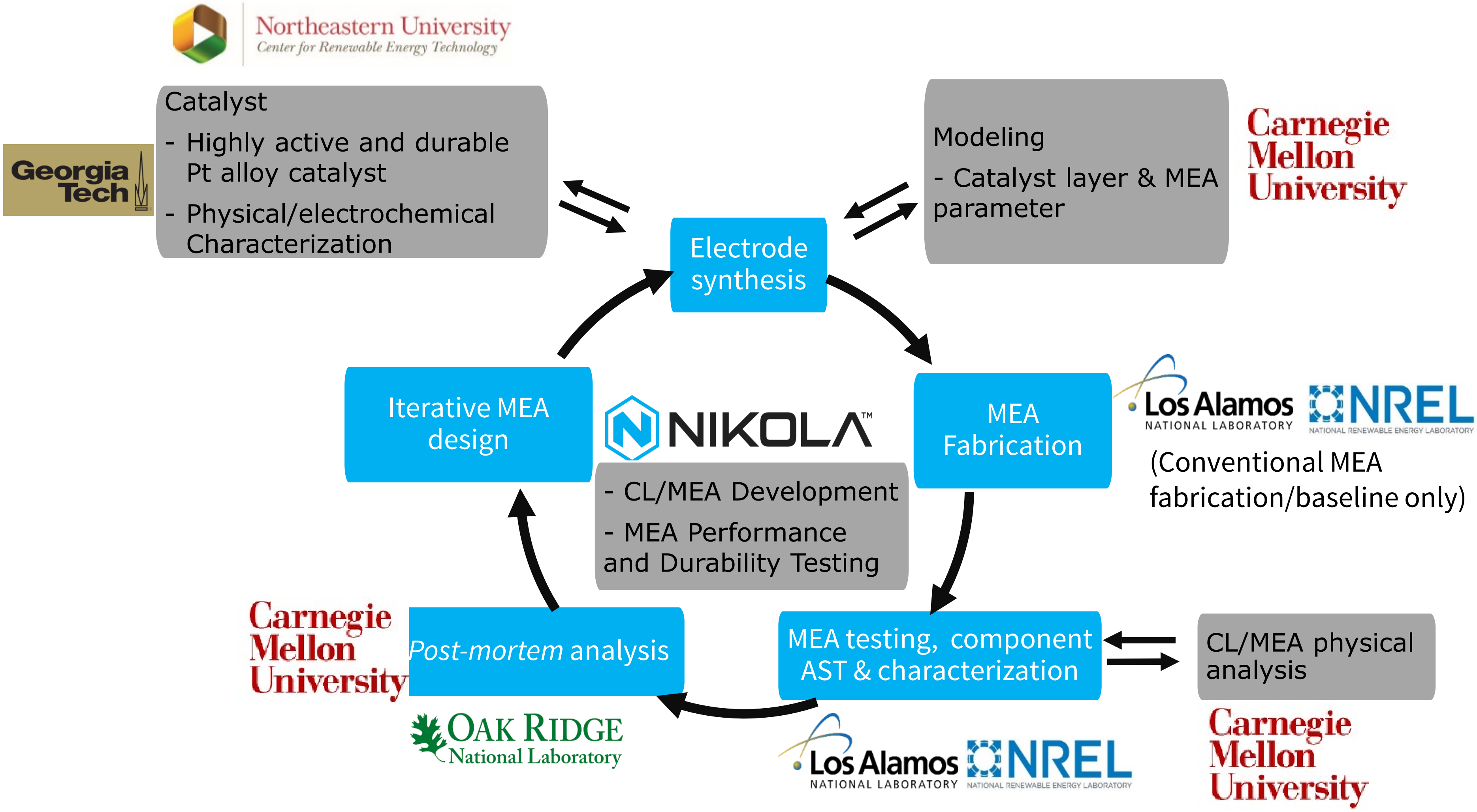
Relevance: Targets and Status

Characteristics	Target	Conditions
Operation hours	≥ 25,000 hours	HDT Drive cycle [1]
Platinum Group metal Loading	0.3 mg/cm^2 (total)	DOE Advanced Truck Technologies: Technical Targets for Hydrogen-Fueled Long-Haul Tractor-Trailer Trucks (released 10/31/2019)
Performance @0.8V	> 350mA/cm^2 (interim) >500 mA/cm2 (end of project)	DOE Technical Target(2016) Table 3.4.5, Table P6
Performance @ rated power	> 700 mW/cm^2 at HD rated power at 80C [2] (Full-size MEA)	- DOE Technical Target(2016) Table P6
Loss in catalytic mass activity	< 25 %[3]	DOE Technical Target(2016) Table 3.4.7, Table P1
Loss in rated power	< 10 %	Heavy-duty Drive-Cycle

1. Not relevant for HD; however used as initial target in the absence of HD specific ASTs.
2. Analysis of Fuel Cells for Trucks (TA024), R. Vijayagopal (ANL), 2019 DOE Hydrogen and vehicle technologies AMR
3. Not necessarily relevant for HD; however used as initial target in the absence of validated HD specific ASTs. Nikola HD truck durability targets are more aggressive.

Approach: Project Workflow & Collaborations



Northeastern University
Center for Renewable Energy Technology

Georgia Tech

Catalyst
- Highly active and durable Pt alloy catalyst
- Physical/electrochemical Characterization

Modeling
- Catalyst layer & MEA parameter

Carnegie Mellon University

Electrode synthesis

MEA Fabrication

Los Alamos NATIONAL LABORATORY

NREL NATIONAL RENEWABLE ENERGY LABORATORY

(Conventional MEA fabrication/baseline only)

Iterative MEA design

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- CL/MEA Development
- MEA Performance and Durability Testing

MEA testing, component AST & characterization

CL/MEA physical analysis

Carnegie Mellon University

Post-mortem analysis

Carnegie Mellon University

OAK RIDGE National Laboratory

Los Alamos NATIONAL LABORATORY

NREL NATIONAL RENEWABLE ENERGY LABORATORY

Project Participants and Roles

Institution; Personnel	Key Role(s)
Nikola Motor Company (Nikola); • Dr. Vivek Murthi (PI) • Dr. John Slack (Lead Project Scientist)	• Benchmarking MEA components • Fundamental MEA architecture design, fabrication, optimization, testing and analysis • MEA development and scale-up strategies and supplier collaboration • Single-cell testing and MEA validation • Program Management
Georgia Institute of Tech. (Georgia Tech.); Prof. Younan Xia	• Durable supported catalyst development and sub-scale testing • Catalyst physical and electrochemical characterization
Northeastern University (NEU); Prof. Sanjeev Mukerjee	• Catalyst development based on Ion Beam Assisted Deposition (IBAD) on commercial and tailored carbon supports; • Fundamental studies to elucidate electrocatalyst structure
Carnegie Mellon University (CMU); Prof. Shawn Litster	• Catalyst layer nano-structure and MEA micro-structure 3D imaging and analysis • Catalyst aggregate scale modelling • O_2 and proton (H^+) transport modelling to provide for MEA optimization
FC-PAD National Laboratory Consortium Members	• Advanced MEA fabrication, sub-scale performance testing and evaluation (NREL, LANL) • Component Diagnostics and Characterization (ORNL) • Operando Evaluation— Accelerated Stress Testing (AST) of MEA components and new AST protocol development for HD FCET (LANL)

Approach: Collaborative Project Team

- Northeastern: **Dual IBAD** technique to produce **durable** catalyst powders
- Georgia Tech: Synthesize supported **durable cuboctahedral PtCo** catalyst
- Nikola: **Novel durable CL/MEA structure** *(Nikola IP)* based on methods similar to nanoencapsulation, to form layered structures of catalyst powders and ionomer
- CMU: Create **CL transport models** and recommend MEA formulation/optimization based on feedback from X-ray computed tomography and pFIB-SEM imaging

Approach: Durable Catalyst Development

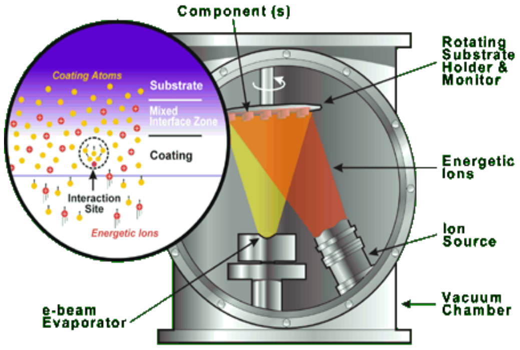
- ❖ **<u>Pt-M (M = Fe, Co, and Ni) Alloy Catalysts – Georgia Tech.</u>**:

- ❑ Colloidal synthesis of Pt–M alloyed nanocrystals
  - ➢ Prior result: 4-nm Pt-Co truncated octahedra
  - ➢ Project Focus:
    1. Controlled compositions, shapes, and sizes
    2. <u>Continuous</u> and <u>Scalable</u> production

AST Cycles	**Mass Activity (A/mgPt at 0.9 V vs RHE)**	
	GT PtCo	Umicore PtCo
BOL	0.294	0.186
BOL + Recovery	0.384	0.394
30000	0.224	0.123
30000 + Recovery	0.335	0.164

- ❖ **<u>Dual-Ion-Beam-Assisted catalysts – Northeastern Univ.</u>**:

- ❑ Line of sight technology referred as dual-ion-beam-assisted deposition (IBAD) technique
  - ➢ Project Focus:
  1. Pt-M alloy
  2. Ti doped interlayer to improve stability and conductivity, both deposited using dual IBAD technique
  3. graphitic and corrosion resistant carbons



Schematic of dual IBAD for creating ultra-thin amorphous coating of mono or mixed metal deposits.

Approach: MEA Development

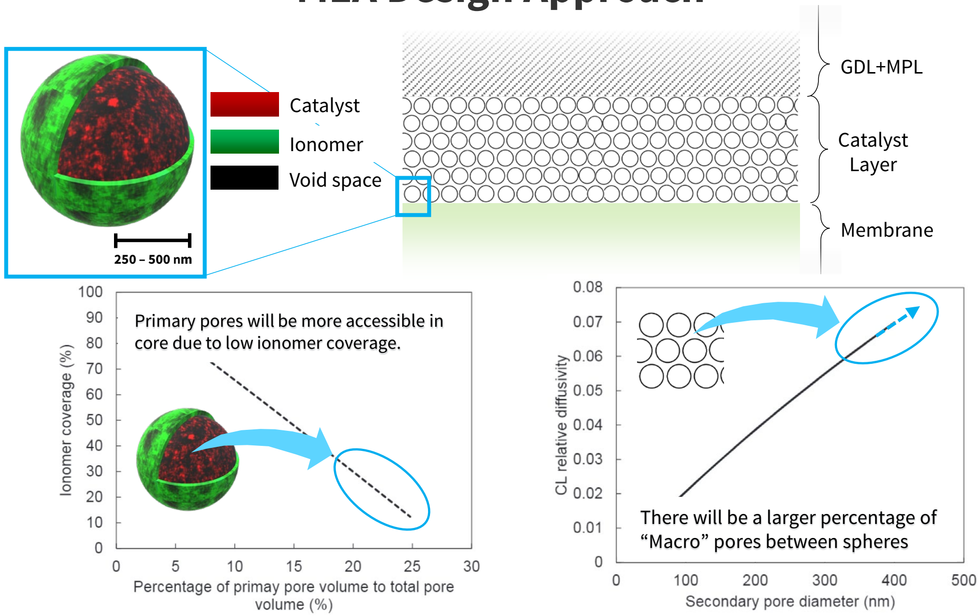
❖ The goal of this project is to:

- Reduce/eliminate crack formation during deposition/operation → **improve durability**
- Improve catalyst utilization → **Use less PGM**
- More effectively orient ionomer, reducing SO_3^- poisoning → **better mass transport and ORR activity**
- Accurately control pore-size distribution → **better water management and mass transport**

❖ Technology Improvement Targets:

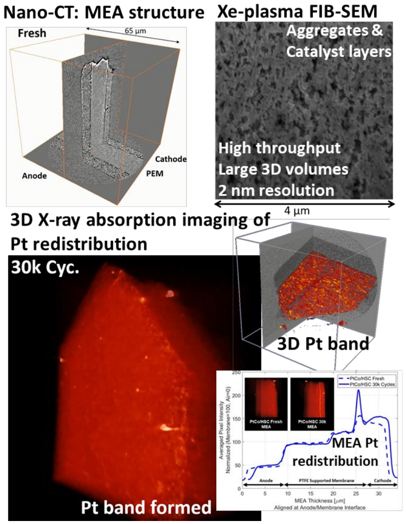
- At high RH: Reduction in flooding from "macro" pores
- At low RH: Improvement in ionic conductivity from water condensation in primary pores

MEA Design Approach



Modeling Approach

- Provide suggested optimal sphere size and ionomer thickness to maximize oxygen transport while maintaining the minimum ionic conductivity required to avoid increases in ohmic overpotential
- Provide insights about ion conduction in ionomer-free domains
- Model Pt dissolution and migration in the Nikola CL structure and compare this to SOA MEAs.
- Determine the impact of pore diameter and catalyst loading on proton conduction and oxygen diffusion, and the impact of internal catalyst morphology on stabilizing Pt particle size with voltage cycling
- Provide baseline membrane-Pt-band data and compare this to Nikola MEAs



Catalyst and electrode imaging at CMU

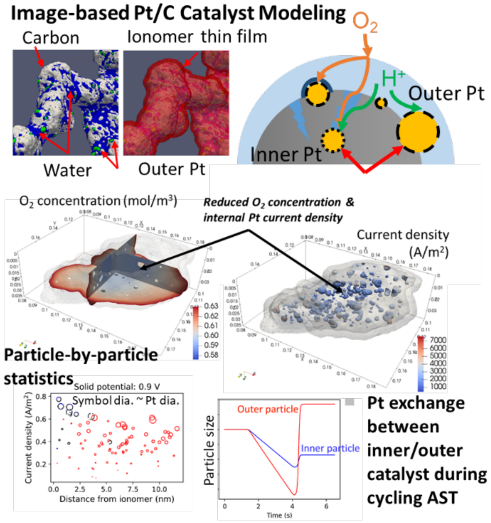
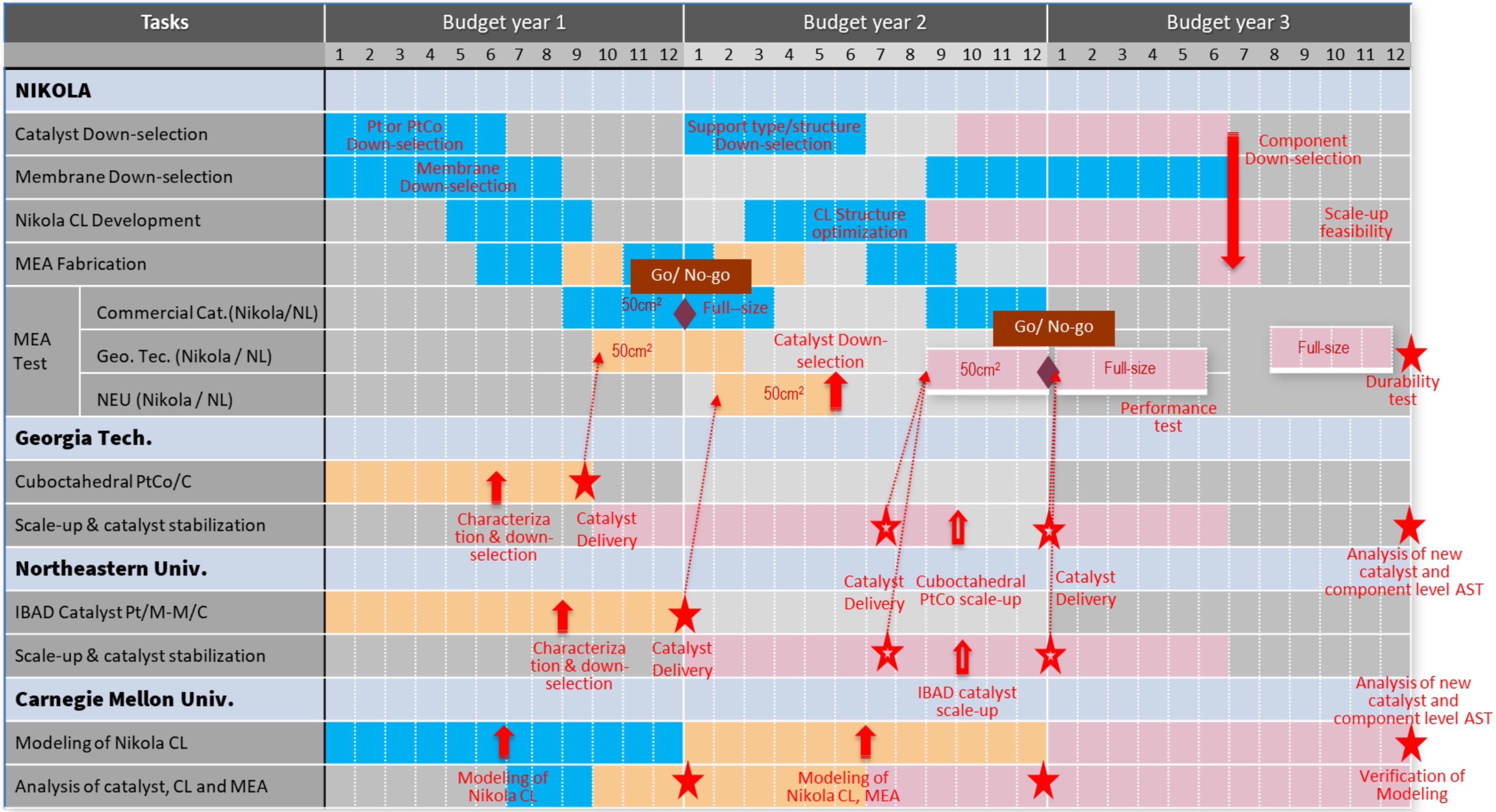


Image-based catalyst performance and durability modeling

Milestones and Go/No-Go Decisions

- 🟦 Commercial catalyst
- 🟧 Experimental catalyst, year 1
- 🟪 Experimental catalyst, year 2/3



Tasks		Budget year 1	Budget year 2	Budget year 3
NIKOLA				
Catalyst Down-selection		Pt or PtCo Down-selection	Support type/structure Down-selection	Component Down-selection
Membrane Down-selection		Membrane Down-selection		
Nikola CL Development			CL Structure optimization	Scale-up feasibility
MEA Fabrication		Go/ No-go		
MEA Test	Commercial Cat.(Nikola/NL)	50cm²	Full--size Go/ No-go	
	Geo. Tec. (Nikola / NL)	50cm²	Catalyst Down-selection 50cm² Full-size	Full-size Durability test Performance test
	NEU (Nikola / NL)		50cm²	
Georgia Tech.				
Cuboctahedral PtCo/C		Characteriza tion & down-selection Catalyst Delivery		
Scale-up & catalyst stabilization			Catalyst Delivery Cuboctahedral PtCo scale-up Catalyst Delivery	Analysis of new catalyst and component level AST
Northeastern Univ.				
IBAD Catalyst Pt/M-M/C		Characteriza tion & down-selection Catalyst Delivery		
Scale-up & catalyst stabilization			IBAD catalyst scale-up	Analysis of new catalyst and component level AST
Carnegie Mellon Univ.				
Modeling of Nikola CL				
Analysis of catalyst, CL and MEA		Modeling of Nikola CL	Modeling of Nikola CL, MEA	Verification of Modeling

Milestones Summary: Budget Period 1

Task Number	Task or Subtask	Milestone Type/number: Milestone Description	Anticipated Month (from Start of the Project)
Subtask 1.1.1	MEA component down-selection	• M 1.1.1: down-select commercial catalyst • M1.1.2: down-select membrane	M6, M9
Subtask 1.1.2	Nikola CL optimization and fabrication	• M 1.1.3: Nikola CL optimization with commercial catalyst • M 1.1.4 : MEA Benchmarking Baseline Definition	M12
Task 1.2	Development of cuboctahedral PtCo/C catalyst for Nikola CL	• M 1.2.1: Metal deposition optimization for cuboctahedral PtCo/C • M 1.2.3: fabrication and delivery of 6g of catalyst	M6, M9
Task 1.3	Development of IBAD Pt/M-M/C catalyst for Nikola CL	• M 1.3.1: Metal deposition optimization for IBAD Pt/M-M/C • Milestone 1.3.3: Perform IBAD catalyst degradation analysis and delivery of 6g of catalyst	M8, M12
Task 1.4	Modeling and analysis of Nikola CL	• M 1.4.1 Modeling of MEA CL • M1.4.2: structural analysis of Nikola CL and MEA	M6, M12
Task 1	Nikola CL MEA fabrication and performance (≤50cm^2 MEA active area)	• **Go/No-Go 1:** Nikola CL MEAs will be fabricated with commercial catalyst • MEA -Performance ≥ 350 mA/cm^2 at 0.8V, 200 kPa$_{ab}$, 80 °C , 0.3 mg/cm^2 PGM total	M12

Milestones Summary: Budget Period 2

Task Number	Task or Subtask	Milestone Type/number: Milestone Description	Anticipated Date (Months from Start of the Project)
Task 2.1	Nikola CL MEA fabrication using new catalyst	• M 2.1.7: Nikola CL MEA performance and durability (full -size) using commercial catalysts at a loading of 0.2~0.4 $mgPt/cm^2$	M15
Task 2.2	Synthesis of PtCo catalyst on durable carbon supports	• M 2.2.2: Fabrication of cuboctahedral PtCo/durable carbon and delivery • M 2.2.3: Scale-up if down-selected	M17 (M20)
Task 2.3	IBAD Pt/M-M/ /C optimization and fabrication for Nikola CL	• M 2.3.2: Fabrication of IBAD Pt/M-M/ on durable carbon and delivery • M 2.3.3: Scale-up of IBAD Pt/M-M if down-selected	M17 (M20)
Task 2.4	Modeling and analysis of Nikola CL and MEA	• M 2.4.1: Refine modeling of Nikola CL and MEA through sub-unit structure parameter confirmation and Electrode-scale modeling • Milestone 2.4.2: Analysis of Nikola CL & MEA using Nano-CT imaging of ionomer distribution Oxygen transport in CL. Etc.,	M18, M24
Task 2	Nikola CL MEA evaluation with Durable catalyst	• **Go/No-Go 2:** Nikola CL MEA using down-selected experimental catalyst with a durable carbon. • MEA -Performance ≥ 350 mA/cm^2 at 0.8V, 200 kPa_{ab}, 80 °C, 0.3 mg/cm^2 PGM total • ≤25% drop in rated power after load cycling (DOE catalyst ASTs)	M24

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Conclusions and Acknowledgements

1. Develop a CL architecture to maximize oxygen transport with the minimum ionomer necessary to maintain the percolation limit of protons
2. Compare new CL structure to SOA MEAs to
   a) Decrease SO_3 adsorption and improve related catalyst activities & electrochemically active surface areas
   b) Reduce Crack formation after a protocol designed to accelerate this degradation mode
   c) Increase Lifetime efficiencies (BOL, average lifetime, and EOL) at nominal operating voltages
3. CMU and ORNL together will provide data to help Nikola correlate structure with performance and durability
4. GA Tech and NEU will develop catalysts that Nikola will compare to commercial catalysts. The catalyst which provides highest BOL, average lifetime, and EOL efficiencies at nominal operating voltage will be down-selected.

Acknowledgements

- EERE Award: DE-EE0008820; FCTO – Greg Kleen, Dimitrios Papageorgopoulos, Elliot Padgett
- Collaborators – Prof. Mukerjee, Prof. Litster, Prof. Xia
- Nikola Team – Dr. John Slack, Dr. Jason Lee, Dr. Andrew Baker, Dr. Bahareh Tavakoli, Christian Appel, Jesse Schneider

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