



## FY2015 – FY2020

## FY2021 - FY2025

# Durability Developments from FC-PAD to M2FCT Rod Borup and Rangachary Mukundan







## **Biden Administration Targets CO2 Emissions**



FACT SHEET: President Biden Sets 2030 Greenhouse Gas Pollution Reduction Target Aimed at Creating Good-Paying Union Jobs and Securing U.S. Leadership on Clean Energy Technologies Building on Past U.S. Leadership, including Efforts by States, Cities, Tribes, and Territories, the New Target Aims at <u>50-52 Percent Reduction</u> in U.S. Greenhouse Gas Pollution from 2005 <u>Levels in 2030</u>

On Day One, President Biden fulfilled his promise to rejoin the Paris Agreement and set a course for the United States to tackle the climate crisis at home and abroad, reaching net zero emissions economy-wide by <u>no later than 2050.</u> As part of re-entering the Paris Agreement, he

- \$1 Trillion Infrastructure Bill Passed Senate (69-30) Nov House (228-206), signed by President Biden Nov 15
- Department of Energy has 180 days to issue proposal calls for:
  - > 4 Hydrogen regional HUBs \$8B over 5 years
  - \$1B for Electrolysis
  - \$500M for Clean Hydrogen Manufacturing and Recycling

# Transition from FC-PAD to M2FCT Target Comparison between Light- and Heavy-Duty

Table 1. Technical Targets for Automotive-Scale (80 kWe net Fuel Cell System Operating on Hydrogen<sup>a</sup>

Characteristic	Units	Status	2020 Target	2025 Target
Peak Energy Efficiency <sup>b</sup>	%	60 °	65	65
Specific power	W/kg	659 <sup>d</sup>	650	900
Cost <sup>f</sup>	\$/kWe	45 e	40	35
Cold start-up time to 50% of rated power				
@ -20°C ambient temp	sec	20 f	30	30
@ +20°C ambient temp	sec	<10 <sup>f</sup>	5	5
Durability in automotive load cycle	hours	4130 <sup>g</sup>	5,000	8,000
Unassisted start from h	°C	-30 <sup>i</sup>	-30	-30

#### Table 1. Technical System Targets: Class 8 Long-Haul Tractor-Trailers (updated 10/31/19)

Characteristic	Unite	Targets for Class 8 Tractor-Trailers		
Characteristic	Onits	Interim (2030)	Ultimate <sup>9</sup>	
Fuel Cell System Lifetime <sup>1,2</sup>	hours	25,000	30,000	
Fuel Cell System Cost <sup>1,3,4</sup>	\$/kW	80	60	
Fuel Cell Efficiency (peak)	%	68	72	
Hydrogen Fill Rate	kg H <sub>2</sub> /min	8	10	
Storage System Cycle Life <sup>5</sup>	cycles	5,000	5,000	
Pressurized Storage System Cycle Life <sup>6</sup>	cycles	11,000	11,000	
Hudrogon Storago System Cost4.7.8	\$/kWh	9	8	
nyurugen storage system cost 42-	(\$/kg H <sub>2</sub> stored)	<mark>(</mark> 300)	(266)	

Condition	Traditional	M2FCT Focus
Operating temperature	60 - 80 °C	$\sim$ 90 °C
Catalyst	Random alloy Pt <sub>90</sub> Co <sub>10</sub>	Tailored and ordered alloys, annealed Pt
Membrane	Ultra-thin, reinforced with mobile Ce	Stabilized, durable, high selectivity (H+ conductance/H <sub>2</sub> permeance)
Operating voltage	0.6 - 0.9 V	>0.7 V
Durability	5,000 hrs	25,000 hrs
Pressure	150 kPa	250 kPa
Catalyst loading	$0.15~g_{Pt}/cm^2$	$0.3 \text{ g}_{\text{Pt}}/\text{cm}^2$

# **M2FCT Consortium - Overview**

### **Timeline**

- Project start date: 10/01/2020
- Project end date: 09/30/2025

### Budget

- FY20 project funding: \$10M
  - Planned \$1M external partners
  - State Support FOAs \$1.5M Effort to Support FOAs
  - 5-year consortium with yearly milestones & Go/No-Go

### **Partners/Collaborations**

- DOE DE-FOA-0002044:
  - 🌭 GM, Nikola, Carnegie Mellon
- **DOE DE-FOA-EE0009244:** 
  - 🌭 3M, Lubrizol, Nikola, UT Knoxville
  - Scummins, Plug Power
- No-cost collaborations

### Heavy-Duty Transportation (2025)

- Durability: 25,000 hour lifetime
- 68% peak efficiency
- \$80/kW fuel cell system cost
- Overall Target: 2.5 kW/g<sub>PGM</sub> power (1.07 A/cm<sup>2</sup> current density) at 0.7 V after 25,000 hour-equivalent accelerated durability test

### **Heavy-Duty Transportation (2030)**

- Durability: 30,000 hour lifetime
- 72% peak efficiency
- \$60/kW fuel cell system cost

# **M2FCT Approach**

Million Mile Fuel Cell Truck (M2FCT) will tackle these challenges through a "team-ofteams" approach featuring main teams in analysis, durability, integration, and materials development. By coming together as sets of dynamic teams, the integrated consortium will provide rapid feedback, idea development, and information exchange, resulting in an effort that is more than the sum of its parts.



2.5 kW/g<sub>PGM</sub> power (1.07 A/cm<sup>2</sup> current density) at 0.7 V after 25,000 hour-equivalent accelerated durability test

# **Organization Chart**



## M2FCT: National Labs in Partnership with Universities and Industry



### "Team-of-teams" approach that allows for rapid feedback, idea development, and information exchange





Main Laboratories LOS Alamos rerere BERKELEY LAB CAK RIDGE Argonne

#### Affiliate Laboratories





# M2FCT: R&D Priorities

#### M2FCT Research Priorities



1 is highest priority



integration

# **International Durability Working** Group (iDWG)

## **8** Countries

from America, Europe, and Asia

### **30 Institutions**

participants representing governments, universities, industry and labs

**80 Researchers** 

facilitating data sharing, exchanging materials, promoting AST development

MILLION MILE **FUEL CELL TRUCK** U.S. DEPARTMENT OF ENERGY



NEDO

New Energy and Industrial Technology **Development Organization** 





#### Characterization



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cea

**Benchmarking and Protocols** 

#### Stressors related to Heavy Duty

# **M2FCT System Modeling**



#### **Salient Features**

- 275 kW net (70-kWh ESS) at EOL
- Multiple stacks: 4
- Electrodes
  - Cathode: a-Pt/C, 0.25  $mg_{Pt}/cm^2$ , 50 wt.% Pt Anode: Pt/C w IrO<sub>2</sub> (TBD), 0.05  $mg_{Pt}/cm^2$
- Membrane: 14 mm, chemically stabilized, mechanically reinforced
- Single air system with expander
- Single anode system with recirculation blower
- Cathode humidifier: No (TBD)
- Rated power conditions at EOL: 2.5 atm, 87-95°C, 660-700 mV
- Control valves for startup and shutdown, cold start and OCV

# **Determining typical HDV relevant condition**

Autonomy simulation results provided by Vincent Nicolas Freyermuth, Argonne National Laboratory

#### Real World Drive Cycle

- Gross Vehicle Weight (GVW): 36.3 MT (80,000 lbs)
- 10.5-h drive segment, 3 extended idles with engine off
- Truck speed around 65 mph

#### **Motor Power**

- Maximum power on drive cycle: 500 kW<sub>e</sub>
- Motor input power on 6% grade at 30 mph, 36.3-MT GVW: 400 kW
- Motor input power at 65-mph, 0% grade: 160 kW

Jason Lustbader and Kenneth Kelly: HD/MD duty cycles from NREL



Time, %

### **Operating Temperatures Statistics**

- Stack temperature remains below 75°C throughout the drive cycle
- Median stack temperature maintained within 65±1 °C for 34.3% time. Can be further adjusted by controlling the fan power and/or the thermostatic valve.
- Stack temperature below 70°C for >98% time.





### **FCS Heat Rejection**

- Fuel cell dominant propulsion system rated at 275-kW $_{\rm e}$  net, 35-kWh ESS
  - ♥ Hill climb at 35 mph, 6% grade, 20-min duration
  - Battery supplies balance of propulsion power and operates in a charge depleting mode
  - ♦ 25°C ambient temperature
- FCS heat load and fan power
  - ♦ Heat load (Q) is mainly a function of cell voltage

  - ✤ Higher cell voltage, smaller fan power
  - Seed cell voltage > 0.75 V at rated power for stacks that operate below 80°C
  - Sample result at 0.7 V: 87°C, 265 kW radiator heat load, 30 kW fan power



# **FCS Heat Rejection on Hill Climb**

Modeled system equivalent to radiator fan for 450-hp diesel engine, 52°C air-to-boil temperature



#### **Study Conclusions**

Study parameters: Hill climb at 30 mph, 6% grade, 20-min duration, 25°C ambient temperature

- 1. Largest FCS for which waste heat can be rejected using radiators in a 450-hp diesel trucks: 275 kW $_{\rm e}$  net
- 2. Smallest battery operated in charge depleting mode during hill climb: 35 kWh
- 3. Lowest cell voltage at EOL for exit coolant temperature below 95°C: 660  $\,mV$

Heat Exchangers	Dimensions and Details	Heat Loads	Radiator Fan 37 kW
HT Radiator	40″ (W) x 42″(H) x 2″ (D) Fins: louvered, 12-fpi, 10-mm height Tubes: 2-mm height	280 kW	Vehicle Speed: 30 mph
LT Radiator	40" (W) x 35" (H) x 2.5" (D) Fins: louvered, 8-fpi, 20-mm height Tubes: 10-mm height	49 kW	Ambient T: 25⁰C
AC Condenser	40″ (W) x 28″ (H) x 0.75″ (D) Fins: plain, 12-fpi, 10-mm height Tubes: 2-mm height	12 kW	Air Flow Rate: 9.3 kg/s





## **FCS-HDV Degradation Adjusted Stack Size**

Lifetime defined by ECSA loss as marker of aging (275  $kW_e$  at EOL)



#### 85-90°C coolant



- Stack size defined by efficiency target
  - Stack coolant exit temperature at peak power
- Stack active area defined for various catalyst degradation for 700 mV (EOL cell voltage)

#### **Modeled Electrode Degradation Mechanisms**

- $\backsim$  ECSA loss due to Pt dissolution and growth in particle size
- ✤ Degradation in ORR kinetics
- ✤ Increase in oxygen transport resistance
- ♥ Change in Pt accessibility
- Relaxing EOL cell voltage to 660 mV decreases the initial stack active area by 10-15%
  - Stack coolant exit temperature at peak power, ∼ 50°C higher

2.5 atm, 1.5 SR(c), 0.3 mg/cm<sup>2</sup> Pt, a-Pt/HSC cathode catalyst



# **Durability**

#### Understanding, evaluating, and mitigating durability concerns with materials-based solutions





## **Flowchart**



![](_page_16_Picture_2.jpeg)

# Plan for ASTs: Catalyst followed by Membrane and MEA

- Currently performing ASTs in H<sub>2</sub>/Air on integral cell @ 90 °C, 100%RH with square wave cycling from 675 mV to 925 mV 30s each at 250 kPa
- Perform ASTs in H<sub>2</sub>/Air on integral cell @ 90 °C, 90%RH with square wave cycling from 675 mV to 925 mV 30s each at 250 kPa
- Perform the same at an LDV relevant condition (Temp, pressure, RH, potential limits)
- Perform the same at an HDV relevant condition (Temp, pressure, RH, potential limits)
- Calculate acceleration factors from 90 °C, 100% RH, 90%RH test to HDV conditions and prescribe test duration to obtain 25,000 hour durability.
  - **Solution** Introduce RH switching and a drier condition to accelerate membrane durability
  - Sept 2021
- Calculate acceleration factor from HDV to LDV conditions and multiply that by 3X to change the targets for all component specific ASTs. If times get too long, then we can further accelerate it. Will be supported by modeling work.

### ♦ 2 years +

• Refine HDV cycle by including other stressors like high temp, high voltage, dry operation.

## Durability Performance: High Loading - Effect of Loading

![](_page_18_Figure_1.jpeg)

80C, 100% RH, 150 kPa

![](_page_18_Picture_3.jpeg)

## Durability Characterization: Effect of loading (0.05 to 0.15 mg<sub>Pt</sub>/cm<sup>2</sup>)

![](_page_19_Figure_1.jpeg)

Mean diameter after AST (S	AXS
0.05 mg/cm <sup>2</sup> : 4.5 nm	
0.10 mg/cm <sup>2</sup> : 5.2 nm	
0.15 mg/cm <sup>2</sup> : 5.5 nm	

![](_page_19_Figure_3.jpeg)

EKAT-05 181113 180828 180529

BOL :	Pt:Co = 75:25
30K (0.05) :	Pt:Co = 80:20
30K (0.10) :	Pt:Co = 85:15
30K (0.15) :	Pt:Co = 87:13

- Higher loading results in slightly larger growth in Pt particle size (both SAXS and TEM)
- On average Co is retained better in the lower loaded electrode
- Larger lattice contraction loss for lower loading (EXAFS data, not shown) coupled with smaller particle growth indicate larger particles retain Co better than smaller particles

![](_page_19_Figure_8.jpeg)

PtCo particle size = 4.2 nm

> 0.10mg<sub>Pt</sub>/cm<sup>2</sup> 30,000 cycles

![](_page_19_Picture_11.jpeg)

PtCo particle size = 5.5 nm

![](_page_19_Picture_13.jpeg)

PtCo particle size = 5.1 nm

> 0.15mg<sub>Pt</sub>/cm<sup>2</sup> 30,000 cycles

![](_page_19_Picture_16.jpeg)

PtCo particle size = 5.6 nm

![](_page_19_Picture_18.jpeg)

## **Durability Performance: High Loading - Effect of Alloying**

![](_page_20_Figure_1.jpeg)

**UEL CELL PERFORMANCE** 

Loading = 0.25mg/cm<sup>2</sup> 80C, 100% RH, 150 kPa

SW AST = Square Wave Catalyst AST (0.6 - 0.95V)

## Durability Characterization: High Loading - Effect of Alloying

![](_page_21_Figure_1.jpeg)

- PtCo has a larger initial particle diameter and grows less
- Pt has a smaller initial particle diameter and grows more (Pt has larger particle size than PtCo after 75,000 cycles)
- Particle size increase does not have an effect on high current performance at high loadings

![](_page_21_Figure_5.jpeg)

Particle Diameter (nm)

	Mean o (r		
Sample	BOL	EOT	$\Delta d$ (nm)
Pt/HSC			
0.25mgPt/cm <sup>2</sup>	4.3	7.5	3.2
PtCo/HSC			
0.27mgPt/cm <sup>2</sup>	5.2	6.8	1.6

TEM shows 6.8 nm Pt and 6.4 nm PtCo particles after 75,000 cycles

![](_page_21_Picture_9.jpeg)

# **ASTWG: Catalyst Durability Protocol**

- Develop H<sub>2</sub>/Air tests to measure degradation rates under accelerated conditions
- Operate cell @ 90°C to accelerate all degradation mechanisms for the longer life time
  - Potential cycling between 0.65V and 0.925V under 100%RH to accelerate catalyst degradation
  - Introduce RH cycling and dry operation to accelerate membrane degradation
- Degradation rates under accelerated conditions will be compared with rates at typical LDV and HDV operating conditions to determine duration of AST to yield 25,000 hours equivalent durability

Pt		PtCo		
Hours	A/mg-Pt	Hours	A/mg-Pt	
0	0.31	0	0.47	
50	0.23	50	0.33	
100	0.21	100	0.23	
200	0.17	200	0.18	
300	0.13	300	0.14	

![](_page_22_Figure_7.jpeg)

- PtCo has mass activity = 0.47 A/mg<sub>Pt</sub> at BOL while
  Pt = 0.31 A/mg<sub>Pt</sub>
- At 300 hours both have almost the same mass activity of 0.135 A/mg<sub>Pt</sub>

![](_page_22_Picture_10.jpeg)

# Catalyst ASTs (90,000 cycles)

![](_page_23_Figure_1.jpeg)

![](_page_23_Figure_2.jpeg)

- Increasing the current trapezoidal AST from 30K to either 75K or 90K cycles seems to serve the purpose
- Maybe decrease UPL to 0.925V instead of 0.95V to prevent elimination of potentially viable catalysts

## **Durability Performance : High Loading: Effect of Alloying**

![](_page_24_Figure_1.jpeg)

SW AST = Square Wave Catalyst AST (0.6 - 0.95

## Durability Characterization: High Loading - Effect of Alloying

![](_page_25_Picture_1.jpeg)

- Larger PtCo particles exhibit a "spongy" or "core-shell" morphology
- Large PtCo particles (Core shell)
- CCL porosity is similar
- Ionomer more aggregated in PtCo than Pt (I/C 0.95 in PtCo and 0.83 in Pt)
- Both Pt and Co measured ITM (In The Membrane)

![](_page_25_Picture_7.jpeg)

CCL thickness =  $7\mu m$ Pt-depleted zone =  $1\mu m$ PITM ~ $4\mu m$  from CCL

![](_page_25_Picture_9.jpeg)

![](_page_25_Picture_10.jpeg)

# Changes in Electrode Microstructure (AST)

AND DURABILITY

![](_page_26_Figure_1.jpeg)

### **Carbon Corrosion During Drive Cycle** –

**Corrosion Rate Depends upon Changing Potential Amplitude** 

![](_page_27_Figure_2.jpeg)

CO2 Corrosion rate depends on the amplitude of the square potential cycle

![](_page_27_Picture_4.jpeg)

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# Membrane Durability – Radical Scavenger Migration

![](_page_28_Figure_1.jpeg)

![](_page_28_Picture_2.jpeg)

# Stabilization of Cerium

Strong complexing agents for cerium

Strong complexing agents for metal ions, Principle: Size selectivity

![](_page_29_Picture_3.jpeg)

A stronger basicity anion than the counter ion in Nafion side chain, and which can assemble around cerium.

![](_page_29_Figure_5.jpeg)

![](_page_29_Picture_6.jpeg)

# **Transport of Cation Contaminant into the Fuel Cell**

**1. Understanding Cation Transport Mechanism** 

![](_page_30_Figure_2.jpeg)

**2.** Development of GDL to suppress the Cation Transport

![](_page_30_Figure_4.jpeg)

Babu, S.K., et al. Journal of The Electrochemical Society 168.2 (2021): 024501.

### **3.** Estimation of Cation Transport Rates

![](_page_30_Picture_7.jpeg)

![](_page_30_Picture_8.jpeg)

# **GDLs for Cation Transport Suppression**

- Cations are present in PFSAs as radical scavengers (Ce and Mn), contaminants (Ca), byproducts of catalyst dissolution (Co and Ni), and from corrosion of BOP components (Fe)
  - ✤ Fe transport through GDL (from BPP corrosion or BOP components) problematic for membrane durability

#### Studied GDL/MPL morphology effect on cation transport

![](_page_31_Picture_4.jpeg)

MPLs with cracks and hydrophilic GDLs show enhanced cation transport

### **GDL** modification to suppress the Cation transport rates

![](_page_31_Picture_7.jpeg)

Addition of hydrophobic layer Addition of hydrophobic layer

- Novel GDLs suppress cation transport from the flow field
  - Smaller Fe<sup>2+</sup>/Fe<sup>3+</sup> redox couple post- flowfield injection of Fe
  - Solution AST corrodes the 29BC and H23C8 MPL significantly and increases the Fe transport rates

#### Flowfield injection of Fe to study cation transport

![](_page_31_Figure_13.jpeg)

- Subtracting the after curve from the before curve to obtain the Fe redox curve
- Peak current used to calculate the Fe loading in the catalyst layer
- Modified 29BC shows significant suppression of Fe transport even after AST

# **Effect of GDLs on Fe Transport**

GDL/MPL morphology effect on Fe transport mechanism

- GDL modification to suppress the Fe transport rates
- Ex-situ experiment for Fe transport rates estimation

![](_page_32_Picture_4.jpeg)

Hydrophobic layer/ Pseudo MPL

**29BC** 

![](_page_32_Picture_7.jpeg)

![](_page_32_Picture_8.jpeg)

![](_page_32_Picture_9.jpeg)

# **Future Work**

- M2FCT consortium aimed at delivering MEAs and components that meet 2.5 kW/gPGM power (1.07 A/cm<sup>2</sup> current density) at 0.7 V
  - ✤ Targets are end-of-life performance
  - In the second secon
  - ⅍ High durability (1,000,000 miles; 25,000-30,000 hrs)
  - ⅍ Material down-selects ~ year 3
    - $\circ$  Catalyst areas Go/No-Go at Q6
- Analysis
  - Refine models, characterization, and diagnostics for heavy-duty operating conditions
  - **befine operating conditions efficiency and durability trade-offs**
  - Scoordinate and harmonize truck platforms and duty cycles
  - Compare systems with different ratios of fuel cell power and battery energy storage
  - Sensitivity of performance, durability and cost to cell voltage target at EOL
  - ✤ Incorporate membrane durability in system analysis
- Machine learning / Data analysis
  - **Source Correlations of metadata for material and integration studies**

- High-Performance Computing
  - ♥ Unsteady FCS simulations on truck drive cycles
  - **Solution** Electrode and agglomerate structure
  - Upscaling physics-based micro- and nano-scale models to cell models and optimization
- **Durability** 
  - Develop refined ASTs for life-time prediction with heavy-duty materials and operating conditions
    - Refine existing LDV ASTs
    - Develop new ASTs/protocols specific for HDV
  - Propose new protocols in collaboration with ASTWG by end of FY21
  - Electrode stability
  - ♦ Membrane and ionomer durability with additives
  - ✤ High-temperature operating time effect on durability
    - Membrane
    - Catalyst
  - Understand long-term durability effects on other components (GDL, contamination, reversible degradation, carbon corrosion at operating potentials)

![](_page_33_Picture_31.jpeg)

# **Future Work**

### Integration

- Saseline SOA
  - Establish benchmark performance and cost of state-of-art MEA
- Incorporate advance catalyst ink understanding into R2R manufacturing
- Integrate newly developed materials into optimized MEA structures
  - Membranes
  - o lonomers
  - Catalysts
  - Catalyst supports
  - o GDLs
- **& Catalyst layer studies** 
  - Understand cation migration effects on catalyst layer performance
  - Catalyst layer porosity
  - Catalyst ink to structure formation models
- Stransport Properties (Gas phase, water, cations)
  - Catalyst Layer
  - GDL

### Material Development

- ♦ Catalysts & Catalyst supports
  - Pt-Co Intermetallics
  - Metal oxide-metal-carbon junction to stabilize PtM NPs catalysts
  - Nitrogen-Doped PtMN Catalysts and Supports
  - Tailored Pt-nanomaterials, supports, and interfaces
- ✤ Membranes & Ionomers
  - High-conductivity Novel Perfluorinated Ionomers
  - Low Molecular Weight Oligomeric Electrode and Membrane lonomers
  - Composite PFSA Membranes and Ionomer EW and Side-Chain Chemistry
- ♦ Other components (GDLs, Bipolar plates...)
- **Solution** Material and characterization studies

Planned activities include discretionary funding for additional collaborators on identified gaps and needs

![](_page_34_Picture_31.jpeg)

# Who is M2FCT? National Lab Contributors

![](_page_35_Picture_1.jpeg)

Rajesh Ahluwalia Firat Cetinbas Nancy Kariuki John Kopasz Debbie Myers Jaehyung Park Voja Stamenkovic (UCI) Xiaohua Wang Andrew Star

![](_page_35_Picture_3.jpeg)

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![](_page_35_Picture_5.jpeg)

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**DOE EERE HFTO** 

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![](_page_35_Picture_11.jpeg)

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![](_page_35_Picture_13.jpeg)

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![](_page_35_Picture_15.jpeg)

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![](_page_35_Picture_17.jpeg)

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# **Acknowledgements**

DOE EERE Hydrogen and Fuel Cell Technologies Office

**Technology Managers:** 

#### **Greg Kleen, Dimitrios Papageorgopoulos**

![](_page_36_Picture_4.jpeg)

http://millionmilefuelcelltruck.org

![](_page_36_Picture_6.jpeg)

#### **User Facilities**

DOE Office of Science: SLAC, LBNL-Advanced Light Source, LBNL-Molecular Foundry, ANL-Advanced Photon Source, LBNL-Molecular Foundry, ORNL-Center for Nanophase Materials Sciences, ANL-Center for Nanostructured Materials, NIST: BT-2

![](_page_36_Picture_9.jpeg)