

# 2010 DOE Hydrogen Program Annual Merit Review

## ***SPIRE***

*Sustained Power Intensity with Reduced Electrocatalyst*

(aka: Durability of Low Pt Fuel Cells Operating at High Power Density)

Scott Blanchet (PI)

Nuvera Fuel Cells

6/8/2010

FC014

# Overview

## Timeline

- Start: October 1, 2009
- End: September 30, 2012
- 18% Complete (4/9/2010)

## Budget

- \$5.096M Total Project
  - \$3.875M DOE Share
  - \$1.221M Contractor Share
- \$0.231M received in FY09
- \$1.769M planned for FY10

## Barriers

- Barriers addressed
  - Stack Durability with Cycling:  
**target: 5000hrs (2015)**
  - Stack Cost:  
**target: \$15/kW (2015)**

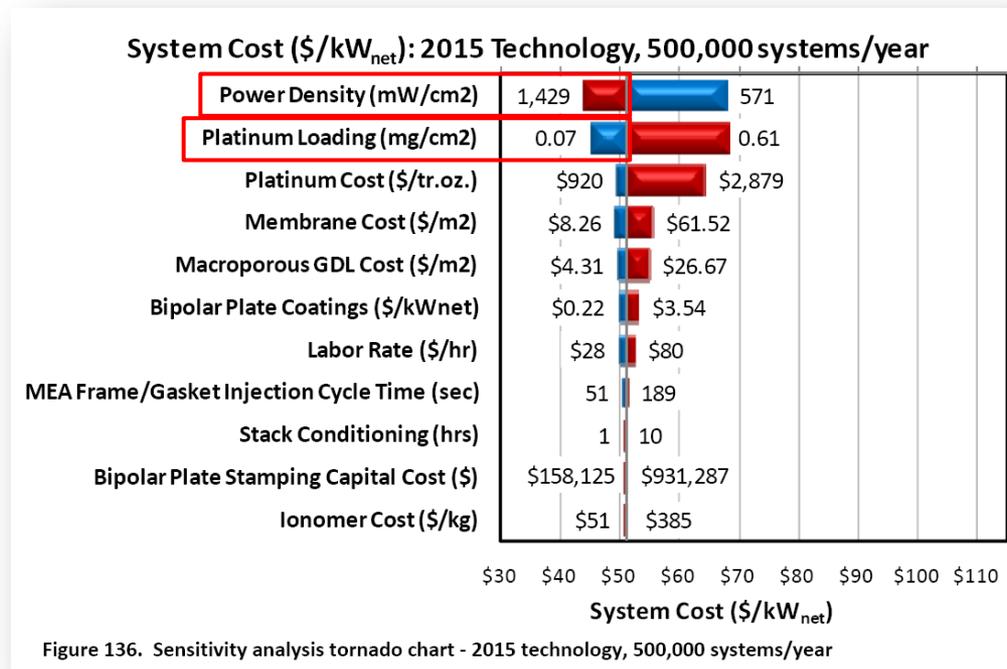
## Partners

- Los Alamos National Laboratory
- Argonne National Laboratory
- Nuvera Fuel Cells (lead)



# Relevance

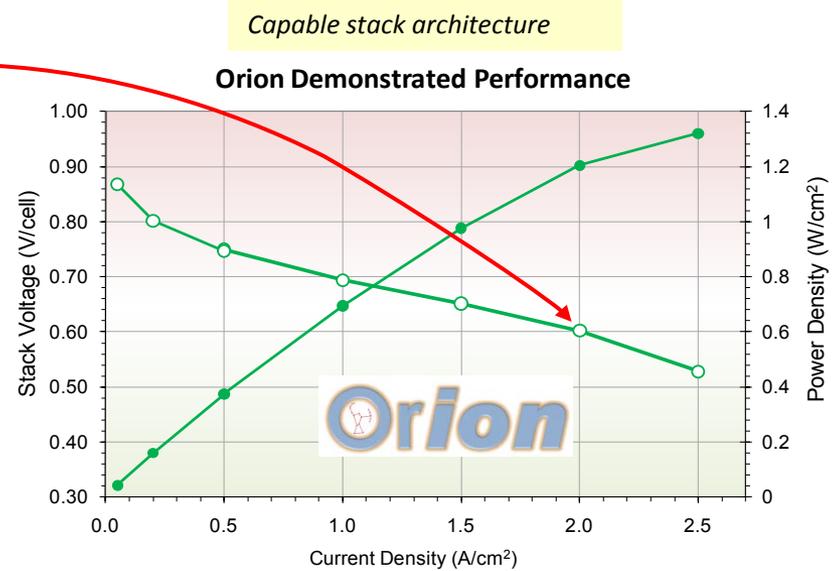
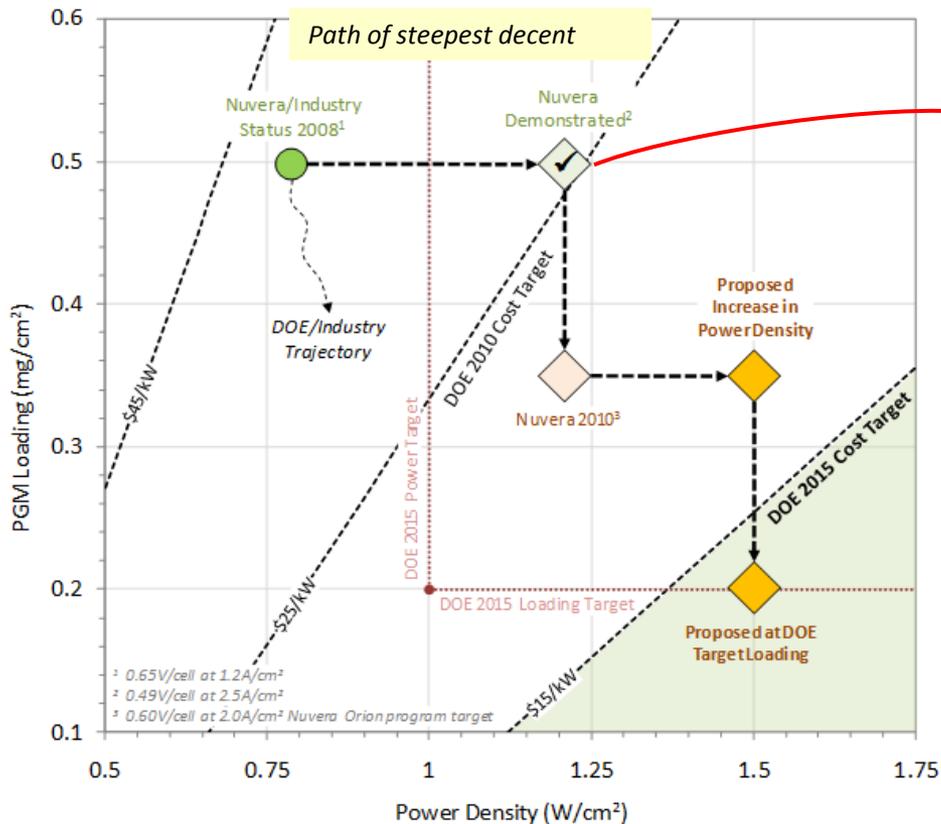
- The objective of the SPIRE program is to study decay mechanisms and identify strategies to assure the durability of fuel cells *capable of achieving* DOE's 2015 cost target.
- The most significant *enablers* for achieving stack cost are increased power density and reduced platinum loading.



Credit: James, Brian D. & Jeffrey A. Kalinoski, "Mass Production Cost Estimation for Direct H<sub>2</sub> PEM Fuel Cell Systems for Automotive Applications: 2008 Update," March 26, 2009

# Approach

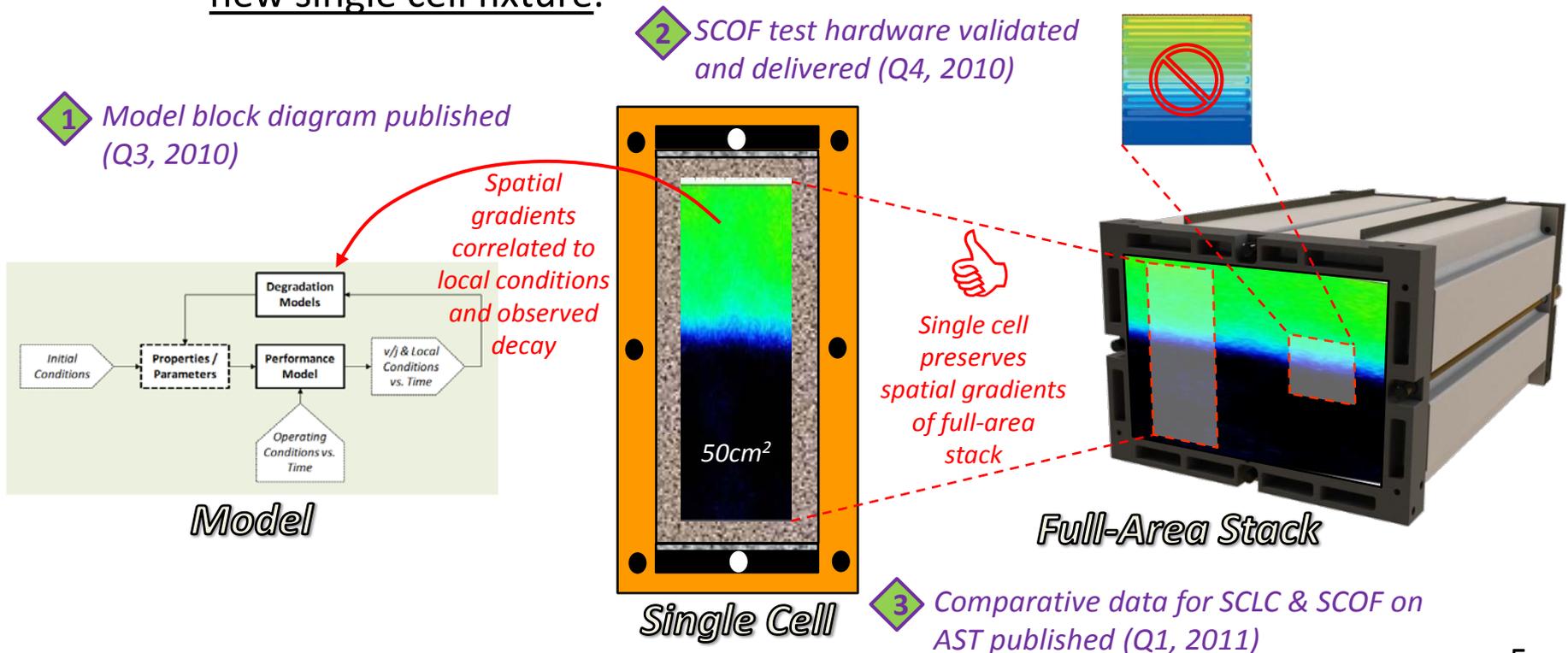
- The technical approach of the SPIRE program is to elucidate the critical durability mechanisms for a stack operating at a power density and Pt loading that can achieve DOE's 2015 cost target.



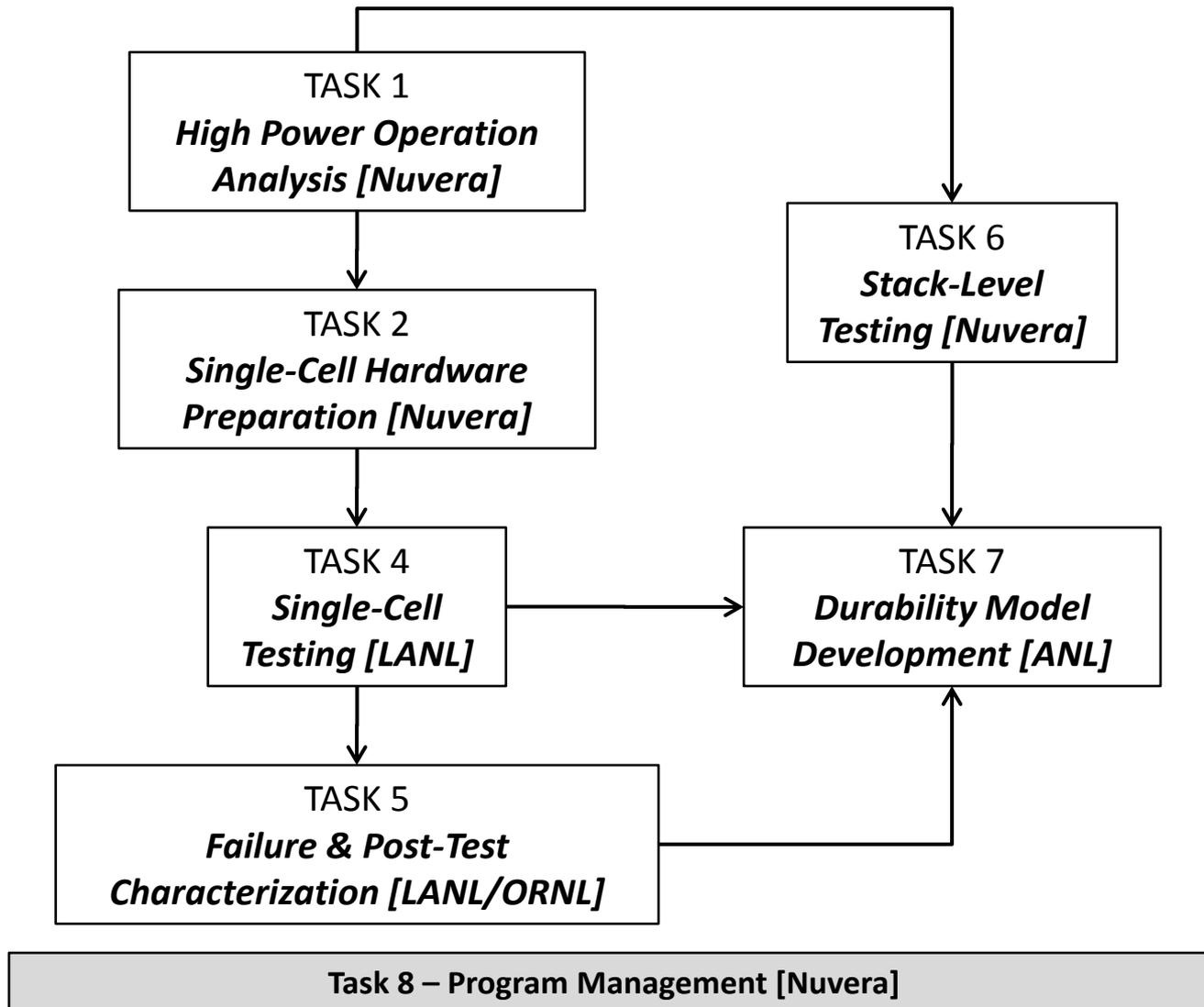
*Iso-cost curves fit to 2008 DTI results for 500k vehicles per year with 2015 technology.*

# Approach (cont.)

- The SPIRE program will balance modeling and experimentation.
  - Mechanistic models of the salient decay mechanisms will be calibrated using single cell experimental results from an advanced fixture capable of high power density operation.
  - Full-area stack testing will be used to validate the results obtained on the new single cell fixture.



# Task Flow Chart

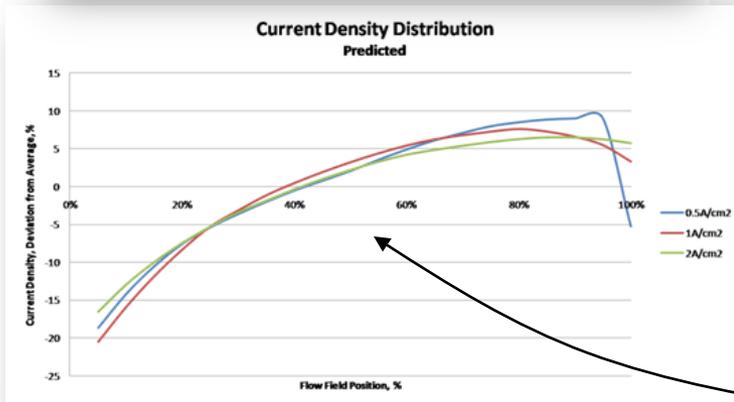
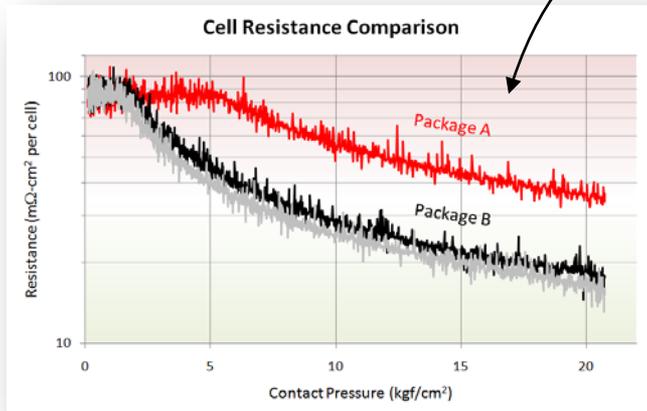
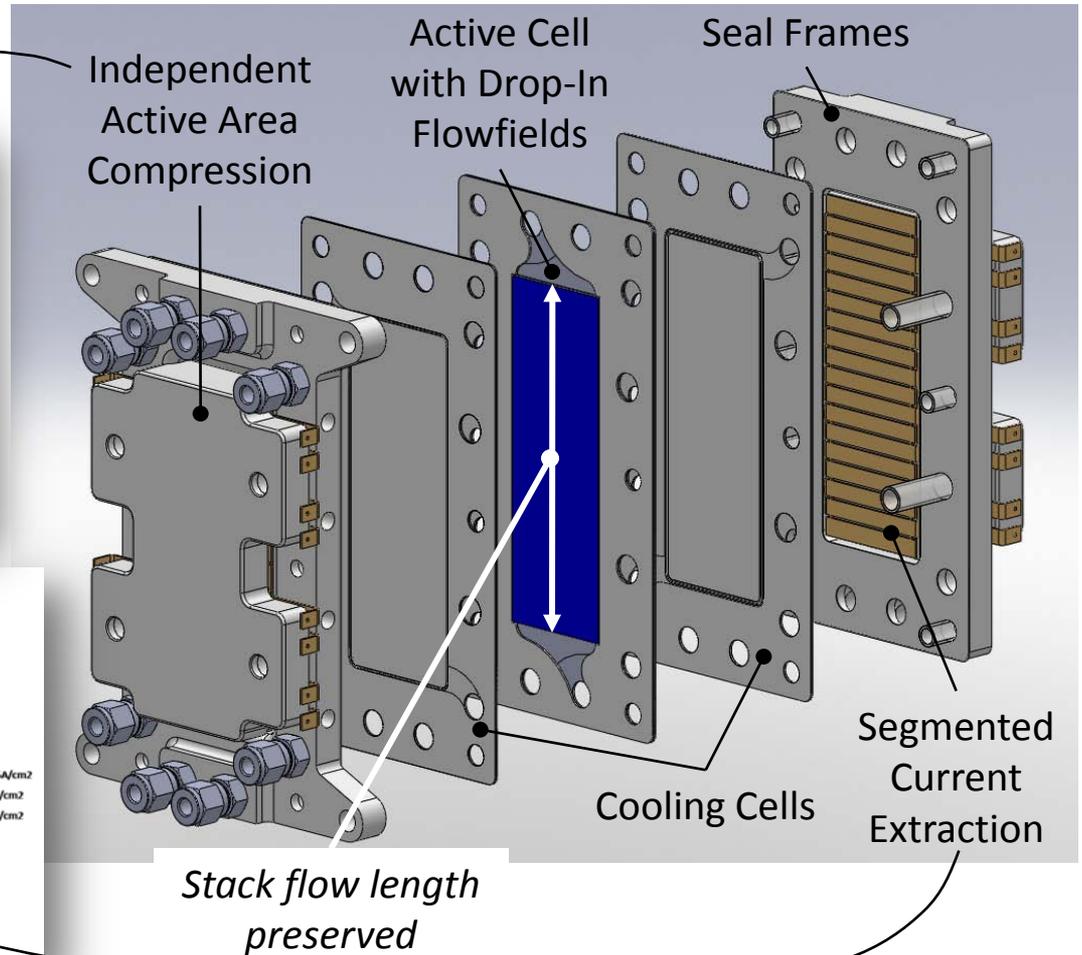


# Technical Progress – Single Cell

## Important Characteristics

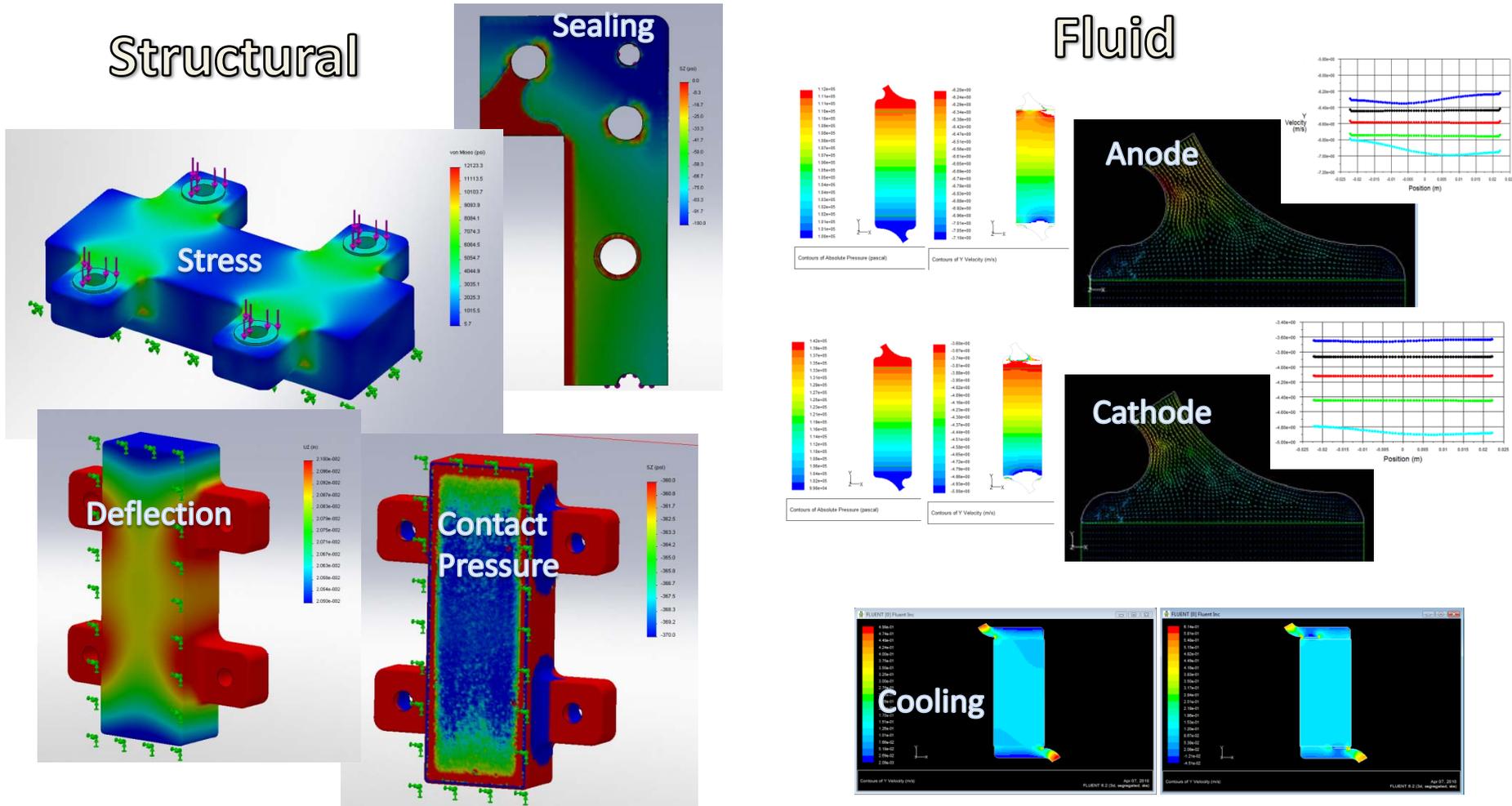
- Capable of high RCD
- Flowfield flexibility.
- Preserve stack-level gradients
- Uniformity & control of AA compression
- Measurement of current distribution

*New single cell fixture critical to program success.*



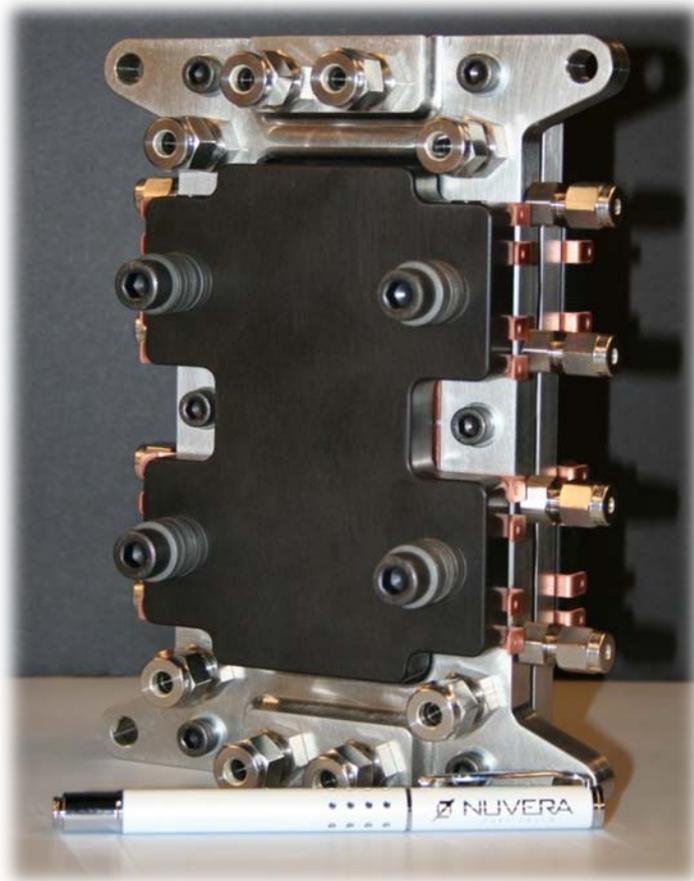
# Technical Progress – Single Cell

*New single cell fixture functionality extensively verified and optimized.*

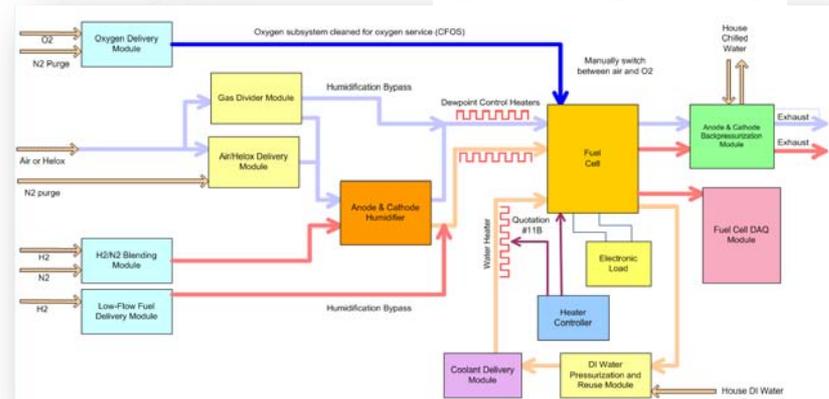


# Technical Progress – Single Cell

*New single cell fixture 1<sup>st</sup> prototypes in-house for qualification and delivery to LANL.*



*Single cell test stand specified and ordered.*

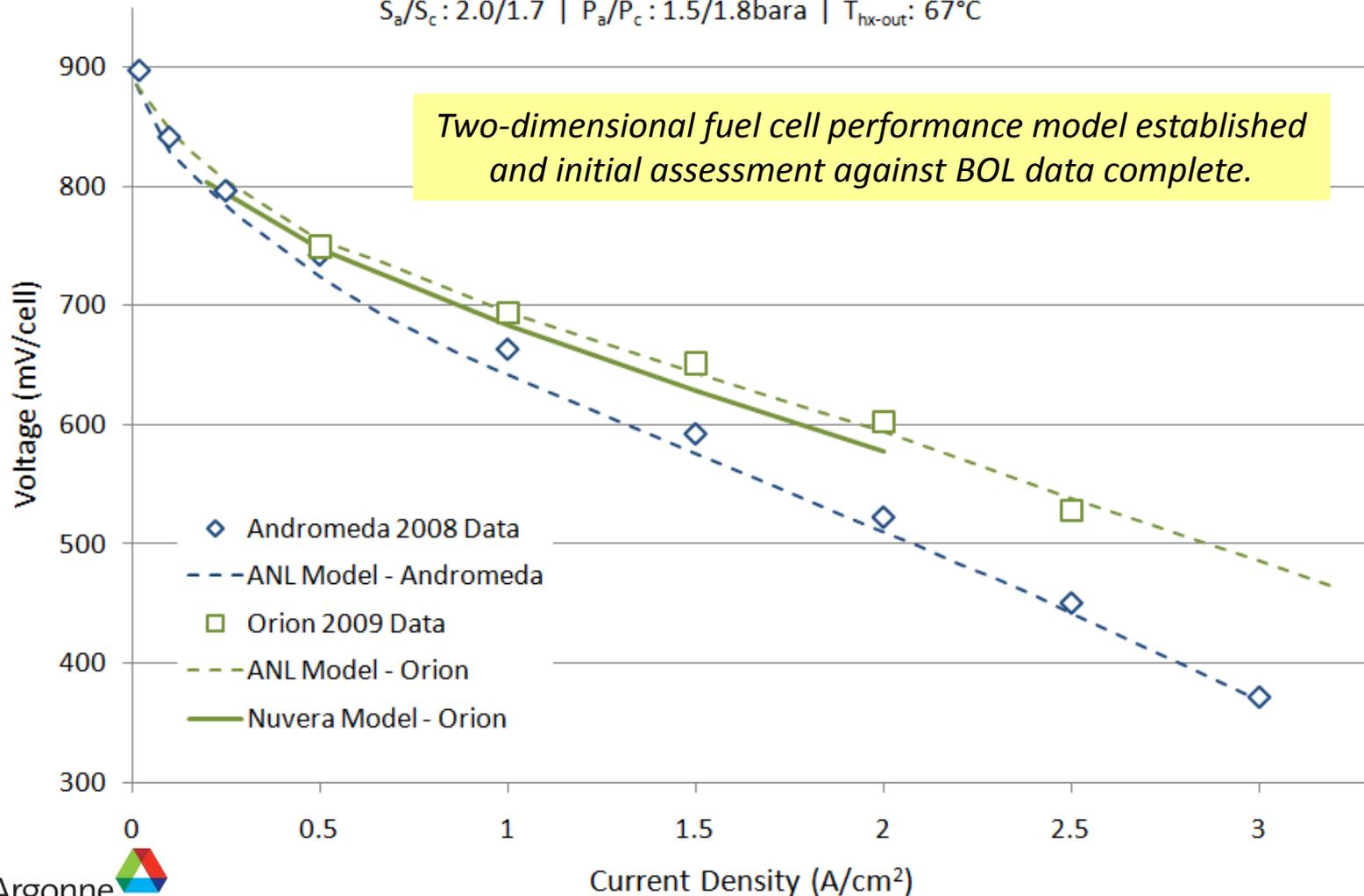


*FCTT combined load/humidity cycle specifies fast transients between wet & dry conditions and requires a special humidification configuration.*

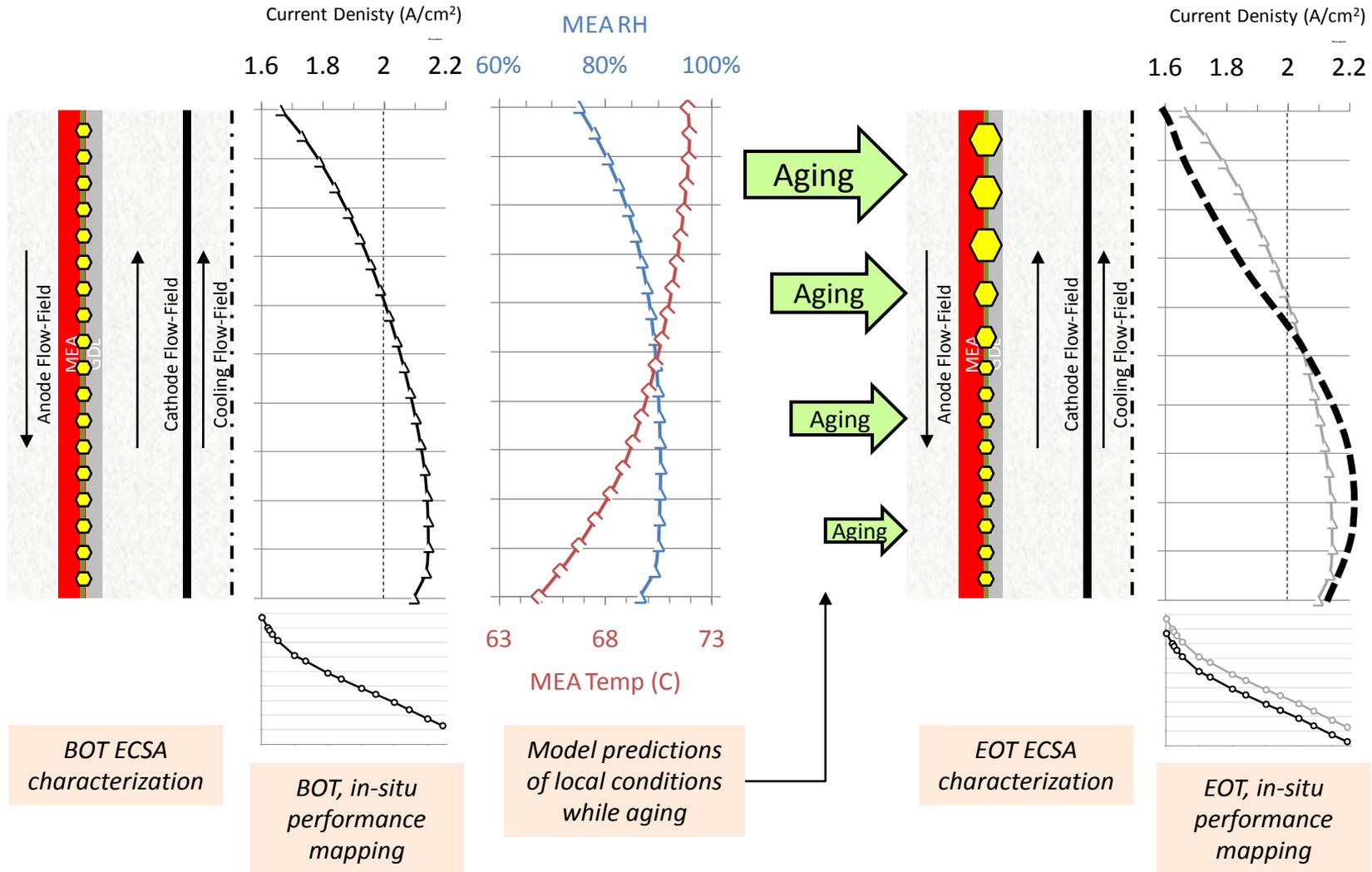
# Technical Progress – Performance Model

## Open Flowfield Performance Model Results

$S_a/S_c$ : 2.0/1.7 |  $P_a/P_c$ : 1.5/1.8bara |  $T_{hx-out}$ : 67°C



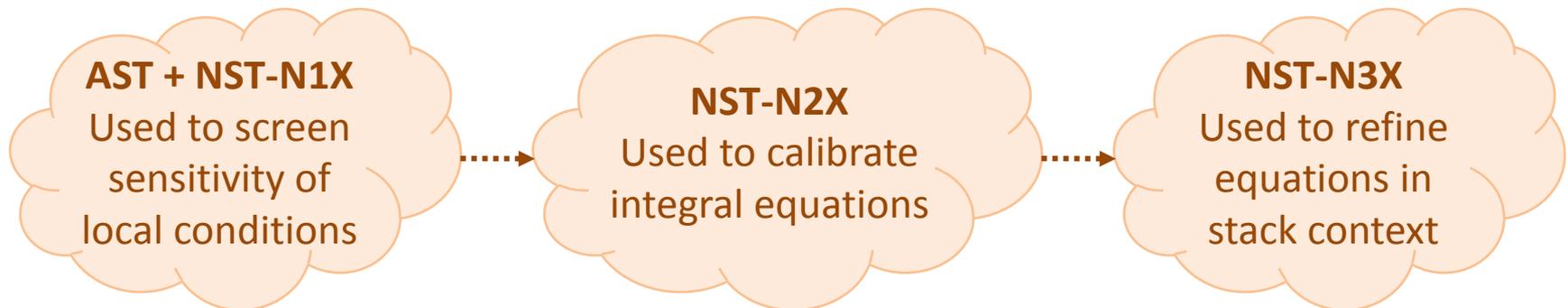
# Technical Progress – Decay Model (Concept)



$$\Delta_{ECSA} = \int f(\text{temp, RH, current, ...}) dt \rightarrow \Delta V_{BOT-EOT} = f(\Delta_{ECSA}, \Delta_{activity})$$

# Technical Progress – Decay Model (Approach)

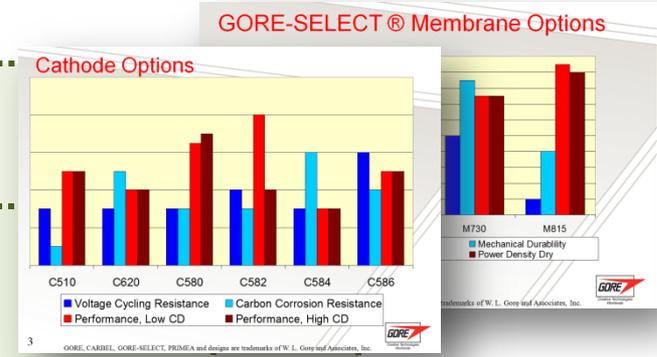
- Enormous suite of potential decay variables
  - Modeling focus will be impact of current and local conditions on catalyst – **ECSA, activity**
- Hypothesis: Change in ECSA and activity is an integral function of exposed time to local conditions (temp, humidity, potential, cycling, ...)
  - Physically-based, empirically calibrated, integral equations will be developed from the single-cell database



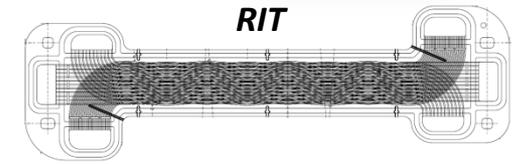
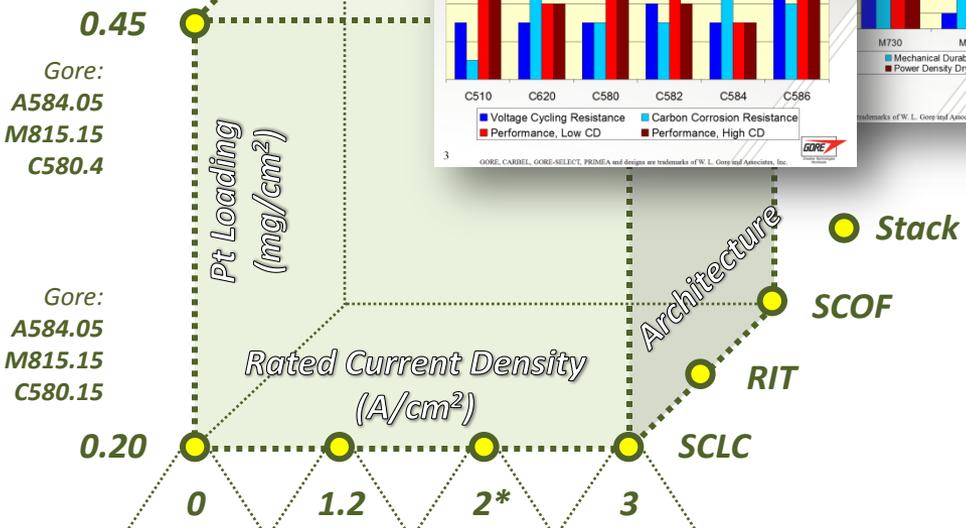
# Technical Progress – Experimental Design

Design of the experimental campaign has been established.

Materials selected based on Nuvera historical experience and availability from Gore



Architectures selected to maximize applicability across industry



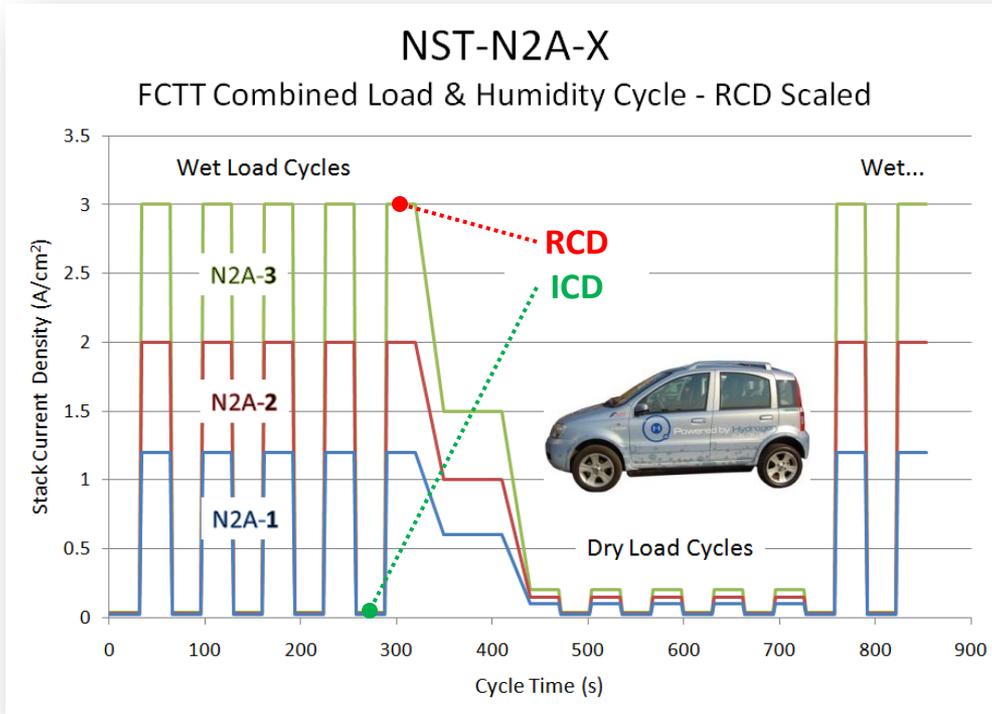
AST	NST			Purpose
B1				Benchmarking+Electrocatalyst cycle stability with and without current <i>B1* potential limits equivalent to N1A-2</i>
B1*		N1A-2		
B4		N1B-2		Membrane mechanical stability with and without current
	N2A-1	N2A-2	N2A-3	FCTT combined load & humidity over range of RCD/ICD
		N3A-2	N3A-3	Combined city/highway drive cycle over range of RCD/ICD



\* mid-current point added based on FCTT recommendations

# Technical Progress – Test Cycles

Combined test cycles scaled based on proposed stack **rated current density (RCD)** and constant turndown ratio of stack power to establish **idle current density (ICD)**.



**RCD**  
**ICD**

**3.0 A/cm<sup>2</sup>**  
**0.04 A/cm<sup>2</sup>**



**No. Cells**  
250cm<sup>2</sup>, 87kW<sub>DCgross</sub>

**258**  
(0.45V/cell @ RCD)

**2.0 A/cm<sup>2</sup>**  
**0.03 A/cm<sup>2</sup>**



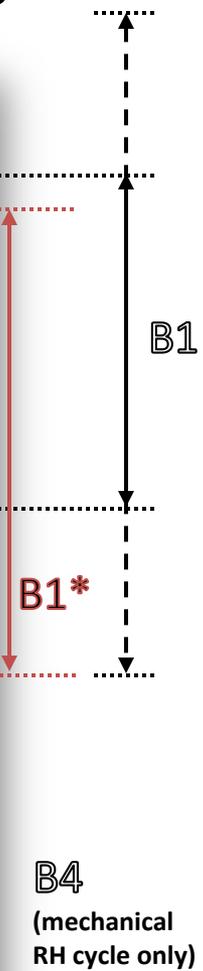
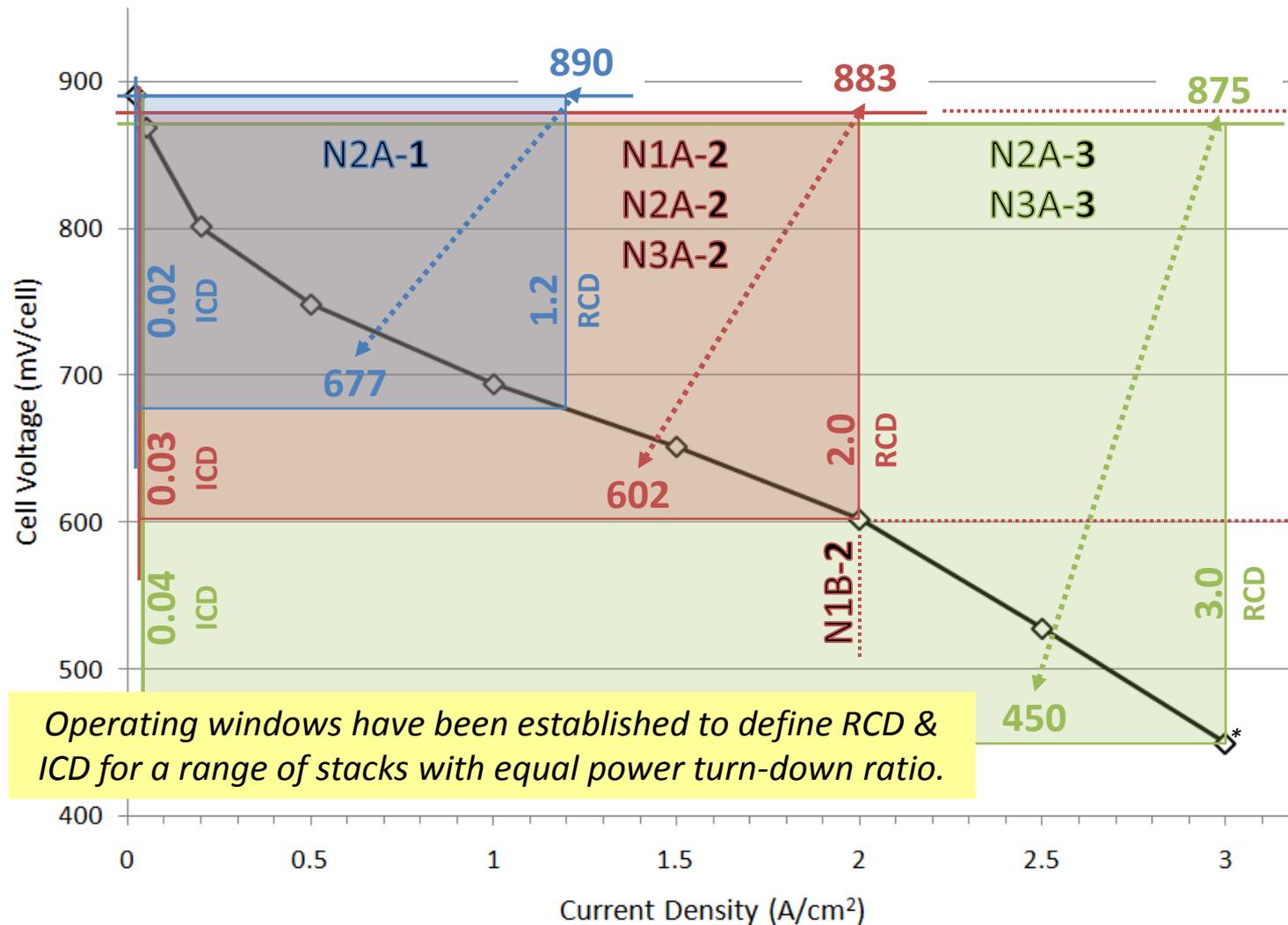
**290**  
(0.60V/cell @ RCD)

**1.2 A/cm<sup>2</sup>**  
**0.02 A/cm<sup>2</sup>**



**427**  
(0.68V/cell @ RCD)

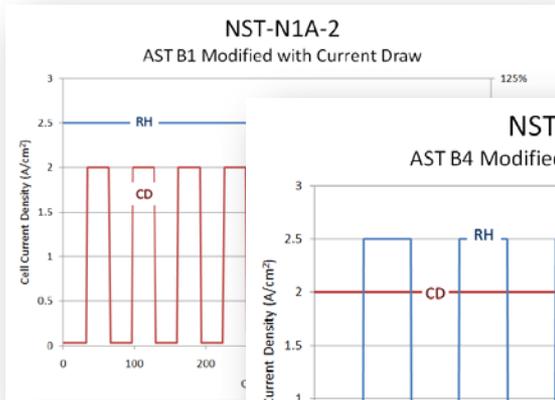
# Technical Progress – Operating Windows



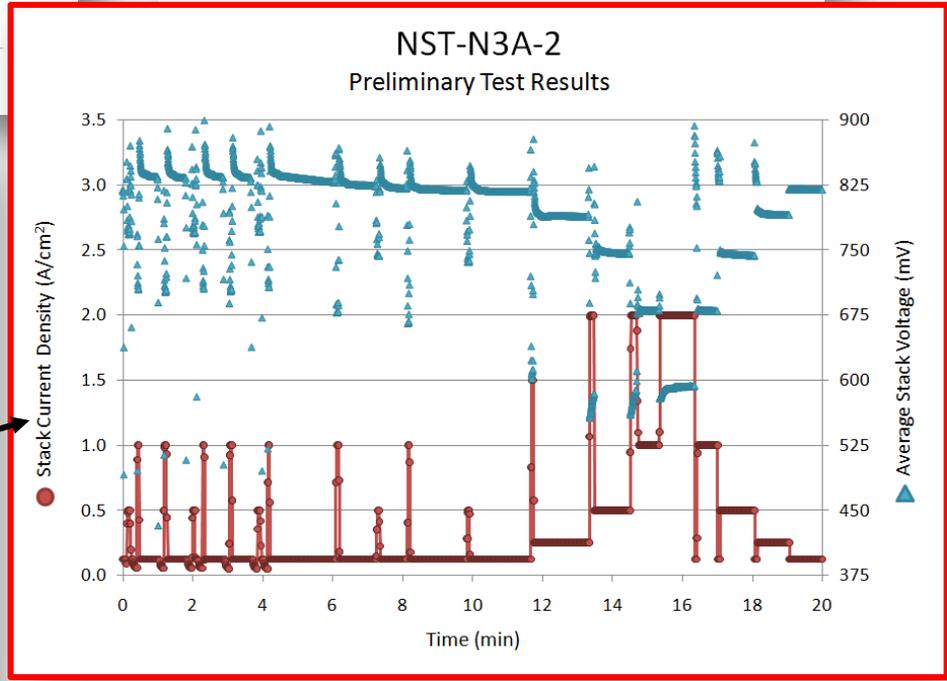
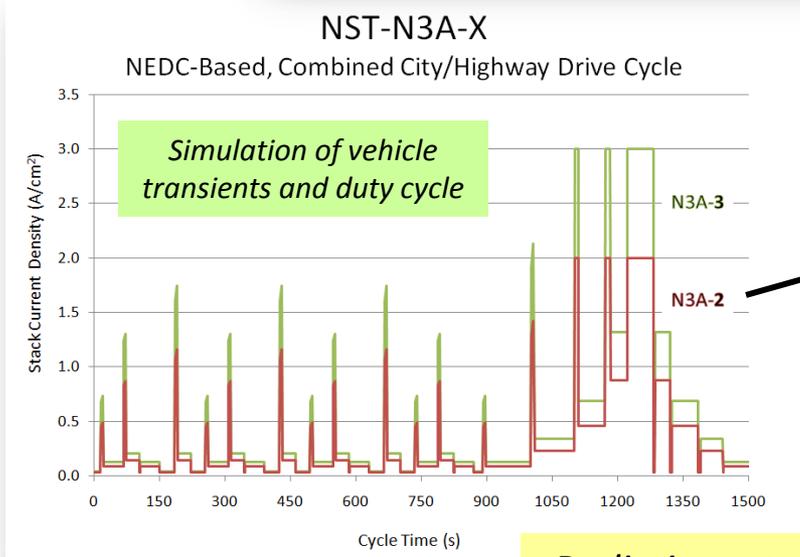
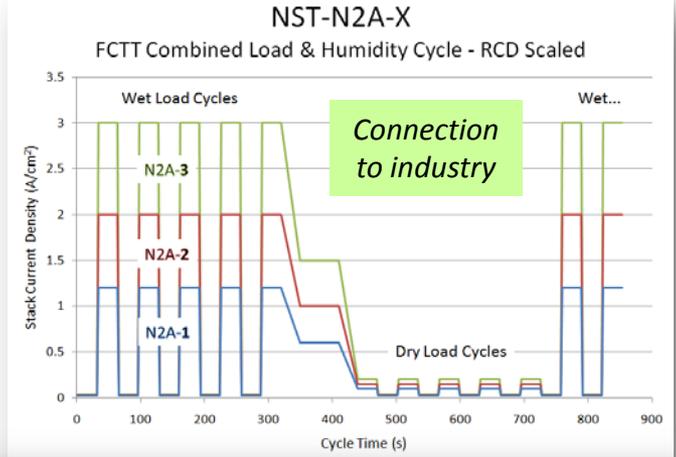
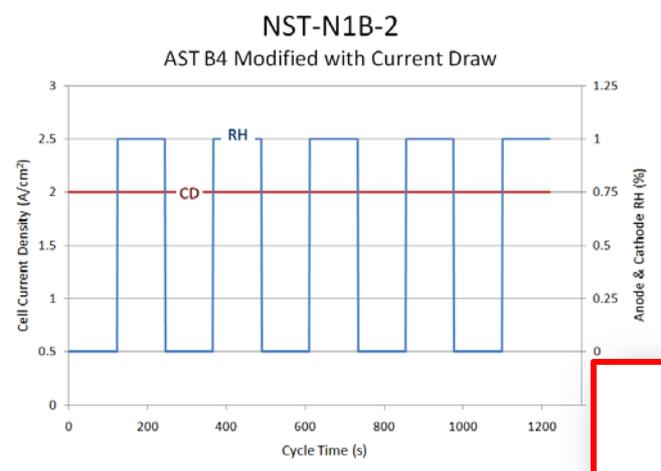
Operating windows have been established to define RCD & ICD for a range of stacks with equal power turn-down ratio.

\*3A/cm² point extrapolated from data up to 2.5A/cm²

# Technical Progress – Test Cycles



Connection to AST



Preliminary test cycles established and under evaluation.



# Collaborations

- Nuvera Fuel Cells – prime contractor
- Los Alamos National Lab – partner
  - Single cell AST/NST testing, post-test characterization
  - Several telecons held to refine test matrix and NSTs
- Argonne National Lab – partner
  - Fuel cell reference and durability model development
  - Several telecons held to review vehicle thermal management strategies/constraints, definition of stack RCD and model architecture
- W.L. Gore & Associates – vendor
  - Materials supply
  - Supply arrangements finalized and baseline material sets selected
- Fuel Cell Tech Team – reviewer
  - FCTT SPIRE Project review: January 13, 2010
  - FCTT interaction on vehicle thermal management at DOE Fuel Cell Pre-Solicitation Workshop: 3/16/2010

# Proposed Future Work

- 3 Milestones in the next period:
  - Model block diagram published (Q3, 2010)
  - SCOF test hardware validated and delivered (Q4, 2010)
  - Comparative data for SCLC and SCOF on AST protocols published (Q1, 2011)
- Initiate and complete AST tests (SCLC)
- Install, validate and begin NST testing (SCOF, RIT – LANL, Nuvera)
- Initiate post-test characterization
- Screen materials and test cycles and begin stack durability testing
- Establish initial decay equations and initiate calibration with single cell results

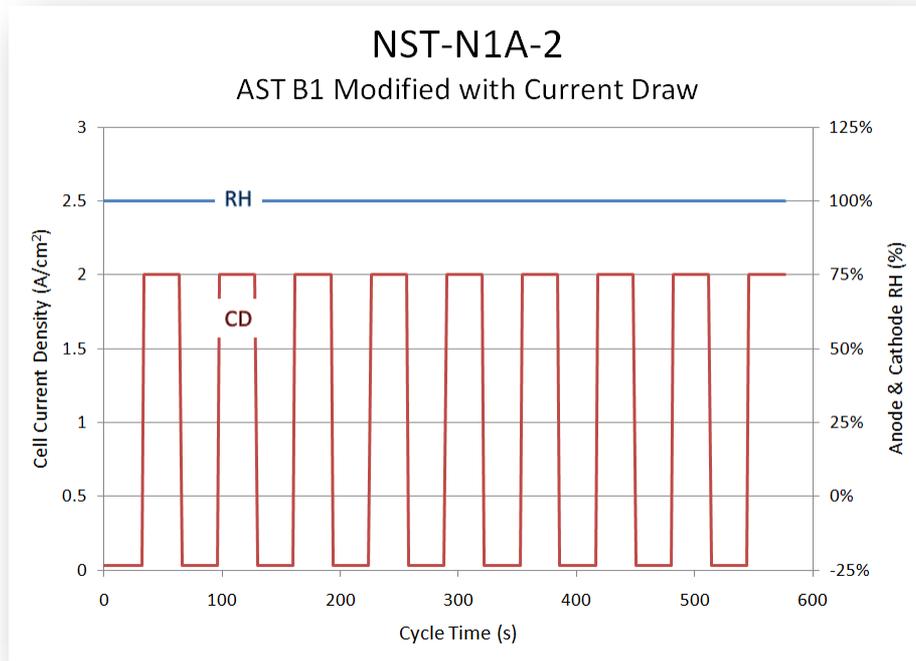
# Summary

- SPIRE is tackling two of the most elusive targets in the hydrogen program – cost and durability
- Good progress has been made on the single cell hardware and it should provide a fundamentally new platform for study
- The relationship of the SPIRE “model” to other DOE durability projects is still fuzzy – We need DOE’s leadership! (*durability task force*)
- The experimental campaign is robust, but our goals are ambitious we must take care not to overextend and lose focus
- The team is excited by ongoing industry interaction and looks forward to continued debate and learning



# Supplemental Slides

# NST-N1A-2



## Goals:

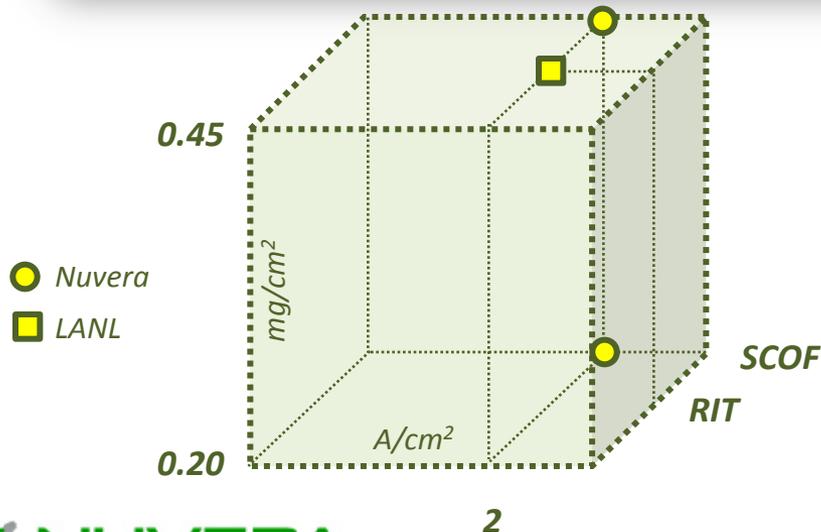
- N1A-2 vs. B1\* → Direct assessment of current draw on catalyst decay.
- SCOF vs. RIT → Direct assessment of architecture on catalyst decay w/ current draw.

## Measurements:

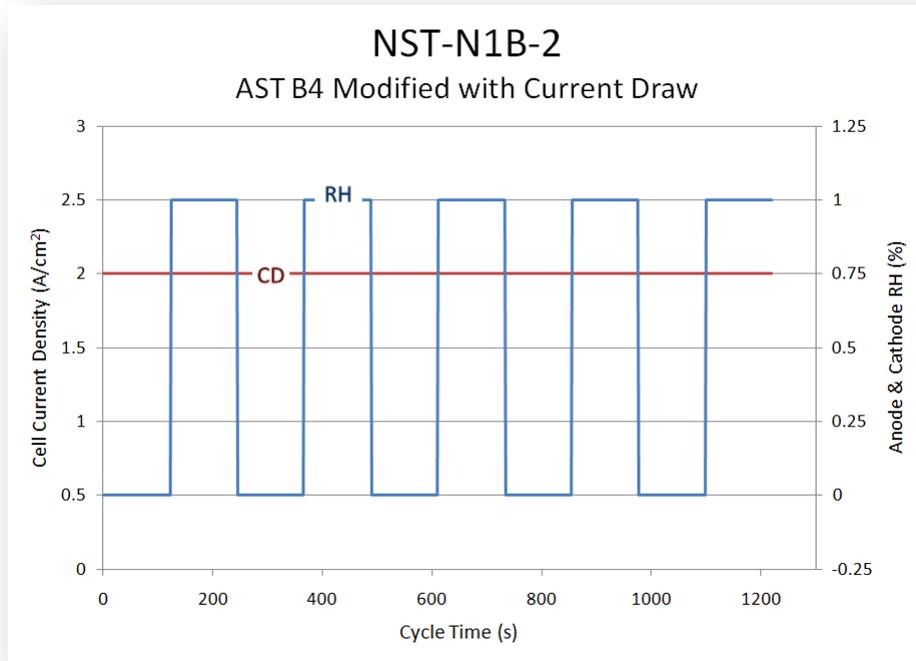
- All per B1 protocol
- All process parameters vs. time
- Current distribution vs. time
- EIS vs. time
- O<sub>2</sub> & Helox pol curves

## Characterization:

- BOT / EOT Areal Variation of thicknesses/morphology (SEM)
- Other (TBD)



# NST-N1B-2



## Goals:

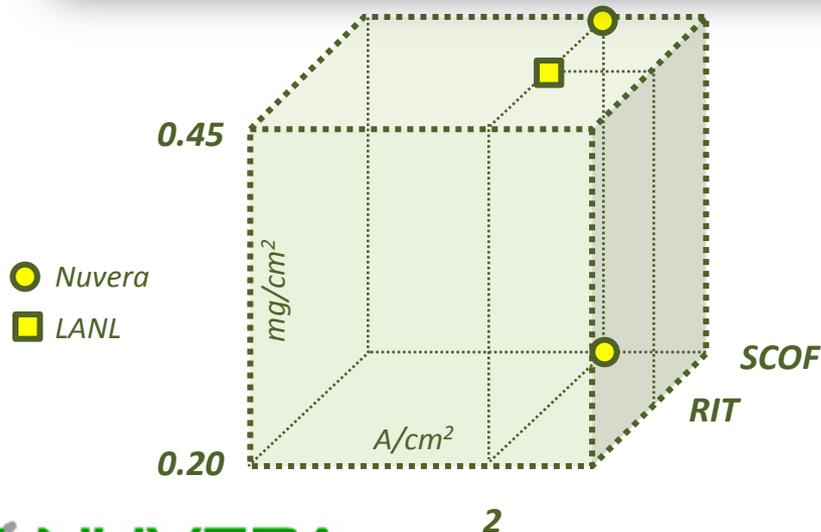
- N1B-2 vs. B4 → Direct assessment of current draw on membrane durability.
- SCOF vs. RIT → Direct assessment of architecture on membrane durability w/ current draw.

## Measurements:

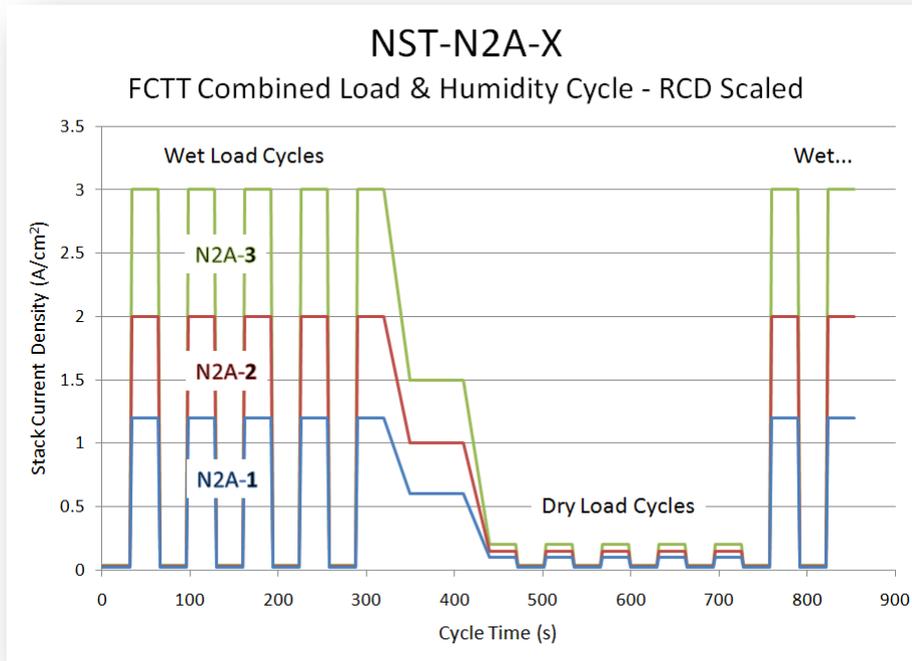
- Electrochemical cross-over
- All process parameters vs. time
- Polarization curve vs. time
- Current distribution vs. time
- EIS vs. time
- O<sub>2</sub> & Helox pol curves

## Characterization:

- BOT / EOT Areal Variation of thickness/morphology (SEM)
- Other (TBD)



# NST-N2A-X



## Goals:

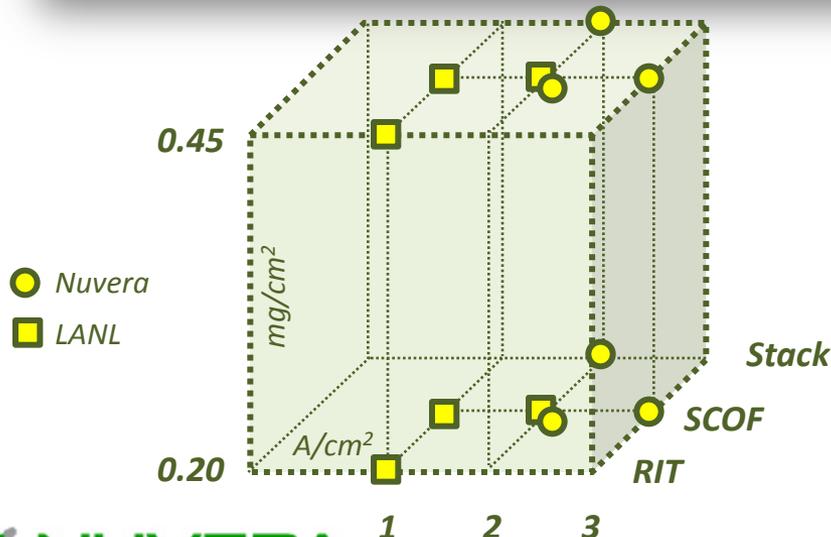
- SCOF vs. RIT → Direct assessment of architecture and Pt loading on combined cycle durability at low-current point.
- 1 vs. 2 vs. 3 A/cm<sup>2</sup> → Direct assessment of current density on combined cycle durability for high and low Pt loading.
- Nuvera vs. LANL → Direct assessment of lab-to-lab variation.
- SCOF vs. Stack → Validation of new fixture (GNG, end Q8)

## Measurements:

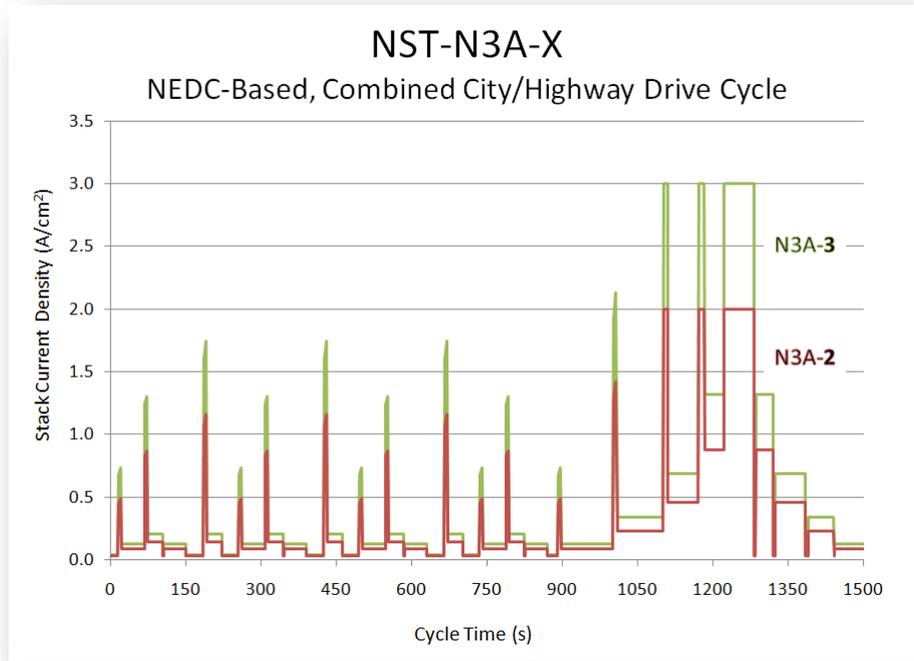
- All process parameters vs. time
- Polarization curve vs. time
- Current distribution vs. time
- EIS vs. time
- O<sub>2</sub> & Helox pol curves
- Electrochemical cross-over

## Characterization:

- BOT / EOT Areal Variation of thicknesses/morphology (SEM)
- Other (TBD)



# NST-N3A-X



## Goals:

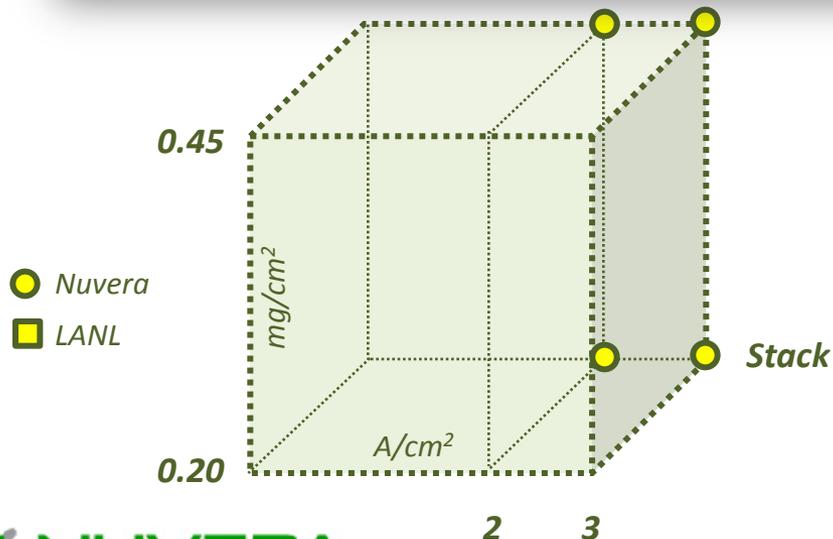
- 2 vs. 3 A/cm<sup>2</sup> → Direct assessment of current density on combined drive cycle durability for real stack.
- 0.20 vs. 0.45 mg/cm<sup>2</sup> → Direct assessment of Pt loading on combined drive cycle durability for real stack.

## Measurements:

- All process parameters vs. time
- Polarization curve vs. time
- Current distribution vs. time
- EIS vs. time
- O<sub>2</sub> & Helox pol curves
- Electrochemical cross-over

## Characterization:

- BOT / EOT Areal Variation of thicknesses/morphology (SEM)
- Other (TBD)



# SPIRE Test Definitions

Protocol ID	Definition	Objective
B1	DOE AST B1 - Electrocatalyst Cycle	Benchmarking (4)
B4	DOE AST B4 - Membrane Mechanical Cycle	Benchmarking (4)
B1*	Modified DOE AST B1 - Electrocatalyst Cycle. Potential range adjusted to match N1A-2	RCD (1); Pt load (2); Flowfield (3)
N1A-2	Fully-humidified current cycle to mimic B1*	RCD (1); Pt load (2); Flowfield (3)
N1B-2	Humidity cycle with constant current to mimic B4	RCD (1); Pt load (2); Flowfield (3)
N2A-1	FCTT combined current and humidity cycle as received (0.02 to 1.2 A/cm <sup>2</sup> current range)	RCD (1); Pt load (2); Flowfield (3)
N2A-2	FCTT combined current and humidity cycle scaled to RCD=2.0 (0.03 to 2.0 A/cm <sup>2</sup> current range)	RCD (1); Pt load (2); Flowfield (3)
N2A-3	FCTT combined current and humidity cycle scaled to RCD=3.0 (0.04 to 3.0 A/cm <sup>2</sup> current range)	RCD (1); Pt load (2); Flowfield (3)
N3A-2	Simulated city/highway power cycle based on EUDC drive cycle and engine with RCD=2.0	Demo/Val (5)
N3A-3	Simulated city/highway power cycle based on EUDC drive cycle and engine with RCD=3.0	Demo/Val (5)

Architecture ID	Description
SCLC	Standard 50 cm <sup>2</sup> graphite, serpentine land-channel cell
SCOF	New 50 cm <sup>2</sup> metal, open flowfield cell
RIT	Benchmark 50cm <sup>2</sup> metal, RIT/GM herringbone cell
Stack	Nuvera 250 cm <sup>2</sup> Orion stack (~8 cells, single MEA material set)

Material ID	Description
A	WL Gore A584.05/M815.15/C580.4 MEA with XXX GDL
B	WL Gore A584.05/M815.15/C580.15 MEA with XXX GDL

Objective ID	Objective
1	Determine the impact of rated current density (RCD) on fuel cell decay mechanisms.
2	Determine the impact of low Pt loading on fuel cell decay mechanisms.
3	Determine the impact of flowfield architecture on fuel cell decay mechanisms.
4	Benchmark program MEA materials against industry (LANL) database of AST performance
5	Demonstrate advanced materials and operating protocols under high RCD in realistic automotive

# Technical Progress – DRAFT Test Measurements

Protocol ID	In-Situ Measurements	Basis	EOT Characterization
B1	per DOE AST-B1 protocol definition EIS preceding polarization curve measurements Repeat polarization on O2 and Helox	catalyst, contact & diffusion changes GDL/catalyst diffusion changes	TBD
B1*	per DOE AST-B1 protocol definition EIS preceding polarization curve measurements repeat polarization on O2 and Helox	catalyst, contact & diffusion changes GDL/catalyst diffusion changes	TBD
N1A-2	per DOE AST-B1 protocol definition all process parameters during cycle at high and low potential points all process parameters, 1-second resolution for 10 full cycles preceding polarization curve measurements current density distribution during polarization curve EIS preceding polarization curve measurements repeat polarization on O2 and Helox	assurance of protocol time-series behavior correlation of decay to local conditions catalyst, contact & diffusion changes GDL/catalyst diffusion changes	TBD
B4	per DOE AST-B4 protocol definition electrochemical crossover every 250 hours EIS every 250 hours	membrane thinning/decay catalyst, contact & diffusion changes	TBD
N1B-2	electrochemical crossover every 250 hours all process parameters during cycle at high and low RH points polarization curve every 250 hours current density distribution during polarization curve repeat polarization on O2 and Helox all process parameters, 1-second resolution for 10 full cycles preceding polarization curve measurements EIS preceding polarization curve measurements	membrane thinning/decay assurance of protocol overall health evaluation (standard conditions) correlation of decay to local conditions GDL/catalyst diffusion changes time-series behavior catalyst, contact & diffusion changes	TBD
N2A-1,2,3	all process parameters during cycle at high and low potential points polarization curve every 250 hours current density distribution during polarization curve repeat polarization on O2 and Helox all process parameters, 1-second resolution for 10 full cycles preceding polarization curve measurements EIS preceding polarization curve measurements electrochemical crossover every 250 hours	assurance of protocol overall health evaluation (standard conditions) correlation of decay to local conditions GDL/catalyst diffusion changes time-series behavior catalyst, contact & diffusion changes membrane thinning/decay	TBD
N3A-2,3	all process parameters during cycle at high and low potential points polarization curve every 250 hours current density distribution during polarization curve repeat polarization on O2 and Helox all process parameters, 1-second resolution for 10 full cycles preceding polarization curve measurements EIS preceding polarization curve measurements electrochemical crossover every 250 hours	assurance of protocol overall health evaluation (standard conditions) correlation of decay to local conditions GDL/catalyst diffusion changes time-series behavior catalyst, contact & diffusion changes membrane thinning/decay	TBD

Characterization ID	Method
A	Pt particle size & distribution (XRD)
B	Component thicknesses and degree of lamination (SEM)
C	Pt & electrode morphology (TEM, case-by-case)
D	GDL contact angle
E	Contaminant mapping (XRD?)
Z-"X"	Areal variation in any of the above

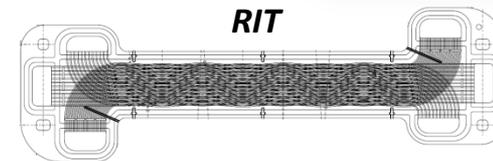
# SPIRE Test Matrix

Test ID	Protocol ID	Architecture ID	Material ID	Objective ID	Test Location
TID-1	B1	SCLC	A	4	LANL
TID-2	B1	SCOF	A	3	LANL
TID-3	B1	SCLC	B	4	LANL
TID-4	B4	SCLC	A	4	LANL
TID-5	B4	SCOF	A	3	LANL
TID-6	B4	RIT	A	3	LANL
TID-7	B1*	SCOF	A	1,2,3	LANL
TID-8	B1*	SCLC	B	1,2,3	LANL
TID-9	B1*	SCOF	B	1,2,3	LANL
TID-10	N1A-2	SCOF	A	1,2,3	Nuvera
TID-11	N1A-2	SCOF	B	1,2,3	Nuvera
TID-12	N1A-2	RIT	A	3	LANL
TID-13	N1B-2	SCOF	A	1,3	Nuvera
TID-14	N1B-2	SCOF	B	1,2,3	Nuvera
TID-15	N1B-2	RIT	A	1,3	LANL
TID-16	N2A-1	SCOF	A	1,2,3	LANL
TID-17	N2A-1	RIT	A	1,2,3	LANL
TID-18	N2A-2	SCOF	A	1,2,3, GNG	Nuvera
TID-19	N2A-2	SCOF	A	1,2,3	LANL
TID-20	N2A-2	Stack	A	5, GNG	Nuvera
TID-21	N2A-3	SCOF	A	1,2,3	Nuvera
TID-22	N2A-1	SCOF	B	1,2,3	LANL
TID-23	N2A-1	RIT	B	1,2,3	LANL
TID-24	N2A-2	SCOF	B	1,2,3	Nuvera
TID-25	N2A-2	SCOF	B	1,2,3	LANL
TID-26	N2A-3	SCOF	B	1,2,3	Nuvera
TID-27	N3A-2	Stack	A	1,2,3,5	Nuvera
TID-28	N3A-2	Stack	B	1,2,3,5	Nuvera
TID-29	N3A-3	Stack	A	1,2,3,5	Nuvera
TID-30	N3A-3	Stack	B	1,2,3,5	Nuvera
TID-31	N2A-2	Stack	B	1,2,3,5	Nuvera

Accounting	
SCLC @ LANL	4
SCOF @ LANL	8
RIT @ LANL	5
SCOF @ Nuvera	8
Stack @ Nuvera	6
LANL Total	17
Nuvera Total	14

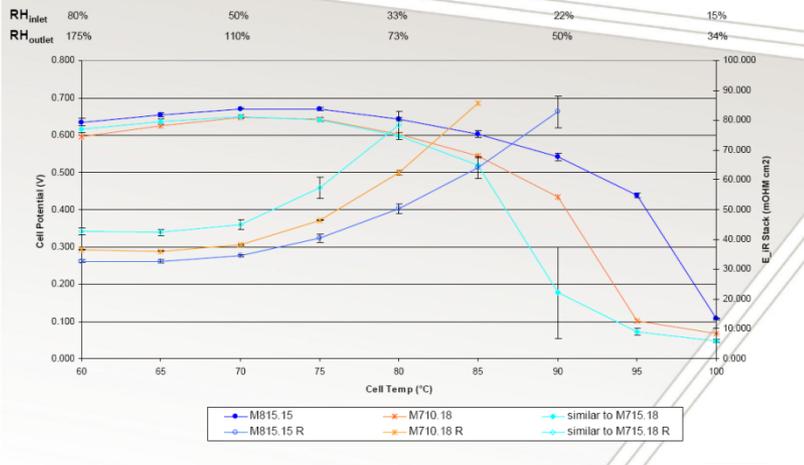
Material A  
Gore: A584.05/M815.15/C580.4

Material B  
Gore: A584.05/M815.15/C580.15

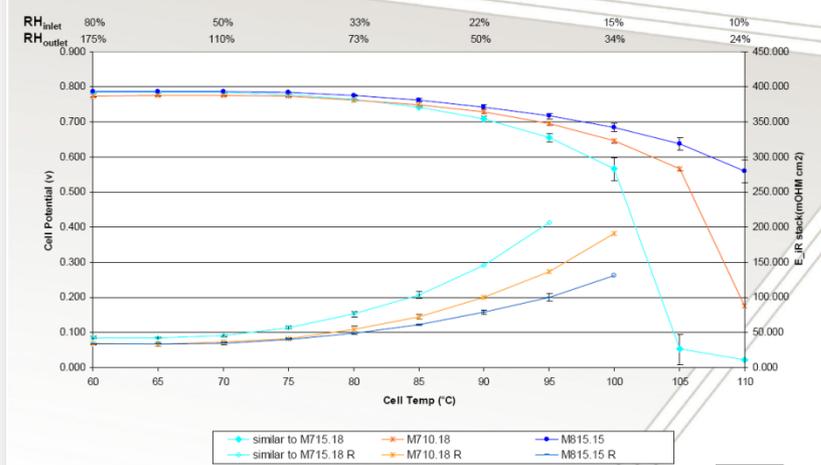


# Material Selection – Basis / Comparison

## Power Density Evaluation: 1200 mA/cm<sup>2</sup>

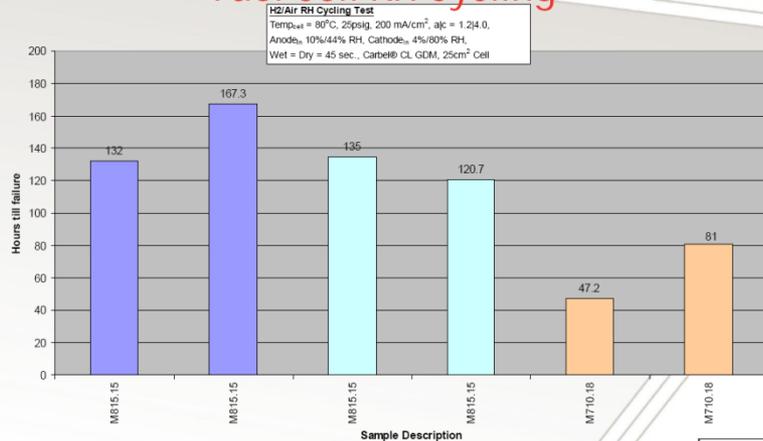


## Power Density Evaluation: 400 mA/cm<sup>2</sup>



M815 Membrane selected for superior dry operability and excellent mechanical durability.

## Durability Evaluation: Fuel Cell RH Cycling

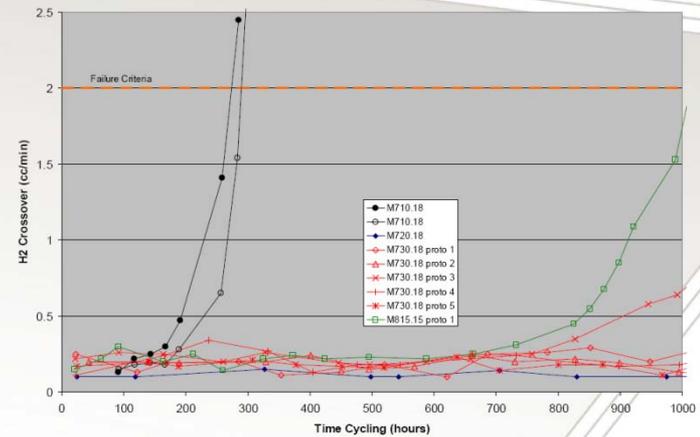


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## Mechanical Durability: Nitrogen RH cycling



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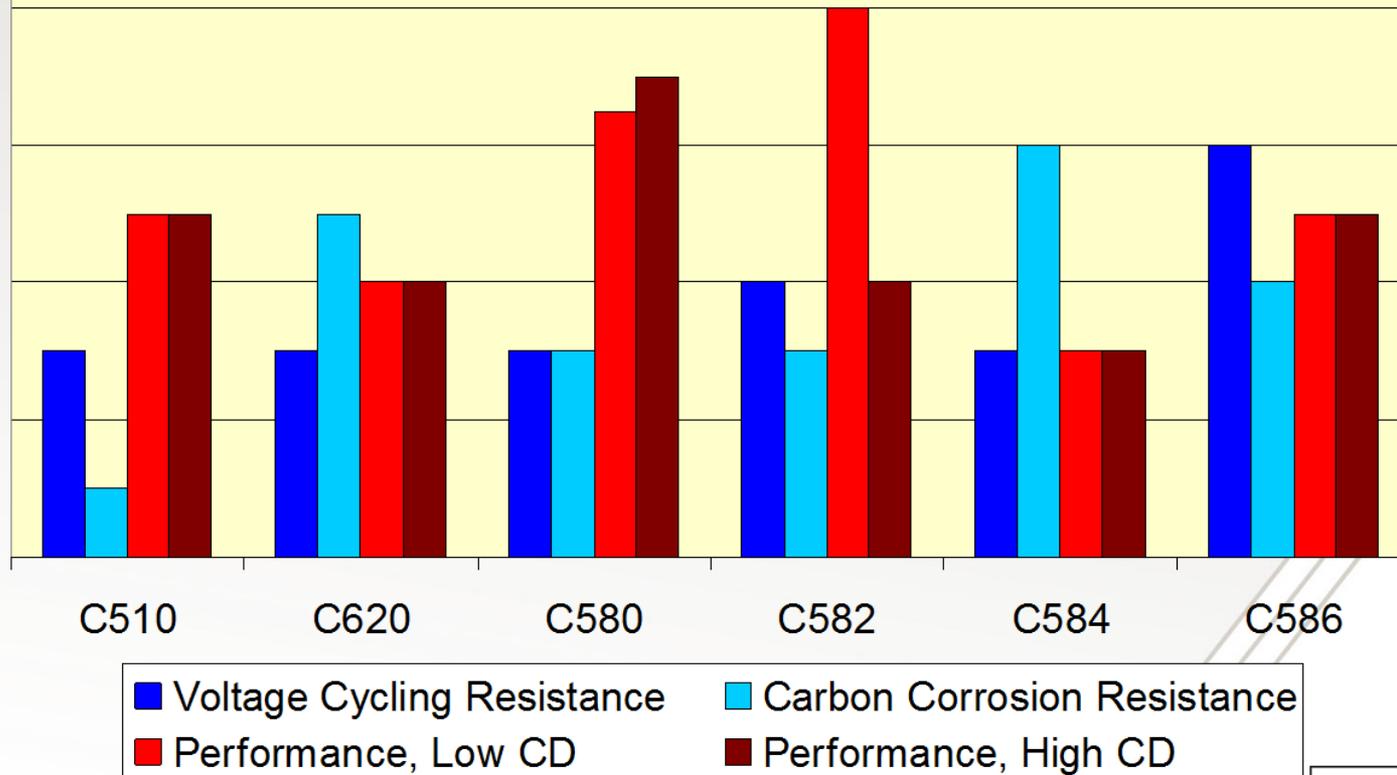
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# Material Selection – Basis / Comparison

## Cathode Options

*C580 Cathode selected for outstanding high current density performance.*

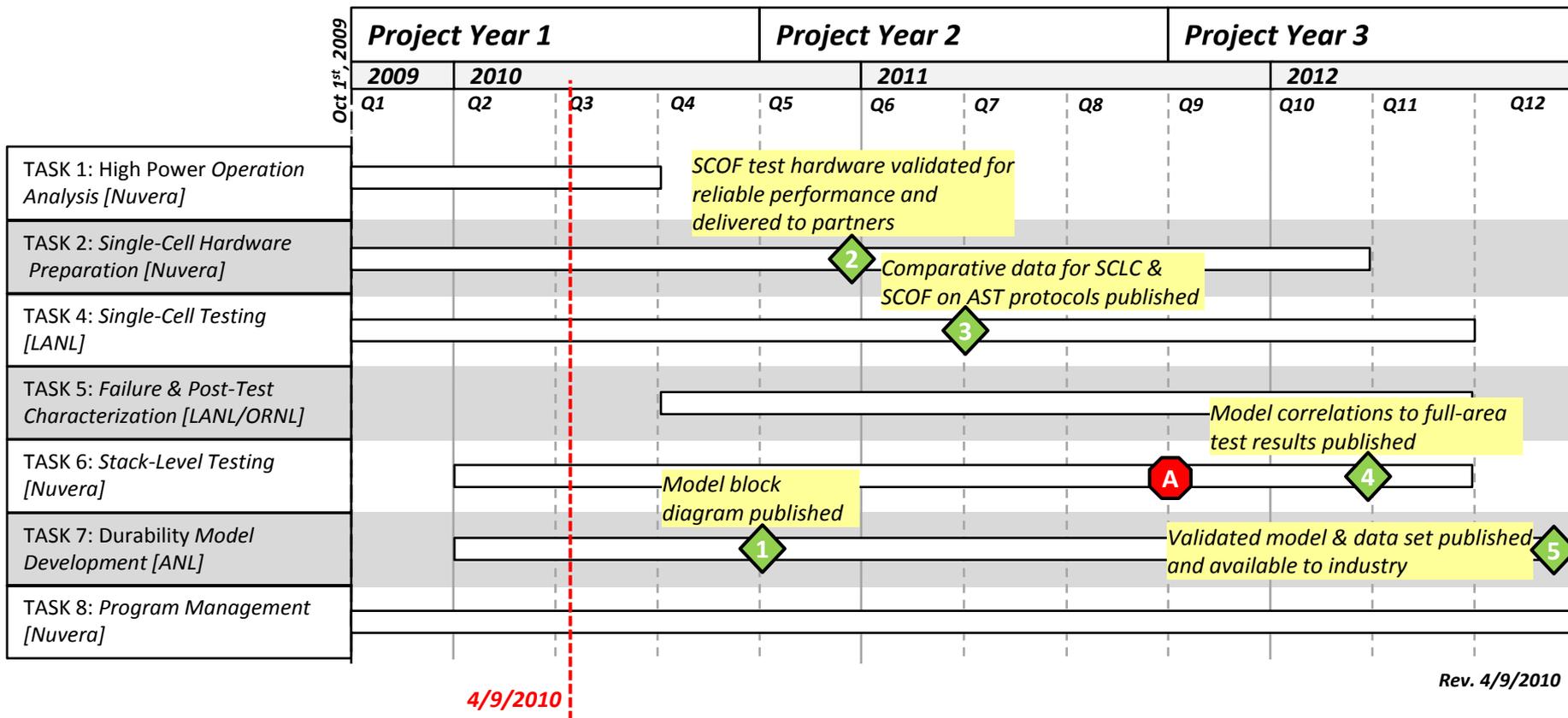


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# Schedule & Milestones



**A** → Demonstrate consistent trends and magnitude of voltage decay, diagnostic and post-test metrics between SCOF and full-area stack durability results for nominal platinum loading (~0.5mg/cm<sup>2</sup>) and rated current density (2A/cm<sup>2</sup>). [End of Q8]