Development of Stable Solid Oxide Electrolysis Cells for Low-cost Hydrogen Production

Contract Number: DE-FE0032105

P.I.: S. Elangovan, **Co-P.I.:** Jenna Pike
Tyler Hafen , Dennis Larsen,
Joseph Hartvigsen, OxEon Energy Team

Dr. Olga Marina, PNNL

Support: NASA, DOE

FECM/NETL Spring R&D Project Review Meeting

23 April 2024

DOE NETL Project Manager: Drew O’Connell

Company Background

Utah, USA R&D/Manufacturing - 2017

- Office, laboratory, and manufacturing facility (24,000 ft^2)
- NASA, DOE, DOD and commercial contracts
- Tape casting, cell and stack production, and testing
- End-to-end power to synfuels pilot plant

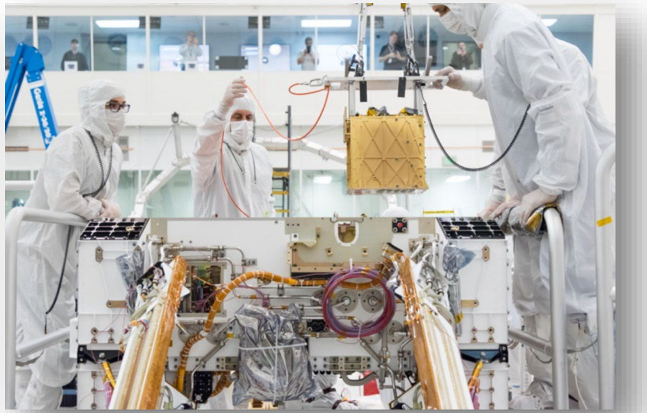


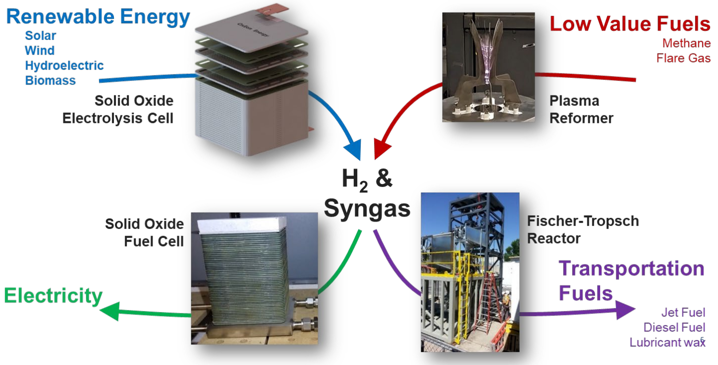
Image credit NASA/JPL-Caltec

Solid Oxide Fuel Cell and Electrolysis Stacks

- Longest running solid oxide fuel cell & electrolysis group
- Only flight qualified, TRL 9 SOEC unit with active NASA demonstration on Mars
- 30kW/10kW and 20kW/10kW reversible SOC system test programs

Fuel Reformation and Generation

- Plasma Reformer – H_2 or syngas from flare gas; digester gas conversion; clean-up bio-gasification
- Fischer-Tropsch Reactors – Modular design for sustainable fuel production from H_2 and syngas



DOE Award Announced (BIL funding) $36 million: SOEC Pilot Manufacturing Plant

Project Objective

Test solid oxide electrolysis cell (SOEC) stack in a laboratory test bed

- Show improved performance over baseline stacks
  - Robustness,
  - Reliability,
  - Endurance,
  - Hydrogen purity, and
  - Hydrogen production at elevated pressure of 2 to 3 bar.

Project Goals

- **Robustness**
  - Capability for thermal cycling of a stack
  - Redox cycling of fuel electrode in a stack
  - Production of hydrogen at elevated pressure

- **Long term stability**
  - Projected lifetime of > 40,000 hours

- **Improved performance over baseline**
  - Reproducibility and lower polarization by electrode modification

Robustness

Redox Tolerant Fuel Electrode - Background

Background: MOXIE Program Overview

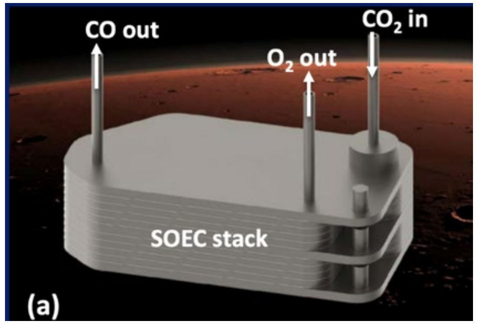
Mars OXygen ISRU Experiment for electrolysis of Mars atmosphere CO_2 to CO + O_2

First of any kind of demonstration of **In-Situ Resource Utilization (ISRU)** technologies to enable propellant and consumable oxygen production from the Martian atmosphere

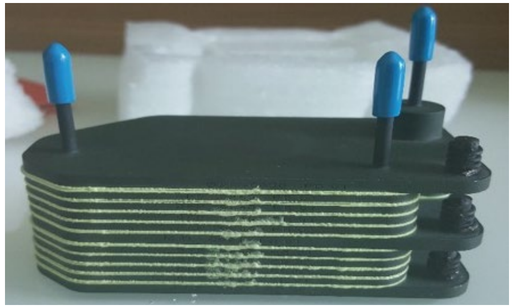
Successfully completed on Perseverance Rover mission

MOXIE is a ~0.5% scale prototype of expected final O_2 production rate

TRL 9 SOEC unit



Solid Oxide Electrolysis (SOXE) Development Team
Supported by NASA through the Jet Propulsion Laboratory (JPL)



NASA support through JPL

Flight Qualification

Baseline Performance

- **21 consecutive stacks** built with *aerospace quality standards and traceability* having consistent baseline performance on dry CO_2 and 99.9%+ O_2 purity

Cycling Performance

- 3 stacks with **21 cycles** of identical test procedure having varying cycle-to-cycle flow rates and final cycle averages of 10.11 g O_2/hr production and 99.8% purity – Targets exceeded
- 1 stack to **61 cycles** with >99.6% purity at a controlled production rate of 6 g/hr at 55g/hr CO_2 feed

Structural Stability Testing

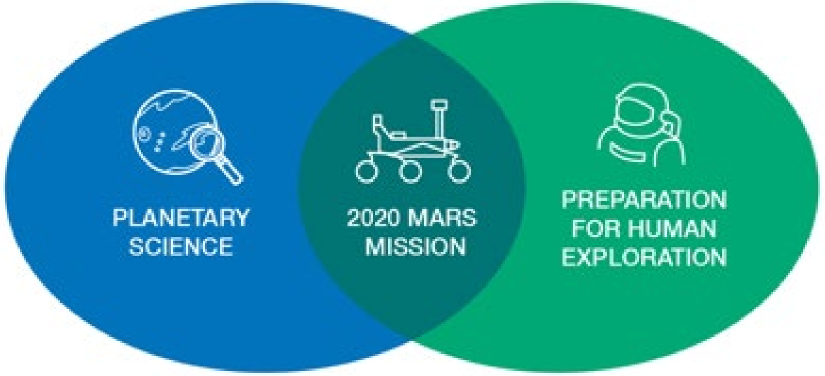
- **No leak or significant performance change after 10kN crush testing**
- Stacks tested to 25kN force with no crossover or external leakage
- *Load to failure required 62.2kN (>30 margin of safety from design)*

Shock/Vibe Testing

- Stacks vibrated at JPL and post vibe tested at OxEon
- **No leak or significant performance change post vibe!**
- **No leak after shock testing, no significant performance change!**

Cryo-Cycling

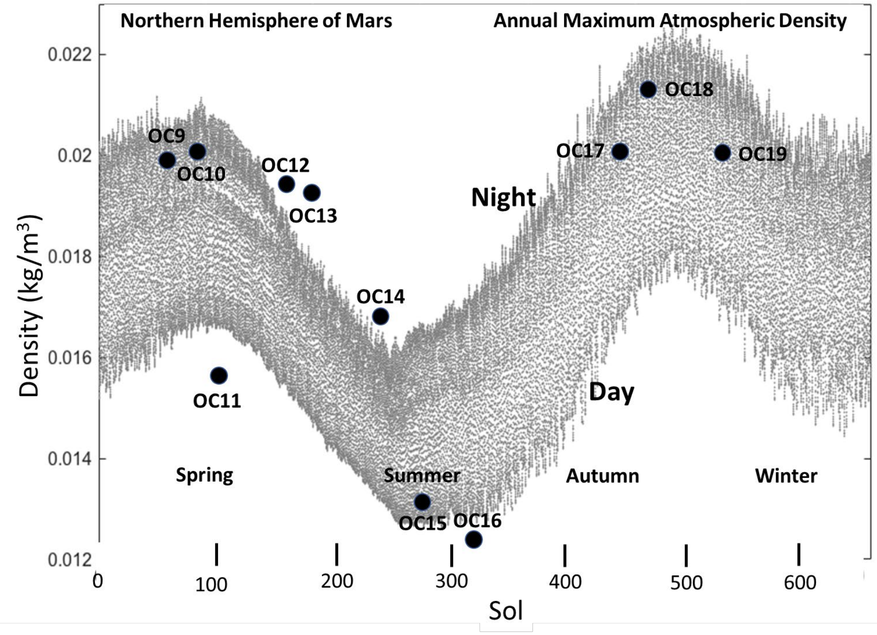
- Vibe stack cryo-cycled to -40°C (40 cycles), -55°C (3 cycles), -65°C
- **Stack performance and purity unchanged in operational cycling post test**

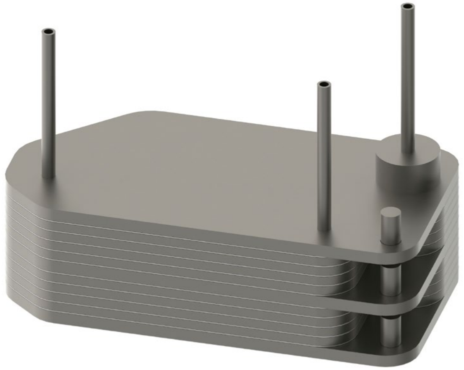


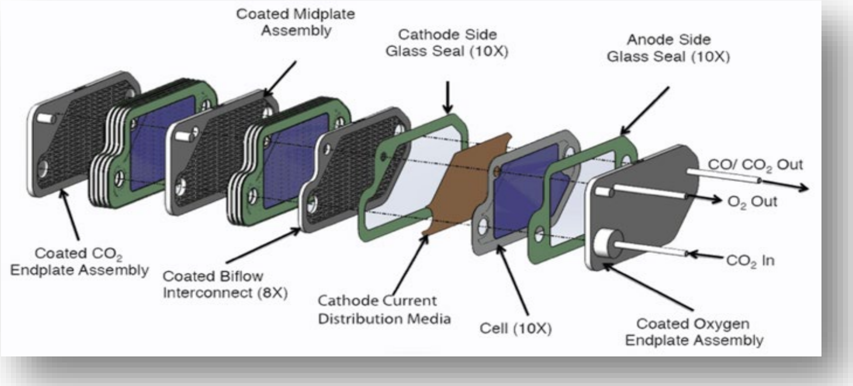
Beyond Current Potential

Flight Test Success –
First Ever ISRU Demonstration

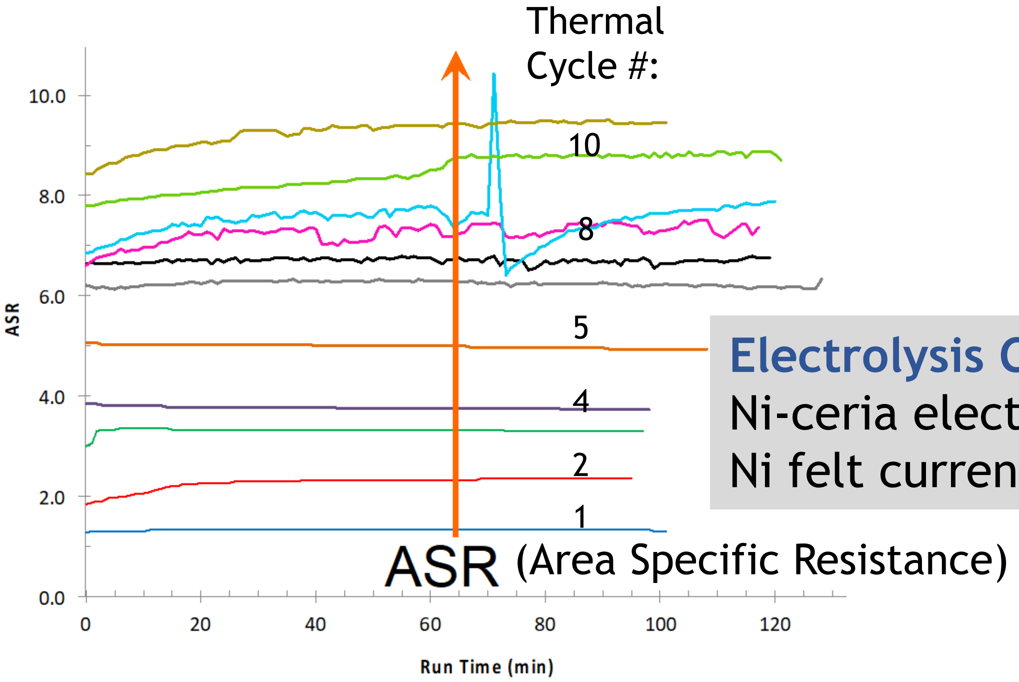
- 16 total operation cycles completed on Mars
- Pressure external to stack ~7 millibar; internal ~1 bar
- >99.6% Oxygen purity
- Operations have spanned the climactic extremes of the Mars year.
- All cycles performed as predicted: lab & models
- The MOXIE Mission completed in Sept 2023
- Basis for a Lunar and a Martian ISRU Demonstration System



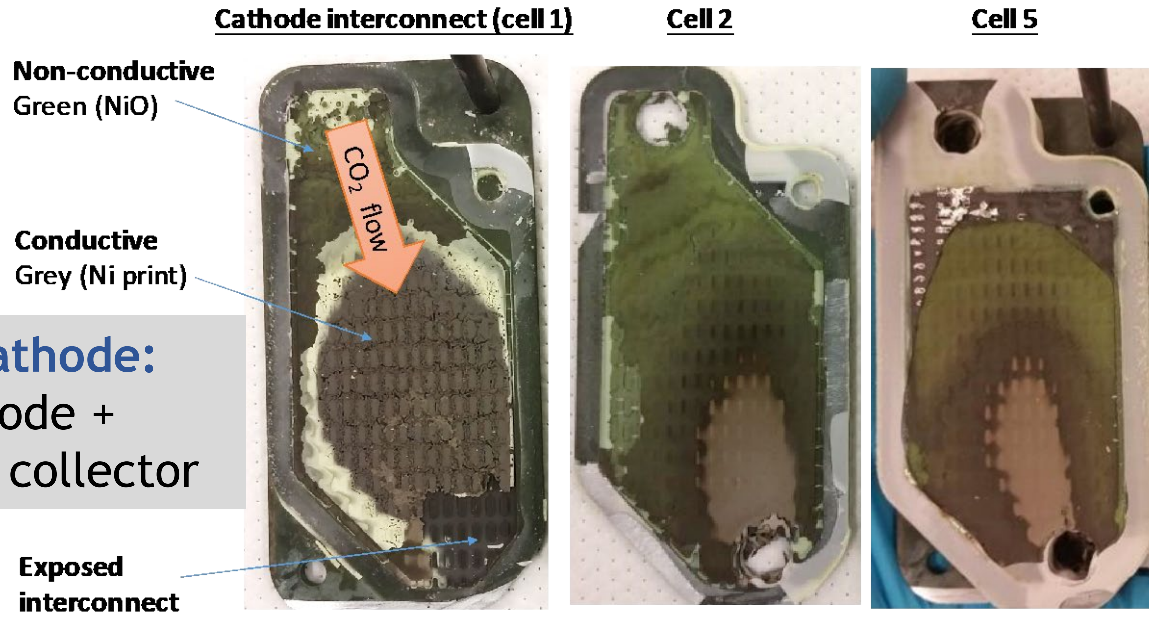




Cathode Challenge: Oxidation in Dry CO$_2$



Electrolysis Cathode:
Ni-ceria electrode +
Ni felt current collector



- Early MOXIE Test Stack:
  - 15 operational cycles - full thermal cycle with 120 min operation on dry CO$_2$
  - Dry CO$_2$ → O$_2$ production ~12% of initial

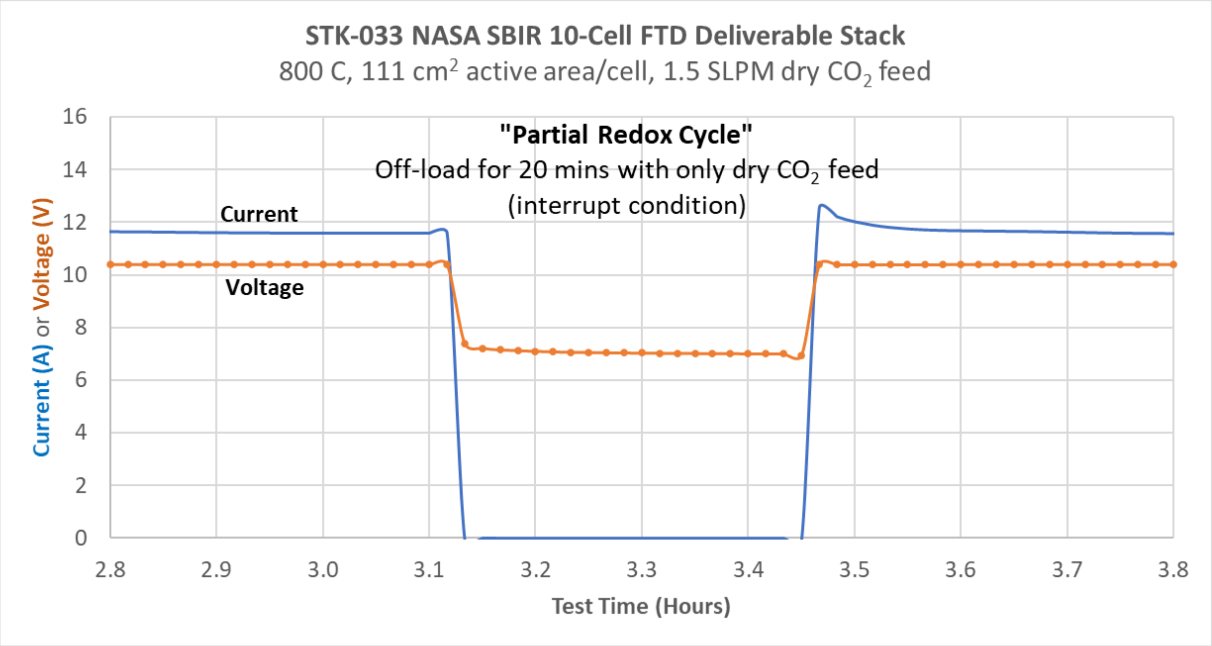
Dramatic degradation resulted from progressive oxidation front

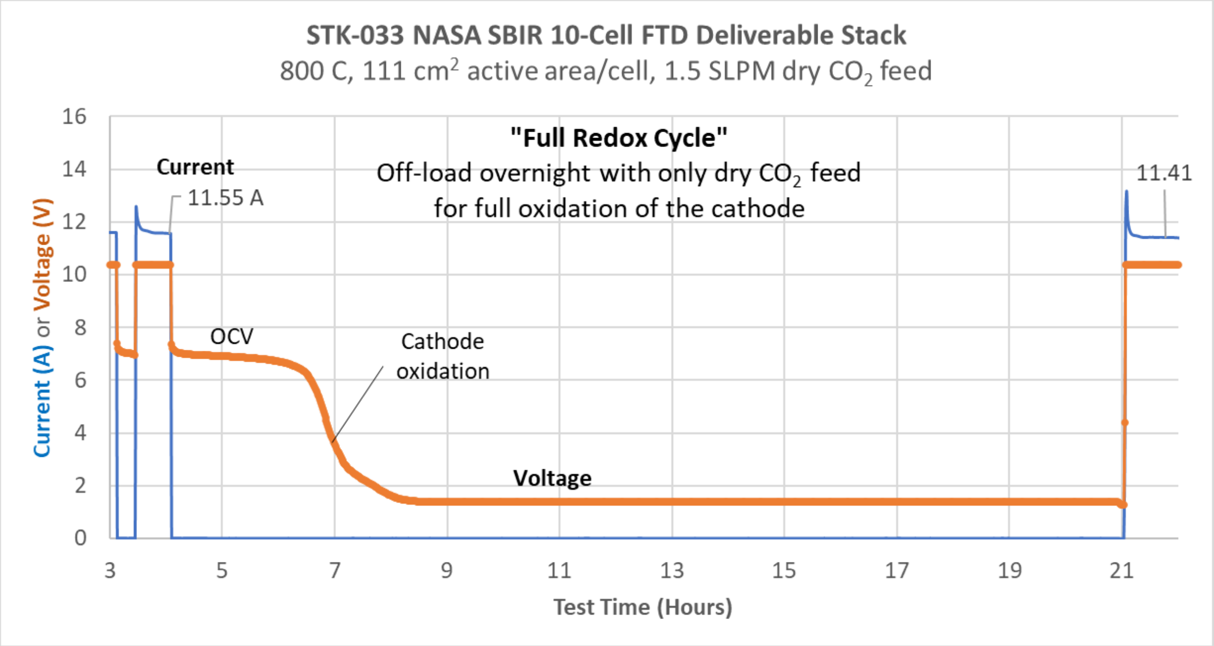
Oxidation of Ni to NiO causes ~24% vol expansion, and in this case, irreversible damage to the electrode & current collector

MOXIE implemented recycle of produced CO to prevent cathode oxidation

NASA support through JPL

Redox Tolerance: Partial and Full Redox Cycles in CO_2 Electrolysis



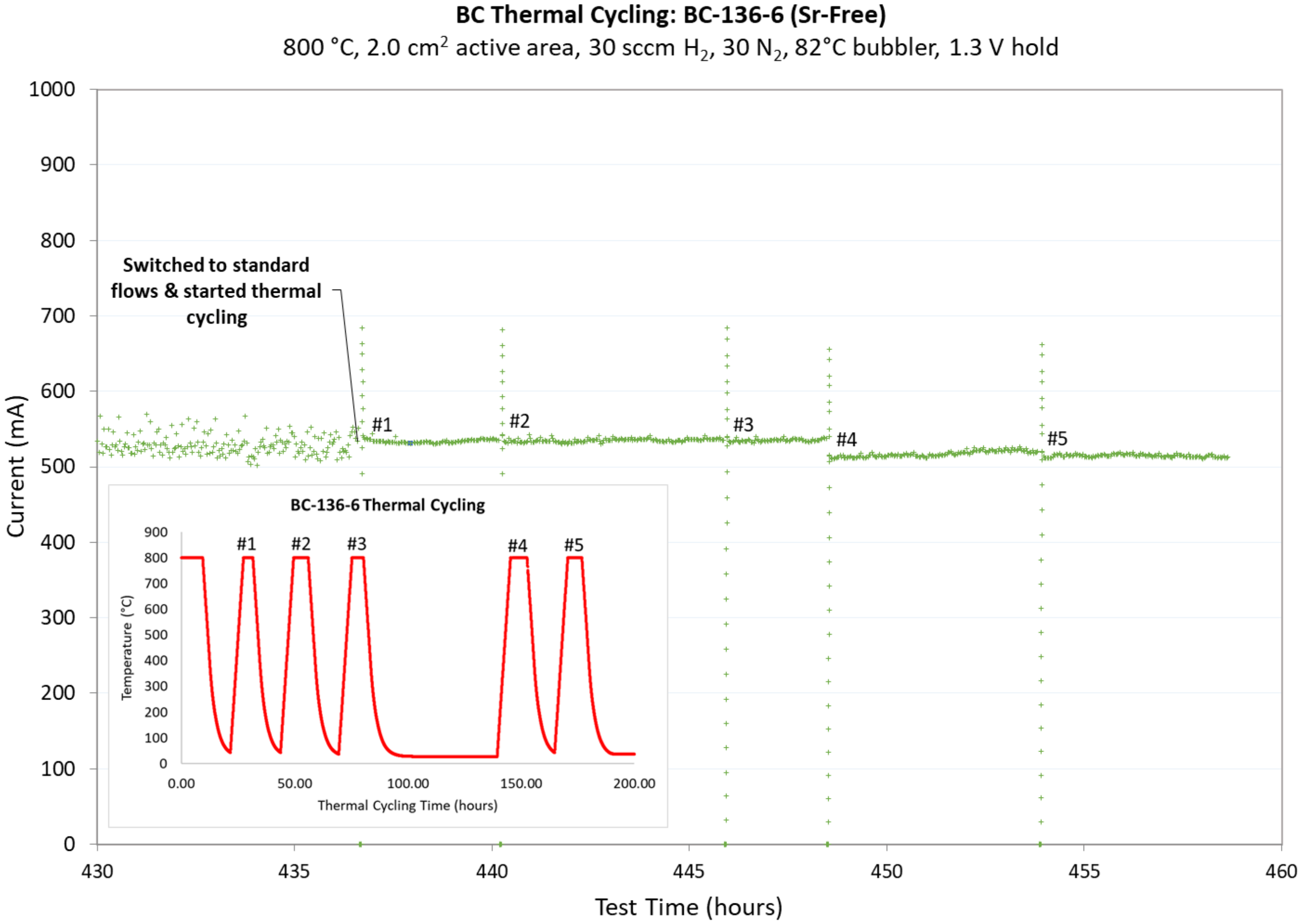


- Stack: Short (20 min) and long (12 hrs) exposure to CO_2
- Application of voltage → Full and immediate recovery of performance

NASA SBIR Funded

Button Cell Thermal Cycling in Steam Electrolysis

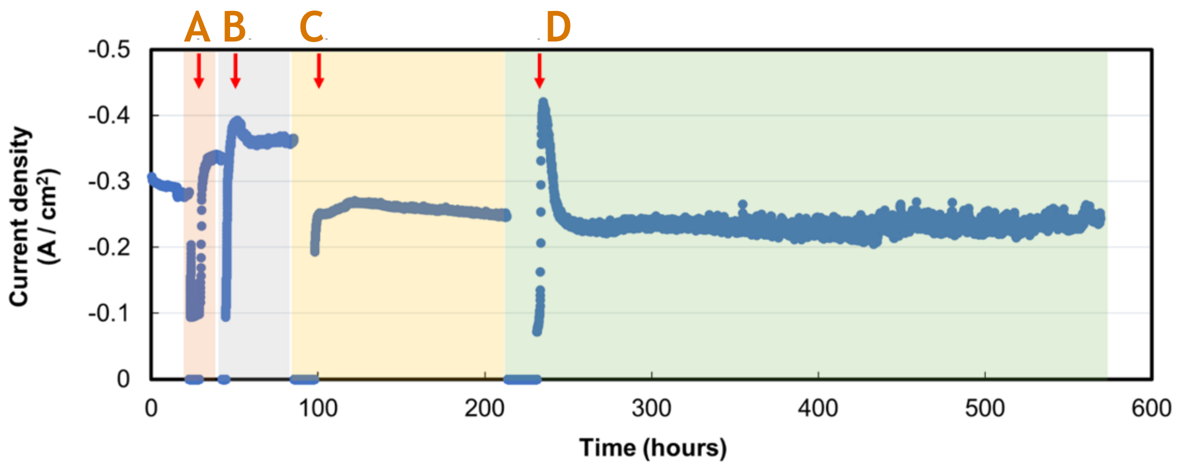
Button cell thermal cycled 5 times→ stable performance



- Sr-free air electrode composition
- Thermal cycle steps:
  - Remove 1.3 V load.
  - Cool to RT, reheat to 800 °C (2°C/min ramps).
  - Reapply voltage.
- Current density
  - Before thermal cycles: 0.266 A/cm^2
  - After 5 thermal cycles: 0.259 A/cm^2

Button Cell Redox Cycling in Steam Electrolysis

Button cell (BC-109-4) performance recovered in redox cycling



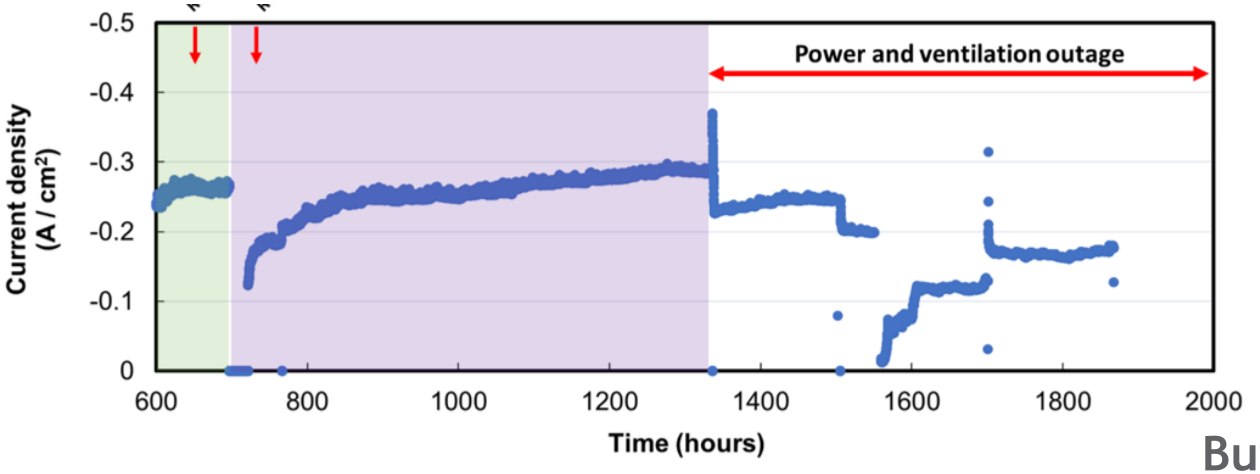
A – Initial performance, 6 30-min redox cycles – **exposure to steam at OCV for 30 minutes; no H2 in feed; apply voltage**

B – 2-hr redox cycle

C –12-hr redox cycles

D – 16-hr redox cycle

Button cell (BC-109-4) performance continually improved over 600 hours after 16 hr oxidation cycle (~700 hrs)

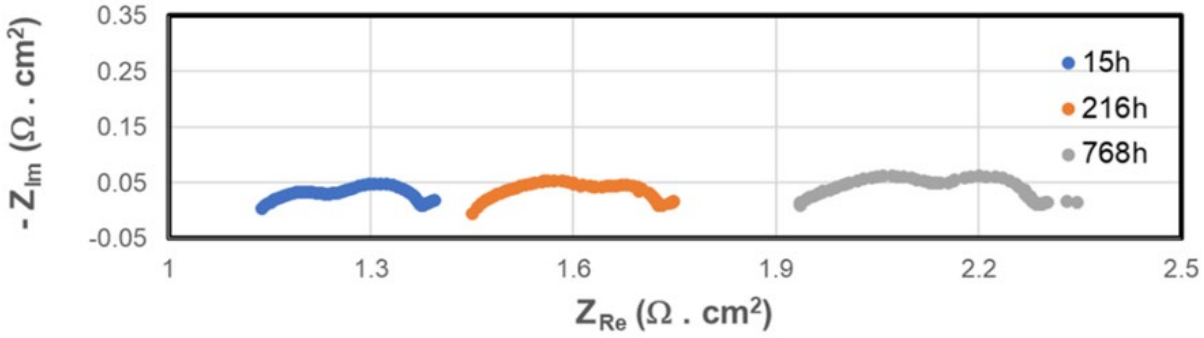


Button Cell Validation at PNNL

Button Cell Redox Cycling in Steam Electrolysis

Ohmic resistance dominates performance loss in redox cycles. Likely results from button cell current collector delamination.

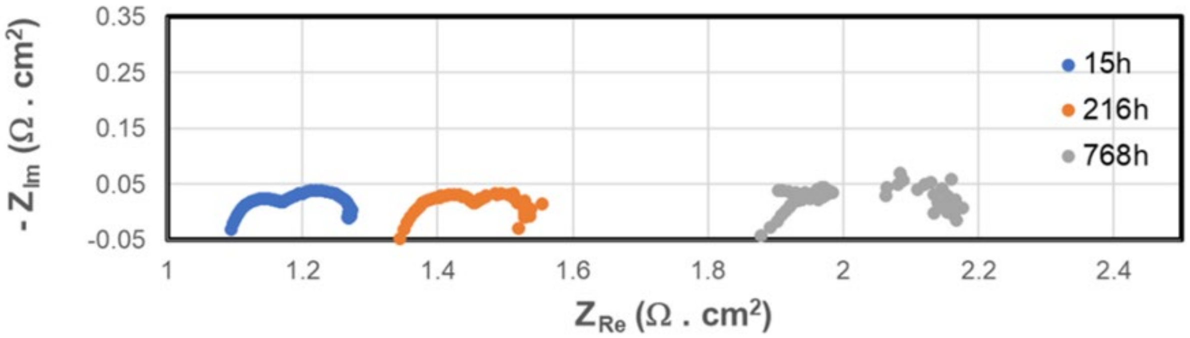
EIS at OCV



Ohmic resistance increased 28%

Polarization resistance increased 19%

EIS at 1.3V

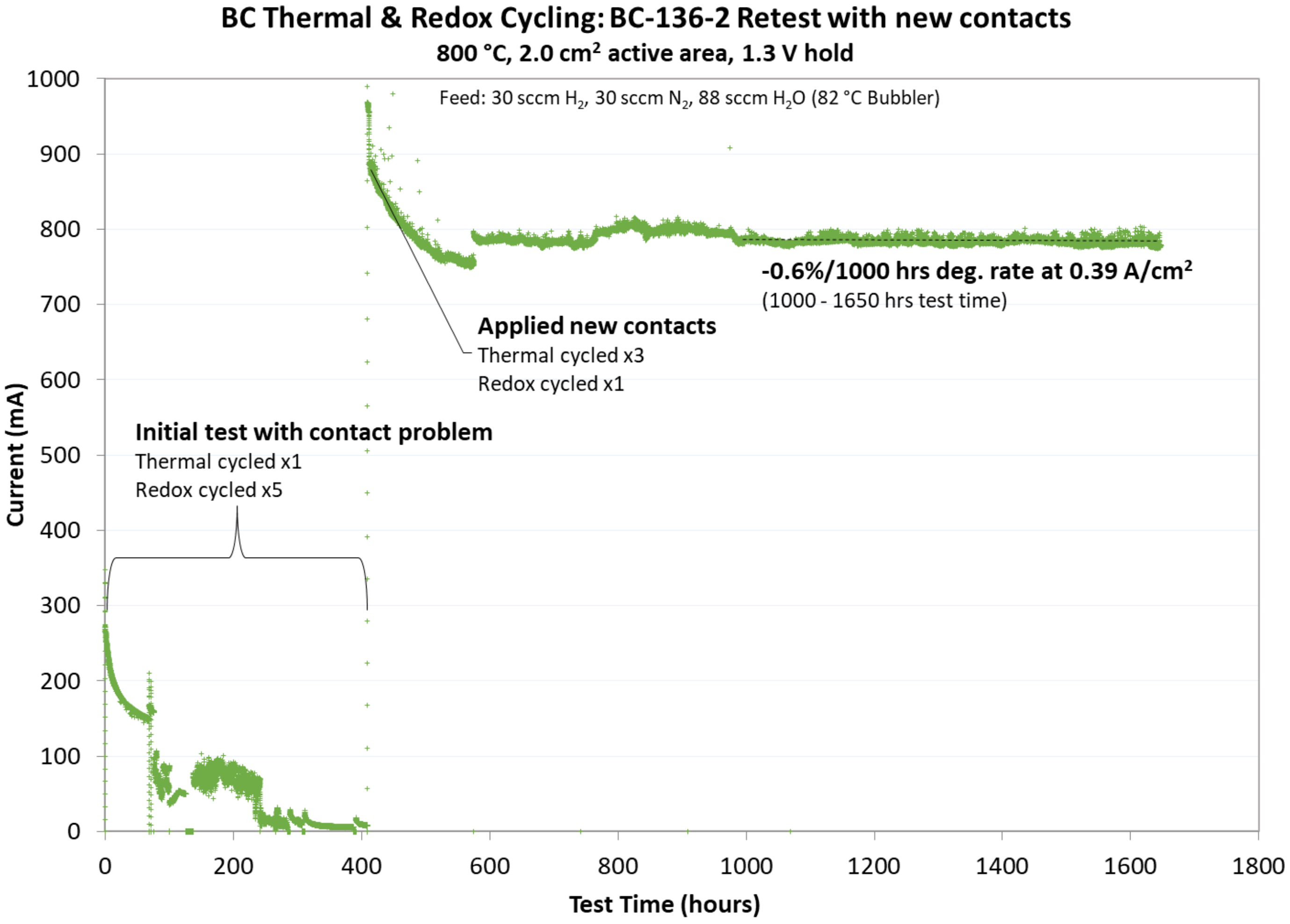


Ohmic resistance increased 23%

Polarization resistance increased 12%

Button Cell Thermal & Redox Cycling in Steam Electrolysis

Button cell thermal cycled 4 time, redox cycled 6 times → stable performance



BC Thermal & Redox Cycling: BC-136-2 Retest with new contacts

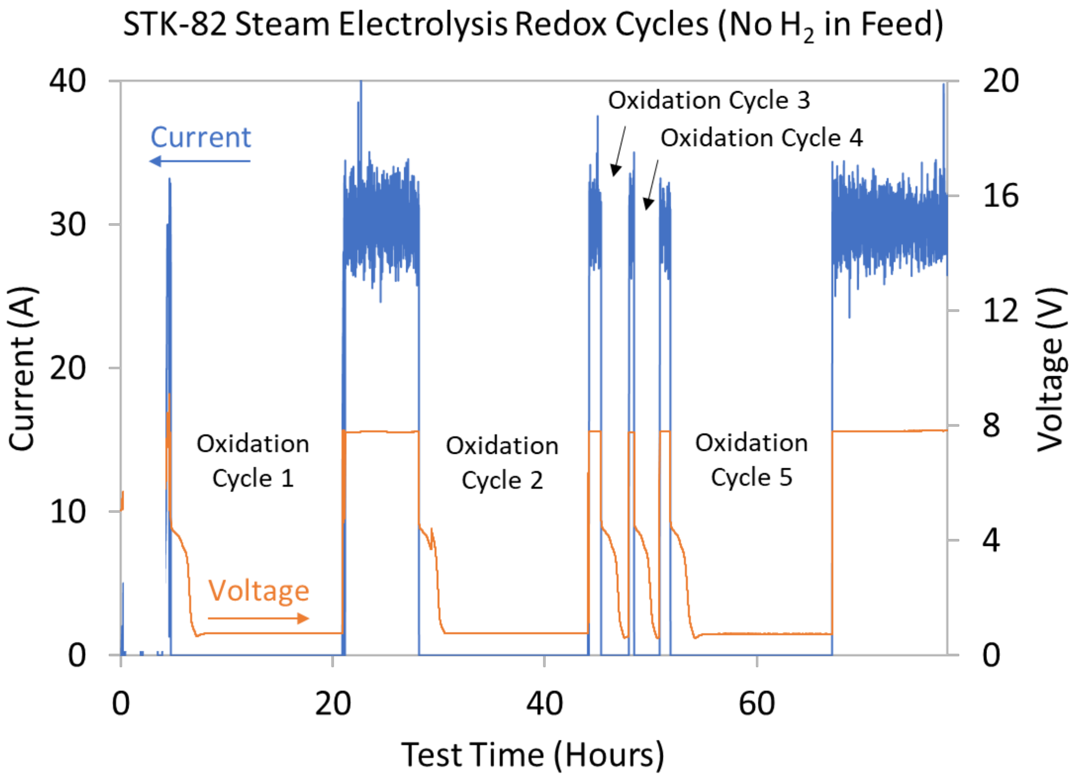
800 °C, 2.0 cm² active area, 1.3 V hold

- Redox cycle steps:
  - Switch to oxidizing feed (N_2/ H_2O).
  - Remove 1.3 V load to stop H2 generation.
  - Wait for full oxidation, then reapply load.
  - Add H2 for reduction recovery if needed.

- Thermal cycle steps:
  - Remove 1.3 V load.
  - Cool to RT, reheat to 800 °C (2°C/min ramps).
    - > 800 °C for new contacts application
  - Reapply load.

- Current density
  - Initially: 0.174 A/cm²
  - At 1,300 hrs: 0.394 A/cm²

STK-82: Full Redox Cycling in Steam Electrolysis



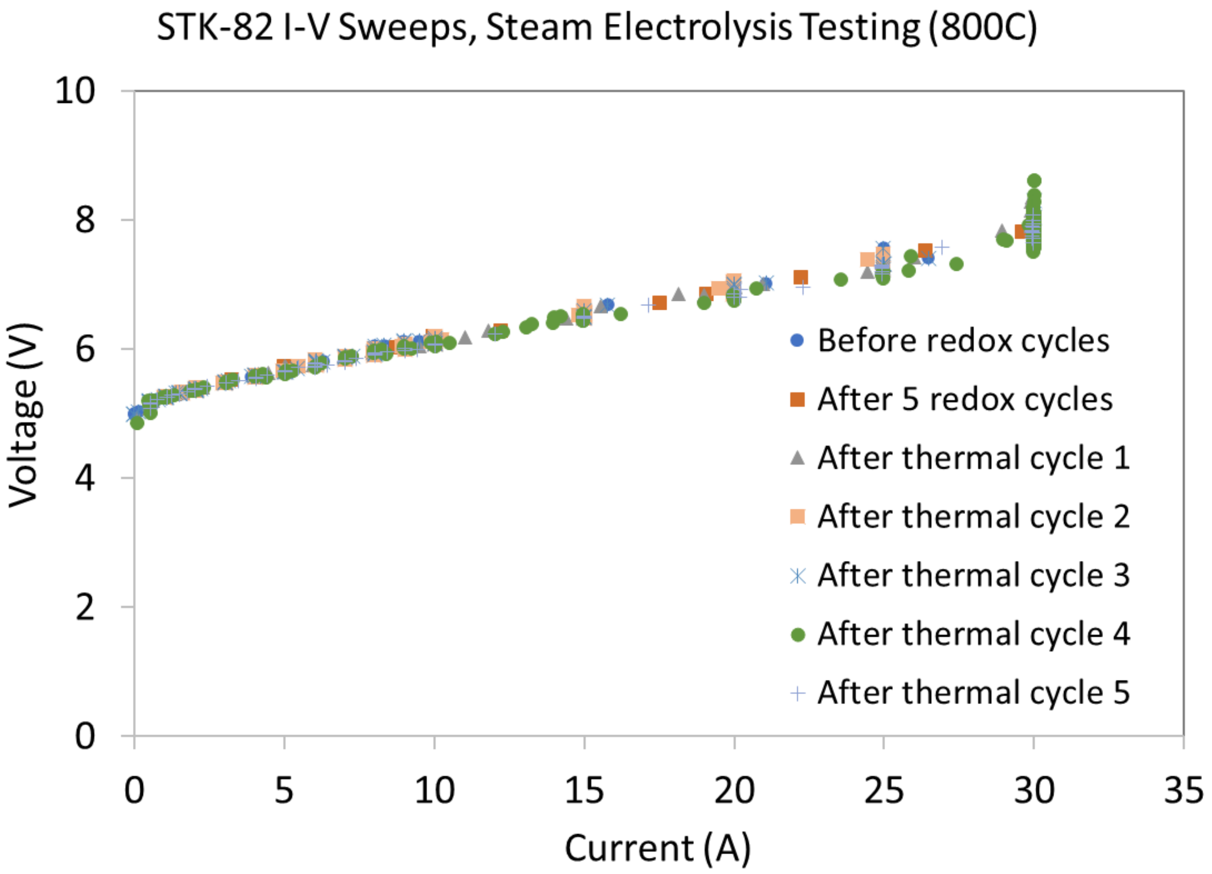
STK-82 (6-cell stack) recovered performance after **6 redox cycles**

STK-82 Redox Cycle Comparison

Cycle	Current (A)	Voltage (V)	Power (W)
1	28.04	7.82	219
2	31.12	7.78	242
3	31.33	7.77	243
4	31.49	7.8	245
5	31.03	7.8	242
6	31.09	7.8	242

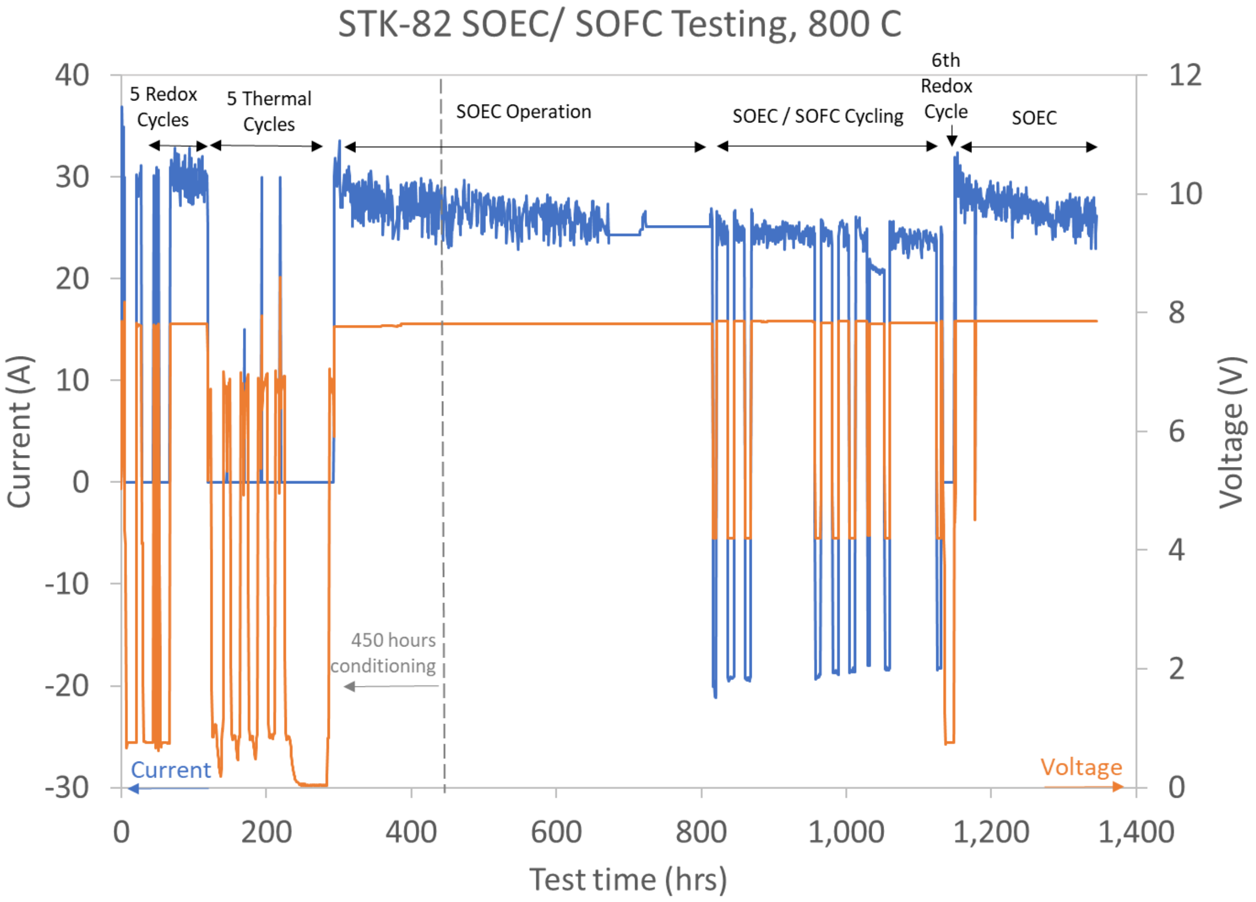
- Fuel electrode was fully oxidized by exposure to steam only
- Application of voltage → Full and immediate recovery of performance
  - Self-generated hydrogen at fuel electrode
  - **No external reducing gas!**

STK-82: Thermal Cycling - 800 °C to Room Temperature



- Stack after 5 redox cycles was cooled to room temperature in dry H_2/N_2 at OCV, then returned to 800 °C (100 °C/min)
  - 5 thermal cycles
- Application of voltage → Full and immediate recovery of performance
- **Demonstrates robust stack and seals**

STK-82: Long Term Stack Stability Testing

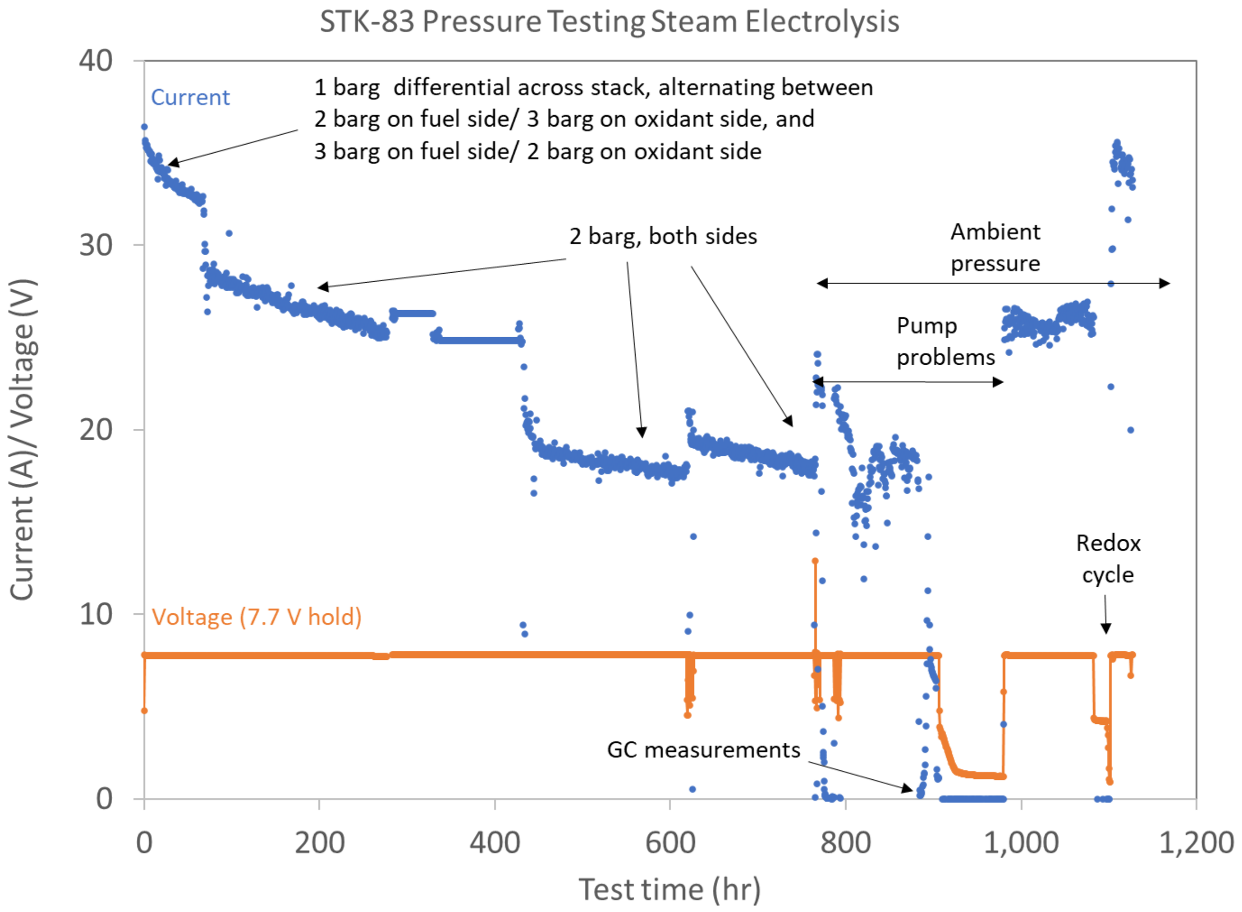


Stack test sequence

- 5 redox cycles
- 5 thermal cycles
- 500+ hour steam electrolysis
- 300+ hours SOEC/ SOFC cycling
- 6th redox cycle
- Steam electrolysis

Degradation = 1.8 %/ 1,000 hrs in SOEC operation after conditioning period.

STK-83: Pressurized Stack Operation



- Stack (6-cell stack) tested at moderate pressure for ~800 hours
  - 3 barg pressure during testing
  - 1 barg differential across stack
- Performance drop is likely caused by poor seal between stack and stack test assembly (external gasket)
- **Hydrogen production at pressure!**
- **Performance (at ambient pressure) recovered to initial value after a redox cycle**

STK-83: Pressurized Stack Operation

Gas chromatograph analysis of anode and cathode tail-gas composition, taken at ~775 hrs

GC Results (%) & Product Calculations (slpm)

Pressure condition	2 barg O_2, 2 barg fuel		2.2 barg O_2, 2 barg fuel		2 barg O_2, 2.5 barg fuel		2 barg O_2, 3 barg fuel	
GC Type	O_2	fuel	O_2	fuel	O_2	fuel	O_2	fuel
H_2		85.8		84.8		84.08	0.01	85.41
O_2	99.84		98.58		99.46		99.58	
N_2	0.16	14.03	1.39	15.03	0.54	15.77	0.41	14.36
H_2 generated (slpm)		0.95		0.86		0.80		0.92
O_2 generated (slpm)		0.47		0.43		0.40		0.46
H_2O Conversion (slpm)		84%		76%		71%		81%
Stack Current, A (calculated)		22.6		20.5		19.2		21.9

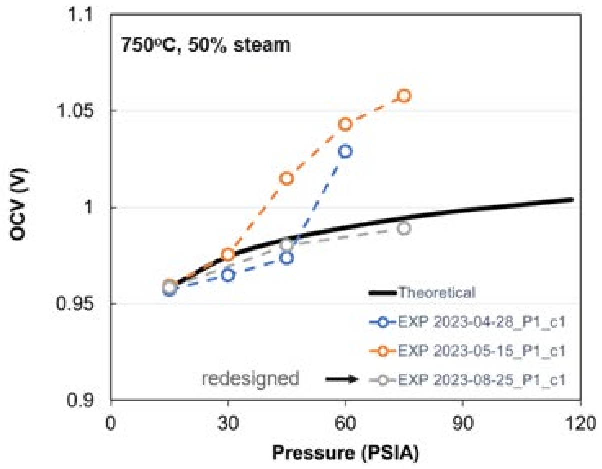
Robust seal: O_2 purity demonstrates lack of cross over leak

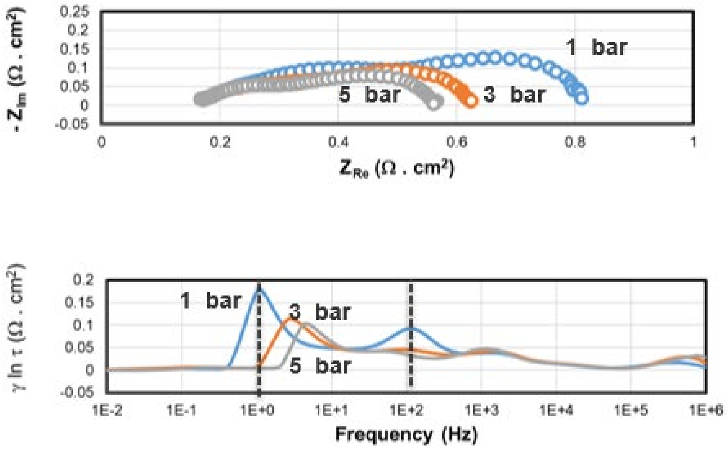
H_2 and O_2 generated at pressure!

Button Cell Operation at Elevated Pressure

SOEC Performance Increases with Pressure

(PNNL-fabricated cells).





Pressurized button cell

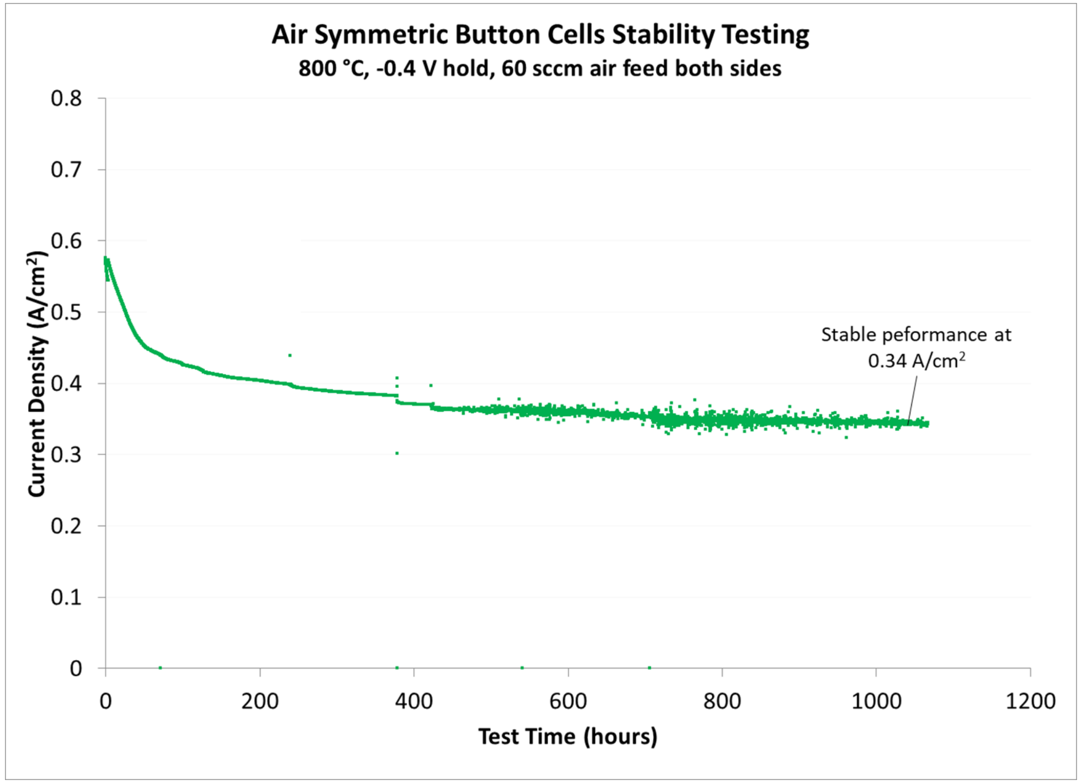
- Low frequency (mass transport) and mid-frequency (diffusion) processes are suppressed
- Troubleshooting seals for long-term tests

Test stand modifications on-going to meet 7 bar pressure target
- Good OCV match to the Nernst voltage, but there is a seal leak somewhere in the test system
- Reached 5 bar.

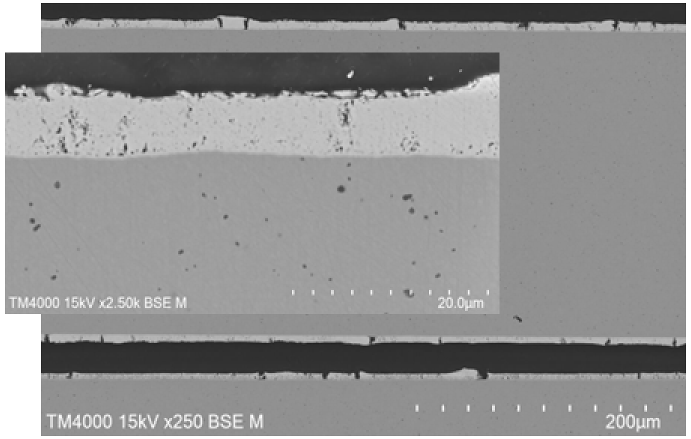
Oxygen Electrode Interface Improvement

Improvements to barrier layer density and continuity.

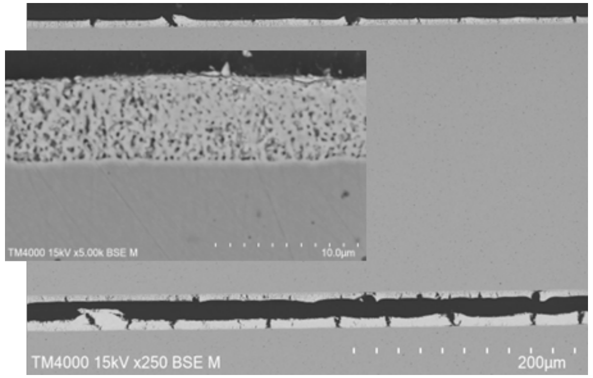
Stable performance at 0.34 A/cm2 measured after 600 hours conditioning that continued through 1,000 testing



Barrier layer density increased using a modified process



Higher porosity with screen printed barrier layer



Oxygen Electrode / Current Collector

Eliminate Sr in current collector layer by replacing LSCF with Sr-free composition

- Ohmic resistance close to expected based on electrolyte thickness
- Polarization resistance indicates Sr-containing LSCF layer is not required for water splitting
  - Sr-free cell measured 58% lower initial polarization resistance relative to the LSCF cell

Electrode CC Layer	**ASR (Ω·cm^2)**		
	Total	**Ohmic**	**Polarization**
Sr-free composition	0.66	0.59	0.08
LSCF	0.71	0.52	0.19

Summary

- Redox tolerance validated for steam electrolysis
  - Stack test complete
  - Oxidized Ni electrode recovery without the need for hydrogen in inlet
- Thermal cycle stability demonstrated (up to 5 cycles)
- Pressurized tests: steam electrolysis
  - Stack showed high purity O_2 and H_2 at 3 barg pressure
  - Button cell test ongoing at PNNL
- Electrode materials modification - validation in progress
  - Composition to improve thermochemical stability
  - Surface/Interface modification for improving catalytic property and performance stability
- Cation migration is still a challenge for long-term stability
- External influences (steam, air, BOP contaminants) still need evaluation and mitigation

Thank you

S Elangovan

elango@OxEonEnergy.com

This presentation contains information from work supported by NASA through JPL's prime contract under JPL subcontract number 1515459 and NASA SBIR contract 80NSSC19C0114.

DOE-NETL support through Contract Number: DE-FE0032105 is acknowledge