

H2NEW: Hydrogen (H2) from <u>N</u>ext-generation <u>Electrolyzers of Water Overview</u>

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Project Goals



<u>Goal</u>: H2NEW will address components, materials integration, and manufacturing R&D to enable manufacturable electrolyzers that meet required cost, durability, and performance targets, simultaneously, in order to enable \$2/kg hydrogen (by 2025 on way to H2 Shot target, \$1/kg by 2031).



H2NEW has a clear target of establishing and utilizing experimental, analytical, and modeling tools needed to provide the scientific understanding of electrolysis cell performance, cost, and durability tradeoffs of electrolysis systems under predicted future operating modes

Overview



Timeline and Budget

- Start date (launch): October 1, 2020
- FY21 DOE funding: \$10M
- Anticipate minimum \$50M over 5 years
- 75% low temperature electrolysis due to higher TRL and intermittency

Barriers

- Durability
- Cost
- Efficiency



- UC-Irvine
- Carnegie Mellon Univ
- Colorado School of Mines

* Expansion to include additional academic and industrial partners in anticipated in the near future.

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Bipartisan Infrastructure Law - Hydrogen Highlights



- Covers \$9.5B for clean hydrogen:
 - \$8B for at least four regional clean hydrogen hubs
 - \$1B for electrolysis research, development and demonstration
 - \$500M for clean hydrogen technology manufacturing and recycling R&D



President Biden Signs the Bipartisan Infrastructure Bill on November 15, 2021. Photo Credit: Kenny Holston/Getty Images

 Aligns with Hydrogen Shot priorities by directing work to reduce the cost of clean hydrogen to \$2 per kilogram by 2026
 https://www.energy.gov/sites

https://www.energy.gov/sites/default/ files/2021-12/h2iq-12082021.pdf

Requires developing a National Hydrogen Strategy and Roadmap

Accomplishments Recent/Planned Workshops



- Workshops supporting the Bipartisan Infrastructure Law (BIL)
 - H2- PACE (Power Electronics) 12/2-12/3, 2021
 - H2 LAWE (Alkaline Electrolysis) Jan 26-27, 2022
 - Bulk storage of gaseous hydrogen (Feb 10-11, 2022)
 - Liquid hydrogen (Feb 22-23, 2022)
 - High Temperature Electrolysis Manufacturing (March 8-9, 2022)
 - Advanced Materials for PEM Electrolysis (March 30-31)
 - Manufacturing and Recycling (H2-MACH, May 24-26)

Bold denotes - H2NEW played leading/co-organizing role in Workshop

https://www.energy.gov/eere/fuelcells/workshop-and-meeting-proceedings

Relevance – H2NEW connection to H2@Scale





Illustrative example, not comprehensive https://www.energy.gov/eere/fuelcells/h2-scale

- Making, storing, moving and using H2 more efficiently are the main H2@Scale pillars and all are needed.
- Making H2 is the inherently obvious, first step to spur the wide-ranging benefits of the H2@Scale vision.
- Electrolysis has most competitive economics and balances increasing renewable generation challenges.
- Timeframe is short, competition intense, coordinated effort critical for domestic competitiveness.

Approach – Consortium Structure and Support Posters



- Task Based Structure Split between:
- Low Temperature Electrolysis (LTE), 75%:
 - Task 1 Durability (<u>P196a</u>)
 - Task 2 Performance (P196b)
 - Task 3 Scale-up (P196c)
 - Task 3c Analysis (P<u>196d</u>)
- High Temperature Electrolysis (HTE), 25%:
 - Task 5 Durability and AST Development (P196e)
 - Task 6 Characterization (P196f)
 - Task 8 Modeling (P196g)

See associated <u>AMR posters</u> for additional details on Approach, Accomplishments, and Future Work



H2NEW Activities: Low Temperature Electrolysis (LTE)



Relevance: Stack Costs (PEM Centric)





Stack Targets	Status	2023	2025
Cell (A/cm ² @1.9V)	2.0	2.5	3.0
Efficiency (%)	66	68	70
Lifetime (khr)	60	70	80
Degradation (mV/khr)	3.2	2.75	2.25
Capital Cost (\$/kW)	350	200	100
PGM loading (mg/cm ²)	3	1	0.5

These 3 areas

- 1. Increased efficiency/current density
- 2. Decreased PGM loading
- 3. Scale-up

Are the strongest levers for addressing stack costs.

https://www.hydrogen.energy.gov/pdfs/review21/p196_pivovar_boardman_2021_o.pdf

Relevance: Hydrogen Levelized Cost (PEM Centric)





Select pathway to \$2/kg and \$1/kg identified.

Much of HLC gains possible through greatly decreasing capital costs and enabling lower cost electricity through variable operation.

These advances can't come with compromised durability or efficiency, so all three areas are linked.

Approach: Select LTE Milestones*



Milestone Name/Description	Due Date	Туре	Status
Assess the impact of the stress tests on electrode, membrane, and PTL through voltage loss breakdown and use ex-situ characterization to identify degradation mechanisms and support AST development. Determine initial stressors to be investigated in detail for potential to accelerate specific degradation mechanisms. Labs – NREL, LBNL, ANL, LANL.	6/30/2021	QPM	Complete
Establish and validate single cell performance testing protocols on SOA MEAs with PGM loading of <0.8 mg/cm ² and membranes <100 μm demonstrating a minimum performance of 2 A/cm ² at 1.8 V and demonstrate ability to perform voltage loss breakdown modeling as verified by agreement within 10% across at least 3 labs.	9/30/2021	QPM	Complete
Targeting increased efficiency through membrane thickness reductions: Perform H2 crossover experiments between atmospheric and 30 bar backpressure, and current densities up to at least 3 A/cm ² to work towards an improved understanding of the mechanism and nature of hydrogen crossover, and its dependency with current density.	12/31/21	QPM	Complete
Targeting cost gains from scaling up processes: Using an automated, scalable coating manufacturing process for MEAs, match or exceed performance of baseline MEA (within 20 mV at current densities up to 2 A/cm ²) and project potential cost reductions through scaleup. (NREL, LBNL, ORNL)	3/30/2022	QPM	Complete
Quantify membrane chemical degradation rates under three different operating conditions including simulated (without current) and electrolyzer operating conditions using fluoride emission rate at the anode and cathode.	6/30/2022	QPM	On Track
Development and validation of AST protocols for PEM LTE/SOEC HTE. Propose at least 3 component-level LTE and HTE ASTs (each) and compare cell and AST performance loss.	9/30/2022	Mile- stone	On Track

*For complete list, see associated <u>AMR posters</u> for additional details

H2NEW: Hydrogen from Next-generation Electrolyzers of Water

Approach: LTE Tasks





- Durability (Task 1, P196a)
 - Establish fundamental degradation mechanisms
 - Develop accelerated stress tests
 - Determine cost, performance, durability tradeoffs
 - Develop mitigation
- Performance (Task 2, P196b)
 - Benchmark performance
 - Novel diagnostic development and application
 - Cell level models and loss characterization
- Scale-up (Task 3, P196a)
 - Transition to mass manufacturing
 - Correlate processing with performance and durability
 - Guide efforts with systems and technoeconomic analysis (Task 3c, P196d)

Lab Scale – Ultrasonic Spray









Task 3





Ex situ characterization

Operando characterization/diagnostics



- Summary of Highest Impact Efforts in Last Year
 - Task 3c Systems/Technoeconomic Analysis
 - System efficiency
 - GPRA
 - Wind/Solar/Battery
 - Task 1 Durability
 - Start/Stop
 - Ir dissolution/modeling
 - Membrane
 - Task 2 Performance
 - H2 Xover
 - Voltage loss breakdown
 - Task 3 Scale-up
 - Low loadings
 - Ink aging
 - PTL (cross task effort)

Low Temperature Electrolysis (LTE)

Task 3c: Analysis (more content in AMR Poster P196d)



Accomplishment: Models Determine System Performance and Degradation

H2NEW U.S. DEPARTMENT OF ENERGY

Efficiency Definitions

- Stack efficiency: LHV of net H₂ produced in the cathode divided by stack input power
- System efficiency: LHV of H₂ supplied to the pipeline divided by the total electrical power supplied to the transformer/rectifier and other BOP components

	Efficiency (%)		Efficiency (kWh/kg-H ₂)	
	BOL	EOL	BOL	EOL
Stack				
Rated Power Efficiency	65.3	59.3	51.1	56.2
Peak Efficiency	76.3	71.2	43.7	46.8
System				
Rated Power Efficiency	60.3	53.9	55.3	61.8
Peak Efficiency	70.2	65.6	47.5	50.8





• Conduct analysis consistent with objectives Q2 milestone, meeting the following:

"Using the baseline PEM electrolyzer system model and the H2A Model, quantify at least three distinct pathways based on improvements to system cost, manufacturing, performance, and lifetime needed to achieve the H2 production cost targets of \$3.60/kg and \$3.10/kg by end of FY22 and FY23, respectively."

- Approach: Use H2A to map pathways to cost targets as a function of key economic inputs
- Capacity factor fixed
- Electricity price fixed
- Capital cost
- Efficiency
- Lifetime

Parameter	Baseline value	Units
Capacity factor	90%	%
Electricity price	0.03	\$/kWh
Stack efficiency	50.8	kWh/kg H2
System efficiency (LHV)	66.6 (50% LHV)	kWh/kg H2 (% LHV)
Replacement lifetime	40,000	hours
Estimated replacement cost	15%	% of direct capital cost

Accomplishment: Mapping possible \$2/kg pathways





- Between efficiency and capital cost, several possible paths exist to meet FY22 and FY23 targets
- Pathways could focus on advancing one parameter or a combination of both

Approach: Optimal Operations Strategy for a Variety of Electricity Sources and Prices

- Task also involves analyzing LTE electrolyzers directly connected to wind and photovoltaic solar electricity generation
- NREL PySAM modeling platform is used to simulate outputs from PV/wind at any location or capacity
- Battery energy storage can also be added to the electrolyzer-renewable system



Low Temperature Electrolysis (LTE)

Task 1: Durability (more content in AMR Poster P196a)



Accomplishment: Accelerated Stress Test - Start-Stop



MEAs: Nafion 117 (to mitigate crossover concerns) a) 3 0.6 • IrO₂ 1.4 V • IrO₂ 1 V 0.1 mg_M cm⁻² anode catalyst loading 2.5 0.5 IrO₂ 0.6 V Dissolution [µg] Start-stop: Voltage control 0 to 2 V triangle (vs. . IrO₂ 0.4 V 0.4 2 intermittent: 1.45 to 2 V triangle) • IrO₂ 0 V Ξ ----Ir 1.4 V 1.5 0.3 Summary of findings: • Higher performance losses for AST cycle through 0.2 catalyst redox (0 to 2 V): major changes in particle 0.1 size, not final oxidation state 0.5 Performance losses correlate with fraction of Ir in metallic state 10 20 30 1.9 1.6 1.8 2 15 17 Loss mechanism: thinning/increased PTL site-access, \checkmark Cycle [# k] E IV vs RHE1 clear agglomeration, increased interfacial tearing



S.M. Alia, Current Opinion in Chemical Engineering, 2021, 33, 100703.

Accomplishment: Degradation studies of IrO₂ anode catalyst

Potential (V_{iR-corr} vs. RHE)





- On-line inductively-coupled plasma mass spectrometry Ir dissolution data modeled with dissolution of two species: IrO₂ and IrO₂OH, with the latter having slower dissolution kinetics
- IrO₂ activity decreases with increasing potential; Change in potential dependence at 1.6 V suggests an appearance of a different IrO_x species – corresponding with in situ X-ray data
- Under steady-state conditions, catalyst is least stable at potentials between 1.5 and 1.55 V; 37.5 mg/g-h peak dissolution rate



Anode Potential, V

- Dissolution increases after stepping to UPL followed by passivation and decreases with continued hold at potential.
- Dissolution increases with increase in step size
- 200 mV > 150 mV > 100 mV > 50 mV



1.8

1.7

Potential, V

Accomplishment: Membrane Degradation





H2NEW: Hydrogen from Next-generation Electrolyzers of Water

Low Temperature Electrolysis (LTE)

Task 2: Performance and Benchmarking (more content in AMR Poster <u>P196b</u>)





Accomplishments – Hydrogen Crossover (FY'22 Q1 QPM)





- Measured H2Xover between atmospheric and 30 bar backpressure at various current densities up to 4 A/cm²
- Leveraged diagnostic equipment developed within EMN HydroGEN project
- Further advanced method to allow for rapid semi-automated operation
 - 2 min sampling time at most current densities
- \Rightarrow Gas mixture of anode exhaust stream can reach flammable H₂:O₂ ratios
- \Rightarrow Strong dependency of $\rm H_2:O_2$ ratio on
 - Current density
 - Operating pressure
 - Membrane thickness





Harmonized Thermodynamic Equation

$$E_{p,T}^{0} = E^{0} - \frac{S_{R,T^{*}}(T - T^{*})}{2F} + \frac{2.303RT}{2F} \cdot \log\left[\frac{(a_{O_{2}})^{0.5}a_{H_{2}}}{a_{H_{2}O}}\right]$$

- Round Robin evaluation of Voltage Breakdown Analyses across four H2NEW Laboratories
- Deviating results originating from
 - Calculation of thermodynamic reversible cell voltage E_{0}
 - Determination of the "linear" Tafel region
- Harmonization of Voltage Breakdown Analysis through
 - Development of protocol
 - Harmonization of equations
 - Development of statistical tool that supports the selection of the "linear" Tafel region and enables the reproducible determination of the Tafel slope
- ⇒ Successful benchmarking of Voltage Breakdown Analysis of low loading, high performance cell across at all participating four laboratories
- \Rightarrow Publication in preparation for dissemination to community

Low Temperature Electrolysis (LTE)

Task 3: Manufacturing, Scale-up and Integration (more content in AMR Poster <u>P196c</u>)





Accomplishment: Thickening of anode ink by aging

- Objective: Low viscosity of IrOx inks can be problematic for large-scale coating, and the catalyst particles are observed to crash out in ~a day, exacerbating coating issues. We want to further explore and understand parameter effects of the observation that anode ink "resting" can improve processability.
- Anode inks experiencing a multi-day dwell via simple rolling can increase in viscosity by several orders of magnitude
- However, this behavior is only observed for waterrich formulations
 - Observed for water mixtures with both IPA and nPA





Accomplishment: R2R Anode Loading Comparison



- Objective: evaluate R2R anode coating efficacy at different loadings
 - Study R2R gravure-coated anodes at 0.2 and 0.4 mg lr/cm2
 - Compare performance of R2R coatings to spray-coated FuGeMEA at same loading _



- Result:
 - Whereas R2R anode showed comparable performance to FuGeMEA at 0.4 loading, the R2R anode performance at 0.2 loading was much worse
 - Optical microscopy (transmission) showed poor micro-scale uniformity of the lower loaded R2R anode
 - Need additional studies to improve the uniformity of low-loaded anodes via R2R processes

Low Temperature Electrolysis (LTE)

PTLs (Porous Transport Layers)

Cross-task effort (additional information found in Posters 196a, b and c)



Accomplishment: Ti PTL Pore Structure Optimization



- Produced and tested PTLs with range of poreformer size and loading
- <u>Result: 60 v% Ti 40v% 60μm poreformer</u> outperforms baseline commercial PTL





Accomplishments – In-depth Study of PTL / Anode Interface





Accomplishments- Tracking Inhomogeneous Degradation with Electron Microscopy





- Optical images show indentations in anode electrode from PTL fibers
- Similar indentations visible
 in cross-sectional SEM
 images correlate with
 degree of degradation



Low Degradation Regions:

- Limited Ir dissolution
- Similar electrode thickness and density to beginning-of-test MEA

High Degradation Regions:

- Band of dissolved Ir in membrane
- Denser electrode structure
- Curved electrode suggests direct contact with PTL

Responses to Previous Year Reviewers' Comments



- Reviewer Comment: The project is performed solely by national laboratories, so industry participation is largely missing.
- Response: We have done what we can to incorporate this through our Advisory Board and other engagement with industrial partners, but also expect this to change in the near future as additional formal engagement opportunities develop.
- Reviewer Comment: Alkaline electrolysis is not addressed in the project, yet there have been substantial technology advances in recent years, and it is likely to capture a large market share in 2030 and beyond.
- Response: The Bipartisan Infrastructure Law identifies this area as a target area, H2NEW co-sponsored H2-LAWE Workshop/Report in this area, and we anticipate it being included in the near future, we have already begun increasing capabilities to address.
- Reviewer Comment: There is no mention of other emerging technologies, such as HTE proton-conducting technology or LTE anion exchange membrane technology.
- Response: This was by design and is beyond the scope of H2NEW for now and in the realm of HydroGEN (2.0).
- Reviewer Comment: The project is focused on standard technologies and material sets. It would be great to see some investigation into promising alternatives that have the potential to leapfrog over the existing state of the art.
- Response: H2NEW co-sponsored H2-AMP Workshop/Report in this area to help identify research needs in the area of advanced materials for PEM electrolysis, and we anticipate novel material development and incorporation being included in the near future.

Collaborations and Coordination (LTE)





Leverage across other Consortium: HydroGEN 2.0 (HFTO) R2R (AMO) Million Mile Fuel Cell Truck (HFTO) ElectroCAT (HFTO) LTE Strategic Advisory Board Members



Numerous industrial, academia, and international interactions: (IEA, ASTWG, materials suppliers, informal collaborations)

Select group of advisors representing OEMs, tier 1 suppliers, analysis and manufacturing interests.

External Facing Website Established: h2new.energy.gov

Added 3 University participants (Carnegie Mellon, UC-Irvine, Colorado School of Mines), have widely coordinated with other research institutions and industry, expect academia and industry to engage further through FOAs and collaborative research.



Remaining Challenges/Proposed Future Work (LTE) (Refer to P149a,b,c & d for more detail at Task Level)



Task 1 (Durability): Aging Studies; Mitigation Strategies; Ex-situ Characterization of MEAs/components/interfaces; Ex-situ Catalyst Durability; Ex-situ Membrane Durability; Accelerated Stress Test Development

Task 2 (Performance): Performance benchmarking, baselining, validation; Cell performance testing in support of electrode development; Ex-situ studies focused on performance factors; Cell level model development

Task 3 (Scale-up): MEA fabrication, Interface engineering (inks, electrodes, cell integration and interfaces); Components (porous transport layers, recombination layers)

Task 3c (Analysis): Performance, manufacturing, and system models; Durability factors; Energy system integration

Any proposed future work is subject to change based on funding levels

Summary: H2NEW LTE



Degradation and Accelerated Stressor Task 1: Durability: Fundamental Understanding to ASTs to Mitig Operando cell and ex situ component studies ✓ Operando neutron diffraction ✓ Identical location microscopy cell commissioned	ess Test (ASTs) mo established for rrelating with AST ST Working Group Established Mitigation Strategies ✓To be informed by degradation and	d AST		Gas Diffusion L Microporous La Catalyst Layer Pwc Membrane Catalyst Layer IrO ₂ Porous Transpo	ayer ayer Drt Layer
✓ Discretionary funding demonstrated for studying		e e	H ₂ O	Gas/Liquid C	hannel
allocated to operando degradation		Stack Targets	Status	2023	2025
tomography		Cell (A/cm ² @1.9V)	2.0	2.5	3.0
Task 2: Performance: Structure-Property Relations to Optimize	ed Performance	Efficiency (%)	66	68	70
Resoling MEA Cell Modeling		Lifetime (khr)	60	70	80
✓ Identified parameter space and experimental		Degradation (mV/khr)	3.2	2.75	2.25
 Crosstask team matrix that support the model needs of the 		Capital Cost (\$/kW)	350	200	100
established consortium with regards to in-situ, ex-situ, and in-	TEA Sustana Analusia	PGM loading (mg/cm ²)	4	1	0.5
common MEA, testing and prep ✓ Identified and sourced PTL Task 3: Component processing to Scale-up to Systems Analysis Inks, Catalyst layers, porous transport layers ✓ Study of PTL structure property demonstrated ✓ Identified and sourced PTL between 3 labs ✓ Feasibility of PTL fabrication via 3-D printing and tape casting	 ✓ Performance, manufacturing, and system analysis is underway and tools are being developed and implemente ✓ Identified Operating Strategy that Minimizes Cost for Several Electrolyze Costs Identified and sourced PTL ✓ Initial Input to Stack Performance Targets ✓ Cell to system level performance monotonic system several explorements 	d d er del	Minimum cos Capacity fact Mean electric Price ceiling: 00 5000 60 peration per year	HLC HLC Hintmu Electric Capital Fixed O :: \$1.54/kg :: \$0.015 50.029/kWh 000 7000	m cost ity costs costs &M 9/kWh

H2NEW Activities: High Temperature Electrolysis (HTE)

Richard Boardman, H2NEW Deputy Director, and HTE Lead





HTE Organization & Tasks





Relevance/Potential Impact



- When strategically coupled with heat sources such as waste heat from nuclear or solar, High Temperature
 [Steam] Electrolysis is a promising technology for achieving the Hydrogen Earth Shot goal of \$1 for 1 kg hydrogen in 1 decade.
- Widespread commercialization of HTE is currently impeded by poor durability. Developing a fundamental understanding of all failure mechanisms is imperative and necessitates acerated aging protocols that mimic the cell behavior of conventionally aged cells.

DOE HTE Targets

HTE Electrolyzer Stack Goals by 2025			
Capital Cost \$100/kW			
Electrical Efficiency (LHV) 98% at 1.5 A/cm ²			
Lifetime 60,000 hr			

 Successfully elucidating failure mechanisms (including validation of accelerated aging protocols) is contingent on robust characterization and modeling of HTE cells across multiple length scales from nanometer to millimeter.

Quarterly Progress Milestones





	Milestone Description	Summary of Criteria
QP1	Initial Round-Robin button cell testing under standard conditions for 500 hours after achieving steady-state	20% Degradation rate standard deviation over at least 500 H operating time after steady-state has been achieved, in both intra-lab and interlab data sets.
QP2	Characterize cells prior to operation, during and following break-in in terms of performance degradation	Achieve steady state operations in < 500 hour; Measure performance, degradation and time-to-steady-state; Characterize virgin and harvested cells to evaluate break-in changes using SEM/EDS, TEM, XRD, XAS, and TXM
QP3	Complete button cell testing under standard operating conditions (1.3 V, 750°C), using aggressive steam% concentrations (high and low)	Complete button cell testing at the thermoneutral voltage at 750°C with aggressive steam concentrations (high and low); evaluate aggressive steam as a stressor capable of accelerating degradation by > 20%
QP3	Complete parametric testing matrix of 2 to 3 kH long-duration experiments	Mean, steady-state degradation rate for 2 and 3 kH experiments within 3%/kH of the mean
QP3	Fabricate and test large planar cell degradation test and compare outcomes with button cells	Demonstrate similarities and differences of degradation mechanisms at relevant scales
QP4	Accelerated stress test protocol development and demonstration; aggressive steam as a stressor, comparing performance and degradation at 95%, 70%, 50% and 10%.	Demonstrate positive correlation between mean, steady-state degradation rates from one-, two- and three-segment, aggressive steam% experiments, and steam% value
QP4	Develop predictive model for one or more mechanisms of degradation	Compare model outcomes to test cell characterization results

Relevance/Potential Impact





<\$2/kg-H2 is projected when:</p>

- Nth-of-a-kind HTE project; 95%
 manufacturing learning rate
- 700 tonnes/day capacity (i.e. the capacity of a large nuclear power plant) selling power for \$30/MWh
- Stack cost of \$78/kW-dc; Assumes a current density of 1.5 A/cm²
- Stack service life of 4 years at 87% on-line capacity; Assumes a stack degradation rate of 0.86%/1000 operating hours
- Steam utilization of 80% per pass

H2NEW Potential Impact: Cost Reduction of \$0.5-0.75 /kg-H₂

AMR Poster: Wendt et al. 2022, INL/RPT-22-66117

Known Mechanisms of SOEC Degradation



1. Cathode degradation

- Ni coarsening /sintering
- Ni oxidation
- Ni mobility

Leads to reduced triple point dispersion and activity and delamination from electrolyte; Results in decreased e⁻ conduction

2. Dense electrolyte degradation

- YSZ composition changes
- Zr reduction/NiZr alloying Results in decreased
 O⁼ ion conduction



3. GDC barrier degradation

- Gd exsolution
- New Gd containing phases
- Coarsening
- Interfacial void formation

4. Anode degradation

- Interfacial void formation
- Sr reaction with Zr, SrZrO₃
 Lead to increased pO2 and delamination

High Temperature Electrolysis (HTE)

Task 5: Durability Testing and AST Development (more content in AMR Poster <u>P196E, Olga Marina, et al.</u>)



Approach: Experimental Development & Investigation of AST



- Fabricate identical cells using well known state-of-the-art materials
- Define standard experimental conditions, including temperature, gas compositions, current/voltage, to obtaining baseline
- Initiative testing under standard operations to determine potential degradation phenomena
- Identify the number of test repeats to establish confidence
- Determine degradation mechanisms via extensive post-test characterization; compare findings to literature references for SOFC and SOEC
- Identify potential stressors for Accelerated Stress Testing (AST)
- Define AST matric and conduct experiments using incrementally aggressive conditions
- Perform experimental validation and post-test characterization and compare results to those obtained under standard operating conditions
- Ensure active participation of HTE stakeholder advisory board that includes commercial cell developers and academia; seek feedback and guidance from experts in the field
- Develop AST Protocol

H2NEW Cell Fabrication (PNNL Leads)



20 µm

500 µm Ni-YSZ support

Complete:

- Identified state-of-the-art cell designs and materials
 - Cells are Ni-YSZ electrode supported with the YSZ electrolyte, GDC barrier, and LSCF-GDC oxygen electrode
- Established a batch fabrication process of button cells to minimize the variance between the separate cells; developing QA/QC procedures
- Provided all labs with identical cells for • testing and performance validation
- Optimized start-up, test and shut-down procedures between the labs for improved data comparison



Inter-Lab Standardization Testing Protocol



LSCF electrode paste used to mount platinum mesh current collectors on the oxygen electrode.

Steam/Hydrogen electrode uses nickel paste to mount nickel mesh.



Cell preparation standardization

- Current collectors & attachment
- Precious metal reduction







- Reduction
- Compositions





Button-Cell 2-4 cm² active area



Enlarged Planar Cell 10-25 cm² active area

Test Parameters

- Temperature
- Voltage & Current Density
- Steam/H2 mixture flow and contact with cell

Accomplishment: INL example test results during cell break-in



H2NEW: Hvdrogen from Next-generation Electrolyzers of Water

Accomplishment: PNNL Baseline Testing and Degradation Results

0.35

0.3

0.25

0.2

0.15

0.1

0.05

0.07

cell 1

cm²)

Cell resistance (Ω .

Complete:

- Obtained reasonable reproducibility cell performance data within the labs. Total cell loses are dominated by the electrodic resistance; ohmic losses are 1/3 of polarization losses
- Established baseline performance of multiple button cells under standard operating conditions (750°C, in 50% steam at 1.3 Volt) over 1000-6000 hours
- Two degradation periods are observed: fast degradation over the initial 600 hours (break-in) and a slow degradation during the following testing
- Average degradation rate is below 17 ohm·cm² per 1000 hours
- DRT peak-fit analysis attributes degradation during break-in period to two peaks (at 1kH – 10kH and 100 Hz - 1kHz) associated with diffusion phenomena in the oxygen electrode.







Accomplishment: PNNL Accelerated Stress Testing for Protocol





Development



In Progress:

- To develop AST protocol as compared to standard operating conditions, the following stressors have been applied: higher temperature (800°C), higher voltage (1.5 and 1.6 V), higher steam (90 and 95%), and lower steam (10%)
- Higher voltage and higher temperature allows to shorten the break-in periods
- Higher steam concentration has no impact on break-in time and degradation rates

Accomplishment: LBNL Dynamic Cycling for Protocol Development



R&D SOEC testing typically at fixed conditions (V, I, T, P...)

BUT: anticipated operation is highly dynamic



Conversion & Storage, Technical University of Denmark

Complete:

- Literature survey, discussion with advisory board complete
- Highest priority cycling variables: H2/H2O ratio, and voltage Ongoing:
- Test standard cells under dynamic conditions
- Deliver cells to Task 7 for analysis
- Assess viability as an Accelerated Stress Test



	Votes
Dynamic	from
Variable	industry
Steam ratio	5
Power (I,V)	4
Temperature	1
Redox	1
Pressure	0

Accomplishments: LBNL Dynamic Cycling Button Cell Test Results





Upcoming:

- Wider steam content window
- Long-term cycling at fixed steam content limits

Stable cycling 1.3 to 1.6 V

- 1 h to 10 sec half-cycles

Upcoming:

- Wider voltage window
- Long-term cycling at fixed frequency

1.3 V

25

High Temperature Electrolysis (HTE)

Task 7: Advanced Characterization (more content in AMR Poster P196e and p196f)



Overview: Classical and New Characterization methods for SOECs



Complexity of the cell components and interfaces in SOECs necessitate **multi-technique approach** to understand failure mechanisms over a range of aging conditions

Transmission X-Ray Microscopy(TXM) 30 nm Resolution

- Changes in pore structure
- Layer delamination
- Cation migration
- Identification of secondary phase formation

X-Ray Absorption Spectroscopy Imaging 1 µm Resolution Parts per million elemental sensitivity

- Cation migration
- Identification of secondary phase formation

Fe contaminant in GDC TXM Data

X-Ray Diffraction Atomic sensitivity to crystal structures

- Secondary phase formation
- Identification of contaminants
- In-operando experiments



SEM/EDS 0.1 μm Resolution Parts per hundred elemental sensitivity

- Cation migration
- Large scale
 structural changes

Robust characterization over multiple length-scales is critical



Changes in Oxygen Electrode after 3000 hrs tests at 1.3V

No Significant Changes in Hydrogen Electrode after 3000 hrs in 90% steam

Co variance



Sr migration





Gd enrichment at the interface



No Ni coarsening or migration







Triple-phase boundary determined using PNNL modeling tools

Accomplishment: Prediction of X-ray Attenuation through HTE Cells depth profiling as run button cells with synchrotron x-rays (by NREL)

H2NEW U.S. DEPARTMENT OF ENER

First steps towards designing in operando HTE X-ray scattering measurements



*Density reduced by particle packing



Validated X-ray attenuation predictions and **demonstrated** XRD can be used to resolve crystal structures and defects from individual layers of intact cells (XRD, right) with simultaneous compositional analysis (XRF, below)





High Temperature Electrolysis (HTE)

Task 8: Multiscale Modeling (more content in AMR Poster P196g)





Physics-based models developed at LLNL and NETL are being linked across scales and accelerated using data science techniques to connect properties to performance loss



Linkage between processes at different scales is crucial to understanding performance degradation and designing effective AST protocols

Models to Interrogate Proposed Degradation Mechanisms





Early studies on Sr interdiffusion confirm that Sr cannot penetrate grains but could use grain boundaries as a migration mechanism



We are exploring machine learning to accelerate simulations and implementing protocols to combine with NETL SOEC database to test performance under varied conditions

Collaboration and Coordination

H2NEW U.S. DEPARTMENT OF ENERGY

- A highly coordinated, interactive four-prong strategy – leveraging
- leading expertise, capability at the National Labs
- Stakeholder Advisory Board
 - Scott Swartz, Nexceris
 - Greg Tao, Chemtronergy
 - John Pietras, St. Gobain
 - Peter Voorhees,

Northwestern

- Scott Barnett, Northwestern
- Xiao-Dong Zhou, University of Louisiana-Lafayette

Task 5. Cell fabrication, operation, AST development	PNNL, INL, LBNL
Task 6. Task integration and protocol validation	INL & All Labs, SAB
Task 7. Advanced characterization	NREL, PNNL, ANL, SLAC
Task 8. Multiscale modeling	LLNL, NETL, PNNL

Select group of advisors representing OEMs, Tier 1 suppliers, analysis and manufacturing interests

Synergy with DOE-Nuclear Energy

- Light-Water Reactor Sustainability
- Integrated Energy Systems

Leverages other Consortium

- HydroGEN 2.0 (HFTO)
- R2R (AMO)
- Million Mile Fuel Cell Truck (HFTO)
- ElectroCAT (HFTO)



Task 5: Durability Testing and AST Development

- i. SOEC materials and component degradation mechanisms are the result of multiple, coupled phenomena derived from operating conditions and a comprehensive and accurate understanding of the interplay of these phenomena remains to be established
- ii. More testing is needed to understand how to cause relative accelerated stress outcomes
- iii. Larger planar cell test stand capacity is needed to fully understand cell degradation phenomena

Task 6: Task Integration and Protocol Validation

i. H2NEW is generating large volumes of data that require review, validation, and interpretation

Task 7: Advanced Characterization

- i. In-operando testing is needed to reduce the number of test samples and to expedite AST outcomes
- ii. Implementing HTE experiments operating at beam line user stations is not easy nor inexpensive

Task 8: Multiscale Modeling

- i. Defining and modeling the mechanisms requires the combined talents, experience and capabilities of each of the Lab Consortium
- ii. Degradation processes at the YSZ|GDC|LSCF-SDC interface are probably coupled in a non-linear fashion

Proposed Future Work (HTE)



(Refer to P196e,f & g for more detail at Task Level)

Task 5: Durability Testing and AST Development

- i. Continue development of AST protocols that accelerate degradation with fidelity to mechanisms functioning at standard operating conditions
- ii. Mature & deploy augmented testing capability sufficient to cover the parameter matrix and requisite

Task 6: Task Integration and Protocol Validation

- i. Ensure refined protocols are validated, widely available
- ii. Organize data volume and surmount archival,/curation challenges

Task 7: Advanced Characterization

- i. Refine and implement in situ / in operando analyses at APS, SLAC
- ii. Coordinate interface between testing, modeling and characterization

Task 8: Multiscale Modeling

i. Ramp up modeling and simulation efforts with increased funding to LLBL, NETL and PNNL to project degradation phenomena exhibited by AST testing

Any proposed future work is subject to change based on funding levels

Summary H2NEW HTE



- The H2NEW HTE hypothesis is that a systematic, coordinated research program targeting the coupled degradation phenomena will yield refinements to composition, fabrication and operation that will enable HTE technology to overcome current durability barriers
- H2NEW HTE is making progress in developing accelerated stress testing protocol
 - Standard electrode-supported cell has been selected
 - PNNL is fabrication and supply the Consortium the standard cells
 - Inter-Laboratory cell testing is underway, and is closing on consistent test outcomes
 - Initial data from PNNL, and also from LBNL and INL have set a foundation for to establish AST protocol
- Cell characterization using standard microscopy is underway using the existing Consortium state-of-the-art X-ray and electron transmission microscopy.
- NREL has validated X-ray attenuation predictions and demonstrated XRD can be used to resolve crystal structures and defects from individual layers of intact cells (XRD, right) with simultaneous compositional analysis (XRF, below)
- The SAB has provided detailed feedback; a 3 hours seminar of Northwestern University R&D for the past decade
- The HTE Consortium will meet this summer to review progress before ramping up modeling and simulation

Technical Backup and Additional Information Slides



Publications (LTE)



- Alex Badgett, Mark Ruth, Bryan Pivovar, "Economic considerations for hydrogen production with a focus on polymer electrolyte membrane electrolysis," Electrochemical Power Sources: Fundamentals, Systems, and Applications, 2022, 327-364. <u>https://doi.org/10.1016/B978-0-12-819424-9.00005-7</u>
- Bryan S Pivovar, Mark F Ruth, Akihiro Nakano, Hirohide Furutani, Christopher Hebling, Tom Smolinka, "Getting Hydrogen to the Gigaton Scale," Electrochem. Soc. Interface, 30, 85, 2021. DOI: 10.1149.2/2.F19214IF
- Bryan S Pivovar, Mark F Ruth, Deborah J Myers, Huyen N Dinh, "Hydrogen: Targeting \$1/kg in 1 Decade," Electrochem. Soc. Interface, 30, 6, 2021.
- N Danilovic, I Zenyuk, "Hydrogen's Big Shot," The Electrochemical Society Interface 30 (4), 40, 2021.
- K Ayers, N Danilovic, K Harrison, H Xu, "PEM Electrolysis, a Forerunner for Clean Hydrogen," The Electrochemical Society Interface 30 (4), 67, 2021.
- A Kusoglu, "Chalkboard 1-The Many Colors of Hydrogen," The Electrochemical Society Interface 30 (4), 44, 2021.
- A Badgett, M Ruth, B James, B Pivovar, "Methods identifying cost reduction potential for water electrolysis systems," Current Opinion in Chemical Engineering 33, 100714, 2021.
- C Liu, M Shviro, AS Gago, SF Zaccarine, G Bender, P Gazdzicki, T Morawietz, I Biswas, M Rasinski, A Everwand, R Schierholz, J Pfeilsticker, M Müller, P Lopes, R Eichel, B Pivovar, S Pylypenko, K Friedrich, W Lehnert, M Carmo, "Exploring the Interface of Skin-Layered Titanium Fibers for Electrochemical Water Splitting," Advanced Energy Materials, 2002926, 2021.
- Zhenye Kang, Yingying Chen, Hao Wang, Shaun M Alia, Bryan S Pivovar, Guido Bender, "Discovering and Demonstrating a Novel High-Performing 2D-Patterned Electrode for Proton-Exchange Membrane Water Electrolysis Devices," ACS Appl. Mater. Interfaces 2022, 14, 1, 2335–2342. <u>https://doi.org/10.1021/acsami.1c20525</u>
- Xiong Peng, Pongsarun Satjaritanun, Zachary Taie, Luke Wiles, Alex Keane, Christopher Capuano, Iryna V. Zenyuk, Nemanja Danilovic, "Insights into Interfacial and Bulk Transport Phenomena Affecting Proton Exchange Membrane Water Electrolyzer Performance at Ultra-Low Iridium Loadings," Adv. Sci. 2021, 8, 2102950. https://doi.org/10.1002/advs.202102950
- M.R. Gerhardt, L.M. Pant, J.C.M. Bui, A. R. Crothers, V.M. Ehlinger, J.C. Fornaciari, J. Liu and A.Z. Weber, "Methods—Practices and Pitfalls in Voltage Breakdown Analysis of Electrochemical Energy-Conversion Systems", J. Electrochem. Soc. 168, 074503, 2021. DOI:10.1149/1945-7111/abf061.
- Boardman, R.D., High Temperature Steam Electrolysis, Encyclopedia of Nuclear Energy, Volume 3 https://doi.org/10.1016/B9780128197257.002026
- Wendt, D.S., Knighton, L.T., Boardman, R.D., "High Temperature Steam Electrolysis Process Performance and Cost Estimates," IN L/R PT 2266117
- Boardman, R.D., Bragg Sittion, S.M., Otgonbaatar, U., "Developing a low cost renewable supply of hydrogen with high temperature electrolysis," MRS Bulletin, Vol. 47, March 2022, mrs.org/bulleting
- Bao, J., Naveen, K., Rechnagle, K., Wang, C., Koeppel, B., Marina, O., "Modeling Framework to Analyze Performance and Structural Reliability of Solid Oxide Electrolysis Cells," J. Electrochem. Soc, in press https://doi.org/10.1149/1945 7111/ac6f87
- Shen, F., Wang, R., Tucker, M.C., "Long term durability test and postmortem for metal support solid oxide electrolysis cells, J. of Power Sources, Vol, 474, 31 October, 2020, 228618