Energy Materials Network
U.S. Department of Energy

HydroGEN
Advanced Water Splitting Materials

HydroGEN Overview:
A Consortium on Advanced Water Splitting Materials

PIs: Huyen Dinh, Shaun Alia, Bryan Pivovar (NREL); Francesca Toma, Adam Weber (LBNL); Dong Ding, Gary Groenewold, (INL); Anthony McDaniel, Andrea Ambrosini (SNL); Tadashi Ogitsu, Brandon Wood (LLNL)

Presenter: Huyen Dinh, National Renewable Energy Laboratory (NREL)

DOE project (AOP WBS#): 2.7.0.518 (HydroGEN 2.0) & 2.7.0.513 (Node Support)

Date: June 8, 2022

DOE Hydrogen Program

2022 Annual Merit Review and Peer Evaluation Meeting

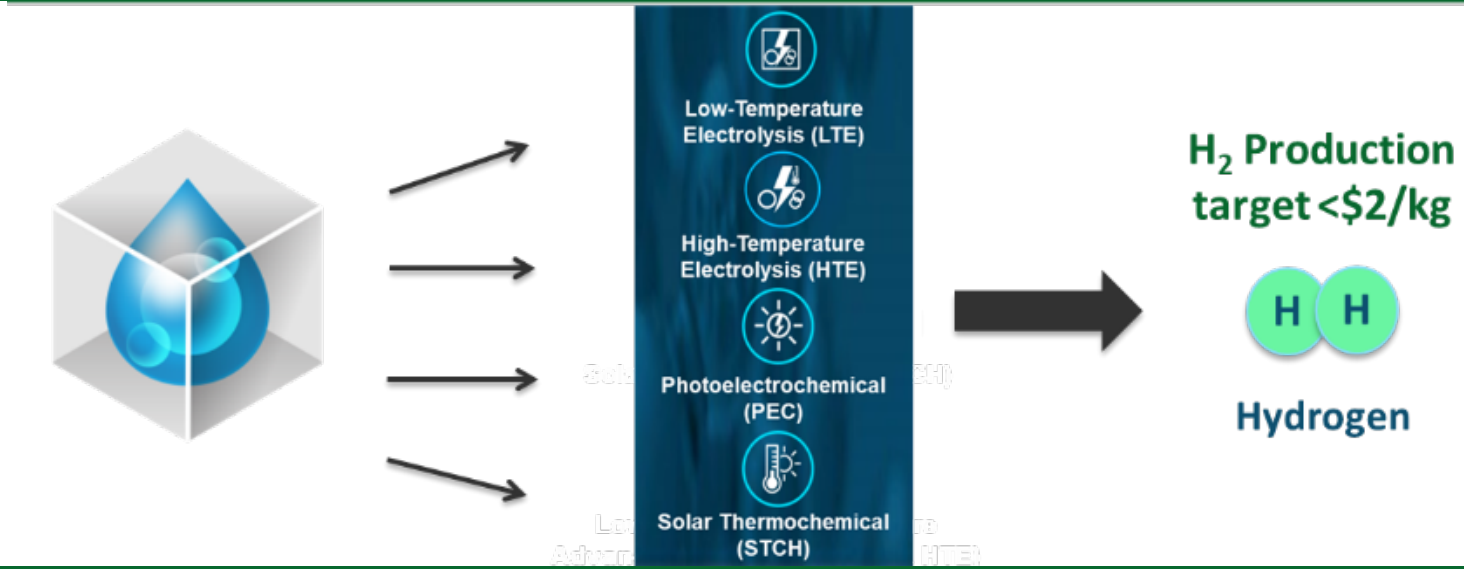
Project ID # P148

NREL Transforming ENERGY
BERKELEY LAB Bringing Science Solutions to the World
Sandia National Laboratories
INL Idaho National Laboratory
Lawrence Livermore National Laboratory

Project Goal

Website: https://www.h2awsm.org/

Goal: Accelerate foundational R&D of innovative materials for advanced water splitting (AWS) technologies to enable clean, sustainable, and low-cost ($1/kg H_2) hydrogen production.



HydroGEN is focused on early-stage R&D in H_2 production and fosters cross-cutting innovation using theory-guided applied materials R&D to advance all emerging water-splitting pathways for hydrogen production

HydroGEN Overview

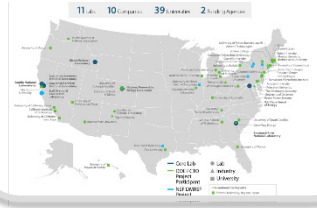
Timeline and Budget

- Start date (launch): **June 2016**
- FY16 DOE funding: **$2M**
- FY17 DOE funding: **$3.5M**
- FY18 DOE funding: **$9.9M**
- FY19 DOE funding: **$8.4M**
- FY20 DOE funding: **$13.175 M**
- FY21 DOE funding: **$5.7 M**
- Planned FY22 DOE funding: **$7.3 M**
- Total DOE funding received to date: **$43.8M**

Barriers

- **Cost**
- **Efficiency**
- **Durability**

Partners:



National Lab Consortium Team

NREL Transforming ENERGY

BERKELEY LAB

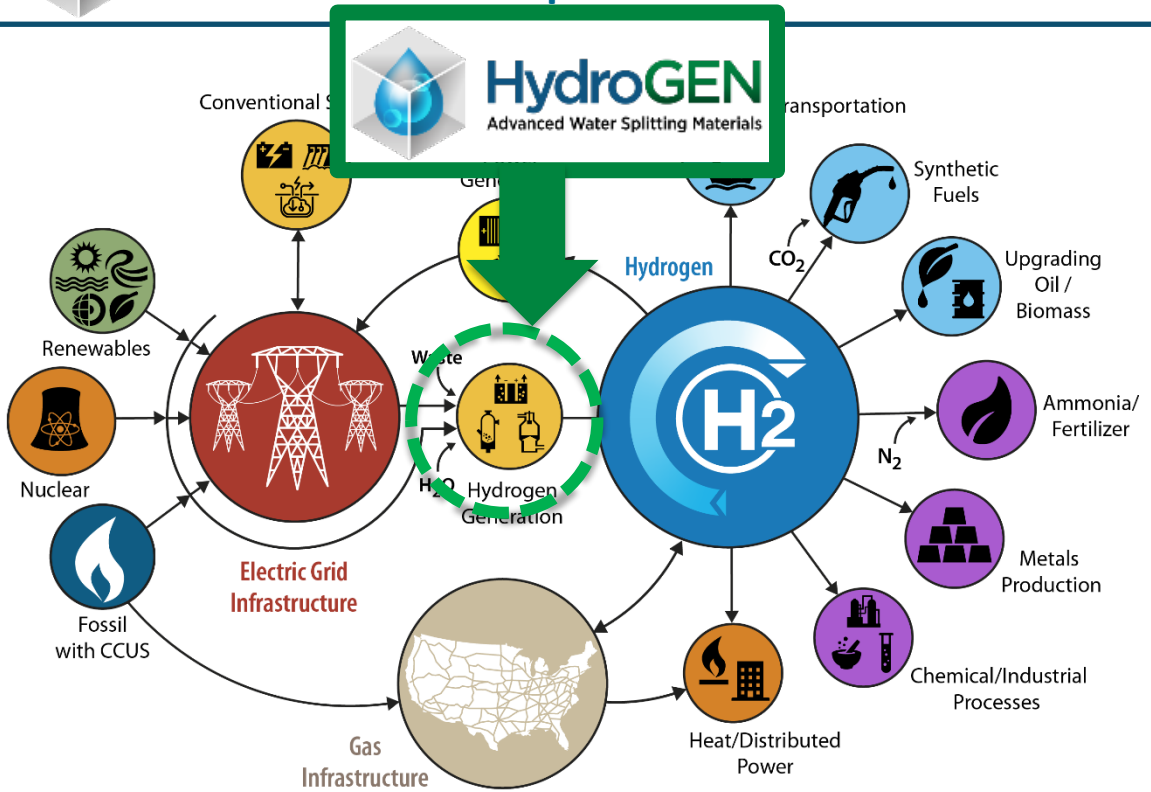
Sandia National Laboratories

INL Idaho National Laboratory

Lawrence Livermore National Laboratory

H2@Scale: Enabling Affordable, Reliable, Clean and Secure energy

Relevance and Impact



Source: DOE Hydrogen and Fuel Cell Technologies Office, https://energy.gov/eere/fuelcells/h2-scale

Transportation and Beyond

Large-scale, low-cost hydrogen from diverse domestic resources enables an economically competitive and environmentally beneficial future energy system across sectors

Hydrogen can address specific applications that are hard to decarbonize

Today: 10 MMT H_2 in the US

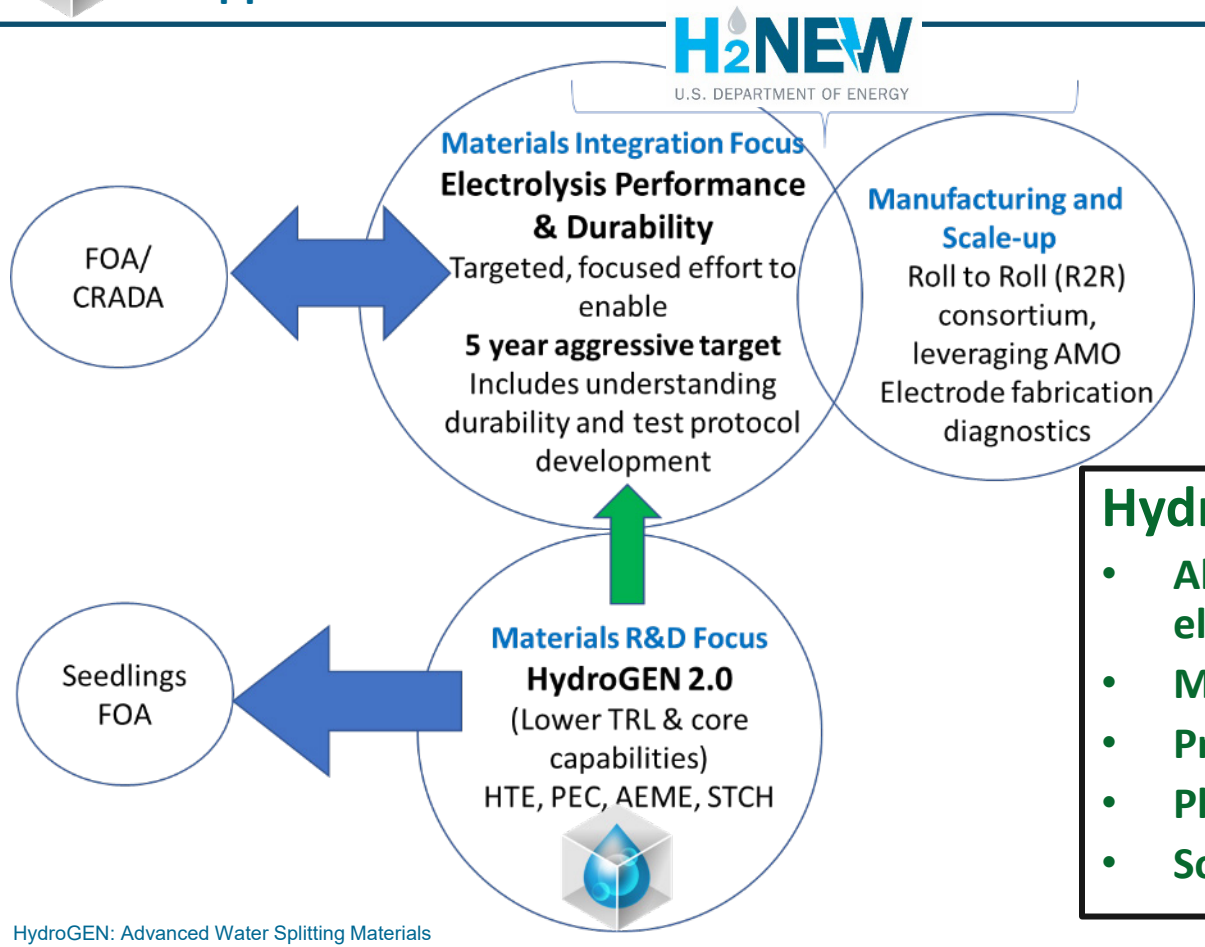
Economic potential: 2x to 4x more

Materials innovations are key to enhancing performance, durability, and reduce cost of hydrogen generation, storage, distribution, and utilization technologies key to H2@Scale

"Hydrogen at Scale (H_2@Scale): Key to a Clean, Economic, and Sustainable Energy System," Bryan Pivovar, Neha Rustagi, Sunita Satyapal, *Electrochem. Soc. Interface* Spring 2018 27(1): 47-52; doi:10.1149/2.F04181if.

HydroGEN Materials R&D Feeds to H2NEW Materials Integration

Approach and Relevance



H2NEW (Hydrogen from Next-generation Electrolyzers of Water):

- Polymer electrolyte membrane (PEM) electrolysis
- Oxygen-conducting solid oxide electrolysis (SOEC)

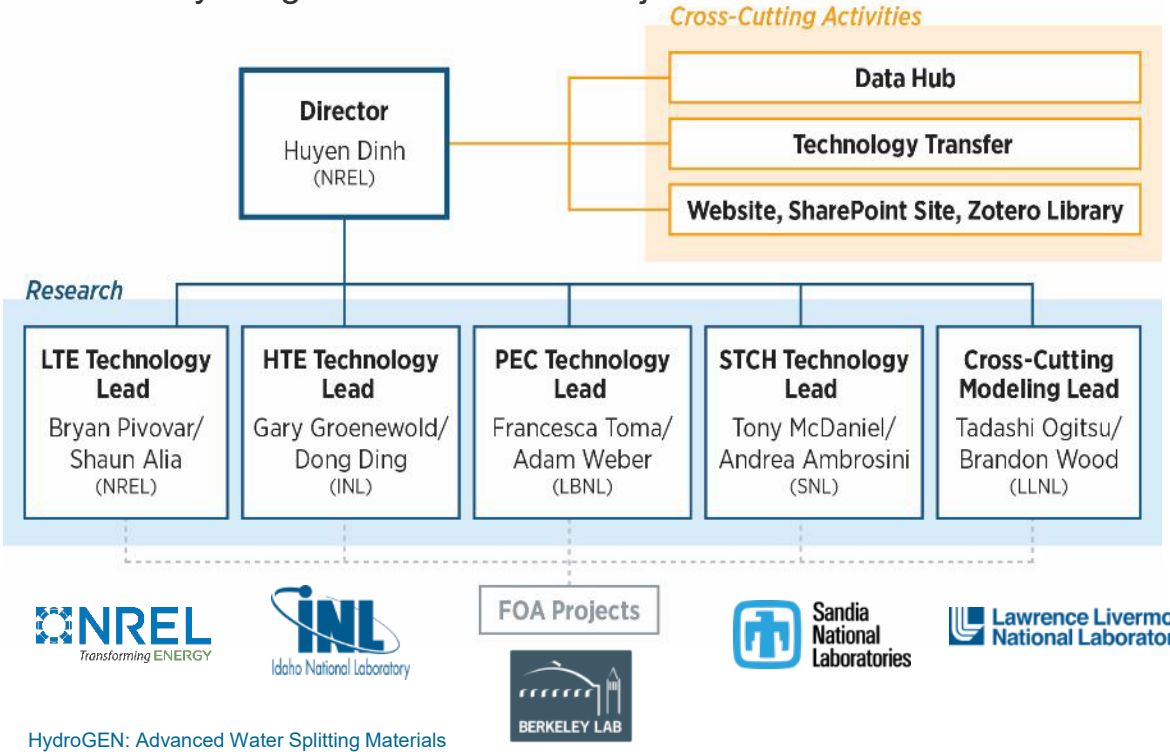
HydroGEN 2.0 (lower TRL AWS):

- Alkaline exchange membrane (AEM) electrolysis
- Metal-supported SOEC (MS-SOEC)
- Proton-conducting SOEC (p-SOEC)
- Photoelectrochemical (PEC)
- Solar thermochemical (STCH)

HydroGEN 2.0 EMN Collaboration and Approaches

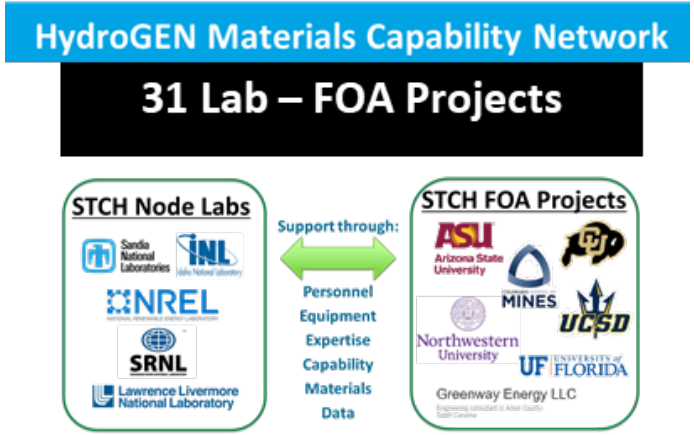
HydroGEN 2.0:

- Total: $4 M/year for 3 years (started on Oct. 1, 2020)
- Early-Stage Materials R&D Projects



HydroGEN 1.0:

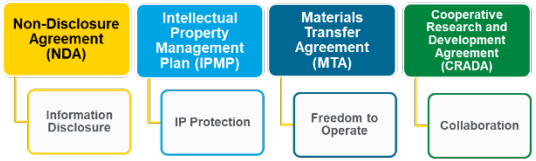
- Lab capabilities and experts support projects



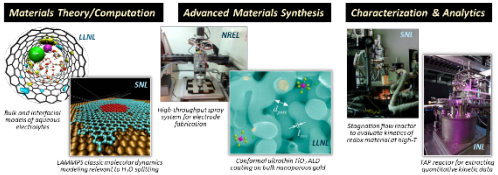
Effectiveness of HydroGEN EMN Framework

Collaboration / Accomplishments, Streamline Access

- 118 Publications, Impact factor* = 2.20
  2,783 citations, 436 authors
- 4 community benchmarking workshops
- 33 project NDAs, 2 MTAs
- 46 capabilities utilized across 6 labs
- STEM Work Force Development
- Diverse leadership and community



https://www.h2awsm.org/working-with-hydrogen



HydroGEN is vastly collaborative, has produced many high value products, and is disseminating them to the R&D community.

*Field-weighted citation impact (FWCI) indicates how the number of citations received by the Publication Set's publications compares with the average number of citations received by all other similar publications in Scopus.

Contributed 2 Articles to ECS Interfaces Winter Issue (Jan. 2022)

Accomplishments

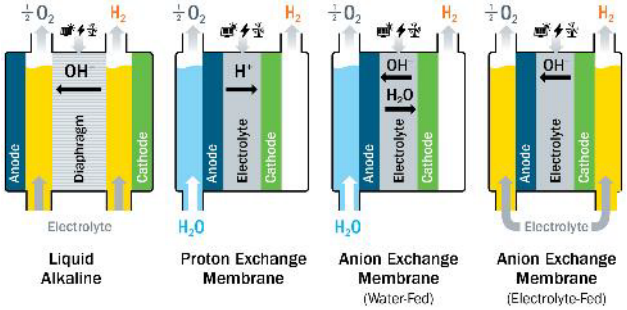
1. **How to Make Clean Hydrogen AWSM: *The Advanced Water Splitting Materials Consortium***

   by Shaun Alia, Dong Ding, Anthony McDaniel, Francesca M. Toma, Huyen N. Dinh

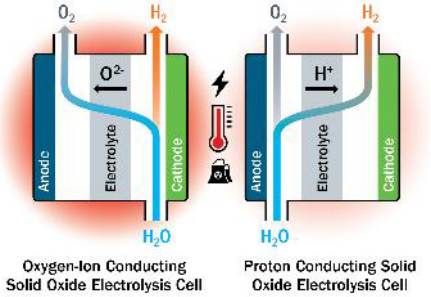
2. **Hydrogen: Targeting $1/kg in 1 Decade**

   by Bryan S. Pivovar, Mark F. Ruth, Deborah J. Myers, Huyen N. Dinh

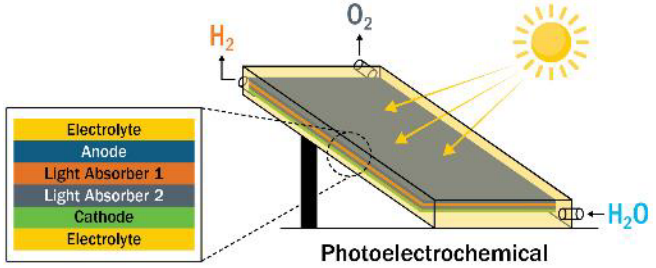
LTE



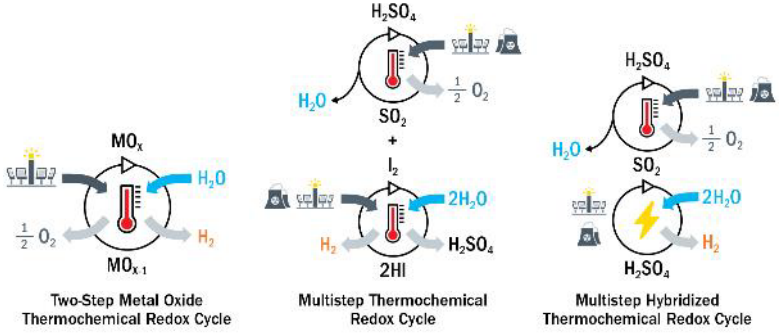
HTE



PEC



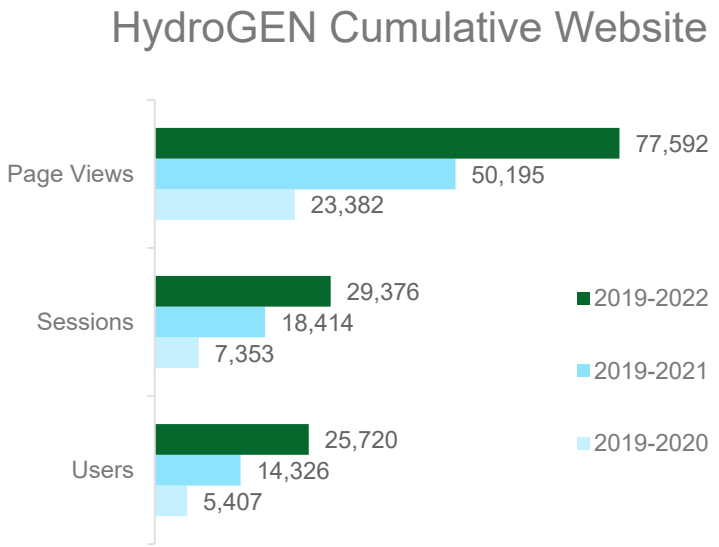
STCH

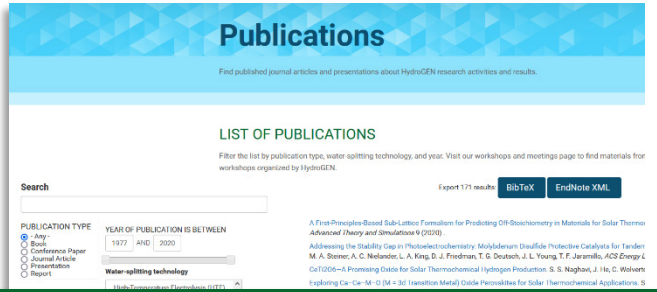


Effectiveness of HydroGEN Framework: Website Outreach

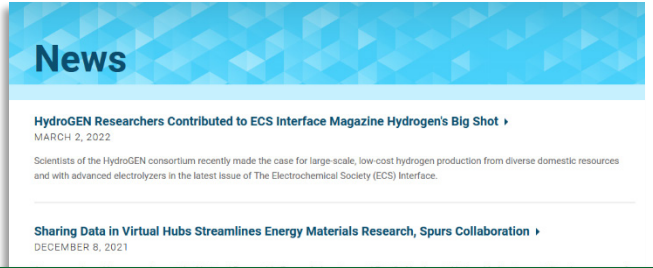
https://h2awsm.org

Website connects users to capabilities, publications, research highlights, contacts, and the Data Hub





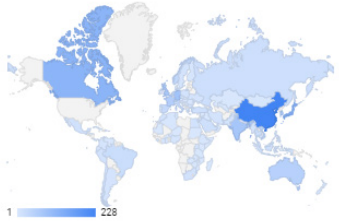
Publications database has search filters for type, year, and advanced water splitting technology, and ability to export results to citation management software



News articles highlight research and accomplishments, including a paper about the Energy Materials Network Data Hub and contributions to ECS Interface Magazine

Effectiveness of HydroGEN Framework: Data Hub Accomplishments

https://datahub.h2awsm.org/

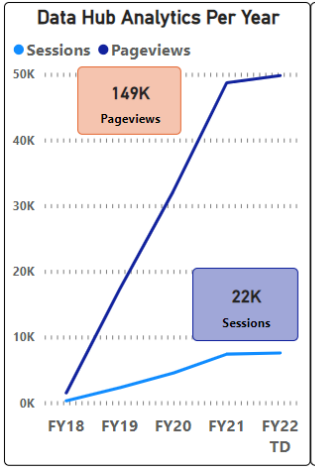


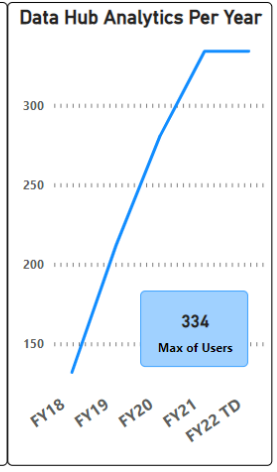
Metric	Projects	Users	Datasets Added	Files added	Site Visitors
FY21-22	7	53	48	2,253	2,459
Total	55	334	416	4,060	4,590

	Country	Visits
1.	China	228
2.	Japan	134
3.	Canada	117
4.	Germany	82
5.	India	76
6.	South Korea	76
7.	United Kingdom	70
8.	France	56
9.	Australia	52
10.	Vietnam	40

Growing & active user community

	Site Visits to Date
Within US	3,309
Outside US	1,757





Data Hub 2021–2022 Year in Review

- Developed **Metadata Service as a solution** to enable more robust metadata curation and validation.
- Defined energy materials **data standards** in metadata for better integration between multiple labs throughout the EMN.
- **Major upgrade of core application infrastructure** to maintain security and cyber standards

Many Types of Experimental Data

Material characterization
- XRD, SFR, XPS, XRF, SEM, TEM, Raman

Device performance
- Electrolysis, PEC J-V, IPCE, Tafel plots

Materials durability data
- TGA, membrane conductivity

XRD = x-ray diffraction; SFR = stagnation flow reactor; J-V = current vs. voltage data; TEM = transmission electron microscopy

XPS = x-ray photoelectron spectroscopy; TGA = thermal gravimetric analysis; IPCE = incident photon to current efficiency

Other = Raman spectroscopy, rheology, helium ion microscope images, conductivity, dilatometry, kinetic, XRF

Data Team:

Community Approach to Benchmarking and Protocol Development for AWS Technologies **Accomplishments**

Goal: Develop best practices in materials characterization and benchmarking: Critical to accelerate materials discovery and development

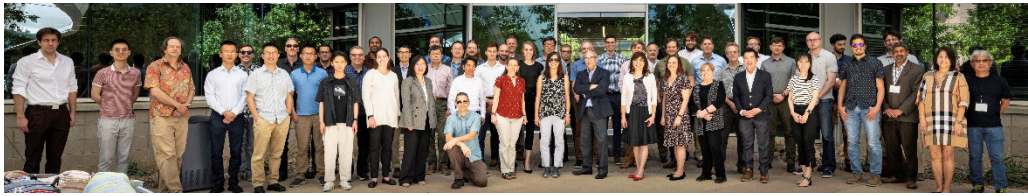
Best Practices in Materials Characterization

PI: Kathy Ayers, Nel Hydrogen (LTE)
Co-PIs: Ellen B. Stechel, ASU (STCH)
Olga Marina, PNNL (HTE)
CX Xiang, Caltech (PEC)
Consultant: Karl Gross, George Roberts

Title	Authors	Technolog
Conductivity and Transference Number Determination Protocols for Solid Oxide Cell Materials	John S Hardy, Aniruddha Pramod Kulkarni, Jeffry W Stevenson and Olga A Marina	HTE
Metal-Supported Solid Oxide Electrolysis Cell Test Standard Operating Procedure	Fengyu Shen, Martha Welander and Michael C. Tucker	HTE
Standard Operating Protocol for Ion Exchange Capacity of Anion Exchange Membranes	Lan Wang, Santiago Rojas-Carbonell, Keda Hu, Brian P Setzler, Andrew R Motz, Matthew E Ueckermann and Yushan Yan	LTE
Assessing the Oxidative Stability of Anion Exchange Membranes in Oxygen Saturated Aqueous Alkaline Solutions	Christopher Arges, Vijay Ramani, Zhongyang Wang and Ryan J Ouimet	LTE
Protocol for Screening Water Oxidation or Reduction Electrocatalyst Activity in a Three-Electrode Cell for Alkaline Exchange Membrane Electrolysis Application	Erin Brahm Creel, Xiang Lyu, Geoff McCool, Ryan Ouimet and Alexey Serov	LTE
Rotating Disk Electrode Standardization and Best Practices in Acidic Oxygen Evolution for Low Temperature Electrolysis	Shaun M Alia and Nemanja Danilovic	LTE
Best practices in PEC: How to reliably measure solar-to-hydrogen efficiency of photocathodes	Francesca Maria Toma, Olivia Alley, Keenan Wyatt, Myles Steiner, Guiij Liu, Tobias Kistler, Guosong Zeng, David Larson, Jason Cooper, James Young and Todd Duetsch	PEC
Long-Term Stability Metrics of Photoelectrochemical Water Splitting	Srinivas Vanka, Guosong Zeng, Todd G Deutsch, Francesca Maria Toma and Zetian Mi	PEC
Comprehensive Evaluation For Protective Coatings: Optical, Electrical, Photoelectrochemical, and Spectroscopic Characterization	Shu Hu, Xin Shen, Rito Yanagi, Devan Solanki, Haoqing Su, Zhaohan Li and Chengxiang Xiang	PEC
Considerations for the Accurate Measurement of Incident Photon to Current Efficiency (IPCE) in Photoelectrochemical (PEC) Cells	David Shai Ellis, Yifat Piekner, Daniel Aldo Grave, Patrick Schnell and Avner Rothschild	PEC
A thermogravimetric temperature programmed thermal redox protocol for rapid screening of metal oxides for solar thermochemical hydrogen production	Michael D Sanders, Anyka M Bergeson-Keller, Eric N Coker and Ryan P O'Hayre	STCH
Performance indicators for benchmarking solar thermochemical fuel processes and reactors	Brendan Bulfin, Miguel Miranda and Aldo Steinfeld	STCH

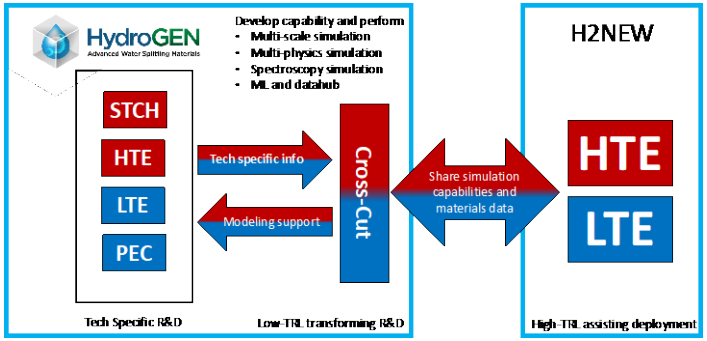
Accomplishments:

- 4 Annual AWS community-wide benchmarking workshop
- 36 test protocols drafted and reviewed
  - 12 test protocols submitted to Frontiers in Energy special issue for publication
  - 11 test protocols reviewed and ready for public dissemination on Data Hub
- 40 additional protocols in drafting process
- Developed high-level roadmaps by AWS technology
- Disseminated information to AWS community through HydroGEN Data Hub, website, SharePoint site, email, quarterly newsletters, and workshops
- Participation from both HydroGEN and H2NEW consortia
- Strong community engagement and participation, nationally and internationally



Science Challenges for HydroGEN 2.0:
Approach

- ***LTE***: improve AEM electrolysis performance and durability by determining the role of supporting electrolyte and the limiting factors behind DI water operation
- ***HTE***:
  - **MS-SOEC**: improve performance and durability with a scale-up cell
  - **p-SOEC**: understand the proton conduction and electronic leakage mechanisms of electrolyte materials in proton-conducting SOEC
- ***PEC***: materials stability and device durability
- ***STCH***: identify and understand how structural features, composition, and defect dynamics engender high capacity–high yield behavior in materials
- ***Cross-Cutting Modeling:*** theory-guided design to analyze performance and durability of materials under simulated operating conditions



Responses to Previous Year Reviewers' Comments

The current form of the consortium is extremely laboratory-heavy. HydroGEN 1.0 had 30 funding opportunity announcement (FOA) projects, and HydroGEN 2.0 appears to have five to date. While the laboratories within the consortium are collaborating very well together, HydroGEN 2.0 should ensure that similar outreach and external interaction to HydroGEN is achieved.

- HydroGEN 2.0 is comprised five specific R&D projects that are carried out by the consortium national labs. In addition to this, the HydroGEN consortium labs continue to support the eleven Round 2 FOA projects. The Round 1 FOA projects have ended. If DOE has another FOA and awards, then HydroGEN consortium national labs will support, interact, and collaborate with the new awarded projects.

Overall, this is a great initiative to bring together all fundamental R&D related to hydrogen. Also, the work makes good use of national laboratory resources.

- Thank you. We agree.

The decision to focus fundamental studies on selected early-TRL areas is a strength.

- Thank you. We agree.

This (collaboration and coordination) is definitely a strong component of the consortium. The external collaboration has greatly improved the productivity of the national laboratory system and energized the academic and industrial partners. This model should be continued and stressed.

- Thank you. We agree.

The community would benefit from more thought around how research in each of the different technology areas can support success in the others. The obvious mechanism is modeling, but it would be good to see this fleshed out more. The "roadmap for cross-cutting modeling" is a great way to start.

- Thank you. We agree.

Responses to Previous Year Reviewers' Comments

A potential weakness is the project's lack of a defined pathway to transfer laboratory knowledge to other researchers. For example, while the super nodes were formed to address key challenges, there is no connection to the balance of the community. It is unclear how the advancements in IrO2 roll-to-roll coating will get to the U.S.-based companies, such as Plug Power and Nel Hydrogen, or a U.S.-based membrane electrode assembly manufacturer. It is unclear how the improved solid oxide electrolysis cell (SOEC) electrode will get utilized by Oxeon or other SOEC companies. In addition, the benchmarking effort appears to disappear from HydroGEN 2.0. It is unclear how the workshops and other activities will be maintained.

- Much of the work within HydroGEN has been published and disseminated to the public and some of the work has been patented and can be licensed by industry. HydroGEN has an intellectual property management plan. Industry partners can work directly with the HydroGEN labs via DOE FOA projects or CRADAs. The component integration and scale up work have been moved to the H2NEW consortium.
- The benchmarking effort has not disappeared from HydroGEN 2.0. HydroGEN continues to work with the community and the benchmarking project and team that organizes workshops and facilitates protocol development. The next step is to publish the protocols and validate them.

STCH is very modeling-heavy. While this is important work, and the focus of the Energy Materials Network is materials, there are cases where infrastructure and equipment are necessary to really understand material performance, such as some of the reactor facilities in Europe. The strategy should include evaluating the need for a domestic resource for these development efforts.

- While not within the scope of this EMN, we agree with the reviewer's statement. Moving STCH technology forward will take a combination of material R&D and hardware (i.e., solar receiver/reactor) R&D. It is very challenging to develop one without the other because concepts for receiver design, cycle operation, and even plant optimization are critically linked to studying material behavior within these systems.

Responses to Previous Year Reviewers' Comments

The demarcation of HydroGEN 2.0 and Hydrogen from Next-generation Electrolyzers of Water (H2NEW) based on current technology readiness levels (TRLs) is smart and practical to move the two consortiums forward. It is a good idea to focus HydroGEN 2.0 on low-TRL areas of AWS R&D, since low-temperature polymer electrolyte membrane (PEM) electrolyzers are far more advanced compared to other AWS routes. Although the HydroGEN 2.0 scope seems to specifically exclude PEM-based LTE technologies, it is not clear why PEM electrolysis projects are still in the portfolio.

- The Chemours seedling (P186, A. Park) is the only HydroGEN EMN effort in PEM-based LTE technologies. The project focuses on developing membranes with low hydrogen cross-over. Materials development falls within HydroGEN. HydroGEN does not specifically exclude PEM-based LTE technologies. It includes material development and excludes integration and scale up. Furthermore, this seedling began in 2019, prior to the formation of H2NEW and the demarcation of HydroGEN/H2NEW, and has met milestone and GNG decisions and is continuing until to the third budget period of the project.

Given the commercial state of LTE PEM technology, it is not clear that the consortium should continue to invest in PEM electrolyzer component integration. The project should consider moving PEM electrolysis work to H2NEW instead. It is not compatible with the rest of the low-TRL efforts.

- HydroGEN does not work on PEM electrolyzer component integration, only materials development. The PEM electrolyzer component integration and scale up work has been moved to H2NEW consortium.

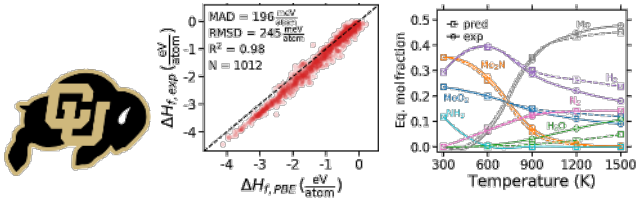
Solar Thermochemical Water Splitting (STCH) Technical Accomplishments: Tony McDaniel

STCH Top Accomplishments

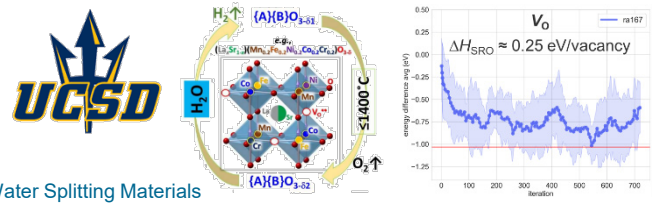
- Theory-based computational tools developed by seedling projects are rapidly identifying new STCH materials.
- Software tools are in place and continue to evolve for critically assessing STCH pathway viability based on measured material properties.
- A high-throughput materials search strategy has been developed to identify STCH materials using DFT and a Machine Learning (ML) model. Workflow applied to 10,000's oxides found in Materials Project, identified ~200 promising new STCH compounds.

Technical Accomplishments: Theory-based Computational Tools Rapidly Identifying New STCH Materials

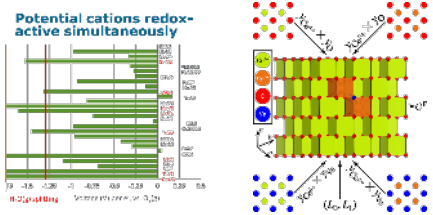
- Use machine-learned models coupled to DFT to discover new redox materials.
  - Rapidly screen materials based on machine-learned predicted stability
  - Formulate descriptor(s) for predicting reaction network energetics and equilibrium



- Develop design rules for high-entropy perovskite oxides.
  - Theoretical screening of HEPO materials using DFT Monte-Carlo
  - Non-stoichiometry of redox cycles studied and results fed back into theoretical calculations



- Incorporate second redox active sublattice to modify thermodynamics.
  - DFT method to predict $\Delta\delta$ a priori using simple sublattice model formulations
  - Discover compounds with optimized thermo (δH, δS)



- Water splitting functionality has been verified in several predicted formulations
- Validated high-throughput computational tools are rapidly expanding the known STCH material space

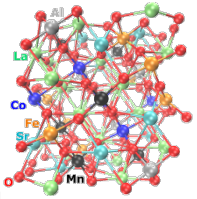
Technical Accomplishments UCSD Seedling Project: HydroGEN Node Support Provided by NREL and SNL

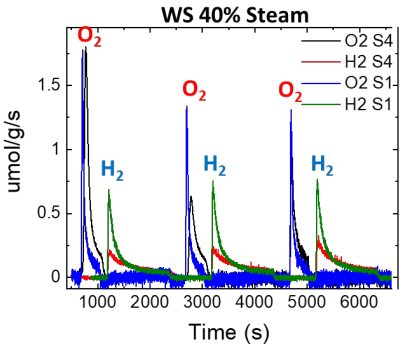
Project ID PD194

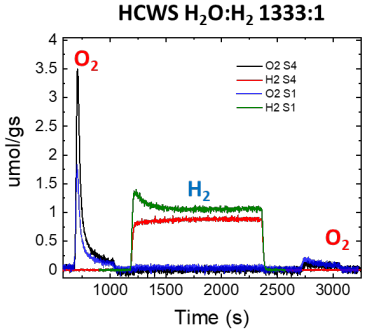
- Subtle compositional variation in $La_{0.8}Sr_{0.2}(Mn_{\alpha}Fe_{\alpha}Co_{(0.16\ or\ 0.40)}Al_{\alpha})O_3$ greatly affects redox behavior.

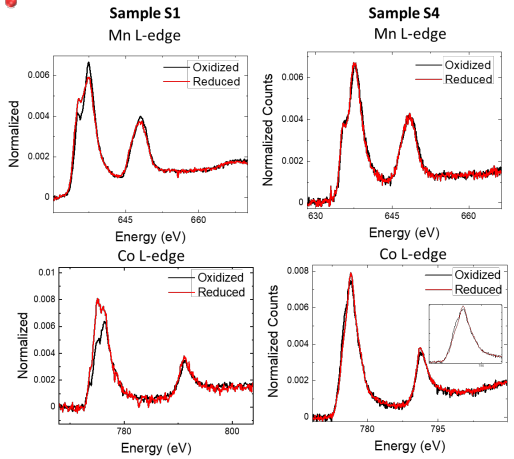
Sample compositions	
HEPO-S1	$(La_{0.8}Sr_{0.2})(Mn_{0.28}Fe_{0.28}Co_{0.16}Al_{0.28})O_3$
HEPO-S4	$(La_{0.8}Sr_{0.2})(Mn_{0.2}Fe_{0.2}Co_{0.4}Al_{0.2})O_3$

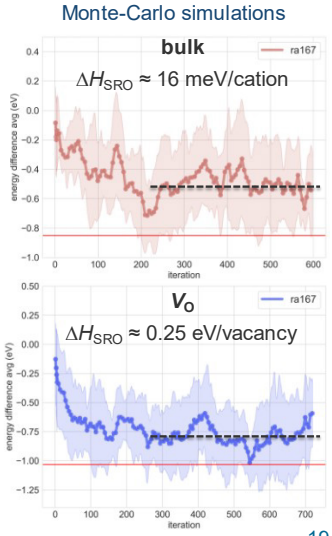
- Sampled configurational disorder via Monte-Carlo simulations for bulk and O vacancy defect structures.
  - Short range ordering (SRO) is moderate in oxidized bulk
  - Co is redox active in reduced oxide, preferential coordination of O vacancies





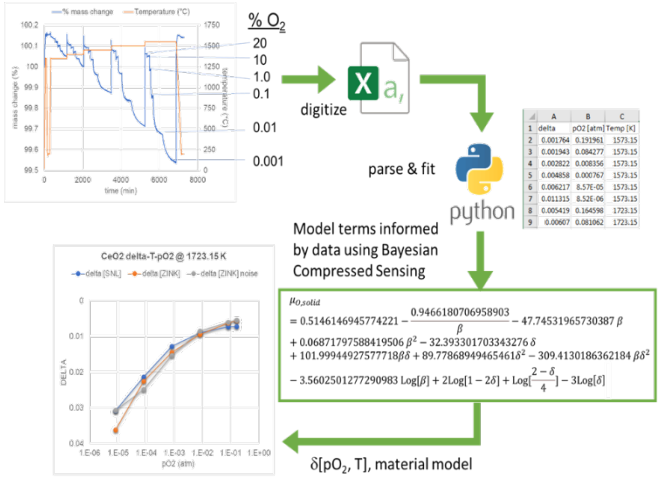






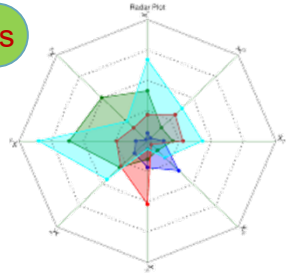
Technical Accomplishments STCH 2.0: Critically Assess STCH Pathway Viability

- Software developed in pure python to formulate a robust material thermodynamic model from TGA data.
  - Jupyter notebooks for interfacing platform
  - ML approach for term selection in compound energy formalism (CEF)
- Cycle efficiency model linked to DAKOTA for automated parametric analysis and optimization.
- Developing a radar plot to graphically depict material performance normalized to CeO_2.
  - coordinating with Benchmarking Group for metrics and choice of exemplar materials ($FeAl_2O_4$, $Ba_4CeMn_3O_{12}$, seedling projects)

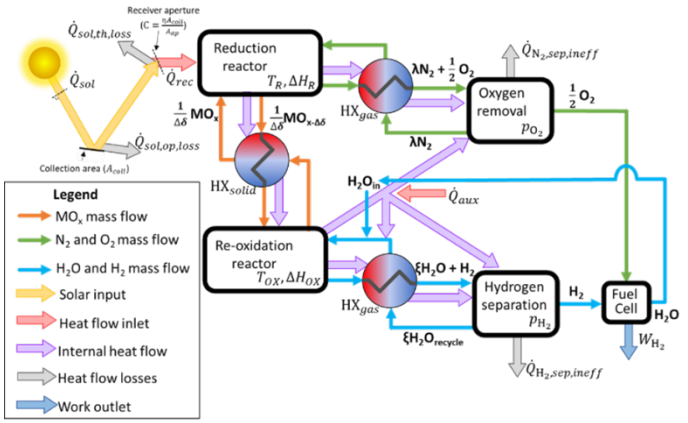


Rigorously assess selected material formulations

METRIC	VALUE
Efficiency	ΔG^o (homolysis) / ΔH_R
Efficiency	optimum derived from model?
Kinetic	Time to 90% of $\Delta\delta$ (H_2O)
O_2 capacity, δ_R	mole O_2/mole atom (10pa O_2)
H_2 capacity, $\Delta\delta$	mole H_2/mole atom (H_2O)
H_2 capacity, $\Delta\delta$	mole H_2/mole atom (H_2O:H_2 = 1000)
H_2 capacity, $\Delta\delta$	mole H_2/mole atom (H_2O:H_2 = 100)
H_2 capacity, $\Delta\delta$	mole H_2/mole atom (H_2O:H_2 = 10)



normalized to CeO_2

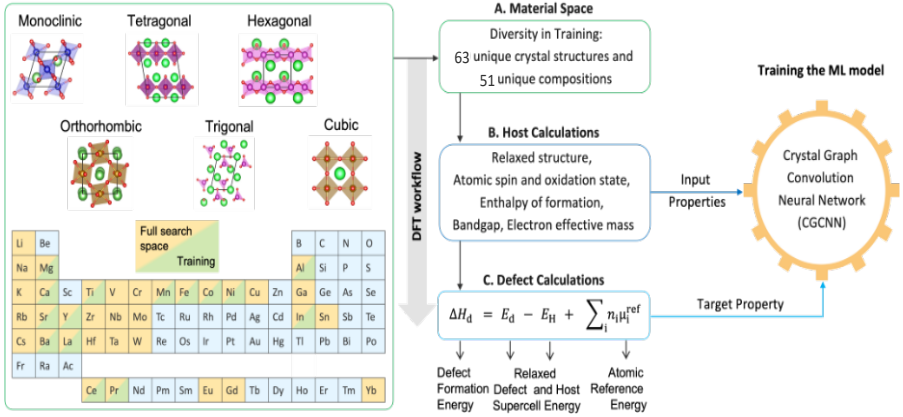


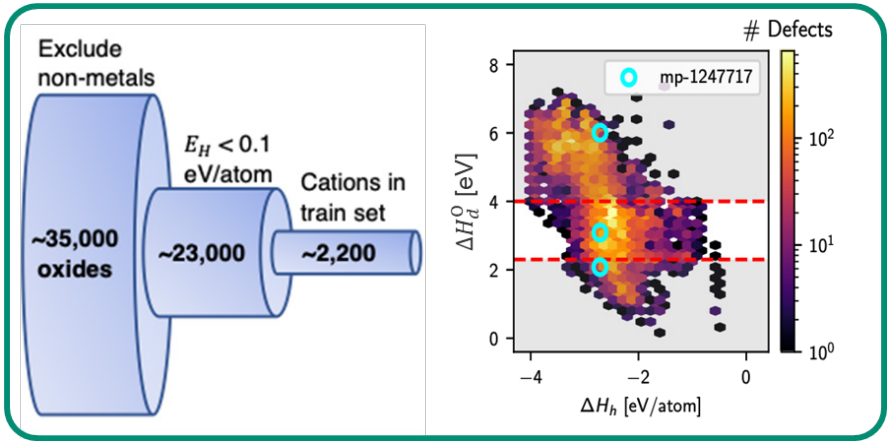
Technical Accomplishments STCH 2.0: Apply Theory-guided Design of Materials to Achieve High Capacity - High Yield H_2 Production

- Developed a DFT-trained Machine Learning model to identify STCH materials.
  - Global defect properties (ΔH^0_d) encoded in ground-state crystal structure
- Diverse and unique DFT training space comprised of 200 binary and ternary oxides.
  - Range of oxidation states (2^+ to 5^+), ionic radii, coordination environments
  - 63 unique crystal structures, 51 unique stoichiometries

Beyond perovskites: 200 host structures – 1500 defect relaxations

Use Crystal Graph Convolutional Neural Network model to screen Materials Project for ALL DEFECTS

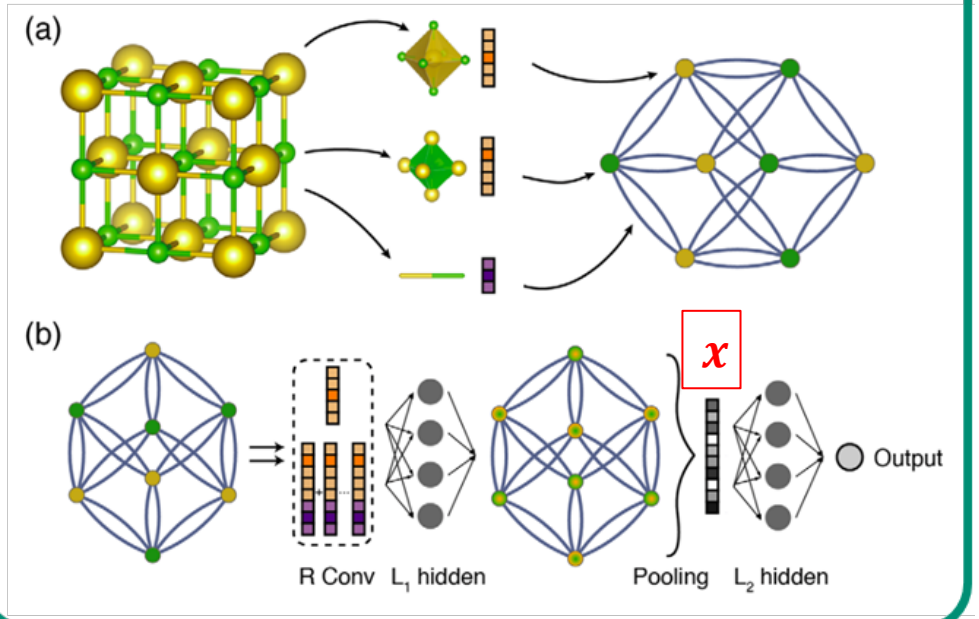




Technical Accomplishments STCH 2.0: Graph Neural Network (GNN) Models Perform "Automated Feature Extraction" Directly from Crystal Structure

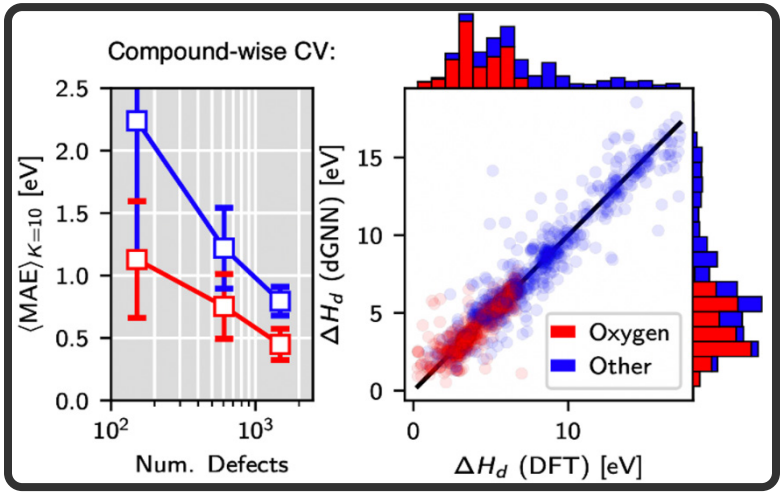
- ***Input*** host properties.
  - relaxed host crystal structure
  - optional: atom oxidation state, atom magnetic moment, compound formation enthalpy, bandgap, e^- effective mass
- Interpret crystal as a graph.
  - nodes = atoms, "bonds" = edges
- Pass information between neighbors.
- Pool atom features to create crystal feature vector (χ).
- ***Output*** atom site-specific defect formation energy without knowing the relaxed defect structure.

Automatically extract the feature vector from the crystal, ***e.g. GNNs***[1]



[1] Xie, et al. *P.R.L.* 120 (14), **2018**

Technical Accomplishments STCH 2.0: Model Validation and High Throughput Screening



- Expected ΔH^0_d MAE for unseen compounds < 450 meV.
- Robust prediction of O and non-O vacancies.

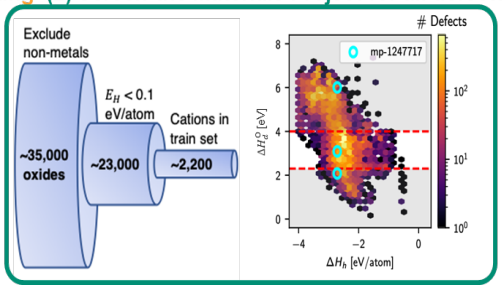
MAE = mean absolute error

ML screens 10,000's of MP structures in minutes that would take 1,000's of DFT months

(1) Co-design of defects and stability for water-splitting

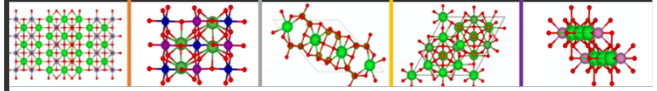
Metric	Requirement
Frac. of defects w/$\Delta H^0_d > 2.3$ eV	$x_{\min} = 1$
Frac. of defects w/$\Delta H^0_d \in [2.3, 4.0]$ eV	$x_{\text{rng}} > 0$
STCH operating range conditions (P_{O_2})	$\Delta\mu'_{O_2}$
Compound stability range	$\Delta\mu_{O_2}^{\phi_H < \{0,\,0.1,\ldots\}}$
Stable in the target range	$\Delta\mu_{O_2}^{\phi_H < X} \cap \Delta\mu'_{O_2}$

(2) Screen the Materials Project for all defects



(3) Identify targets w/increasingly stringent metrics

197 formulas (48 training)	114 formulas (33 training)	34 formulas (17 training)	16 formulas (11 training)	9 formulas (9 training)
$x_{\min,1} = 1$ $x_{\text{rng},1} > 0$ $\Delta\mu_{O_2}^{\phi_H<0.1}$	$x_{\min,2} = 1$ $x_{\text{rng},2} > 0$ $\Delta\mu_{O_2}^{\phi_H<0.1}$	$x_{\min,3} = 1$ $x_{\text{rng},3} > 0$ $\Delta\mu_{O_2}^{\phi_H<0.05}$	$x_{\min,3} = 1$ $x_{\text{rng},3} > 0$ $\Delta\mu_{O_2}^{\phi_H=0}$	$x_{\min,3} = 1$ $x_{\text{rng},3} = 1$ $\Delta\mu_{O_2}^{\phi_H=0}$
$Sr_6Ti_3FeO_{14}$ (mp-1645141)	La_2MnCoO_6 (mp-19208)	$BaSr(FeO_2)_4$ (mp-1228024)	$Ba_6SrLa_2Fe_4O_{15}$ (mp-698793)	$Ba_3In_2O_6$ (mp-20352)



- Identify all candidates satisfying minimum requirements
- Identify candidates with increasingly certain performance
- Mainly IDs known, synthesizable compounds

- Model rediscovers known water-splitting oxides and identifies new ones.

Proposed Future Work for STCH 2.0

- STCH 2.0
  - Use the technology assessment methodology derived during the course of this project to evaluate material viability (a selected group of known and new materials will be evaluated for their potential to meet DOE STCH technology performance targets)
  - Demonstrate theory-guided design of materials using machine learning to identify one or more redox active materials that optimize the capacity/yield tradeoff
- Leverage HydroGEN nodes to enable successful completion and continuation of the seedling projects.

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

High Temperature Electrolysis (HTE) Technical Accomplishments: Dong Ding

HydroGEN 1.0 HTE Projects and Lab Collaboration

Project-driven tech transfer, resulting in

- **Efficiency**
- **Yield**
- **Cost**
- **Durability**
- **Manufacturability**

HTE Node Labs

- Idaho National Laboratory
- NREL Transforming ENERGY
- Sandia National Laboratories
- BERKELEY LAB
- Lawrence Livermore National Laboratory

Support through:

- Personnel
- Equipment
- Expertise
- Capability
- Materials
- Data

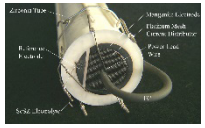
Interactive HTE Projects

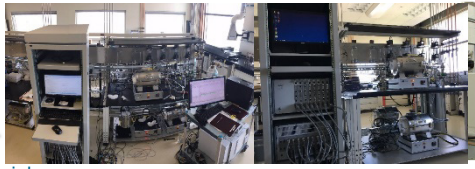
p-SOEC

- Raytheon Technologies
- University of Connecticut
- West Virginia University
- REDOX (Redox Power Systems, LLC)

o-SOEC

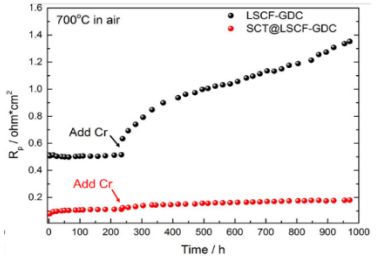
- Northwestern University
- SAINT-GOBAIN
- University of South Carolina
- NEXCERIS (where energy meets environment)

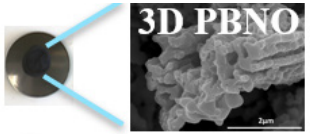


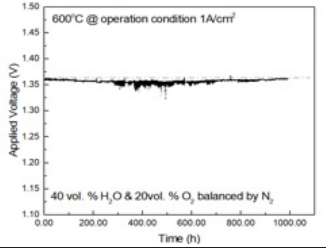



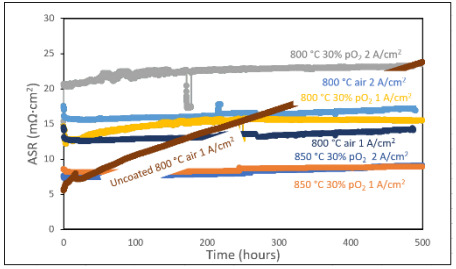

HydroGEN 1.0 HTE (seedling projects with nodes)
Technical Accomplishment Highlights

- **(Northwestern U, LBNL and INL) Developed models to explain and predict Ni migration degradation in Ni-YSZ fuel electrodes:** Analytical calculations and phase-field simulations of Ni migration in Ni-YSZ fuel electrodes have been developed, where the migration is driven by a gradient in interfacial tension. The results provide predictions of the factors that accelerate degradation via this mechanism.
- **(USC and INL) Demonstrated a 650°C-barrier layer free and 700°C-bilayer oxygen electrode for o-SOEC:** Determined exchange current density and transfer coefficient of each oxygen electrode and developed criterion to predict safe operating condition. Established a multi-physics electrochem-chemo-mechano model to predict failure mode.
- **(WVU and INL) Developed 3D hierarchical electrode for durable operation:** Developed highly performing triple conducting electrode with 3D microstructure and demonstrated ~1A/cm^2 at 1.37 V 600°C and durability of <30 mV/1000 h.
- **(Redox, INL and NREL) Developed multilayer electrolyte with compatible electrode:** Stable protective layers, deposited by sputtering, tested >200 hours in 50-100% steam at 500°C, and integrated the INL's stable steam electrode.
- **(Nexceris and INL) Developed advanced interconnect coating at varied testing metrics:** Explored advanced coatings under aggressive SOEC conditions, and demonstrated < 50 mΩ·cm^2 change for 500 hours.





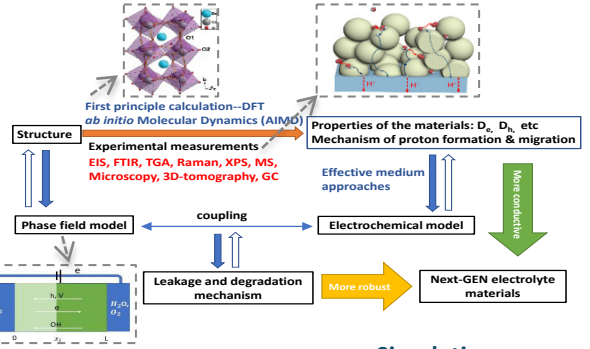


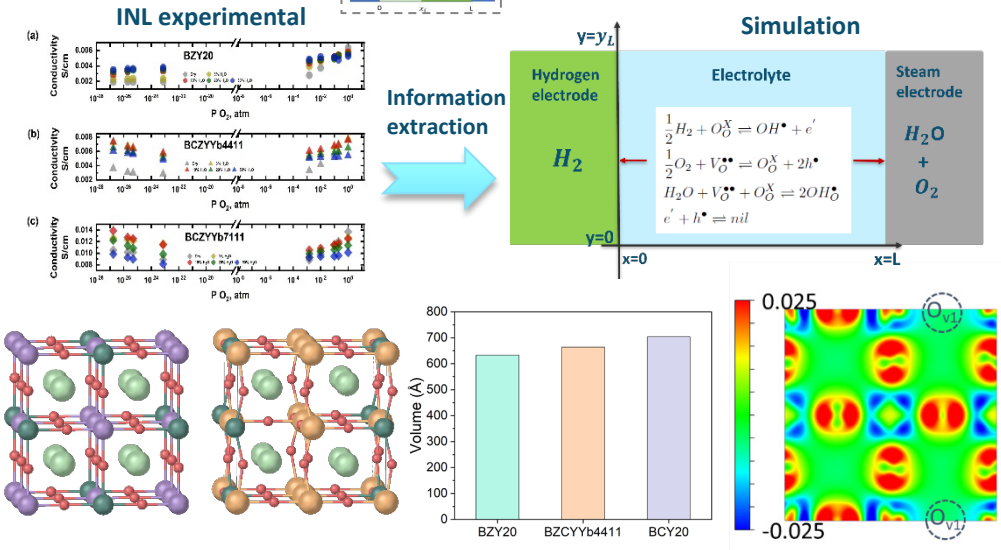


HydroGEN 2.0 HTE p-SOEC Approach:
Combine Multi-Scale Computation and Experiment to Develop Electrolyte Material

- **Develop effective approaches to suppress electronic leakage by understanding the proton conduction and electronic leakage mechanisms.**
- **Develop a robust, energy-efficient, and reliable electrolyte, for p-SOEC at 500-600°C, achieving high Faradaic efficiency (FE) and long durability.**

- **Framework:** Established an efficient framework by integrating experiment and multi-scale simulation (DFT/AIMD, phase-field model) for mechanism study in broad time and size scales.
- **Experiment:** Electrochemical characterization of state-of-the-art electrolytes to disclose thermodynamic information for feeding NWU's modeling study.
- **Phase-field simulation:** Studied transference number of each charge carrier under different operating conditions.
- **DFT/AIMD:** Explored electronic density distribution, proton formation and migration in BZY and BCY based electrolyte materials.



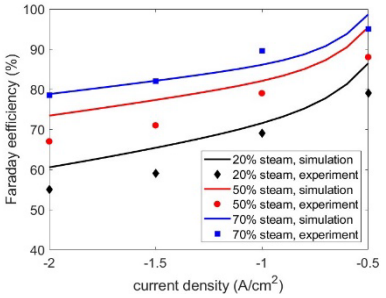


DFT models with different elements on B site and different oxygen vacancies

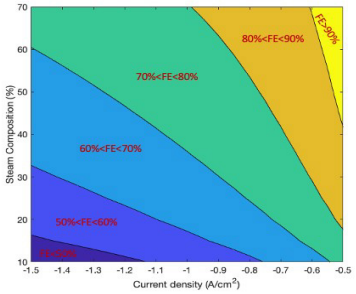
HydroGEN 2.0 Technical Progress:
Accurate Validation of Multi-Scale Framework (*FY2021 Annual Milestone Completed*)

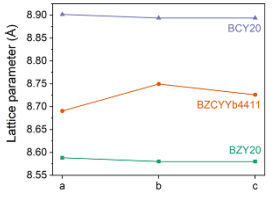
- **High accuracy in experimental data simulation**
  - Electrochemical performance under different operating conditions was simulated by phase-field models.
  - The accuracy of phase-field simulation was validated using electrochemical experiment data.
  - Predictions of optimal conditions will help tune experiment protocol.
- **> 75% agreement in predictions**
  - DFT/AIMD models with different distributions of doped elements were explored and confirmed by comparison with lattice parameters from X-ray diffraction patterns.
  - Proton formation and migration mechanisms were explored. Proton migration in BZY and BCY based electrolyte materials occurs via a Grotthuss mechanism.
  - 90.9% of calculated energy barriers agree with the range of published computational and experimental data.

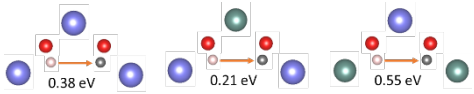
Validate experiment data

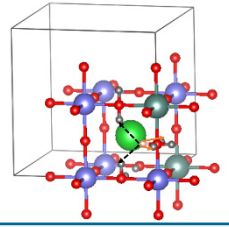


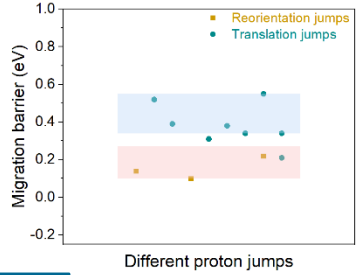
Predict optimal conditions











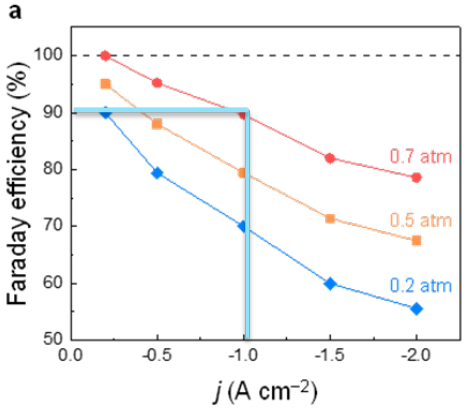
Successfully completed annual milestone by establishing multi-scale framework and validating its accuracy.

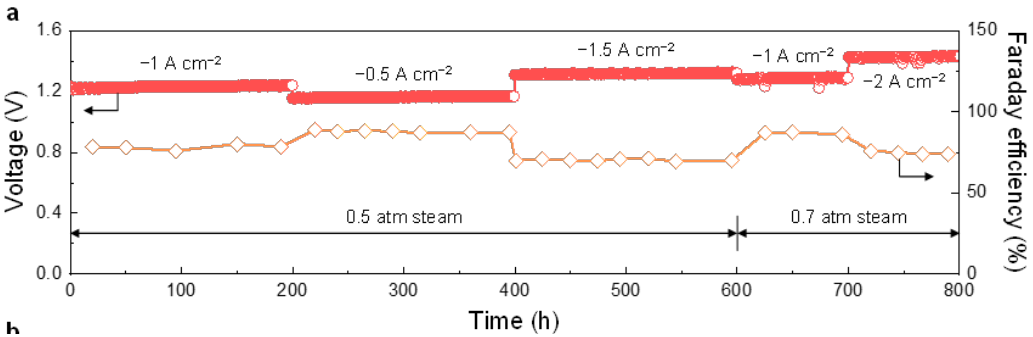
HydroGEN 2.0 Technical Accomplishment:

Achieved GNG point: BZY cell shows >90% Faradaic efficiency at 1.0 A cm^{-2}, 600°C, 70% H_2O

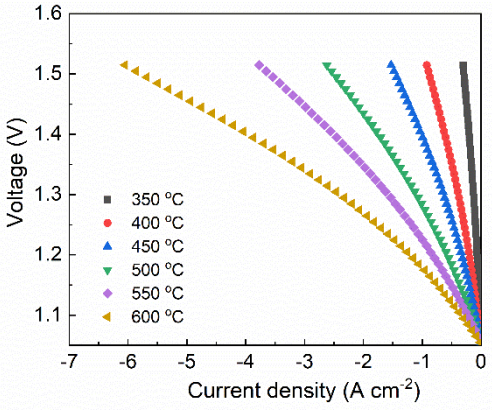
BZY demonstrates an expected FE behaviors with clear pathways:

- **GNG point achieved: at 1.0 A cm^{-2} and 600°C, the Faradaic efficiency is >90%.**
  - Fabricating high-performing p-SOEC is critical by 1) achieving high conductive electrolyte film; 2) using active electrode to reduce polarization.
- FE can be further increased by improving the cell performance (INL recent interface reconstruction demonstrated the highest performance of p-SOEC among those reported)
- 70% steam is practical in long-term testing for BZY material system



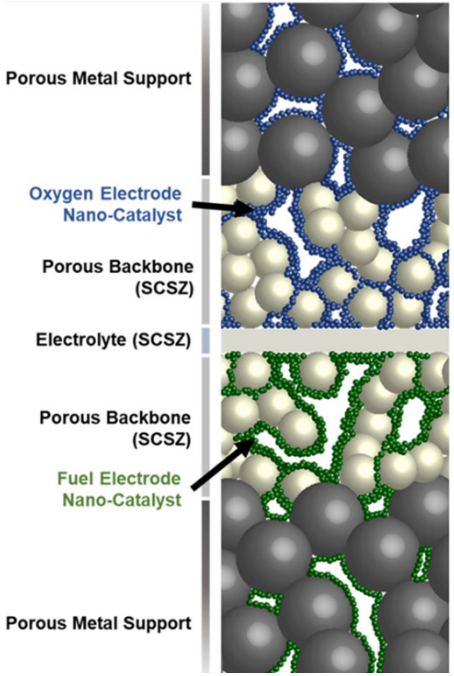


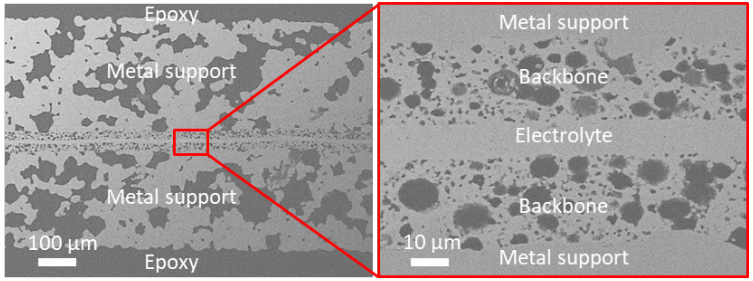
Tang W, Ding H, Ding D, et al. Nature Energy, under review

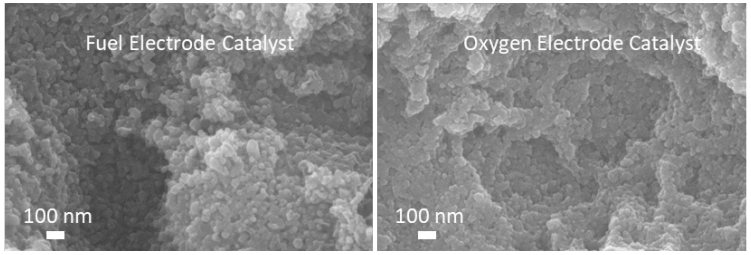


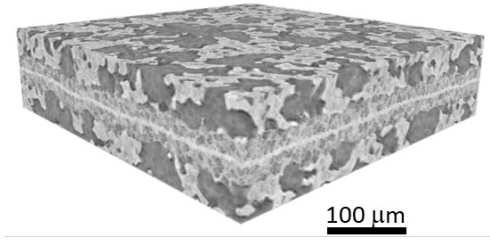
Bian W, Ding D, et al. Nature, accepted

LBNL Metal-Supported Solid Oxide Electrolysis Cell (MS-SOEC): Approach & Impact









➢ Cost

- Stainless steel is order of magnitude less expensive than ceramics
- Single high temperature sintering step

➢ Mechanical

- Rugged, strong cell
- Welded electrical connections
- Tolerates rapid temperature ramp
- Tolerates large temperature gradient

➢ Operational

- Temperature can vary quickly during operation
- Tolerates intermittent fuel/hydrogen
- Imbalanced pressure acceptable

Technical Accomplishment: HTE MS-SOEC

Q2FY22 (GNG) milestone

- Demonstrate improved performance and durability for Gen-2 MS-SOEC cell, with at least 0.8A/cm^2 at 1.4V and <10%/kh degradation at 700°C with at least 50% steam.

Performance

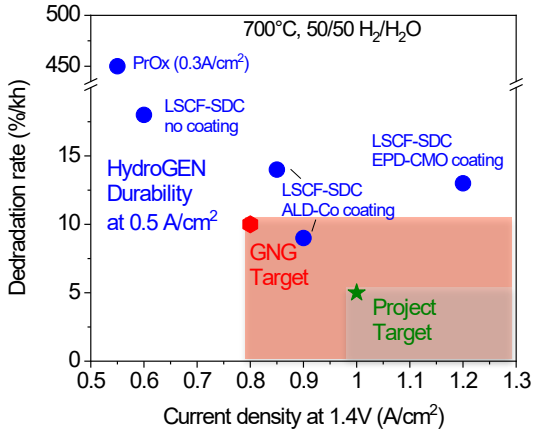
Target: > 0.8 A/cm^2 at 1.4V

Status: 1.2 A/cm^2 at 1.4V

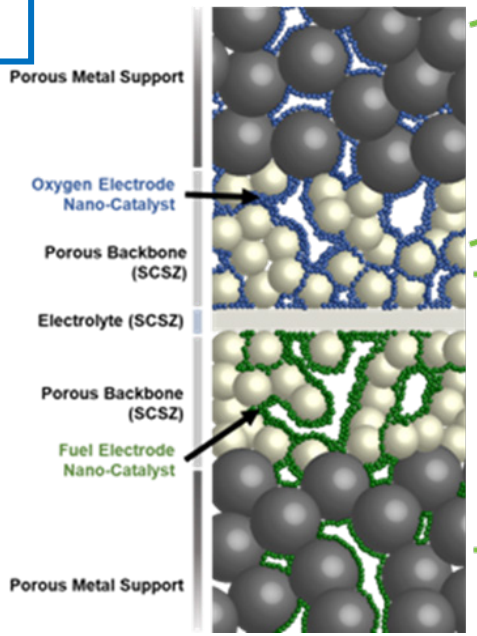
Durability

Target: <10%/kh

Status: 9%/kh at 0.5 A/cm^2



Approaches to improve performance and durability



Protective coatings to block Cr

Catalyst composition and processing

- LSCF-SDC firing temperature
- LSCF-SDC # infiltration cycles
- Alternative OER catalysts
- Ni:SDC ratio

Cell structure and processing

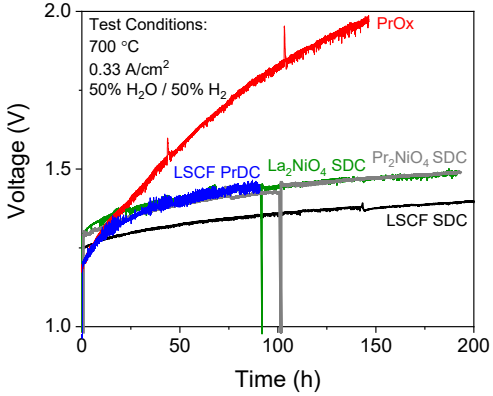
- Support thickness and porosity

Operating conditions

- Temperature
- H_2O:H_2 ratio (high H_2O utilization)
- Dry air

Technical Accomplishment: HTE MS-SOEC

Catalyst screening



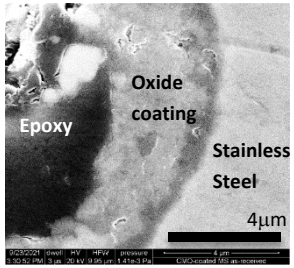
Selected:
LSCF/SDC 11x infiltrations

Future work:

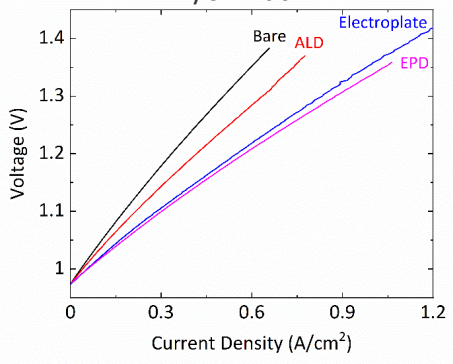
- Optimize coating thickness, composition, uniformity, firing protocol
- Continue catalyst screening
- Optimize catalyst firing
  - higher temp to stabilize SDC-Ni
  - lower temp to avoid Cr on LSCF
- Scale up to 40 cm² cell

Coatings

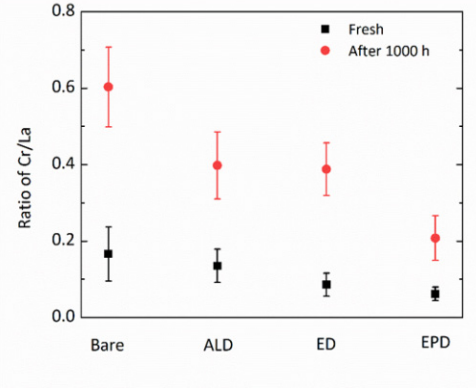
Coatings suppress Cr during cell fabrication (fresh) and after operation (1000h)



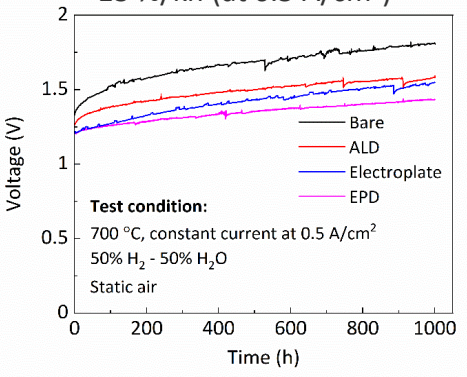
1.2 A/cm² at 1.4 V



Cr in LSCF electrode



13 %/kh (at 0.5 A/cm²)



Proposed Future Work for HTE

HydroGEN 1.0:

- Continue the collaboration between Lab Node and the seedling project

HydroGEN 2.0 p-SOEC:

- Validate the computational framework that can provide more powerful guidance in experiments for materials R&D and operation conditions
- Enhance the composition development and the condition optimization to further improve performance with higher FE;

HydroGEN 2.0 MS-SOEC:

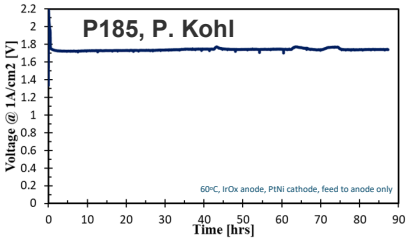
- Further improve the performance and durability of MS-SOEC by applying coatings, optimizing the composition and processing, as well as operation conditions.

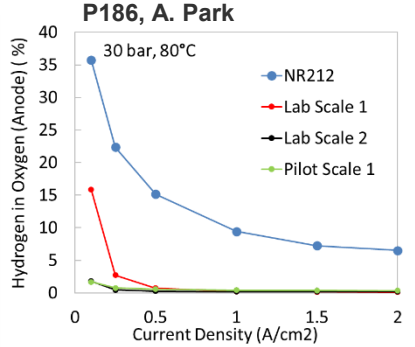
Any proposed future work is subject to change based on funding levels

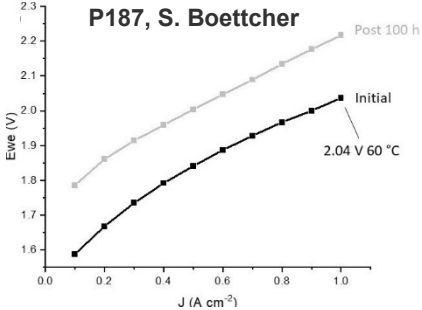
Low Temperature Electrolysis (LTE) Technical Accomplishments: Shaun Alia

LTE Technical Accomplishment Highlights

- **Significantly improved AEM membrane electrode assembly kinetics with water-only feeds. (LTE 2.0).** Compared to PEM baselines (1 A cm^{-2}), reduced kinetic overpotential from 500 to 200 mV, starting to bridge the gap between PEME and AEME performance.
- **Developed fundamental understanding of catalyst-ionomer interactions for AEME (LTE 2.0).** N+R groups can poison activity by blocking sites, degrading, or introduce competing reactions to OER. Found reasonable correlation between ab-initio simulations and cell performance/stability with different ionomer-catalyst combinations.
- **(P185) P. Kohl. Achieved Go/No-Go AEME performance and durability.** PGM-free performance <1.75 V at 500 mA/cm^2; PGM performance <1.75 V at 1A/cm^2; Durability <40 mV/1000 h extrapolated from >100 h tests (see backup slide).
- **(P186) A. Park. Demonstrated up to 50x reduction in effective H_2 crossover** using a 50 µm reinforced PEM membrane with GRC at 80°C, 30 bar p_{H2} (see backup slide).
- **(P187) S. Boettcher. Achieved Go/No-Go AEME durability milestone using impure water.** Tap water AEME operation with < 200 mV loss after 100 h (see backup slide).







OER = oxygen evolution reaction; PGM = Precious group metal; GRC = Gas Recombination Catalyst

LTE 2.0 Approach:

Enabling High Efficiency, Durable AEM Electrolysis Performance

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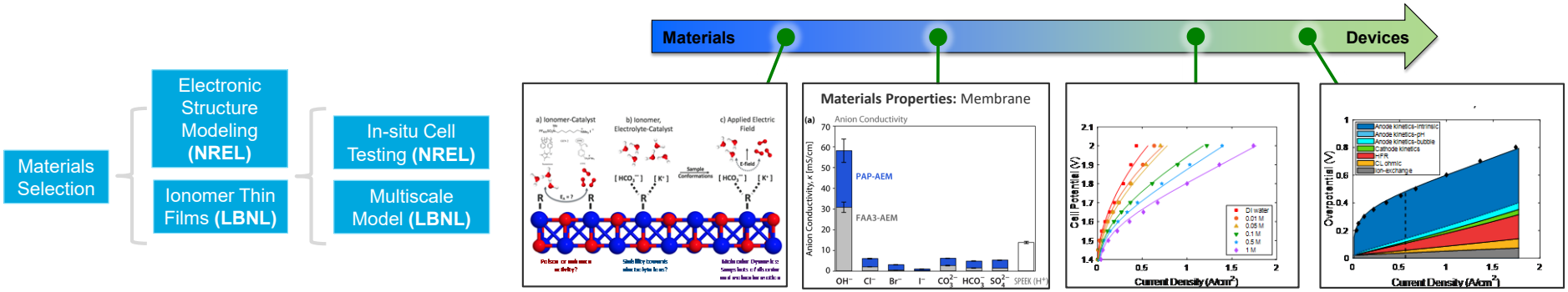
Goals: Determine the role of the supporting electrolyte and the limiting factors behind water operation in AEM electrolysis

- Evaluate AEM's ability to approach PEM performance/durability in water feeds
- Examine ionomer-electrolyte effects, how the ionomer's interaction with charged species enhances or diminishes catalytic activity
- Elucidate the role of conformational disorder and molecular motions to changes in catalyst activity via *ab initio* molecular dynamics simulations of the ionomer in contact with the catalyst surface



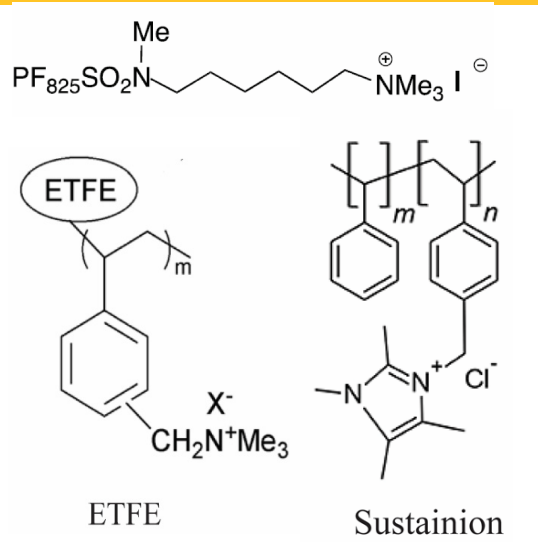
LTE 2.0 Accomplishments:

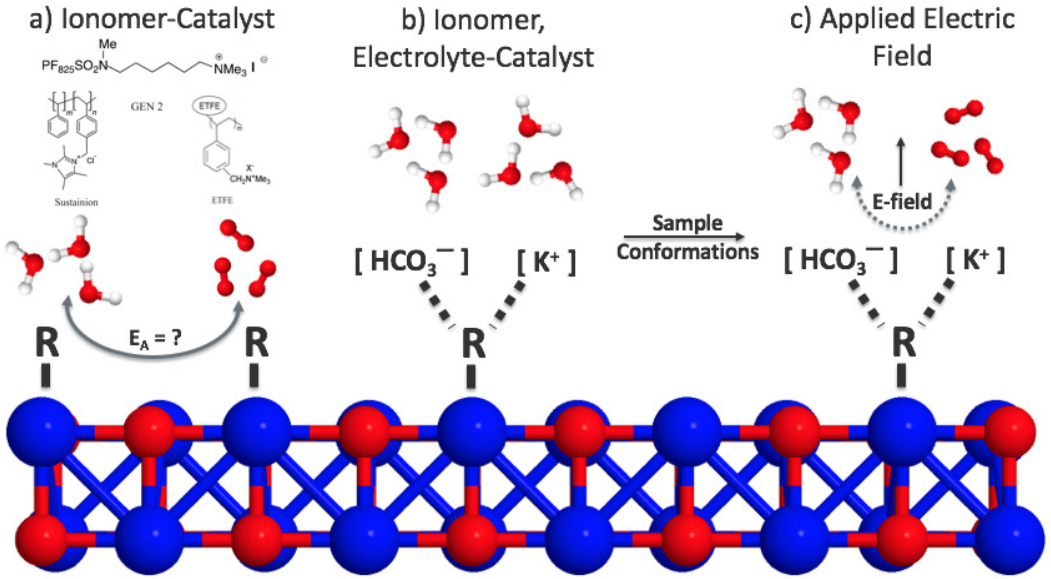
Atomistic Insight into Components and Their Effects on Electrocatalysis

NREL Transforming ENERGY | BERKELEY LAB | 4 Nodes

AEM ionomers: Theory provides atomic and electronic resolution of stability and reactivity

UNDERSTAND AEM catalyst *beyond the binding site* by considering the *in-situ* environment of the ionomer, electrolyte, and applied potential





Poison or enhance activity?

Stability towards electrolyte ions?

Molecular Dynamics: Snapshots of disorder and molecular motion

LTE 2.0 Accomplishments:

Understanding Ionomer-Electrolyte Effects on Oxygen Evolution

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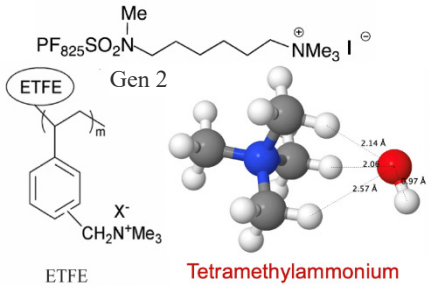
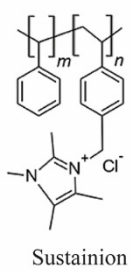
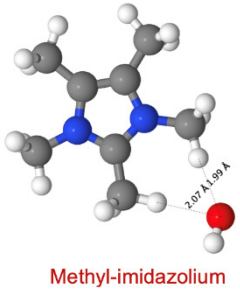
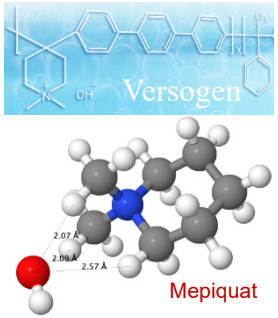
Approximate Ionomer with Smaller Organic Fragments: Theoretical calculations can give critical insights into ionomer-catalyst chemistry







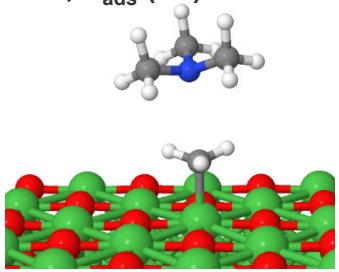


Ionomer/Catalyst Chemistry Can Impact Material Performance and Stability:

- Does the ionomer ***poison the catalyst*** by introducing competing reactions or covering up active sites?
- Does the ionomer ***remain stable or does it degrade*** into other species?
- Ideal: Ionomer stable, metal active sites available for OH adsorption

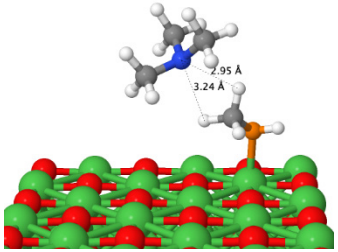
Ionomer-Catalyst Interactions: N^+R group can poison activity by blocking sites, degrade, or introduce competing reactions to OER

I, E_{ads} (eV) = -1.12



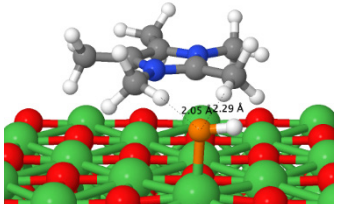
ETFE/Gen 2 → Ionomer Unstable, Degradation

I, E_{ads} (eV) = -1.01



ETFE/Gen 2 + OH → Alcohol Formation Instead of OER

I, E_{ads} (eV) = -0.49



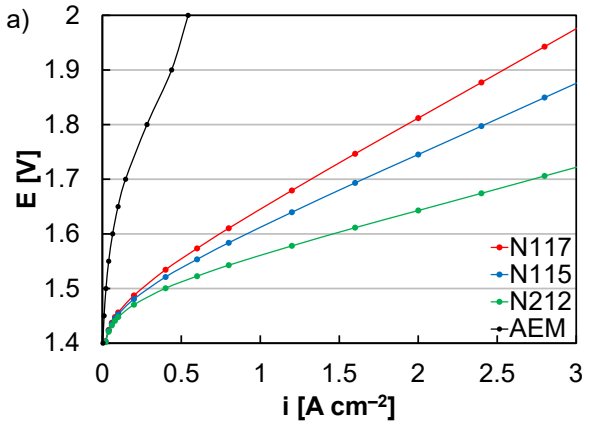
Sustainion → Stable, Active Sites Available for OH

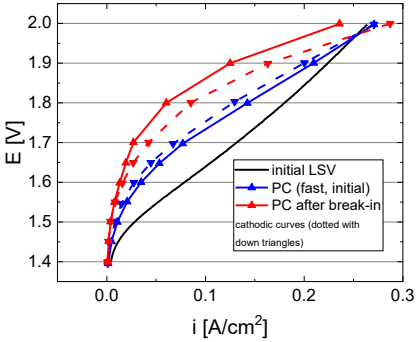
- ***Sustainion Ionomer*** is *stable* and *does not block or poison* Ni sites
- ***ETFE, GEN 2 Ionomers*** are *unstable* and *poison active sites*:
  - Degradation via de-methylation
  - Alcohol formation competes with OER

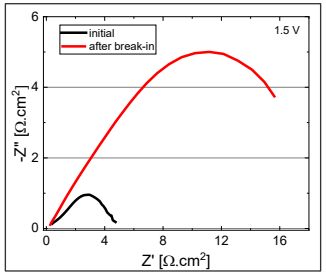
Theory can identify *key limitations or advantages* to specific ionomers, **critical to our understanding of the ionomer/catalyst interface**

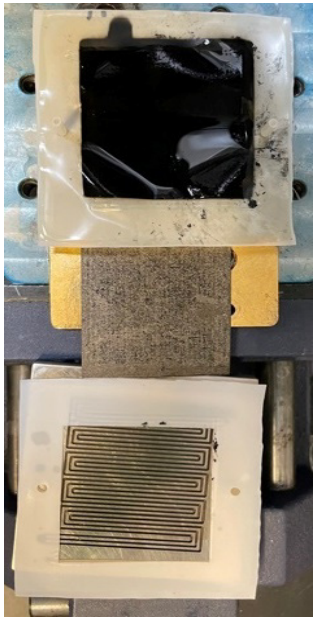
LTE 2.0 Accomplishments:

Decal Transfer, Thermal Restructuring of the Catalyst Layer







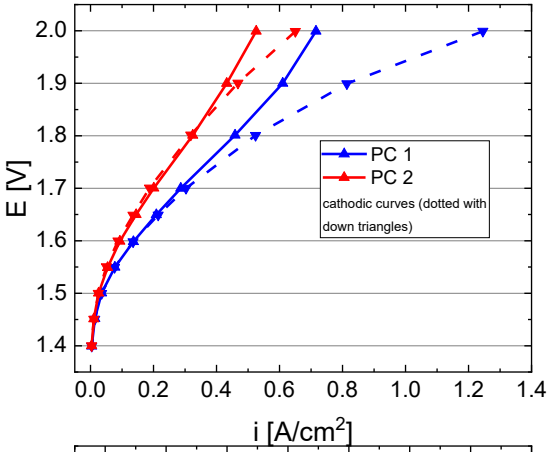


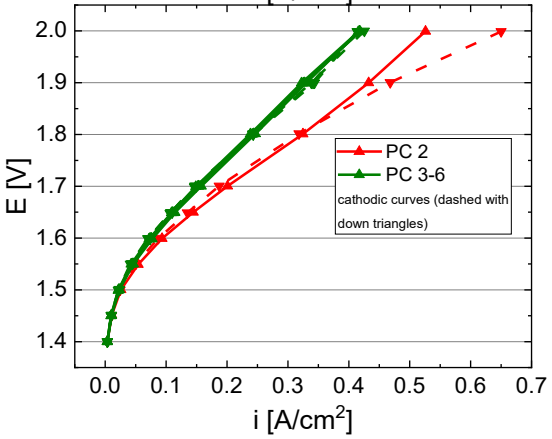
- Catalyst layer delaminates during cell conditioning in water-only operation
- Decal transfer restructures catalyst layer, modifies ionomer integration, slows delamination
- Allows for short-term operation, does not prevent catalyst layer loss
- Integration can partially address limitations

LSV: linear sweep voltammogram; PC: polarization curve

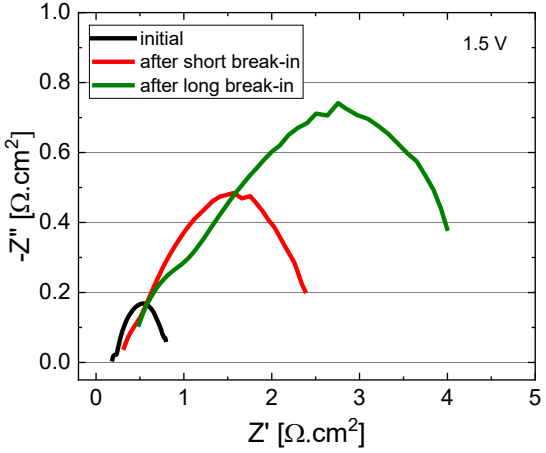
LTE 2.0 Accomplishments:

Porous Transport Layers to Limit Catalyst Layer Loss





- Transport layer with lower porosity, pore diameter at the PTL/catalyst layer interface
- Limits migration/loss of catalyst layer and allows for longer conditioning, consistent performance
- Does not prevent delamination, catalyst transfer to PTL surface



PC: polarization curve

LTE 2.0 Accomplishments:

Go-No Go (GNG) Milestone for Water-Only Feeds

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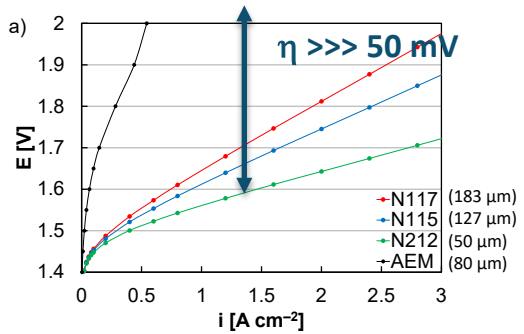
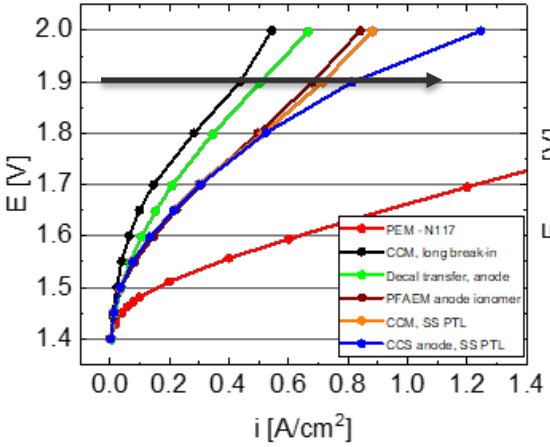


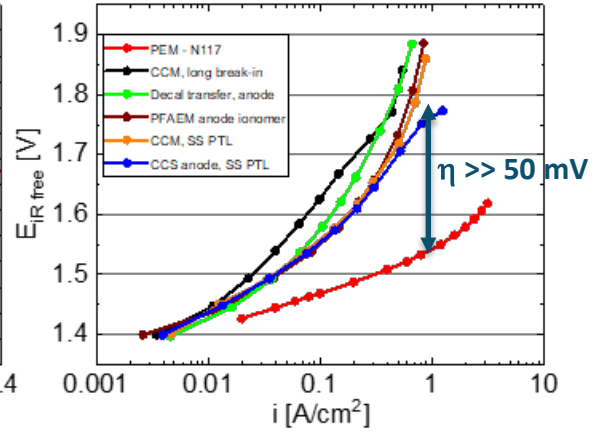
Name/ Description	Incorporating experiment and modeling, down-select AEM operating conditions (e.g., flow configurations, back pressure, temperature) that show water-fed (i.e., no supporting electrolyte) AEMWE is a viable pathway, by demonstrating cell overvoltage within 50 mV (IR-free) of a 50 µm commercial Nafion at 1 A/cm^2. (NREL & LBNL).
Criteria	Demonstrate water-fed AEMWE (i.e., no supporting electrolyte) with a cell overvoltage within 50 mV (IR free) of commercial 50 µm Nafion at 1 A/cm^2.

- GNG related to performance, comparison to Nafion/PEM different thicknesses
- Improvement in kinetics (overpotential from 500 mV to 200 mV at 1 A cm^{-2}), does not reach GNG target
- Pathways forward for supporting electrolyte:
  - Study of materials, integration approaches, and catalyst layer properties to address performance and stability limitations, resolving the role and need of the ionomer in a supporting electrolyte
  - Study of supporting electrolytes to delineate the impact of electrolyte conductivity and alkalinity on AEM electrolysis performance and durability.

Improved AEM performance



Improvements primarily kinetic



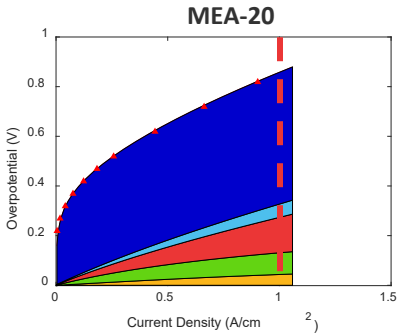
CCM: catalyst coated membrane; CCS: Catalyst coated substrate; SS: stainless steel; PTL: porous transfer layer; PF: perfluorinated

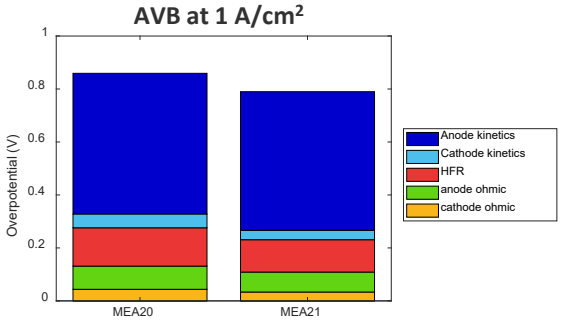
LTE 2.0 Accomplishments:

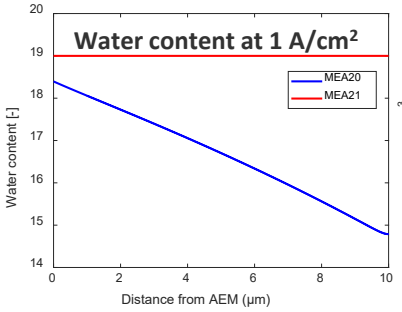
Wet/Dry Cathode Operation

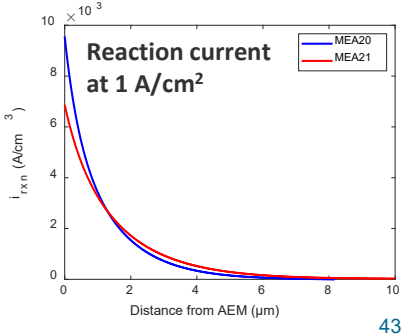
	Feed	T (°C)	AEM (μm)	Ionomer content	Catalyst	PTL
MEA 20	**DI water to Anode (30%RH on cathode)**	80	40	Anode: 10 wt% + 2 wt% overlayer Cathode:30wt% + 2wt% overlayer	Anode: IrO_2 (Pajarito) Cathode: Pt/HSC	Anode: SS PTL (25 AL3)
MEA 21	**DI water to Both electrodes**	80	40	Anode: 10wt% + 2wt% overlayer Cathode:30wt% + 2wt% overlayer	Anode: IrO_2(Pajarito) Cathode: Pt/HSC	Anode: SS PTL (25 AL3)

- Dry cathode operation, cathode relies on water transport through membrane
- Applied voltage breakdown comparison. MEA-20:
  - Higher HFR and cathode ohmic loss due to lower water content, ionic conductivity
  - Higher cathode kinetics loss due to lower water activity
- Low cathode RH results in lower water content and activity in cathode, which leads to lower ionic conductivity and makes reaction current more concentrated next to the AEM
- Quantifies performance loss associated with dry operation









Proposed Future Work for LTE

- LTE 2.0 and a shift in scope to supporting electrolytes
  - Study of materials, integration approaches, and catalyst layer properties to address performance and stability limitations, resolving the role and need of the ionomer in a supporting electrolyte
  - Study of supporting electrolytes to delineate the impact of electrolyte conductivity and alkalinity on AEM electrolysis performance and durability.

- Leverage HydroGEN nodes to enable successful completion and continuation of the seedling projects, depending on which budget period they are in

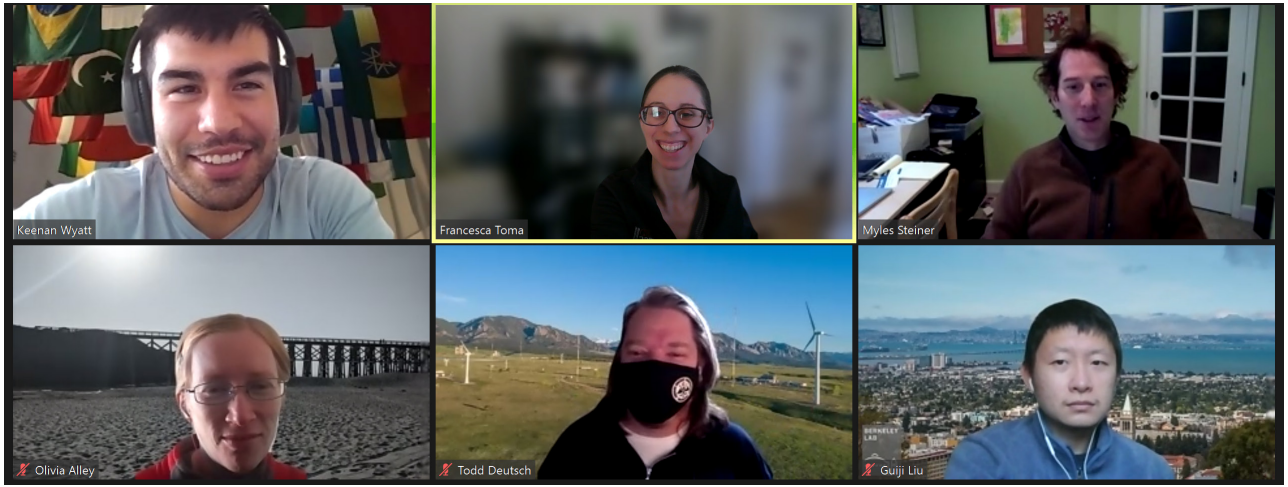
Any proposed future work is subject to change based on funding levels

Collaboration, Effectiveness

- 4th Annual Advanced Water Splitting Technology Pathways Benchmarking & Protocols workshop, May 3-4, 2022, ASU, AZ
- Interfacing between HydroGEN and IEA Annex 30 in benchmarking
- Contributions to the Meta Data development for the HydroGEN Data Center

Seedling Leads	Seedling Teams	LTE 2.0 Teams
• Shannon Boettcher • Paul Kohl • Andrew Park	Chemours, Pajarito Powder (Fuel Cell Catalysts), Advent, nel, Argonne National Laboratory, Oak Ridge National Laboratory, Los Alamos National Laboratory (Est. 1943), University of Oregon, UC Irvine, Georgia Institute of Technology, Rensselaer, Northeastern University, University of South Carolina, University at Buffalo (The State University of New York), University of Delaware	NREL (Transforming Energy) Shaun Alia Huyen Dinh Mai-Anh Ha Saad Intikhab Ross Larsen Bryan Pivovar Berkeley Lab (Bringing Science Solutions to the World) Nemanja Danilovic Julie Fornaciari Ahmet Kusoglu Jessica Luo Adam Weber Guosong Zeng Jeremy Zhou

Photoelectrochemical Water Splitting (PEC): Francesca Toma

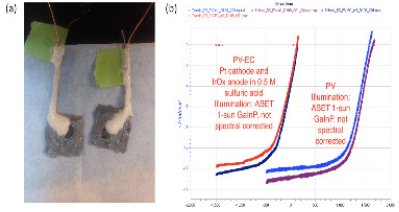


Not in the picture:
James Young (NREL)
Alex King (LBNL)
Adam Weber (LBNL)
Ethan Crumlin (LBNL)
Rebecca Hamlyn (LBNL)
Anh Pham (LLNL)
Tadashi Ogitsu (LLNL)

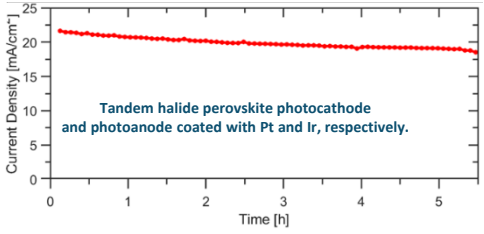
Alumni:
Guosong Zeng – runner up Postdoc Award [currently Assistant Professor at Sustech (China)]
David Larson (now at Twelve)

PEC Technical Accomplishment Highlights

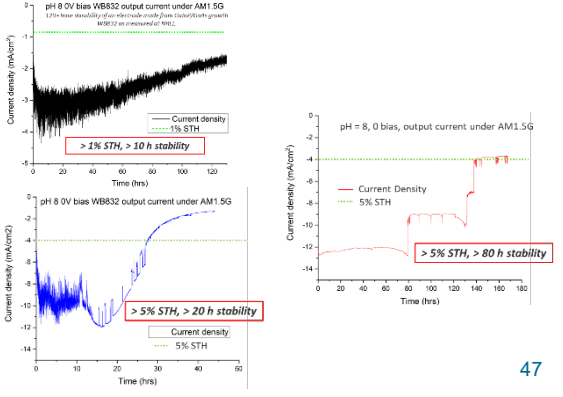
- **NREL,** in collaboration with the **University of Toledo** (Prof. Yanfa Yan), worked on the characterization of halide perovskite/ halide perovskite tandem photoelectrodes for unassisted water splitting demonstrating up to 13.1% in a two-electrode configuration under one sun illumination without an external bias with 50 h stability. *(see backup slides for more detail)*



- **LBNL and NREL** worked with **Rice University** (Prof. Aditya Mohite) to characterize halide perovskite photoelectrodes coated with catalysts and a hydrophobic graphene-based barrier which ensures optimal charge transfer at the light absorber/catalyst interface. We achieved >100 hours stability with peak efficiency exceeding 20% STH. A paper which report this results is under revision. *(see backup slides for more detail)*

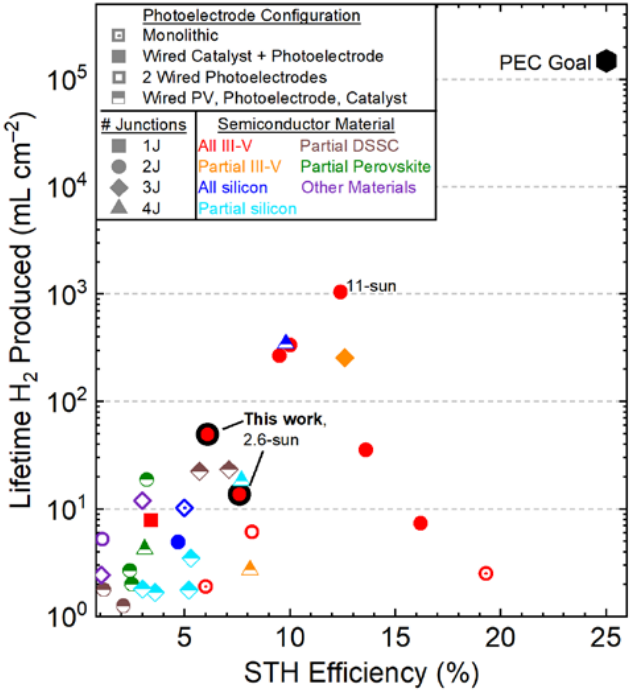


- **LBNL and NREL** worked together to standardize measurements at both Labs at near neutral pH by testing similar samples and comparing results. We designed a novel testing cell and met our Year 1 and Year 2 milestones as well as our Go/No-Go. We actively exchanged and tested several samples across the two Labs. A paper reporting the standardization of our methods has been recently accepted. We also actively participate in the Benchmarking workshop.



HydroGEN 2.0 PEC Approach

- Prioritize durability stressors and establish PEC device durability protocol
- Use density functional theory (DFT) and microkinetic modeling to describe the local environment at the electrode/electrolyte interface under operation
- Provide mechanistic understanding of PEC device degradation guided by theory and in operando characterization

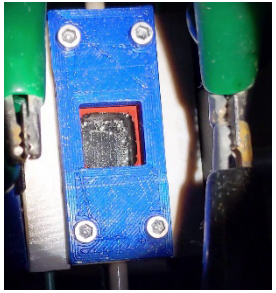


Comparison of the solar to hydrogen efficiency (STH) and lifetime H2 produced for unassisted water splitting devices. The "PEC Goal" point in the upper right was calculated assuming a 20% capacity factor over a 10 year lifetime (15) .

HydroGEN 2.0 PEC Workflow & Lab Collaboration

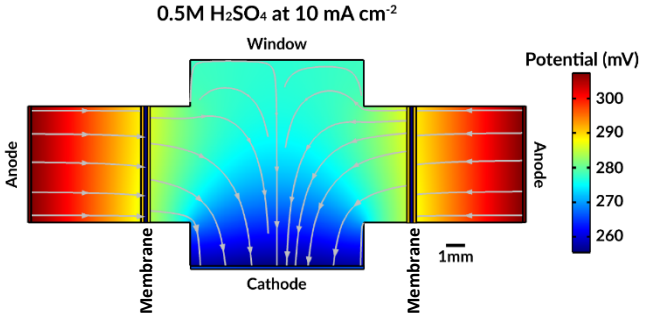
Workflow

Synthesis of III-V structures (NREL) and catalyst deposition



Testing and characterization at NREL and LBNL

Modeling at LBNL and LLNL

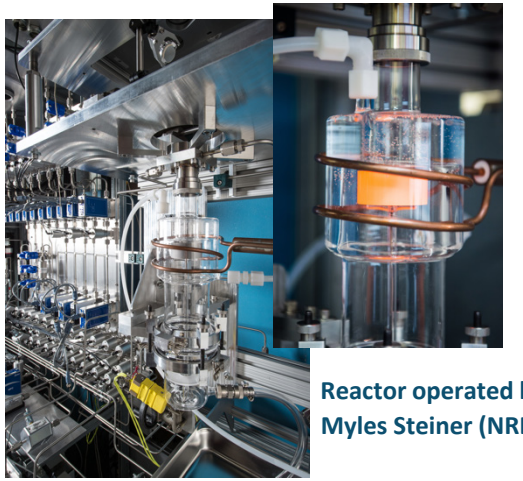


Cell design and shipping of samples

HydroGEN 2.0 PEC Workflow Approach & Accomplishment

***First step**: i) fabrication of high quality III-V semiconductors to obtain the needed photovoltage and photocurrent output for autonomous water splitting with targeted efficiencies*

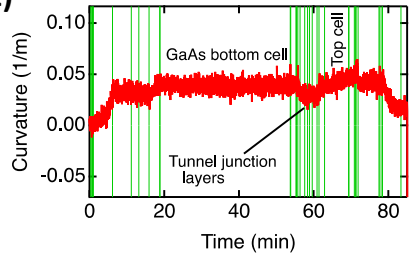
Growth by atmospheric pressure metal organic vapor phase epitaxy (MOVPE)



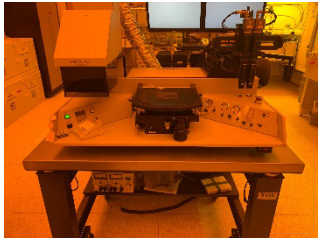
Reactor operated by Myles Steiner (NREL)

MOVPE at 620℃ growth temperature

Characterization and processing

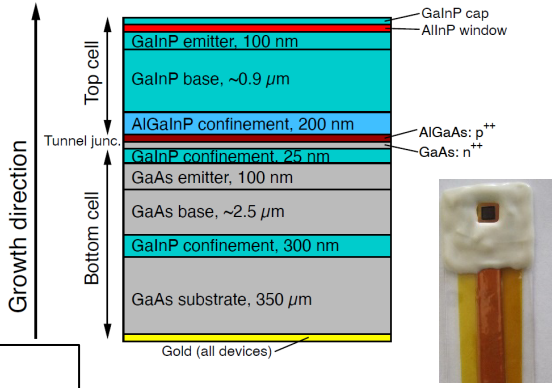


In-situ characterization of stress



Cleanroom processing into devices

Photoelectrode fabrication



Schematic of the III-V (left), high efficiency photoelectrode (right)

Challenges:

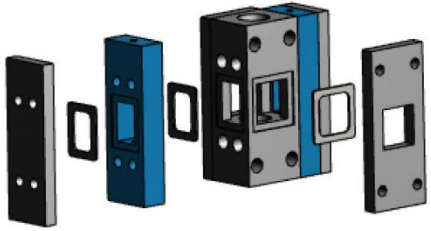
- Exact calibration of compositions, dopants, carriers
- Drift of reactor temperature and pressure
- Thickness uniformity across a 2” wafer
- Processing repeatability and uniformity

ii) hydrogen evolution catalyst deposition is carried out by electrodeposition (Pt) and sputtering (PtRu)

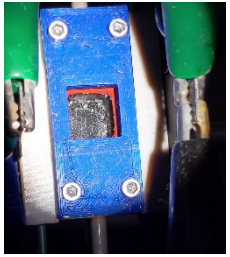
HydroGEN 2.0 PEC Workflow Approach & Accomplishment

***Second step**: Exchange samples and test their photoelectrochemical behavior at NREL and LBNL – to enable this step we designed a novel testing cell*

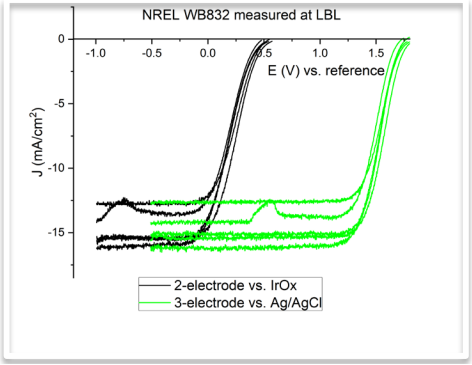
New modular testing cell with two anode compartments

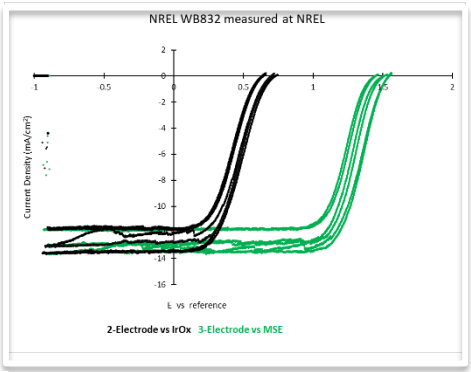


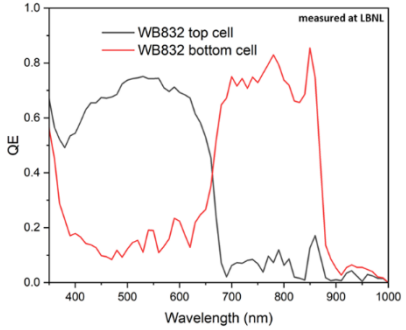
Due to its high modularity and ease of fabrication, we are benchmarking this cell to make it available to the community

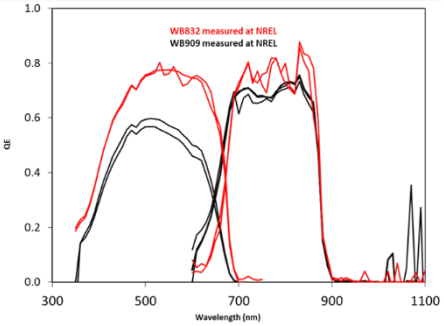


@ LBNL **Characterization** **@ NREL**









Reproducible results are obtained at both Labs

***J-V curves** were measured at LBNL and NREL in a **2- and 3-electrode** configuration.*
All measurements took place in 0.5 M omni trace sulfuric acid with 1 mM Triton X-100 surfactant.

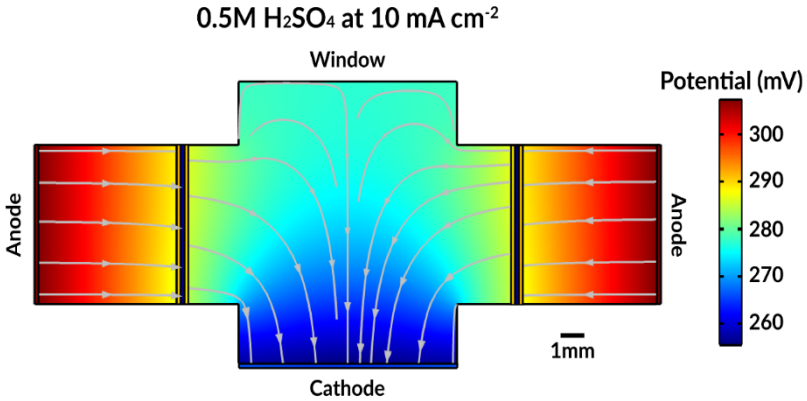
*example of **IPCE measurement** taken on the bottom cell (red) and top cell (black) of an electrode from a portion of sample WB832*

HydroGEN 2.0 PEC Workflow Approach & Accomplishment

***Third step**: Provide theoretical understanding through theory and modeling*

Modeling predicts the kinetics, thermodynamic, and ohmic losses of the cells and inform its design

Nearly constant current and potential near window compartment

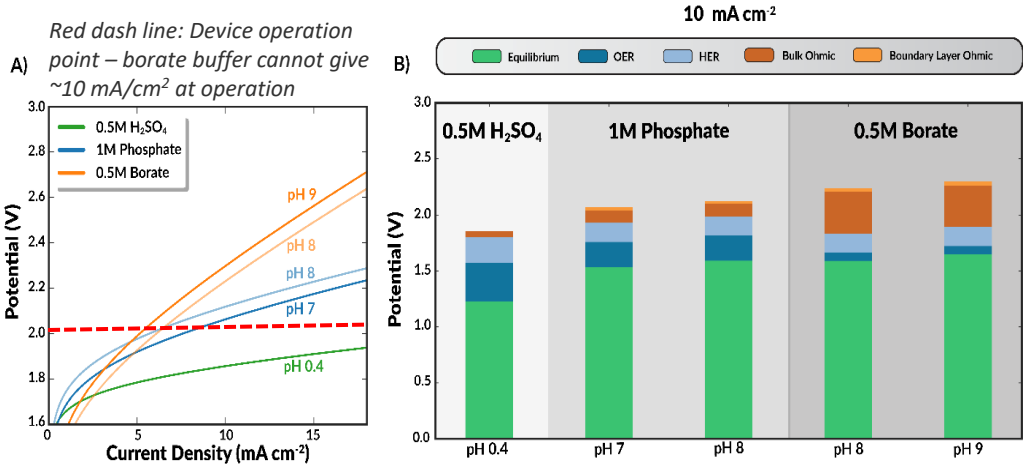


Electrolyte potential surface plot throughout the model and current density streamlines at 10 mA cm^{-2} and 10 mL min^{-1} in 0.5 M H_2SO_4 electrolyte.

Longer ionic path length near "top" of anodes causes distributions across cathode

Only H_2SO_4 can reach 10 mA cm^{-2} with 2 V of photovoltage

Red dash line: Device operation point – borate buffer cannot give ~10 mA/cm^2 at operation

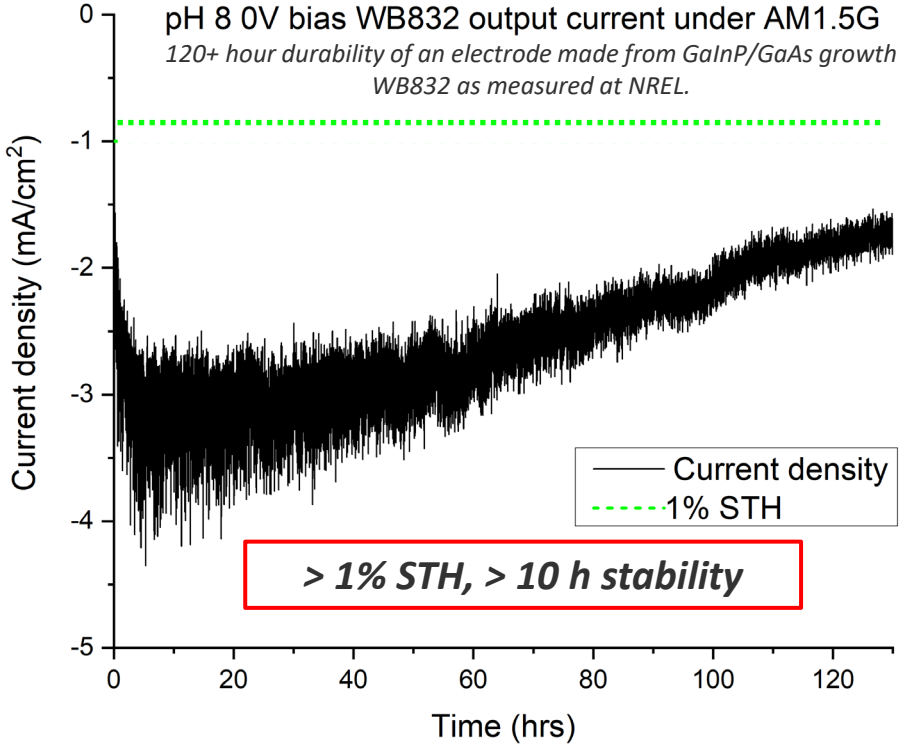


A) Polarization curves of the PEC cell with different electrolytes and pHs.
B) Breakdown of contributions to voltage.

Borate electrolyte offers lower kinetic potential losses but greater ohmic and thermodynamic losses

HydroGEN 2.0 PEC Year 1 Milestone Accomplishment

FY21Q4 Milestone: Demonstrate Type III device with near 1% or better solar-to-hydrogen (STH) efficiency and near 10 h or better stability tested at LBNL and/or NREL with the same absorber architecture and device configuration having near-neutral electrolyte and a membrane but different cells under ambient conditions and the AM 1.5G reference spectrum.

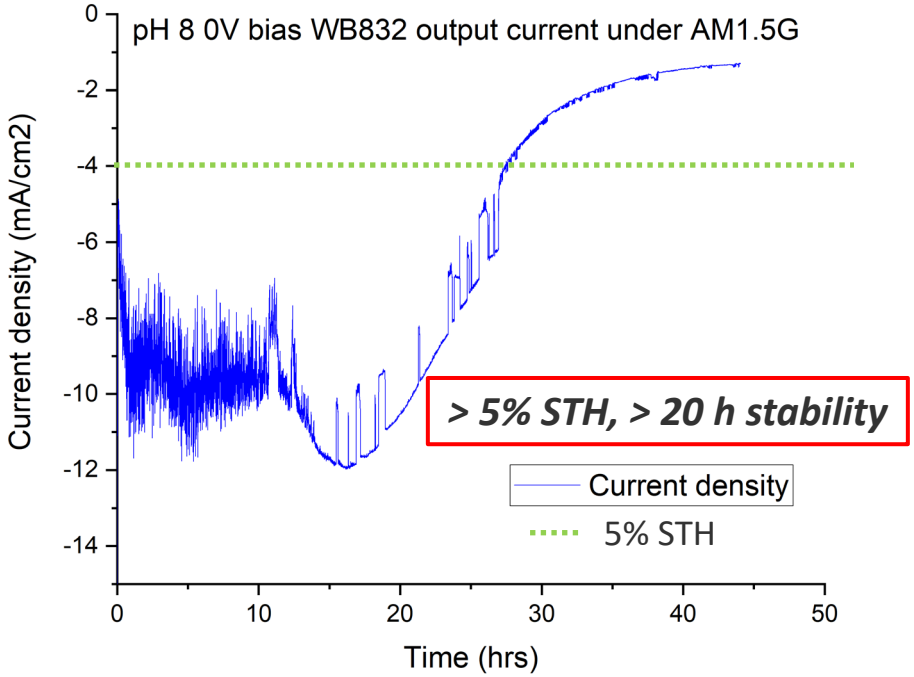


Chronoamperometry meets Year 1 milestone

Potassium phosphate buffer with no additives, a Nafion 117 PEM separating the counter electrode (CE) compartment from the photocathode compartment, and an IrOx CE. The green line represents 1% STH efficiency.

HydroGEN 2.0 PEC Year 2 Milestone Accomplishment

FY22Q1 Progress Milestone – PEC Durability: Demonstrate PEC device that maintains an unbiased STH efficiency greater than 5% over 20 h in near-neutral pH conditions. (LBNL & NREL)

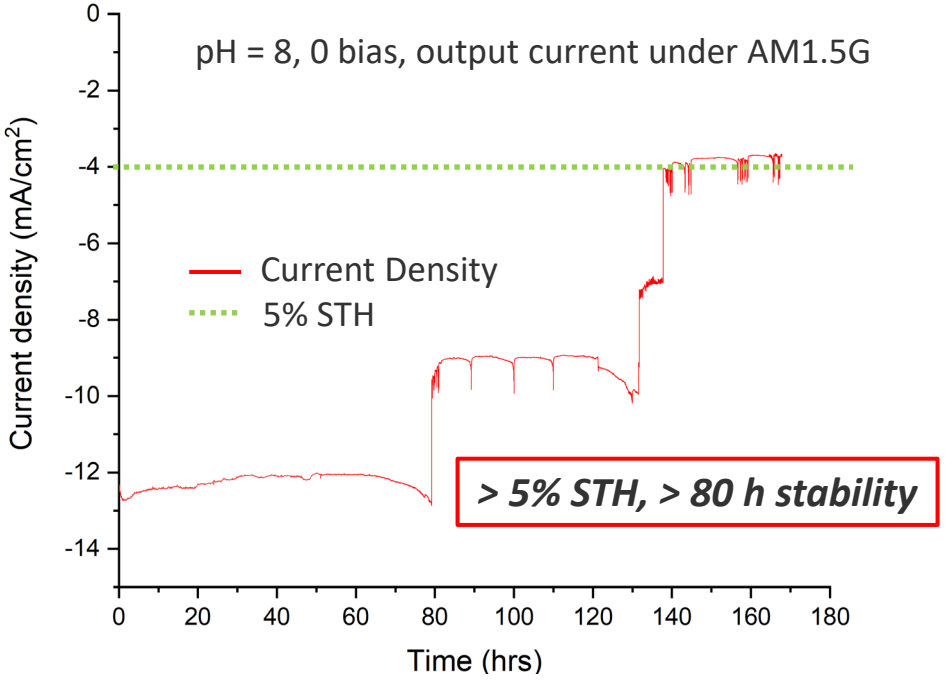


Chronoamperometry meets Year 2 milestone

Potassium phosphate buffer with no additives, Potassium phosphate buffer with no additives, no separator, and a RuO_2 CE.

HydroGEN 2.0 PEC Year 2 Go/No Go Accomplishment

Go/No-Go (FY22Q2): Demonstrate 5% STH or better efficiency with 50 h or better durability (within 20% of initial STH) with down-selected device components and architecture with a membrane or separator, in near neutral pH conditions with development of a protocol with standardized testing conditions between NREL and LBNL. (LBNL & NREL).



Chronoamperometry meets Go/No Go

pH 7 potassium phosphate buffer made in a 0.05 M sodium sulfate solution. IrOx was the CE and was isolated from the photocathode via a Ti mesh separator

Currently around 10 samples reproducibly met >80 h; we consistently observed a decline after that time

Summary of Accomplishments: PEC 2.0

- The team has met Y1, Y2 milestones and the Go/No Go (5% STH or better efficiency with 50 h or better durability).
- The team has finalized a protocol that was accepted in a special issue of *Frontiers in Energy Research - section Hydrogen Storage and Production, on Advanced Water Splitting Technologies Development: Best Practices and Protocols*.
- As a result of this protocol, we are also finalizing a manuscript on the benchmarking of the cell utilized in HydroGEN 2.0 to provide to the community as a reliable cell for PEC testing. In addition to experimental results, we included modeling.
- The team was also an active participant in the 2022 Water Splitting Technologies Benchmarking and Protocols Workshop (May 2022), through presentations and breakout sessions.
- The biggest achievement was for us our collaboration under the hardship of the pandemic and the several issues related to limited access over these two years. We were able to send and test samples through two different labs and kept everybody engaged across labs through frequent meetings and updates.

Proposed Future Work for PEC

Now working to meet the “end of the project milestone”:

Demonstrate a complete device assembly and neutral pH conditions that achieve >220 h durability at short circuit and > 5% STH efficiency. Scale-up to a hand-held > 4 cm^2 active area device that will split water under direct sunlight illumination with appreciable bubble generation. (LBNL, NREL).

- Understand origin of degradation after 80 h
- Deposit different protective layers to enhance durability
- Look into new cell geometries to improve efficiency and durability
- Designing strategies for scale up for a hand-held device

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

Cross-Cutting Modeling Accomplishments: Tadashi Ogitsu

Cross-Cutting Modeling

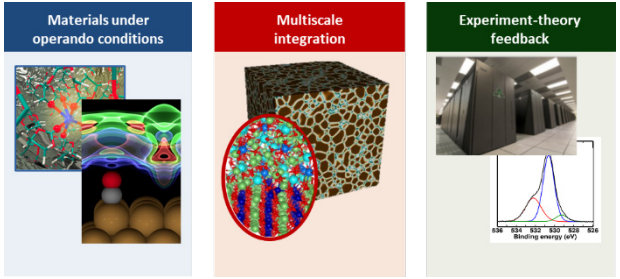
Goal: Exploit similarities in technical challenges across AWS technologies and required simulation capabilities to advance material performance and durability.

Multiscale performance and durability modeling across the technology areas

	Limiting factor	PEC	LTE	HTE	STCH
Performance	Materials thermodynamics			X	X
	Carrier generation and mobility	X	X	X	
	Interfacial (electro)chemical kinetics (solid-solid = S, solid-liquid = L)	L	L	S	
	Mass transport kinetics (solid = S, liquid = L)	L	L/S	S	S
	Phase transformation kinetics			X	X
	Multiphysics device operation	X	X	X	X
Durability	Thermomechanical			X	X
	Electrochemical	X	X	X	
	Photochemical	X			

The modeling and simulation strategy will focus on key factors limiting performance and durability across the four advanced water splitting technology areas.

Priorities for Cross-Cut Modeling Activities



Theory-guided design: models and experimental testing will be combined to analyze performance and durability of the materials under simulated operating conditions. Sensitivity analysis will be utilized to connect the material properties (composition, microstructure, etc.) and operating environment (temperature, pressure, solution composition, current density) to device behavior.

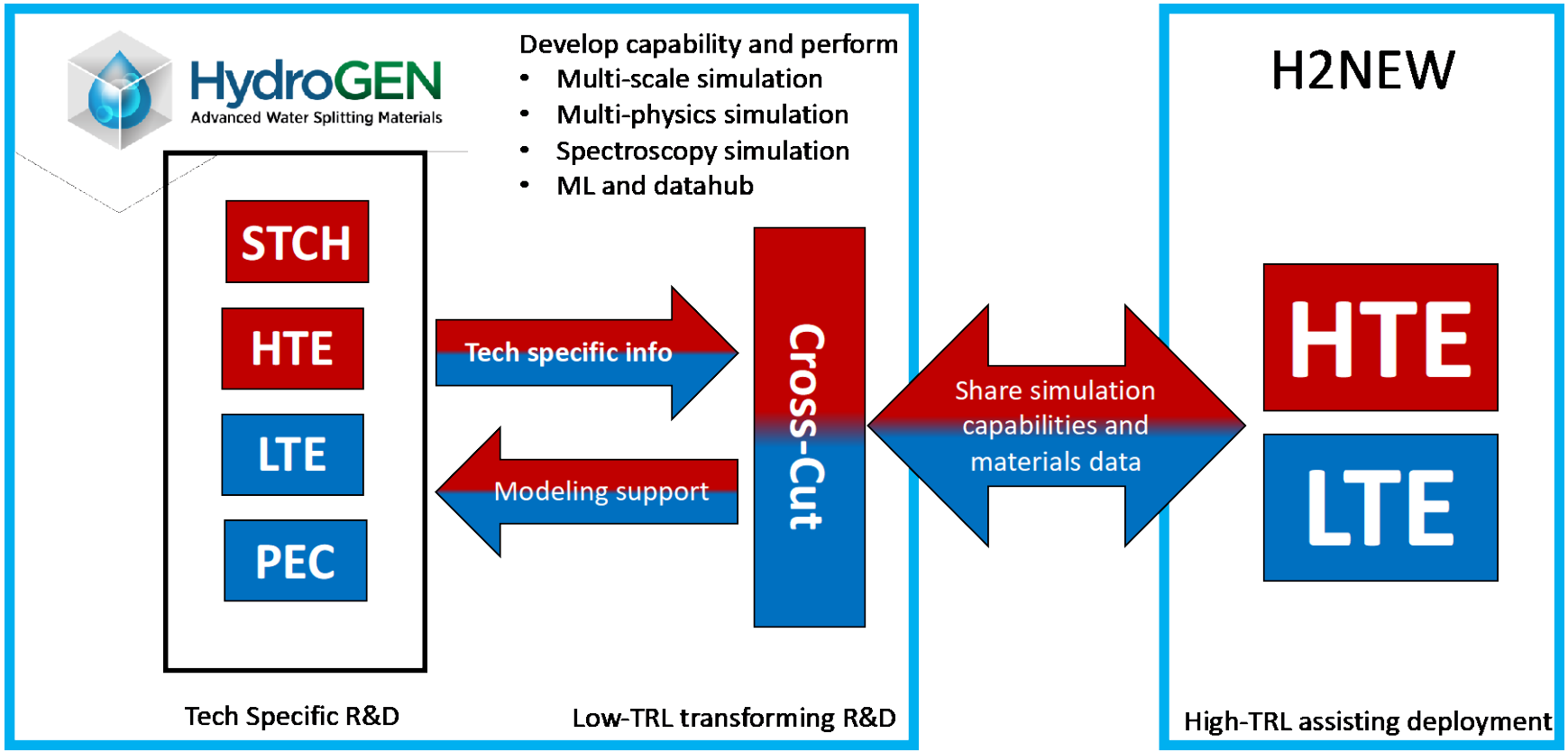
Pushing low-TRL technologies to higher TRL by:

1. **Predictive simulations:** develop and apply multi-scale modeling capability to simulate performance and durability

   Provide clear link between material property and device performance/durability: crucial information for improving the component materials and their assembly
2. **Validation:** theory-experiment integrated approach to ensure reliability of simulation results and interpretation of experiments
3. **Sharing:** help develop accessible energy materials database for scientific community and industry
4. **Collaboration with H2NEW:** assist further advancement of high-TRL hydrogen production technologies (PEME, o-SOEC)

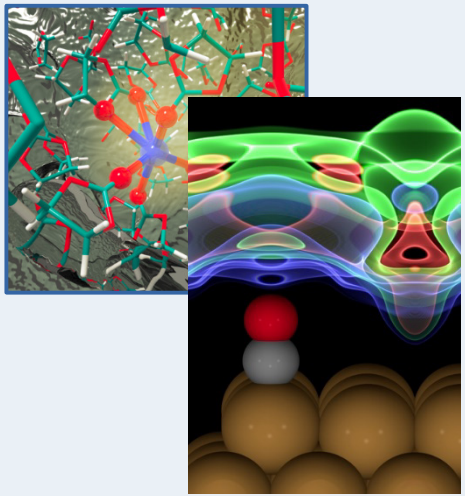
The funding allocated to achieve this task is provided under other technology specific tasks.

Cross-cutting in HydroGEN2.0 and H2NEW

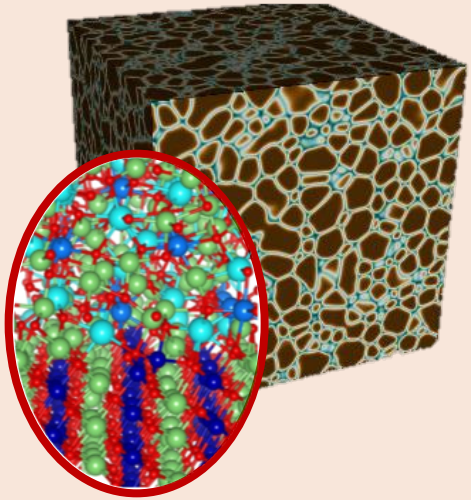


Priorities for cross-cutting modeling activities

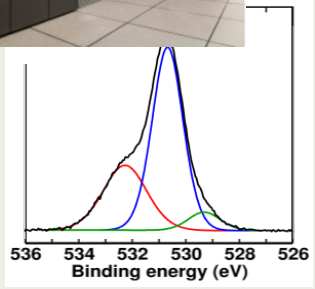
Materials under operando conditions



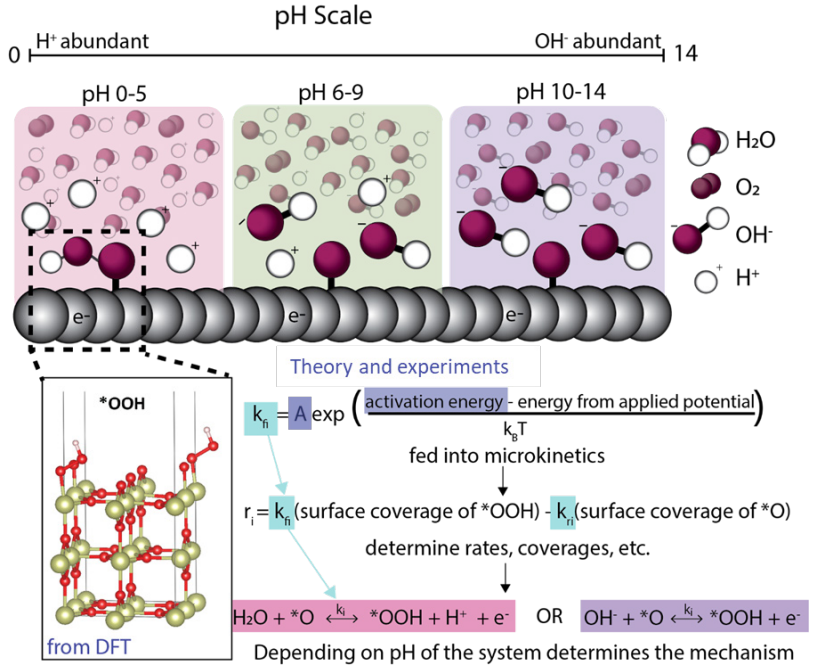
Multiscale integration



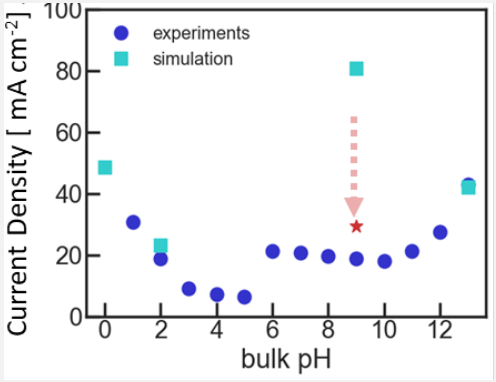
Experiment-Theory-Data integration

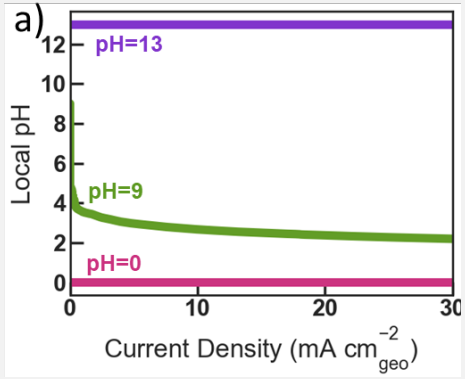


OER Supernode: Multiscale modeling and experimental validation for IrO_2



Fornaciari, Weng, Alia, Zhan, Pham, Bell, Ogitsu, Danilovic, Weber, *Electrochim. Acta* 405, 139810 (2022)

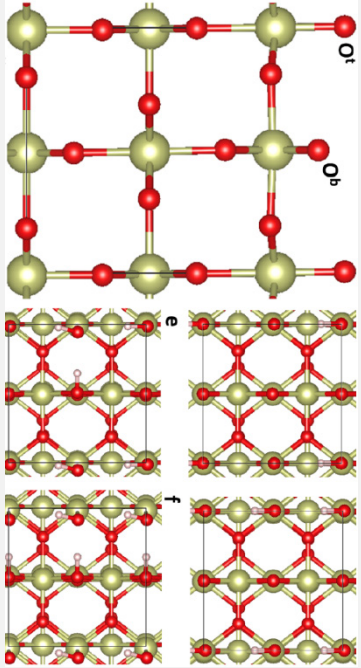




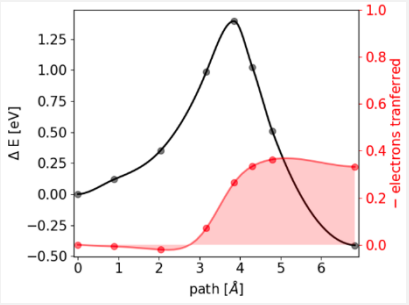
- We established a multi-scale modeling approach to calculate the Tafel plot from ab-initio simulations
- We provided critical insights into the effects of pH and transport on OER kinetics

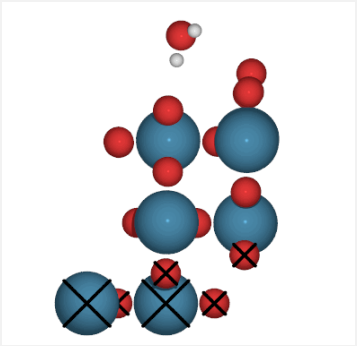
OER Supernode: Realistic simulations of interfacial electrochemical kinetics

Surface thermodynamics
First-principles simulations

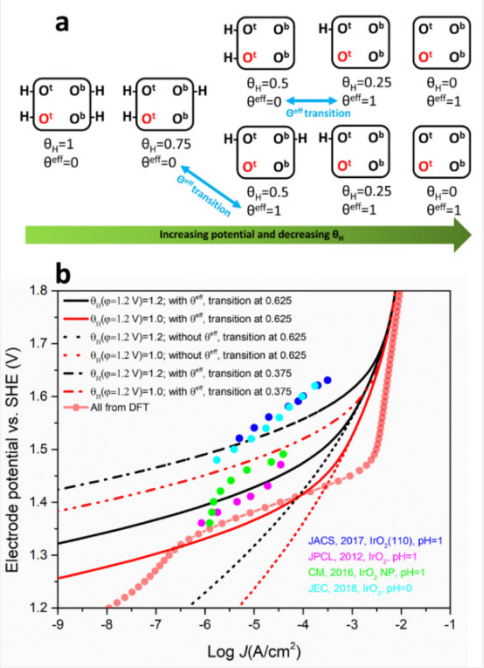


OER kinetics
First-principles simulations





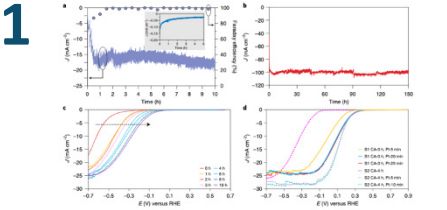
Microkinetic modeling



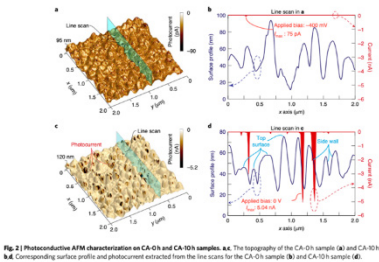
Environmental effects on OER kinetics

PEC: Theory-experiment integrated approach elucidated the origin of GaN performance

FY21

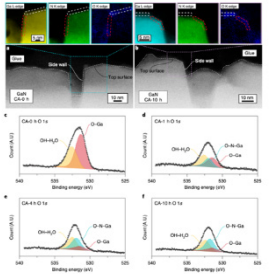


Improve hydrogen evolution reaction (HER) efficiency observed at the initial stage of experiment

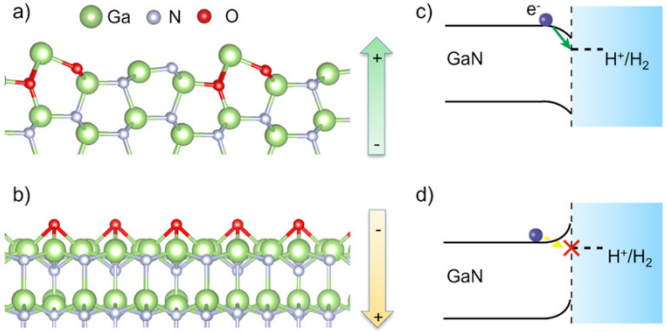


The improvement of photocurrent is localized at the sidewalls, likely (1010) surface

Nature Materials **20**, 1130 (2021)



The sidewall surface develops a composition similar to gallium oxynitride



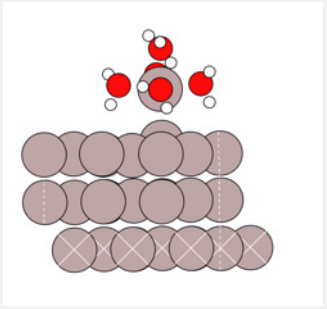
DFT simulations show that alternating substitution of N with O at the first two layers of GaN(1010) surface provides the band-bending that facilitate charge transfer at the interface, which is consistent with improved HER activity of GaN(1010)

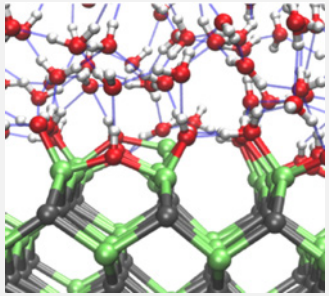
U Michigan seedling project on GaN supported by PEC capability nodes at LLNL, Nato Lett. **22**, 2236 (2022)

Proposed future work: LTE

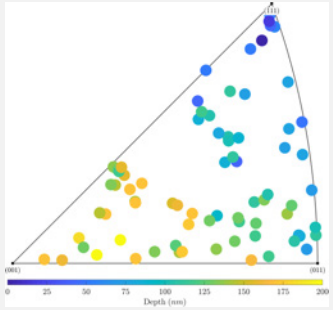
Microstructure-aware interfacial reaction kinetics

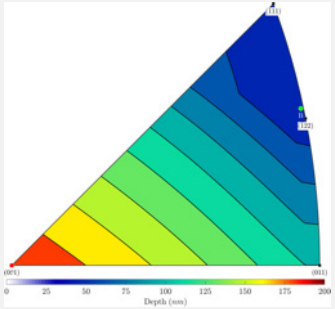
Thermodynamics and Kinetics
First-principles simulations



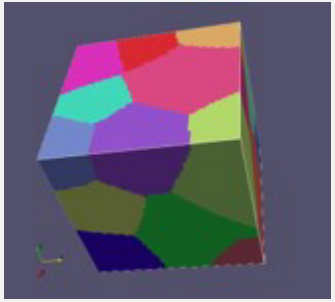


Data science & ML for interpolation





Effects of Microstructure
Phase-field modeling



FY22

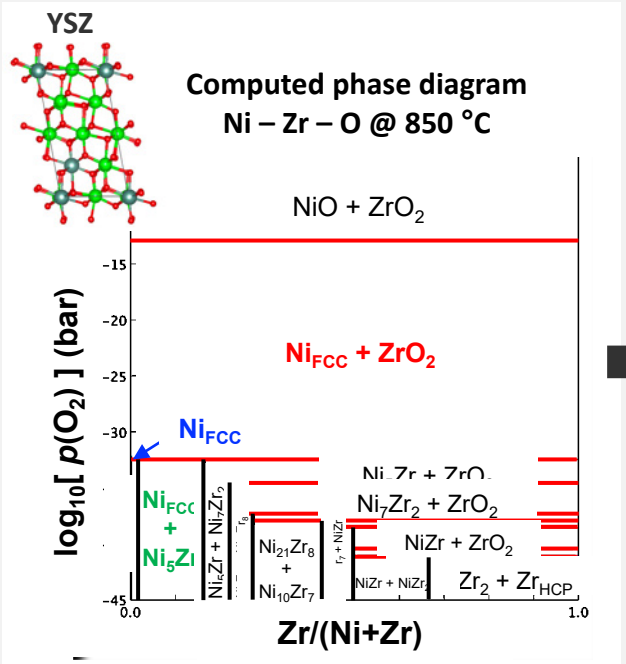
LTE/PEC: Relative degradation kinetics between 2- and 3-electrode setups

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

Proposed future work: HTE

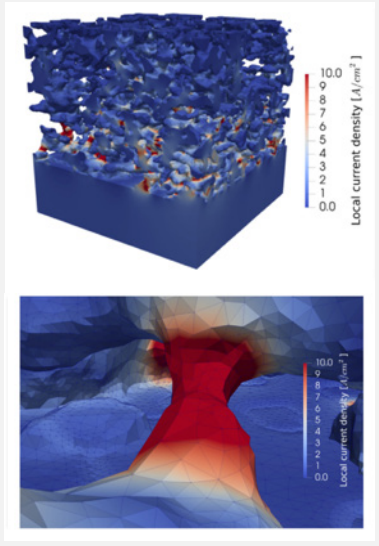
Mesoscale modeling of microstructure evolution

Computational thermodynamics



Phase-field modeling

Degradation-prone hotspots



STCH:
Phase transformation under thermochemical driving force

FY22

HTE:
Cathode phase formation and coarsening

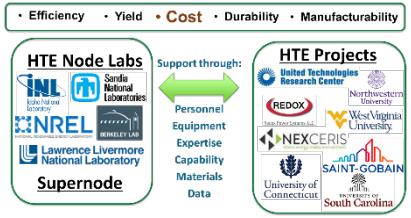
Any proposed future work is subject to change based on funding levels

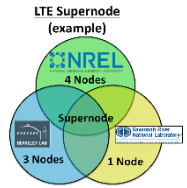
Proposed Future Work

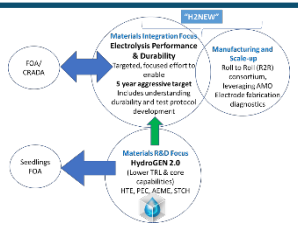
- Continue collaborative and integrated research on the five HydroGEN 2.0 projects
  - Achieve the HydroGEN 2.0 FY22 Annual and FY22 Go/No-Go milestones
- Core labs will execute HydroGEN lab nodes to enable successful phase 2 and 3 FOA-awarded project activities
  - Core labs' interaction with a specific project will end if that project does not achieve its go/no-go decision metric
- Continue to develop a user-friendly, secure, and dynamic HydroGEN Data Hub that accelerates learning and information exchange within the HydroGEN EMN labs, their partners, and other EMN, LTE, HTE, PEC, and STCH communities
  - Perform major infrastructure Data Hub upgrade of additional plugins to maintain application functionality and security standards.
  - Investigate technologies to enhance web application and application program interface (API) capabilities
  - Categorize and prioritize development possibilities based on a variety of research data engineering needs and scientific direction.
- Continue to develop a user-friendly, information rich, and relevant HydroGEN website and implement the publication page
- Conduct outreach via conference organizations, presentations and participation, benchmarking workshops, website updates and news, publications, and generally socializing the HydroGEN EMN concept to the community

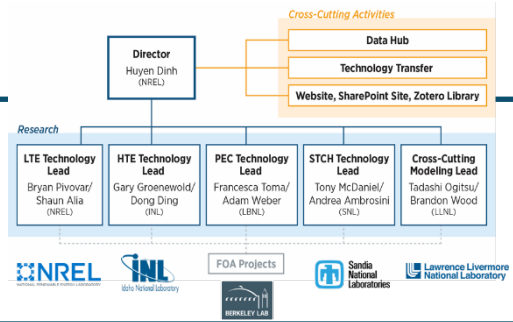
Any proposed future work is subject to change based on funding levels

Summary – HydroGEN Consortium: Advanced Water-Splitting Materials (AWSM)









- Accelerating the early-stage AWS technologies by using the consortium approach to address the critical R&D gaps of each AWS technology with the goal of improving the performance and durability and lower the cost of hydrogen production
- Achieving technical progress towards achieving HydroGEN EMN Phase 2 Go/No-Go milestones
  - 5 HydroGEN 2.0 lab projects (AEME, p-SOEC, MS-SOEC, PEC, STCH)
- Continue to connect with H2NEW consortium via cross-cutting modeling and materials development
- 4th annual benchmarking workshop: developing technology roadmaps and standard protocols for each AWS technology, engaging with and disseminating information to the community, and validating protocols
- Continue to develop Data Hub repository, tools, and metadata; Upgrading application infrastructure to maintain security and cyber standards. (https://datahub.h2awsm.org/)
- Continue to connect users to capabilities, publications, research highlights, contacts, and the Data Hub via the HydroGEN website (https://h2awsm.org)

HydroGEN fosters cross-cutting innovation using theory-guided applied materials R&D to accelerate the time-to-market and advance all emerging water-splitting pathways to enable clean, low cost, and sustainable low-cost hydrogen production

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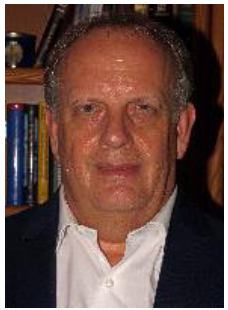

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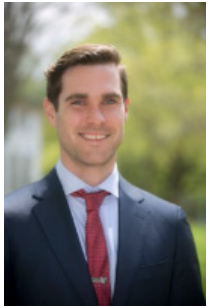
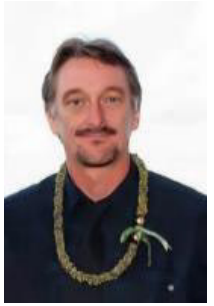


James Vickers

William Gibbons



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Acknowledgements

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Technical Backup and Additional Information Slides

Overview – LTE Technology Relevance / Impact

Proton exchange membrane (PEM)

- Gas Crossover
- Membranes
- Catalyst Materials
- Catalyst Loading
- PTL Materials

Anion exchange membrane (AEM)

- Membranes
- Catalyst
- Ionomer
- Electrolyte feed
- BOP Materials

Common Barriers

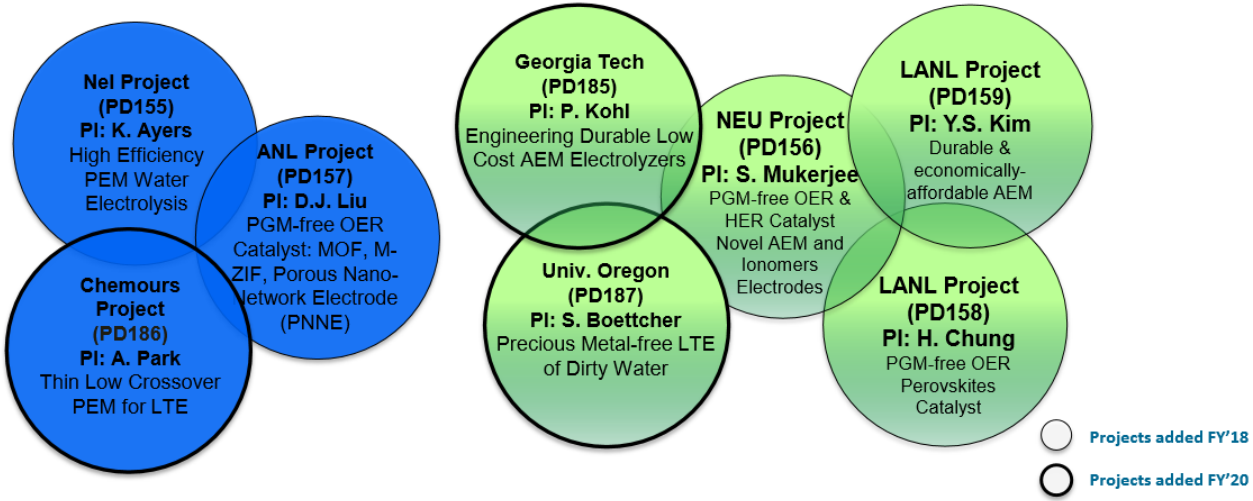
- Material Integration
- Material Cost
- Understanding Interfaces and Interactions

BOP – Balance of Plant
PTL – Porous Transport Layers

Approach: Collaborative HydroGEN LTE Projects

HydroGEN LTE Projects

- LTE 2.0 with 4 nodes
- 8 FOA projects with 41 nodes
  - 3 projects currently supported by HydroGEN labs (in Technical Backup)
  - 5 projects with closeout contributions
- 2 Supernodes with 14 nodes
  - ended last year



LTE Node Labs

NREL, Berkeley Lab, Sandia National Laboratories, Lawrence Livermore National Laboratory

Support through:

Personnel
Equipment
Expertise
Capability
Materials
Data

LTE FOA Projects

nel, Argonne National Laboratory, Northeastern University, Los Alamos (2), Georgia Institute of Technology, Chemours, University of Oregon

Supernodes	A. Weber T. Ogitsu	Understanding OER Across pH Ranges Through Multiscale, Multi-Theory Modeling
	H. Dinh B. Pivovar	Linking LTE/Hybrid Materials to Electrode Properties to Performance
LTE 2.0	S. Alia B. Pivovar	Enabling High Efficiency, Durable AEMWE Performance

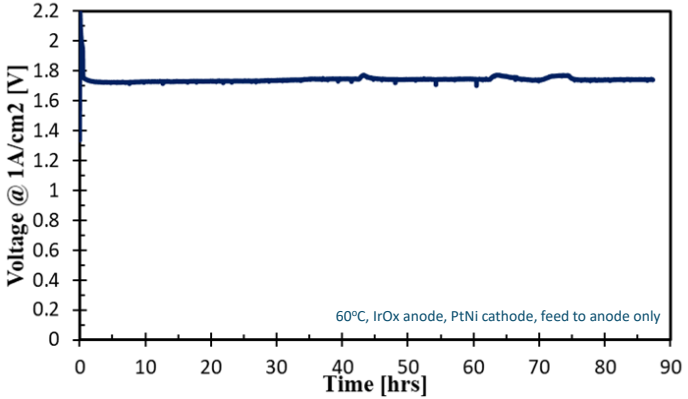
PEM: Understanding and improving materials

AEM: Developing and understanding materials

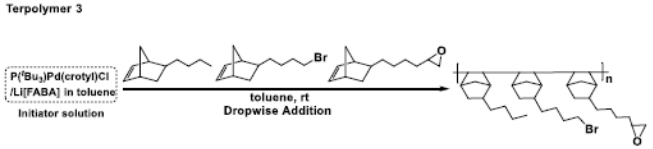
LTE P185 (P. Kohl)

High Performance AEM LTE with Advanced Membranes, Ionomers, and PGM-Free Electrodes

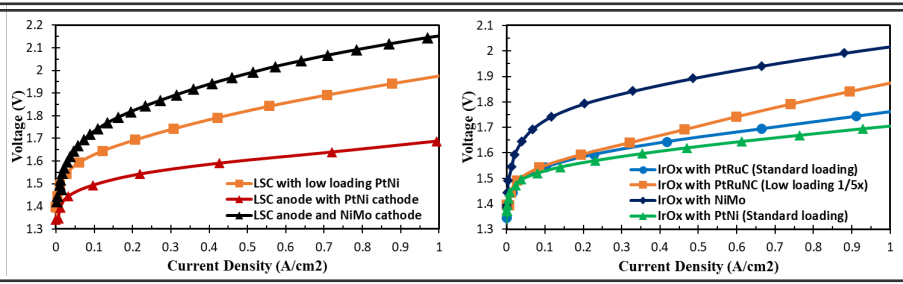
Go/No-Go (Accomplished)
PGM performance <1.75 V at 1 A/cm^2 (PGM), durability <40 mV/1,000 h extrapolated from >100 h tests



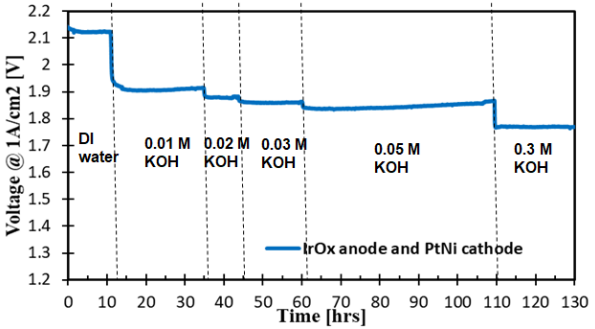
Family of self-adhesive ionomers

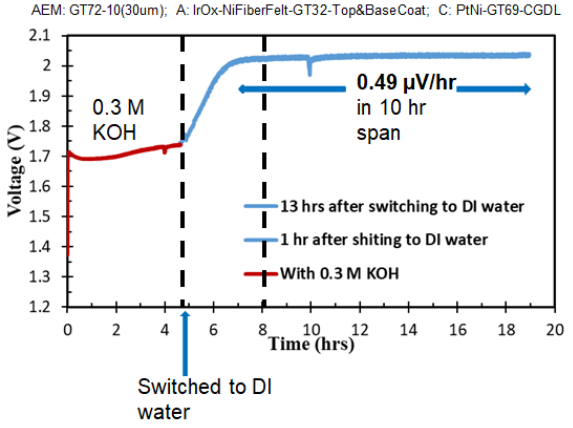


Low- and PGM-free operation



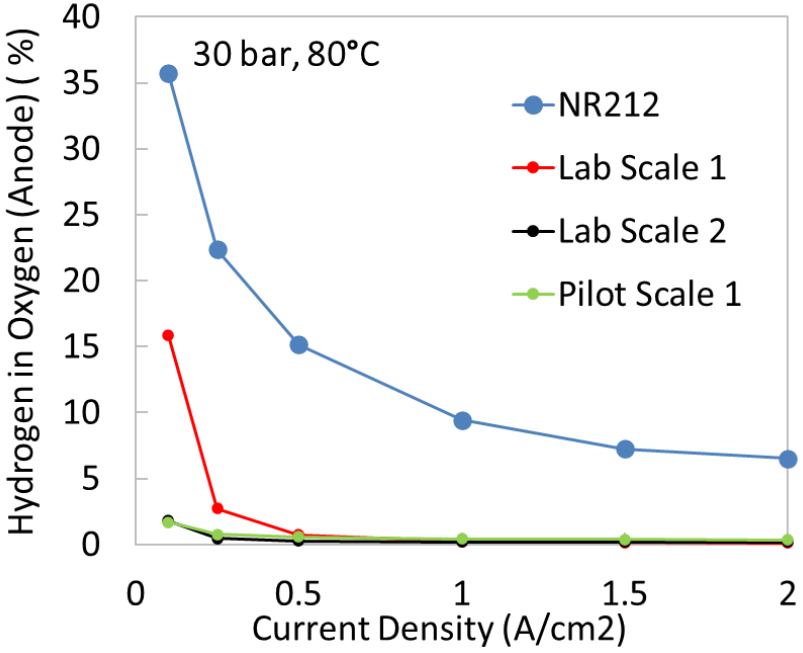
Understanding DI Water Operation





LTE P186 (A. Park)

Performance and Durability Investigation of Thin, Low Crossover PEMs for Water Electrolysis



Active area: 50 cm², Cell temperature: 80°C, Anode: 0.4 mg/cm² IrO_2, Cathode: 0.1 mg/cm² Pt/HSC, partial pressures: ~0.8 bar (O_2), 30 bar (H_2)

H_2 Crossover Mitigation with Gas Recombination Catalyst

- PEM MEAs utilizing 50 µm membranes require hydrogen crossover mitigation for safe operation
- Gas recombination catalysts (GRCs) in membrane are effective tools to mitigate hydrogen crossover
- At 80°C and 30 bar p_{H2} (most extreme conditions yet tested), mitigated prototype membranes exhibit:
  - <20% LFL from 0.25 A/cm² to 2 A/cm²
  - <50% LFL from 0.1 A/cm² to 0.25 A/cm²
- Time effect on membrane performance under evaluation

Membrane	Thickness (µm)	GRC Loading (mg/cm²)	H_2 in O_2 Content in Anode at 0.25 A/cm² (%)
NR212	51	0	22.4
Lab Scale 1	~60	<<0.1	2.7
Lab Scale 2	~60	<<0.1	0.5
Pilot Scale 1	50	<<0.1	0.75

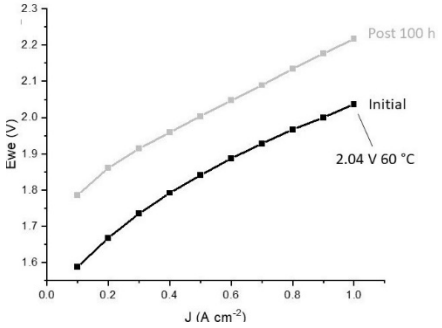
50 µm reinforced membrane with GRC demonstrates up to 50x reduction in effective H_2 crossover at 80 °C, 30 bar p_{H2}

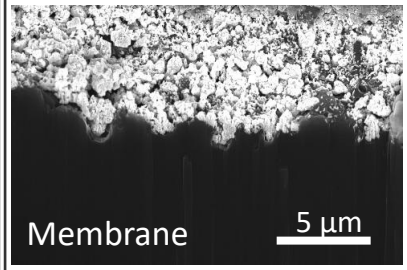
LTE P187 (S. Boettcher)

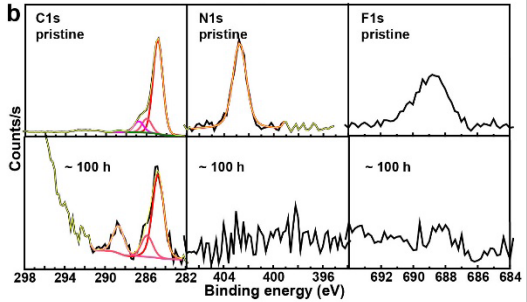
Pure Hydrogen Production through PGM-Free Membrane Electrolysis of Dirty Water

OREGON CENTER FOR ELECTROCHEMISTRY

Go/No-Go Achieved: Tap water operation with <200 mV loss after 100 h

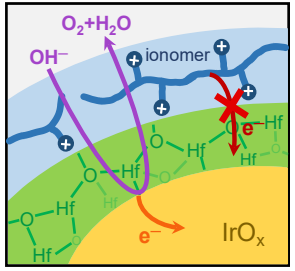




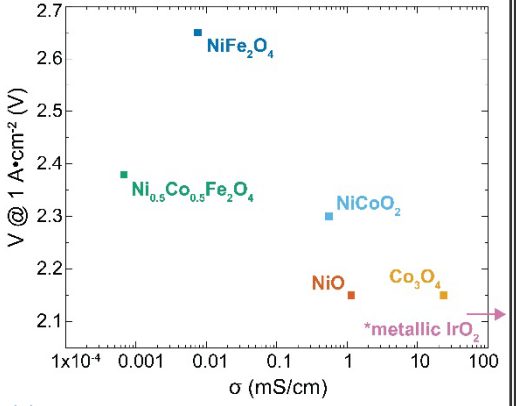


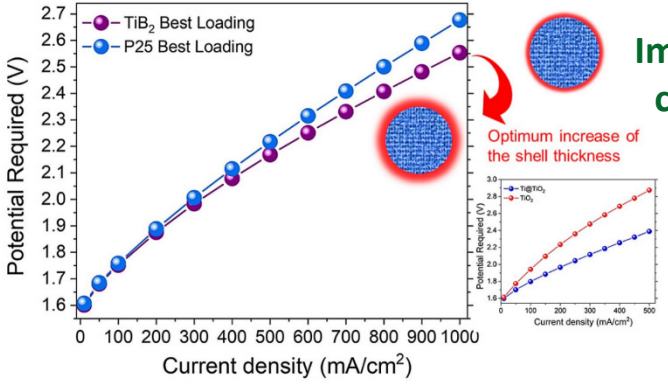
Ionomer oxidation in anode catalyst layer is significant degradation mechanism during pure water operation

HfO_x inorganic protective layer prevents ionomer oxidation



Non-PGM anode catalysts: High electrical conductivity is essential for pure-water MEA operation





Improved BPM WD catalysts achieve η_{WD} <200 mV @ 500 mA cm^{-2}

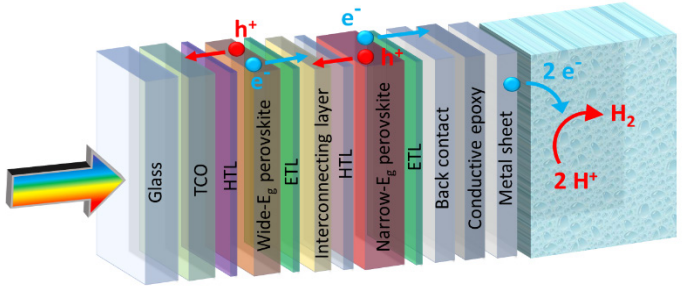
BPM = bipolar membrane
WD = water dissociation

PEC Seedling Projects Accomplishments: Perovskite/Perovskite Tandem Photoelectrodes for Low-Cost Unassisted Photoelectrochemical Water Splitting

University of Toledo PI: Y. Yan

Project Goals:

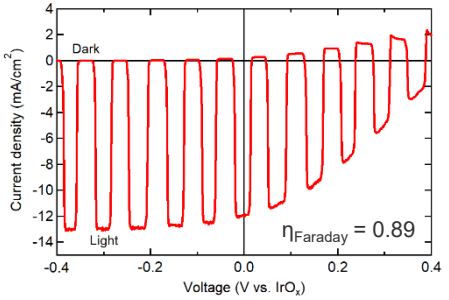
- Demonstrate perovskite/perovskite tandem photoelectrodes for wireless and unassisted water splitting
- Develop water impermeable coating to protect tandem photoelectrodes during the operation in water
- Demonstrate tandem photoelectrodes with an STH efficiency of up to 20%

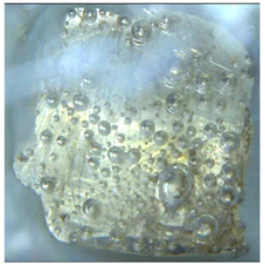


Schematic of a perovskite/perovskite tandem photoelectrode

Highlights for BP2:

- Demonstrated prototype integrated perovskite/perovskite tandem photocathodes for unassisted solar-driven water splitting.
- The perovskite/perovskite tandem photoelectrodes deliver a solar-to-hydrogen efficiency of up to 13.1% in a two-electrode configuration under one sun illumination without an external bias.
- The perovskite tandem photoelectrodes show a continuous operation lifetime in an aqueous acid solution for more than 50 hours.





Chopped-light LSV curve of a perovskite tandem photoelectrode in two-electrode configuration. Photo of solar-driven hydrogen generation on a perovskite tandem photoelectrode

PEC Seedling Projects Accomplishments: Highly Efficient Solar Water Splitting Using 3D/2D Hydrophobic Perovskites with Corrosion Resistant Barriers

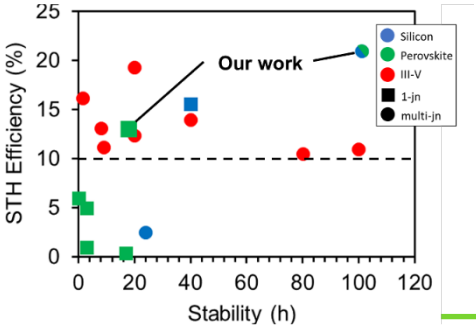
Rice University PI: A. Mohite

RICE

Project Goals:

- Develop high-efficiency 3D/2D halide perovskite solar cells that reach 20% solar to hydrogen (STH) efficiency
- Incorporate HER/OER catalysts into an autonomous PEC platform
- Using corrosion-resistant barriers, including hydrophobic polymers, carbons, and ALD oxides (Al_2O_3, HfO_2), reach 500 hours stability
- Understand degradation mechanisms in PEC and mitigate
- Demonstrate 5x5 in^2 module to show scalability

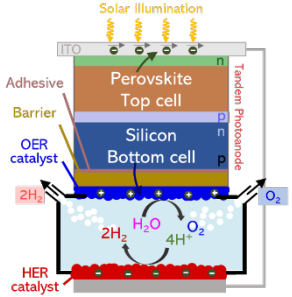
Efficiency/stability comparison to other demonstrated PECs

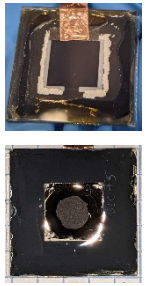


Highlights for BP2:

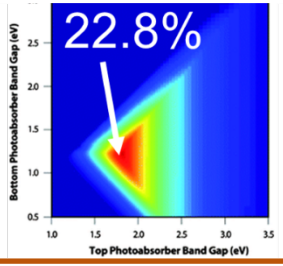
Measured STH 20.8% best literature HaP

Developed and tested a 3D printed PEC reactor incorporating 3D/2D perovskite HER+OER catalysts:





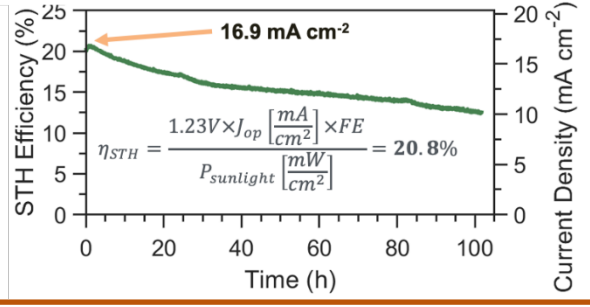
Practical limit for 2-jn PEC with SOA catalysts



Bias-free reactor performance using 2-terminal measurement:

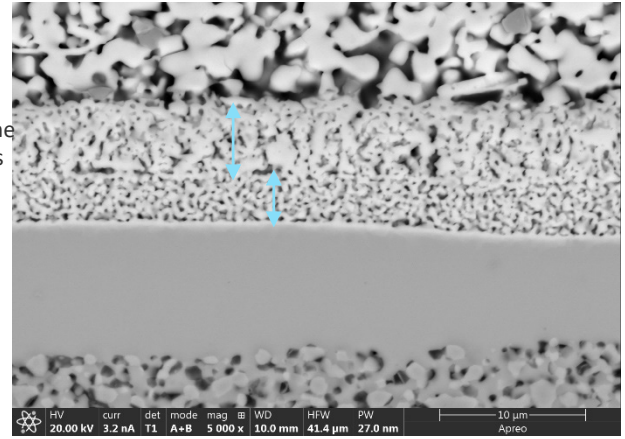
Solar-to-Hydrogen η = 20.8% and 102 hours of durability

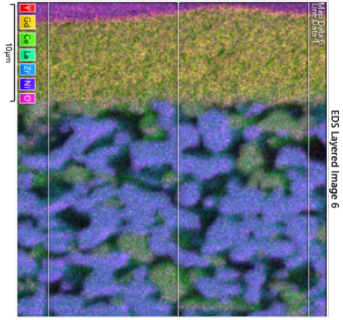
High efficiency, stability via anticorrosion barrier, PM catalysts

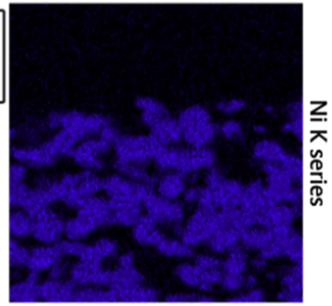


HTE Saint Gobain/BU: Degradation Caused by Decomposition and Pore Structure Changes in Electrode

NNO-NDC/GDC10
The pore structure of the GDC layer changes at its midpoint.

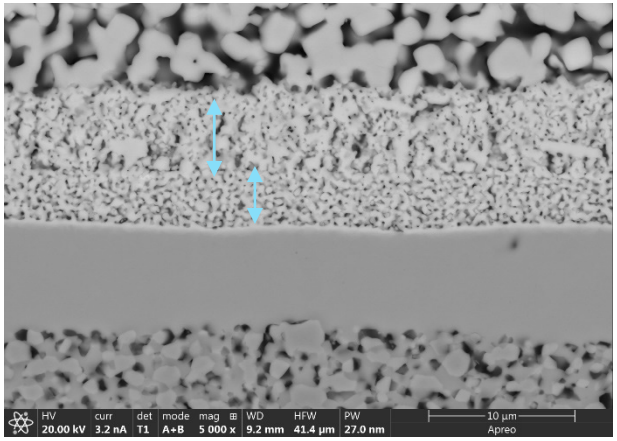


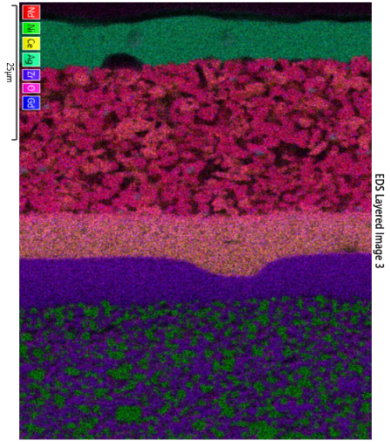


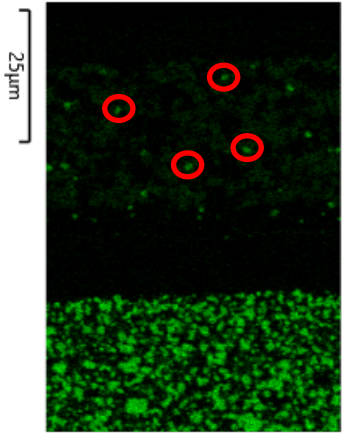


The LNO-LDC does not decompose

The same occurs in the GDC layer adjacent to a LNO-LDC electrode



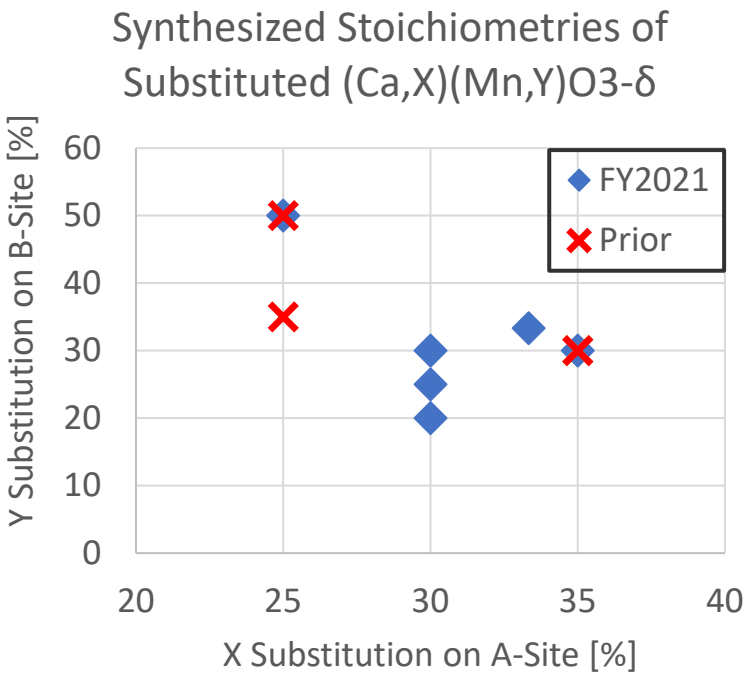




The NNO-NDC decomposes into Ni-O rich inclusions

STCH Seedling Support: Empirical Testing of Two Redox Active Sublattices

- **Motivation:** DFT predicts multiple sublattice participation advantageous for STCH performance
- [NREL, SLAC] Demonstrated wide range of solid solubility by synthesizing six distinct stoichiometries of $(Ca,X)(Mn,Y)O_{3-\delta}$ perovskite in FY 2021.
  - Building off and reproducing four prior stoichiometries
- [NREL, SLAC, SNL] Synchrotron and electron characterization of structure and chemistry
  - Charted range of solid solution stability under redox cycling:
    - All stoichiometries demonstrated solid solution at 1,500˚C
    - Exosolution of X under redox cycling at certain stoichiometries and temperatures
  - Measured degree and reversibility of reduction/oxidation of both active cations (X and Mn) under identical conditions
  - Phase stability and lattice response to redox cycling
- [SNL] Validated water splitting by flow reactor measurements

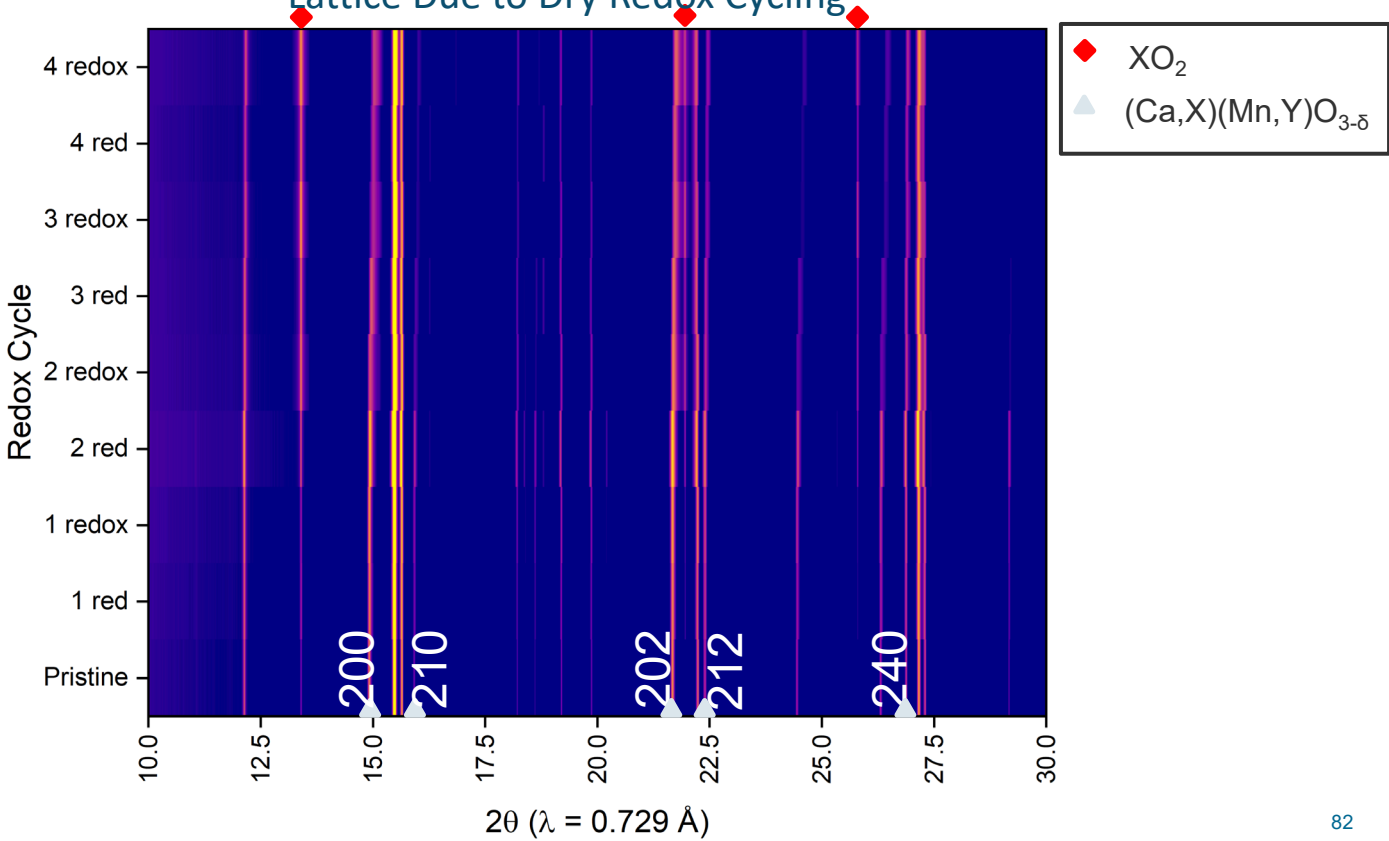


STCH: Demonstrating Cycling Stability of Different Substitutional Stoichiometries

Synchrotron Characterization

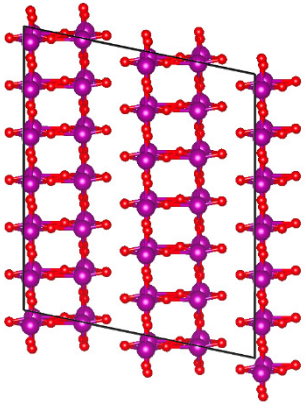
- Confirmed crystal structure match between stoichiometries
- Probed exosolution kinetics and conditions

Ex-Situ X-Ray Diffraction Demonstrates Shift in Perovskite Lattice Due to Dry Redox Cycling



STCH: A *More Generalizable* Approach (like GNNs) is Ideal to Model Vacancy Defects

(1) Compositional models destined to fail



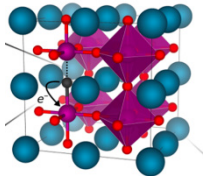
Wyckoff site	ΔH_d [eV]
Mn1	12.2
Mn2	12.1
O1	2.1
O2	2.2
O3	2.6
O4	2.7

$$MnO_{1.5} \mapsto \boldsymbol{x}_{O1} = \boldsymbol{x}_{O4} = \{\bar{v}_{pa}, \bar{r}_{cov}, \bar{\chi}, \ldots\}$$

Limitations: $\boldsymbol{x}_{O1}$ and $\boldsymbol{x}_{O4}$ are identical but…
- Local structures quite different
- Our target value differs by 0.6 eV

(2) Hand-engineered x used for specific material classes

Wexler et al *J. Am. Chem. Soc.* **2021**



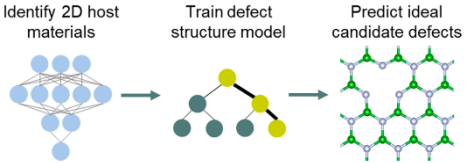
Linear Model works well for ABO3 perovskites:

$$0.1\Sigma E_b - 1.5V_r + 0.4E_g - 55.8E_{hull} + 0.4 \text{ (eV)}$$

Crystal bond dissociation; Crystal reduction; Band gap; Stability; $\boldsymbol{x}$

Limitations:
- Model only validated for O sites in ABO_3 perovskites
- Needs re-derivation for other structure classes

Frey et al. *ACS Nano*. **2020**



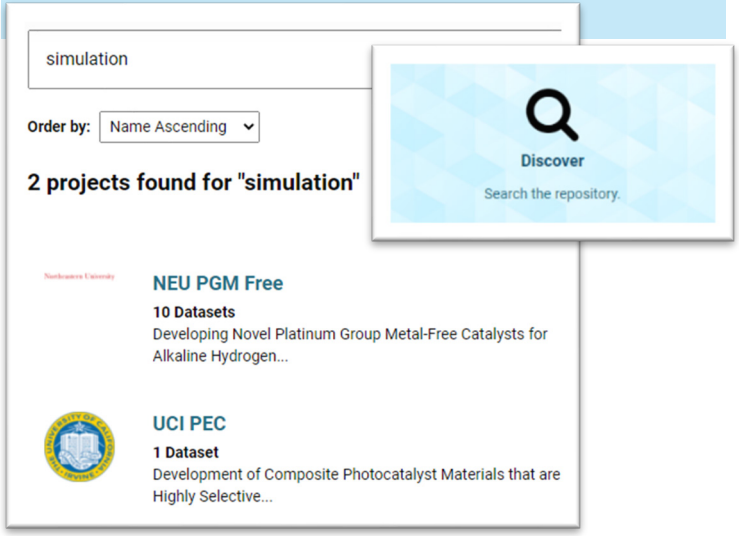
Random forest model works well for certain vacancies in 2D materials

Limitations:
- Model and features specific to 2D material classes
- Needs re-derivation for other structure classes

FY22 Data Hub Quarterly Planning Map

What: *HydroGEN Q4 QPM*

- Investigate technologies and capabilities for increasing web API usability and data searchability
- Categorize and prioritize development based on needs and direction



Why: *In Response to User Feedback*

- Feedback collected from Users and PIs across data hubs for enhancements
- Request for search and find capabilities determined to be beneficial

How: *Approach and Work Outline*

1. **Identify core, top priority enhancements** that would return the highest value.
2. **Investigate technologies and development techniques** for high value feature enhancements to improve searchability:
    1. Research cutting edge technologies
    2. Create implementation plan
    3. Scope efforts
    4. Identify development hot spots for research
    5. Proof of concept hot spots
3. **Categorize new feature development and enhancements.**
    1. Identify number of benefiting research areas
    2. Implementation scheduling
    3. Categorize
4. **Update overall implementation plan and begin feature development**

Technology Transfer Activities

- HydroGEN EMN has developed four standard, pre-approved technology transfer agreements between all consortium partners to enable streamlined access:
  - Non-Disclosure Agreement (NDA)
  - Intellectual Property Management Plan (IPMP)
  - Materials Transfer Agreement (MTA)
  - Cooperative Research and Development Agreement (CRADA)
- HydroGEN has executed 33 project NDAs and 2 MTAs
- Tech transfer in HydroGEN occurs organically via collaboration between lab capability nodes and industry partners in the different AWS technologies
- Continue to develop IP and follow the IPMP developed by the HydroGEN consortium
- Patent Applications:
  - T. He, D. Ding, W. Wu. Methods and systems for hydrogen gas production through water electrolysis, and related electrolysis cells. US Patent (16/483,631), Granted, 2021.
  - D. Ding, W. Bian, W. Wu. Facile methods to rejuvenate electrolyte surface for high-performing protonic ceramic electrochemical cells. US Patent Provisional Application (63/298,084), 2022.
  - A. Mohite, M. Wong, A. Fehr, A. Agrawal, F. Mandani, Conductive Adhesive-Barrier Enabling Integrated Photoelectrodes for Solar Fuels. Provisional Application for U.S. Letters Patent (22511 Patent Trademark Office).

Special Recognitions and Awards

- Dong Ding: INL-EEST Leadership Award, 2021
- Francesca M. Toma: Oppenheimer Fellow 2022
- Aditya Mohite: Outstanding Faculty Research Award 2022, Rice University
- Savannah Tiemann (undergraduate researcher): James S. Waters Creativity Award, Rice University

Publications

1. W. Bian, W. Wu, B. Wang, W. Tang, M. Zhou, C. Jin, H. Ding, W. Fan, Y. Dong, J. Li, and D. Ding, “Revitalizing Interface in Protonic Ceramic Cells by Acid Etch,” Nature 604 (2022): 479–485, https://doi.org/10.1038/s41586-022-04457-y.
2. Y. Zhang, N. Xu, Q. Tang, and K. Huang, “Intermediate Temperature Solid Oxide Cell with a Barrier Layer Free Oxygen Electrode and Phase Inversion Derived Hydrogen Electrode,” *Journal of the Electrochemical Society* 169 (2022): 034516, https://doi.org/10.1149/1945-7111/ac565a.
3. K. Cook, J. Wrubel, Z. Ma, K. Huang, and X Jin, “Modeling Electrokinetics of Oxygen Electrodes in Solid Oxide Electrolyzer Cells,” *Journal of the Electrochemical Society* 168 (2021): 114510, https://doi.org/10.1149/1945-7111/ac35fc.
4. Q. Zhang, S. Barnett, and P. Voorhees, “Migration of inclusions in a matrix due to a spatially varying interface energy,” *Scripta Materialia* 206 (2022), https://doi.org/10.1016/j.scriptamat.2021.114235.
5. Q. Zhang, Q.-Y. Liu, B.-K. Park, S. Barnett, and P. Voorhees, “The oxygen partial pressure in solid oxide electrolysis cells with multilayer electrolytes,” *Acta Materialia* 213 (2021), https://doi.org/10.1016/j.actamat.2021.116928.
6. B.-K. Park, D. Cox, and S. A. Barnett, “Effect of Nanoscale $Ce_{0.8}Gd_{0.2}O_{2-\delta}$ Infiltrant and Steam Content on Ni–$(Y_2O_3)_{0.08}(ZrO_2)_{0.92}$ Fuel Electrode Degradation during High-Temperature Electrolysis,” *Nano Letters* (2021), https://doi.org/10.1021/acs.nanolett.1c02937.
7. Q. Zhang, B. K. Park, P. W. Voorhees, and S. A. Barnett, “On The Role Of The Zirconia/Ceria Interface In The Degradation Of Solid Oxide Electrolysis Cells,” *Applied Physics Letters* 117 (2020): 123906, https://doi.org/10.1063/5.0016478.
8. W. Bian, W. Wu, Y. Gao, J. Gomez, H. Ding, W. Tang, M. Zhou, and D. Ding, “Regulation of cathode mass and charge transfer by structural 3D engineering for protonic ceramic fuel cell at 400 C,” *Advanced Functional Materials* 31 (2021): 2102907, https://doi.org/10.1002/adfm.202170244.
9. C. Vera, H. Ding, D. Peterson, W. Gibbons, M. Zhou, and D. Ding, “A mini-review on proton conduction of $BaZrO_3$-based perovskite electrolytes,” *Journal of Physics: Energy* 3 (2021): 032019, https://doi.org/10.1088/2515-7655/ac12ab.

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10. A. Bayonet, et al., “Operational limits of redox metal oxides performing thermochemical water splitting,” *Energy Technology* 10 (2022): 2100222.
11. A. M. Bergeson-Keller, M. D. Sanders, and R. P. O’Hayre, “Reduction Thermodynamics of $Sr_{1-x}Ce_xMnO_3$ and $Ce_xSr_{2-x}MnO_4$ Perovskites for Solar Thermochemical Hydrogen Production,” *Energy Technology* 10 (2022): 2100515.
12. M. D. Sanders, A. M. Bergeson-Keller, E. N. Coker, and R. P. O'Hayre, “A Thermogravimetric Temperature Programmed Thermal Redox Protocol for Rapid Screening of Metal Oxides for Solar Thermochemical Hydrogen Production,” *Frontiers in Energy Research* 10 (2022): 856943, https://doi.org/10.3389/fenrg.2022.856943.
13. B. Bulfin, M. Miranda, and A. Steinfeld, “Performance Indicators for Benchmarking Solar Thermochemical Fuel Processes and Reactors,” *Frontiers in Energy Research* 9 (2021): 677980, https://doi.org/10.3389/fenrg.2021.677980.
14. S. L. Millican, et al., “Predicting Oxygen Off-Stoichiometry and Hydrogen Incorporation in Complex Perovskite Oxides,” *Chemistry of Materials* 34 (2022): 510–518.
15. S. L. Millican, et al., “Redox Defect Thermochemistry of $FeAl_2O_4$ Hercynite in Water Splitting from First-Principles Methods,” *Chemistry of Materials* 34 (2022): 519–528.
16. Z. Bare, R. J. Morelock, and C. B. Musgrave, “A Computational Framework to Accelerate the Discovery of Perovskites for Solar Thermochemical Hydrogen Production: Identification of Gd Perovskite Oxide Redox Mediators,” *Advanced Functional Materials* (2022): 2200201.
17. J. E. Park, et al., “Computationally Accelerated Discovery and Experimental Demonstration of $Gd_{0.5}La_{0.5}Co_{0.5}Fe_{0.5}O_3$ for Solar Thermochemical Hydrogen Production,” *Frontiers in Energy Research* 9 (2021).
18. R. B. Wexler, et al., “Factors Governing Oxygen Vacancy Formation in Oxide Perovskites,” *Journal of the American Chemical Society* 143 (2021): 13212–13227.

Publications, continued

19. S. J. Heo, et al., “Double-site substitution of Ce into (Ba, Sr) MnO_3 perovskites for solar thermochemical hydrogen production,” *ACS Energy Letters* 6 (2021): 3037–3043.
20. X. Qian, et al., “Outstanding Properties and Performance of $CaTi_{0.5}Mn_{0.5}O_{3-\delta}$ for Solar-Driven Thermochemical Hydrogen Production,” *Matter* 4 (2021): 688–708.
21. N. A. Strange, J. E. Park, A. Goyal, R. T. Bell, J. A. Trindell, J. D. Sugar, K. H. Stone, E. N. Coker, S. Lany, S. Shulda, and D. S. Ginley, “Formation of 6H-Ba3Ce0.75Mn2.25O9 during Thermochemical Reduction of 12R-Ba4CeMn3O12: Identification of a Polytype in the Ba(Ce,Mn)O3 family,” *Inorg. Chem.* 61 (2022): 6128, https://doi.org/10.1021/acs.inorgchem.2c00282.
22. V. K. Budama, N. G. Johnson, I. Ermanoski, and E. B. Stechel, “Techno-economic analysis of thermochemical water-splitting system for co-production of hydrogen and electricity,” *International Journal of Hydrogen Energy* 46 (2021): 1656–1670.
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24. O. Alley, et al., “Best practices in PEC water splitting: How to reliably measure solar-to-hydrogen efficiency of photocathodes,” *Frontiers in Energy Research* (2022), accepted.
25. Y. Xiao, S. Vanka, T. A. Pham, W. J. Dong, X. Liu, I. A. Navid, J. B. Varley, H. Hajibabaei, T. W. Hamann, T. Ogitsu, and Z. Mi, “Crystallographic Effects of GaN Nanostructures in Photoelectrochemical Reaction,” *Nano Lett.* 22 (2022): 2236.
26. S. Hu, X. Shen, R. Yanagi, D. Solanki, H. Su, Z. Li, and C. Xiang, “Comprehensive Evaluation For Protective Coatings: Optical, Electrical, Photoelectrochemical, and Spectroscopic Characterization,” *Frontiers in Energy Research* 9 (2022): 799776, https://doi.org/10.3389/fenrg.2021.799776.

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27. D. S. Ellis, Y. Piekner, D. A. Grave, P. Schnell, and A. Rothschild, “Considerations for the Accurate Measurement of Incident Photon to Current Efficiency (IPCE) in Photoelectrochemical (PEC) Cells,” *Frontiers in Energy Research* 9 (2022): 726069, https://doi.org/10.3389/fenrg.2021.726069.
28. A. R. Motz, D. Li, A. Keane, L. D. Manriquez, E. J. Park, S. Maurya, H. Chung, C. Fujimoto, J. Jeon, M. K. Pagels, C. Bae, K. E. Ayersa, and Y. S. Kim, “Performance and Durability of Anion Exchange Membrane Water Electrolyzers using Down-selected Polymer Electrolytes,” *Journal of Materials Chemistry A* 9 (2021): 22670–22683.
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30. H. R. Colon-Mercado, S. A. Mauger, M. B. Gorensek, C. H. Fujimoto, A. A. Lando, P. Ganesane; B. H. Meekins, and N. D. Meeks, “Electrode optimization for efficient hydrogen production using an SO2-depolarized electrolysis cell,” International Journal of Hydrogen Energy 47 (2022): 14180–14185.
31. J. Liu, Z. Kang, D. Li, M. Pak, S. M. Alia, C. Fujimoto, G. Bender, Y. S. Kim, and A. Z. Weber, “Elucidating the Role of Hydroxide Electrolyte on Anion-Exchange-Membrane Water Electrolyzer Performance,” *Journal of The Electrochemical Society* 168 (2021): 054522, https://doi.org/10.1149/1945-7111/ac0019.
32. A. Kiessling, J. C. Fornaciari, G. Anderson, X. Peng, A. Gerstmayr, M. R. Gerhardt, S. E. Mckinney, A. Serov, Y. S. Kim, B. Zulevi, A. Z. Weber, and N. Danilovic, “Influence of Supporting Electrolyte on Hydroxide Exchange Membrane Water Electrolysis Performance: Anolyte,” *Journal of the Electrochemical Society* 168 (2021): 084512, https://doi.org/10.1149/1945-7111/ac1dcd.
33. J. C. Fornaciari, L.-C. Weng, S. M. Alia, C. Zhan, T. A. Pham, A. Bell, T. Ogitsu, N. Danilovic, and A. Z. Weber, “Mechanistic understanding of pH effects on the oxygen evolution reaction,” *Electrochim. Acta* 405 (2022): 139810, https://doi.org/10.1016/j.electacta.2021.139810.

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34. A. G. Divekar, M. R. Gerhardt, C. M. Antunes, L. Osmieri, A. C. Yang-Neyerlin, A. Z. Weber, B. S. Pivovar, G. Bender, and A. M. Herring, "Spatially Resolved Performance and Degradation in a Perfluorinated Anion Exchange Membrane Fuel Cell," *Electrochimica Acta* 405 (2022): 139812, https://doi.org/10.1016/j.electacta.2021.139812.
35. C. Arges, V. Ramani, Z. Wang, and R. J. Ouimet, "Assessing the Oxidative Stability of Anion Exchange Membranes in Oxygen Saturated Aqueous Alkaline Solutions," *Frontiers in Energy Research* 10 (2022): 871851, https://doi.org/10.3389/fenrg.2022.871851.
36. S. Alia, D. Ding, A. McDaniel, F. M. Toma, and H. N. Dinh, "Chalkboard 2 - How to Make Clean Hydrogen," *The Electrochemical Society Interface* 30 (2021): 49, https://doi.org/10.1149.2/2.F13214IF.
37. B. Pivovar, M. F. Ruth, D. J. Myers, and H. N. Dinh, "Hydrogen: Targeting $1/kg in 1 Decade," *The Electrochemical Society Interface* 30 (2021): 61, https://doi.org/10.1149.2/2.F15214IF.
38. R. R. White, K. Munch, N. Wunder, N. Guba, C. Sivaraman, K. M. Van Allsburg, H. Dinh and C. Pailing, "Energy Material Network Data Hubs," *International Journal of Advanced Computer Science and Applications(IJACSA)* 12 (2021), http://dx.doi.org/10.14569/IJACSA.2021.0120677.

Presentations

1. F. M. Toma, “Photoelectrochemical Fuel Generation: Light Absorbers Under the Spotlight,” Invited talk, Renewable Energy: Solar Fuels (Gordon Research Conference) Advancing Complexity, Selectivity and Efficiency in Artificial Photosynthesis (May 8–13, 2022, Barga, Italy).
2. F. M. Toma, “Understanding the mechanism of photo electrochemical transformations in functional architectures for artificial photosynthesis,” Invited talk, Symposium #173 Recent Trends and Advances in Artificial Photosynthesis, Pacifichem (December 16–21, 2021).
3. F. M. Toma, “Chemical transformations of (photo)electrocatalytic materials,” Invited talk, EPFL Open Science Workshop on Solar Fuels Standards and Benchmarking (December 7, 2021).
4. F. M. Toma, “(Photo)electrocatalysis at work,” Invited department seminar,” Department of Chemical and Biochemical Engineering, Rice University (September 14, 2021).
5. A. Fehr, et al., “Highly-Efficient, Stable, and Scalable Integrated Hybrid Perovskite Photoelectrochemical Cells with STH>12.4% for Water-Splitting,” ECS Fall Meeting (2021).
6. Z. Song, C. Li, L. Chen, S. Rijal, J. L. Young, T. G. Deutsch, and Y. Yan, “High-Photovoltage All-Perovskite Tandem Solar Cells for Photovoltaic-Electrolysis Water-Splitting Applications,” 2021 IEEE 48th Photovoltaic Specialists Conference (PVSC) (June 20–25, 2021).
7. Z. Song, C. Li, L. Chen, and Y. Yan, “Air-stable all-perovskite tandem solar cells,” Solar Power Tech Conference, Virtual Conference (July 5–8, 2021).
8. Z. Song, C. Li, J. L. Young, T. G. Deutsch, and Y. Yan, “High-Photovoltage Pb-based All-Perovskite Tandem Solar Cells for Unassisted Water-Splitting,” AIP Publishing Horizons: Energy Storage and Conversion, Virtual Conference (August 4–6, 2021).

Presentations, continued

9. Z. Song, C. Li, L. Chen, and Y. Yan, “Inorganic Lead Halide Perovskites Based Tandem Photoelectrodes for Unassisted Water-Splitting,” MRS Spring Meeting (May 9–13, 2022).
10. E. Dogdibegovic, G. Arkenberg, D. Kopechek, A. Wallace, and S. Swartz, “Development of Solid Oxide Cell and Stack Technologies at Nexceris: From Fuel Cells to Electrolyzers and Reversible Operation,” Invited Keynote Lecture at TMS (February 2022).
11. K. Huang, “High Performance Bilayer Oxygen Electrode for Solid Oxide Water Electrolyzers: Experimental and Modeling Results,” Invited talk, ICACC virtual meeting (2022).
12. D. Ding, “Is proton conducting solid oxide electrolysis cell (p-SOEC) ready for scalable electrochemical hydrogen production? An overview of research advancement of p-SOEC at Idaho National Laboratory,” Invited talk, 2021 Materials Research Society (MRS) Fall Conference, virtual meeting, Boston, MA (December 6–8, 2021).
13. T. A. Pham, “First-Principles Simulations of Heterogeneous Interfaces for the Water-Energy Nexus,” Invited, APS March Meeting, Chicago (March 15, 2022).
14. T. A. Pham, “First-Principles Simulations of Heterogeneous Interfaces for the Water-Energy Nexus,” Invited (Virtual), CECAM Workshop: Recent Advances in Machine Learning Accelerated Molecular Dynamics (March 18, 2022).
15. B. C. Wood and T. Ogitsu, “Multiscale Modeling and Theory Experiment Integration for Understanding Complex Interfaces in Materials for Hydrogen Production and Storage,” ECS Meeting (October 2021).
16. T. Ogitsu, “Multi-scale computational modeling of photoelectrochemical solar-to-fuel conversion processes,” Pacifichem (December 2021).
17. T. Ogitsu, “Probing complex interfaces using ab-initio simulations and experimental characterizations,” Catalysis and Chemical Engineering Conference (March 2022).

Presentations, continued

18. A. Goyal and S. Lany, “Ab Initio Study of Atomic and Electronic Structure of Promising $Ba_4XMn_3O_{12}$ (X = Nb, Ce, Pr) Oxides for Solar Thermochemical Hydrogen Production,” Contributed talk, Symposium EN05, MRS Spring Meeting, virtual (April 17–23, 2021).
19. S. L. Millican, J. M. Clary, C. B. Musgrave, and S. Lany, “Redox defect thermochemistry of $FeAl_2O_4$ hercynite in water-splitting from first principles methods,” Contributed talk, Symposium EN14, MRS Fall Meeting, virtual (December 6–8, 2021).
20. A. Goyal, M. Sanders, R. O’Hayre, and S. Lany, “Thermodynamic Modeling of Redox Behavior of Complex Oxides for Solar Thermochemical Hydrogen (STCH),” Contributed talk, Symposium EN14, MRS Fall Meeting, virtual (December 6–8, 2021).
21. S. Lany, “Computational Approaches to Defects and Doping in Non-Ideal Semiconductors,” Invited talk, Session B67, APS March Meeting 2022, Chicago, IL (March 14–18, 2022).

HydroGEN 2.0 FY21 Annual Milestones and Go/No-Go Milestones

AWS	FY21 Annual Milestones (Q4)	Go/No-Go Milestone (in 18 months)
LTE AEM	LBNL and NREL will measure electrolysis performance using 50 micron commercial Nafion and one AEM at 40, 60, and 80°C and compare results to establish baseline required for Q2 FY22 LTE go/no-go decision. (NREL, LBNL, Q2) **- COMPLETE**	Description: Incorporating experiment and modeling, down-select AEM operating conditions (e.g., flow configurations, back pressure, temperature) that show water-fed (i.e., no supporting electrolyte) AEMWE is a viable pathway, by demonstrating cell overvoltage within 50 mV (IR free) of a 50 µm commercial Nafion at 1 A/cm^2. (NREL and LBNL) Criteria: Demonstrate water-fed AEMWE (i.e., no supporting electrolyte) with a cell overvoltage within 50 mV (IR free) of commercial 50 µm Nafion at 1 A/cm^2. **- Did not achieve; No-Go water-fed AEMWE approach**
HTE	**MS-SOEC (Q4 QPM):** Gen 2 cell operated for >500 h at constant inlet steam content of 50% or higher, current density (chosen to give initial voltage around 1.3 to 1.4 V), and temperature around 700°C. Gen 1 status is: 0.6 A/cm^2 at 1.4V, 700°C, 50:50 H_2O:H_2; 15%/kh degradation at 0.33 A/cm^2. (LBNL) **- COMPLETE** **p-SOEC:** Establish correlations between the computational framework and experiments including electrochemical, FTIR, TGA, and TPD to determine primary mechanisms affecting formation and proton/polaron migration. Demonstrate >75% agreement in prediction of proton formation and/or migration barrier for BZY and Ce-doped BZY systems. (INL) **- COMPLETE**	**MS-SOEC:** Description: Demonstrate improved performance and durability for Gen-2 MS-SOEC cell, with at least 0.8 A/cm^2 at 1.4 V and <10%/kh degradation at 700°C with at least 50% steam. (LBNL) Criteria: Button cell MS-SOEC operated at 700°C with at least 50% steam provides at least 0.8 A/cm^2 at 1.4 V and displays <10%/kh degradation over at least 200 h continuous operation after initial transients are complete. **- COMPLETE** **p-SOEC:** Description: Develop effective approaches including tuning dopant elements and their concentrations, manipulating electronic structure of Ce, optimizing operating condition to achieve high Faradaic efficiency at 1.0 A/cm^2 by suppressing electronic leakage of electrolyte materials. (INL) Criteria: Demonstrate ≥2 strategies combined to effectively suppress electronic leakage and achieve an improved Faradaic efficiency >90% at 1.0 A/cm^2 at 600°C by optimization of electrolyte composition and operating conditions. **- COMPLETE**

HydroGEN 2.0 Go/No-Go Milestone (in 18 months)

AWS	FY21 Annual Milestones	Go/No-Go Milestone (in 18 months)
PEC	Demonstrate Type III device with near 1% or better solar-to-hydrogen (STH) efficiency and near 10 h or better stability tested at LBNL and/or NREL with the same absorber architecture and device configuration having near-neutral electrolyte and a membrane but different cells under ambient conditions and the AM 1.5G reference spectrum. (NREL, LBNL, LLNL) **- COMPLETE**	Description: Demonstrate 5% STH or better efficiency with 50 h or better durability (within 20% of initial STH) with down-selected device components and architecture with a membrane or separator, in near neutral pH conditions with development of a protocol with standardized testing conditions between NREL and LBNL. (LBNL and NREL) Criteria: Demonstrate 5% STH or better efficiency with 50 h or better durability in near neutral pH conditions. **- COMPLETE**
STCH	Demonstrate a comprehensive STCH material down-select process that combines detailed thermodynamic data (e.g., 248 kJ/mol O <reduction enthalpy <500 kJ/mol O, reduction entropy that trends more positive with increasing enthalpy) with computational methods that incorporate necessary and sufficient reactor conditions (e.g., maximum reduction temperature <1,500°C, minimum oxidation temperature >500°C, moles H_2/mole oxide determined by reactor modality and solar-to-hydrogen conversion efficiency trade-offs, moles H_2O/mole H_2 <10) needed to predict best-case material performance. Analysis will result in a rigorous assessment and ranking of a material's likelihood to meet DOE STCH technology performance targets, which will ultimately inform the down-select process. Demonstration will consist of analyzing one or two exemplar materials. (SNL, NREL, LLNL) **- COMPLETE**	Description: Incorporate experiment and thermodynamic cycle analysis on exemplar and predicted material formulations, determine cycle conditions (TRED, TOX, reduction pO2, etc.) necessary to achieve 0.1%, 5%, and 10% reaction yields. Assess material viability based on physical limitations exposed in the cycle analysis.(NREL & SNL) Criteria: 4–5 known STCH oxide materials synthesized and benchmarked relative to CeO_2 using new performance/technology viability metric. Document which rise above or fall below CeO_2. Also synthesize and assess 1–2 materials discovered by computational ML platform. Description: Complete data collection from high throughput first principles calculations and database queries for the reduction/oxidation properties for more than 100 oxides. Demonstrate progress towards machine learning capabilities to correlate composition/ structure features with STCH relevant thermochemical properties. Criteria: Identify at least 1 new STCH material formulation based completely on first principles defect structure calculations and machine learning. **- Due in FY22 Q3**

HydroGEN 2.0 End of Project Milestone (in 3 years)

AWS	State of the Art (Point A)	End of Project Milestone (Point B)
LTE AEM	Overvoltage for AEMWE operation with DI water is in excess of 100 mV of Nafion 112 at 2 A/cm^2.	Demonstrate overvoltage for AEMWE operation with deionized water that is within 50 mV of Nafion 112 at 2 A/cm^2. Demonstrate an overvoltage increase for AEMWE operation with deionized water that is within 0.15 mV/h during 500 h of operation at 1 A/cm^2. (NREL, LBNL)
HTE	**MS-SOEC:** Metal-supported oxygen-conducting SOEC (MS-SOEC) button cells have demonstrated 0.6 A/cm^2 at 1.4 V, 700°C, 50:50 H_2O:H_2; and 15%/kh degradation at 0.33 A/cm^2 over 1,000 h of operation. **p-SOEC:** The proton conduction is not well understood and a low Faradaic efficiency (<80%) has been observed in p-SOEC when the state-of-the-art electrolyte materials are used for a given current density (e.g., > 0.5 A/cm^2 at 1.3 V)	**MS-SOEC:** Demonstrate metal-supported oxygen-conducting SOEC (MS-SOEC) cells with >1 A/cm^2 at 1.4 V, 700°C, 50:50 H_2O:H_2, and <5%/kh degradation with a minimum of 500 h testing. Validate performance for a large-format cell of >40 cm^2. (LBNL) **p-SOEC:** Develop a robust, energy-efficient, and reliable electrolyte, for proton-conducting SOEC (p-SOEC) at 500°–600°C, that has proton conductivity >0.02 S/cm^2 at 600°C, by understanding the proton conduction and electronic leakage mechanisms. Achieve >95% Faradaic efficiency when steam concentration is >50% at 600°C or lower (>1 A/cm^2 at 1.3 V) tested for a minimum of 500 hours. (INL)
PEC	Tens of hours of durability is common for high efficiency PEC systems. For short-circuit demonstrations in neutral pH at STH >10%, a photocathode has operated for 200 h in pH 7 without a membrane separator—setting a component baseline—and a photoanode for 110 h in pH 9.3 with a bipolar membrane is considered the system baseline.	Demonstrate a complete device assembly and neutral pH conditions that achieve >220 h durability at short circuit and >5% STH efficiency (system). Scale-up to a hand-held >4 cm^2 active area device that will split water under direct sunlight illumination with appreciable bubble generation. (NREL, LBNL, LLNL)

HydroGEN 2.0 End of Project Milestone (in 3 years)

AWS	State of the Art	End of Project Milestone (in 3 years)
STCH	Materials evaluation protocols are largely absent as is a rigorous assessment of the potential for the known pool of candidate materials to meet DOE STCH technology performance targets such as \$2/kg H_2, 26% solar to hydrogen (STH) energy conversion ratio, and 2.1E-06 kg/s per m^2 1-sun hydrogen production rate. Materials that efficiently and cost effectively produce H_2 remain elusive because increasing capacity ($\Delta\delta$ at lower T_{RED}) and reaction yield (oxidation @ minimum H_2O:H_2 ratio) in non-stoichiometric oxides has not been demonstrated. These properties are anti-correlated and deeply rooted in thermodynamic behavior dictated by a material's composition and electronic structure.	Use the technology assessment methodology derived during the course of this project, and baselined or "calibrated" against the CeO_2 cycle, to evaluate material viability. The new metric natively incorporates material thermodynamic behavior (such as reduction enthalpy, reduction entropy, H_2 production capacity ($\Delta\delta$), and H_2 yield) weighted by pertinent factors that impact solar-to-hydrogen conversion efficiency. The metric accounts for material-specific cycle dynamics and plant operational modality. A selected group of known and new materials (i.e., best materials produced by HydroGEN seedling projects) will be evaluated for their potential to meet DOE STCH technology performance targets. Demonstrate theory-guided design of materials using machine learning (ML) by establishing the correlations between thermochemical properties and the underlying structure/composition features for a large number (>1,000) of compositions and structures. Identify and validate materials that optimize the capacity/yield tradeoff via high reduction entropy mechanisms.