



HydroGEN: High-Temperature Electrolysis

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Project ID # P148B

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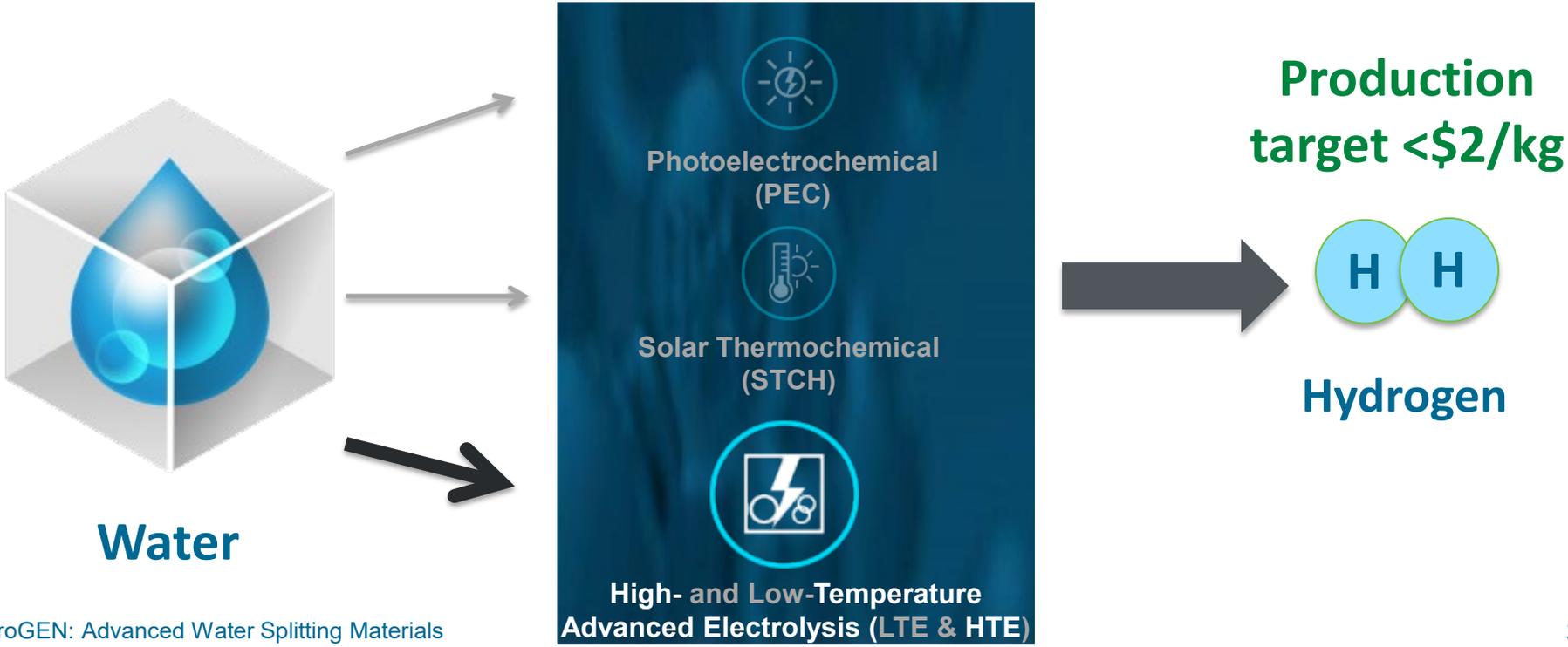


Advanced Water-Splitting Materials (AWSM) Relevance, Overall Objective, and Impact

AWSM Consortium 6 Core Labs:



Accelerating R&D of innovative materials critical to advanced water splitting technologies for clean, sustainable & low cost H₂ production, including:

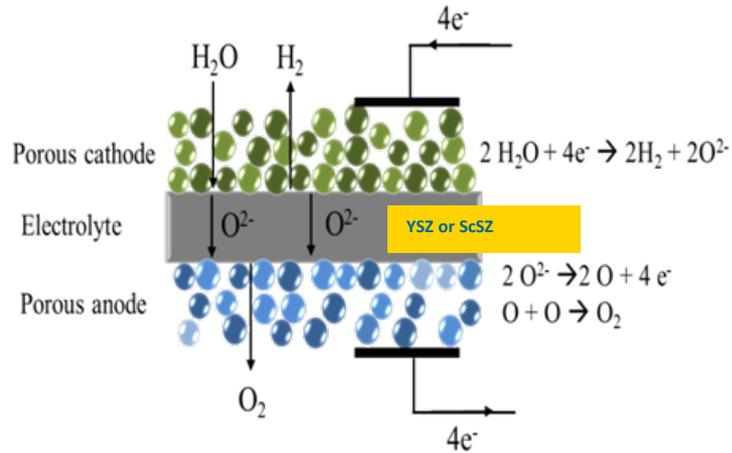




Overview – HTE Technology

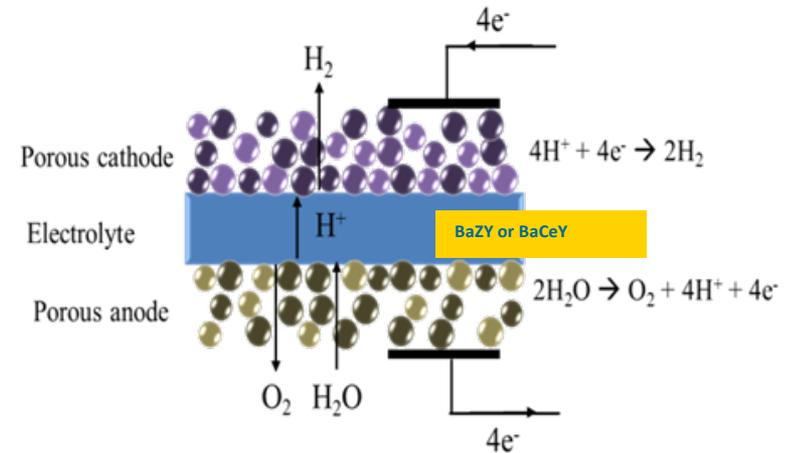
Oxygen Ion Transport Solid-Oxide Electrolysis*

(O^{2-} -SOEC; Unresolved R&D Material Barriers Remain)



Proton-Conducting Solid-Oxide Electrolysis*

(H^+ -SOEC; Early-Stage Research Needed)



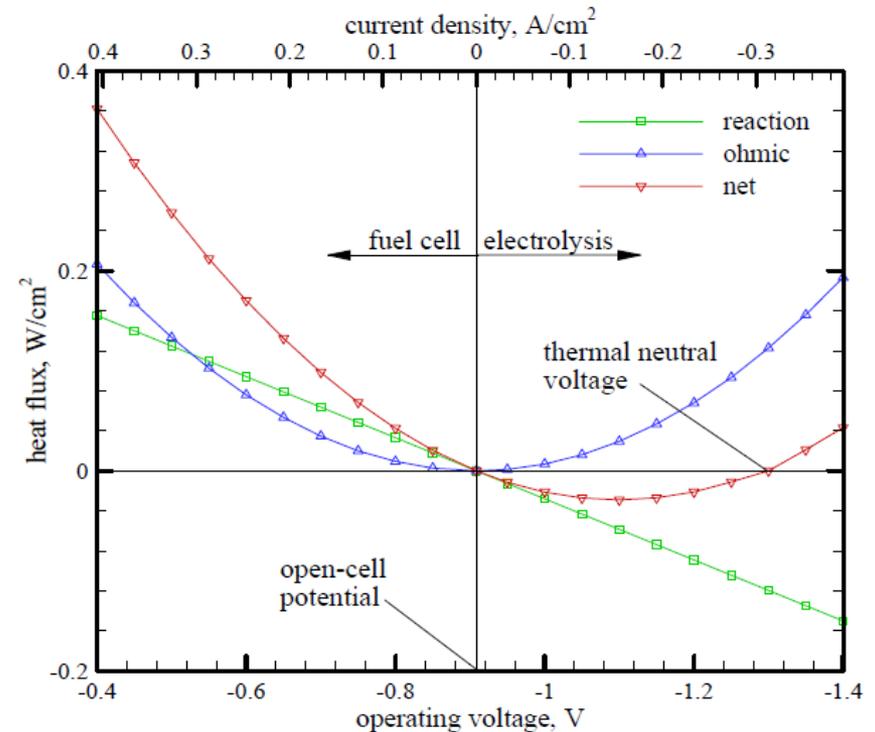
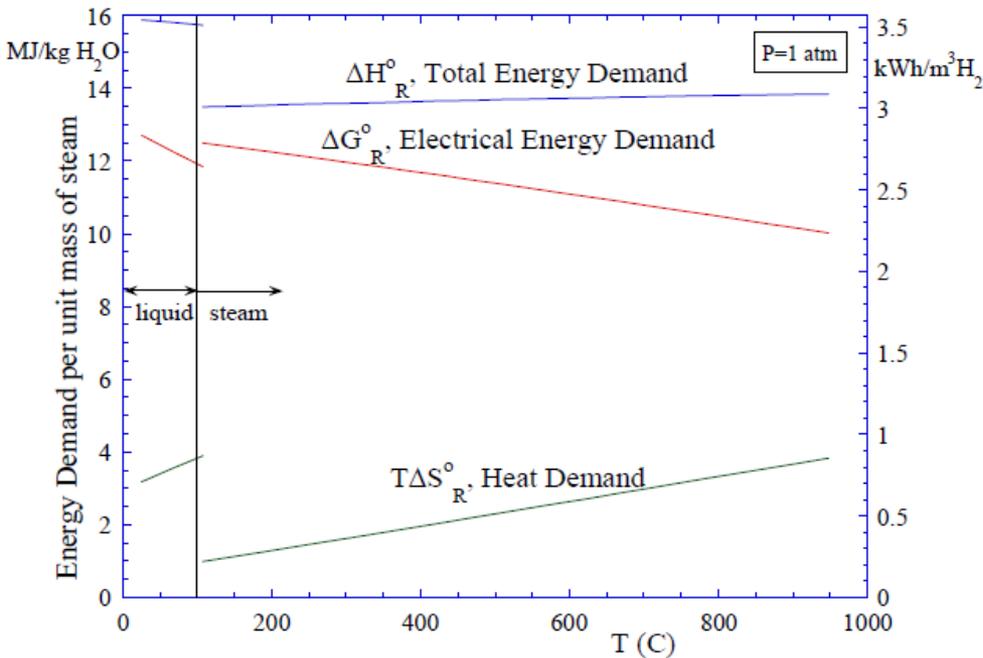
* Figure and Table Adapted from: Singh and Hu, UConn

Attributes	H^+ - SOEC	O^{2-} - SOEC
Operating Temperature	550-750°C	650-850°C
Electrolyte Conductivity	0.01 S.cm ⁻¹ at 650°C	0.015 S.cm ⁻¹ at 850°C
Cathode Products	Pure H ₂	H ₂ O + H ₂
Anode Products	O ₂ + sweep gas	H ₂ O + O ₂

- Thermodynamic efficiency and solid oxide conduction and kinetics increase at higher temperature, but materials durability decreases due to progressive micro-structure evolution, thermal stresses, and chromium migration
- Solid oxide is purely proton conducting < 600°C, but kinetics are slower
- Electrode and electrolyte materials synthesis, densification, and enhancement for proton-conduction efficiency is in progress



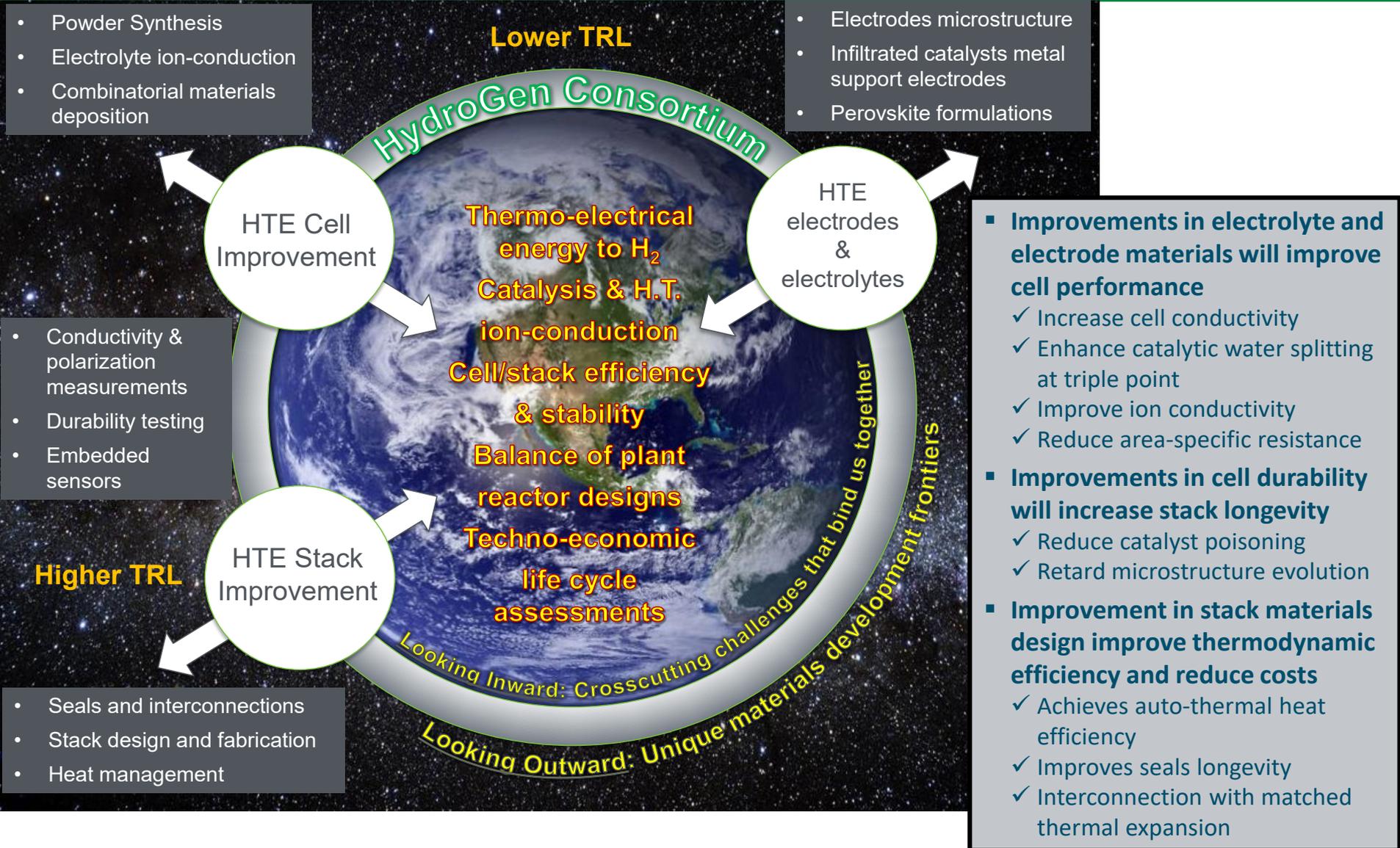
Advantages/Disadvantages of HTE



- 30-50% higher thermodynamic efficiency is possible for steam compared to water splitting (combined free energy and electricity use)
- Reversible operation is possible with optimal design of cells, stacks and modules
- Does not require highly precious metals
- Major concerns are rapid cell degradation (sintering, pore consolidation, Cr migration/poisoning, catalyst deactivation (Ni hydridation), delamination)

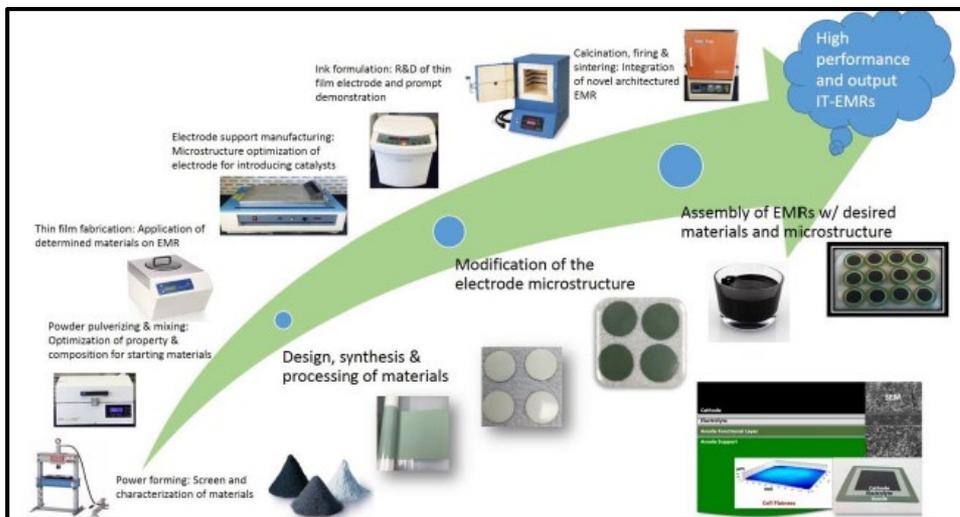


Overview - HTE Technology Relevance / Impact

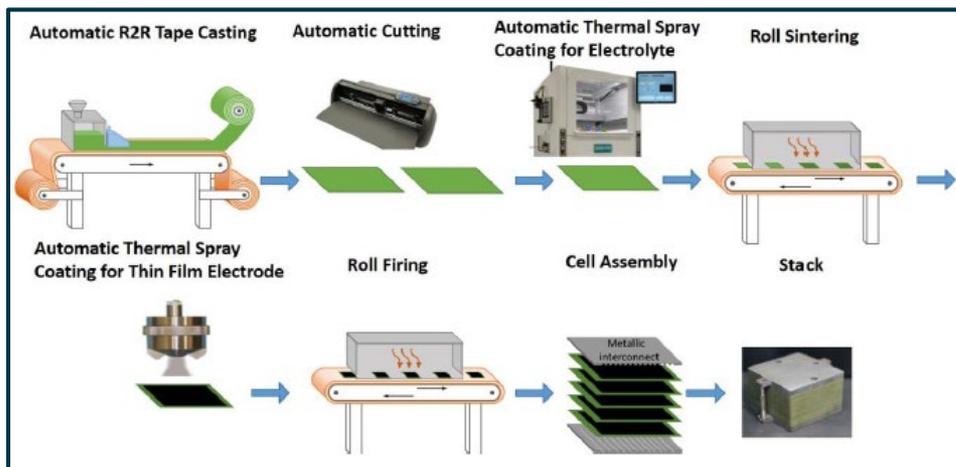




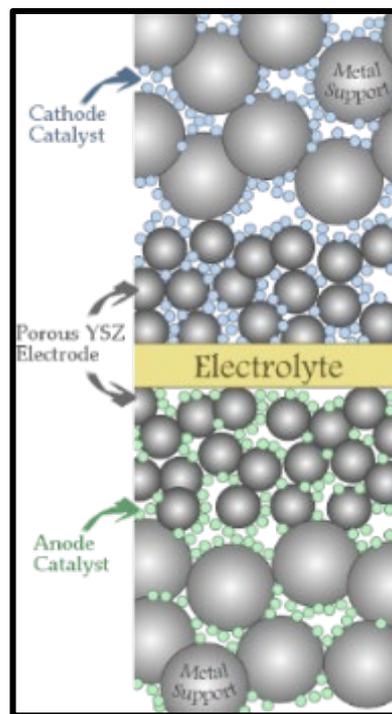
Overview - HTE Technology Relevance / Impact



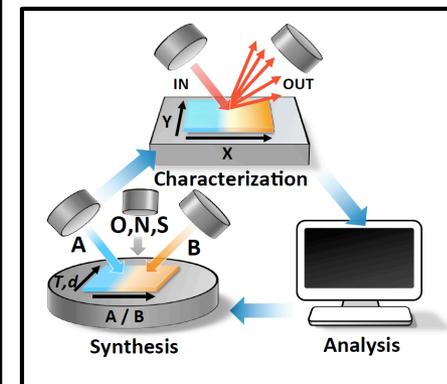
- Advanced HTE Electrode and Electrolyte Materials
- Fabricating cells for stack assembly
- Button Cell Fabrication and testing
- Metal-supported cells reduce cost while managing thermal transients and reducing fabrication steps
- Thermal Spray and Thin-Film Synthesis reduce defects and improve performance



INL Node for Process for Button Cell Fabrication (Upper)
 INL Node for Super Proton-Conducting cells (Lower)



LBL Node for Metal-Supported SOEC and Coatings (Left)



NREL Node for Thin Film Coatings and Combinatorial Testing (above)



Approach – HydroGEN EMN

DOE

EMN

HydroGEN

Core labs
capability
nodes

Data Hub

FOA Proposal
Process

- Proposal calls out capability nodes
- Awarded projects get access to nodes

<https://www.h2awsm.org/capabilities>



Approach: HTE Projects & Collaboration

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

HTE Node Labs



Supernode

Support through:



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

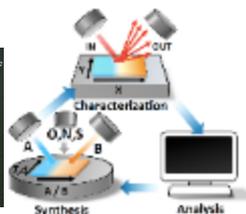
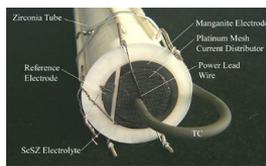
HTE Projects



Northwestern University



University of Connecticut





Accomplishments and Progress: Established Nodes for Project Support

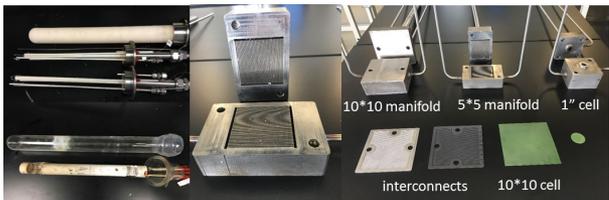
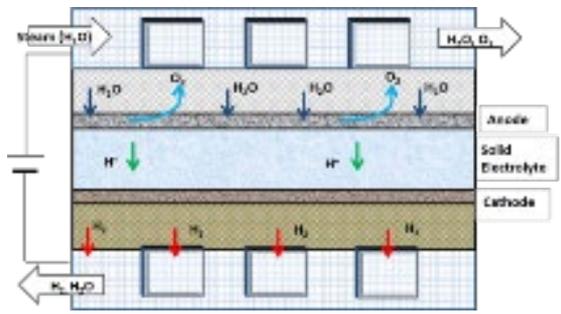
44 nodes for HTE

- 8x readiness level 1
- 24x readiness level 2
- 11x readiness level 3

**7 nodes used by
current HTE projects**

Node Classification

- 6x Analysis
- 6x Benchmarking
- 20x Characterization
- 13x Computation
- 6x Material Synthesis
- 5x Process and Manufacturing Scale-Up
- 5x System Integration

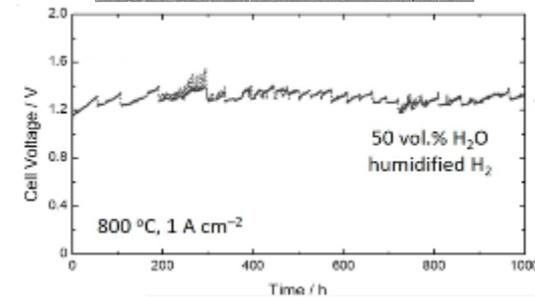
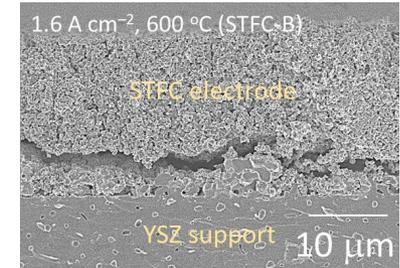




Goal and approach



- Degradation mechanisms in solid oxide electrolysis cells (SOECs) are studied using accelerated life testing with varying conditions, materials, and cell designs
- Theory used with experimental data to develop a basic understanding of degradation mechanisms
- Development of improved SOECs that provide stable long term operation at high current density



Accomplishments in BP1

- Oxygen electrode overpotentials > 0.19 V shown cause cell fracture
- Theory predicts where internal oxygen pressure exceeds fracture strength solid oxide cells providing stable electrolysis operation at 1 A cm^{-2} have been demonstrated
- Novel metal-supported cells have been demonstrated for electrolysis

Focus of BP2

- Advanced solid oxide electrolysis cells will be life tested with electrochemical and microstructural characterization
- Achieving improved long-term stability
- Electrode and electrolyte degradation theory will be further developed by comparison with experiment
- Refine optimized cell designs for BP3



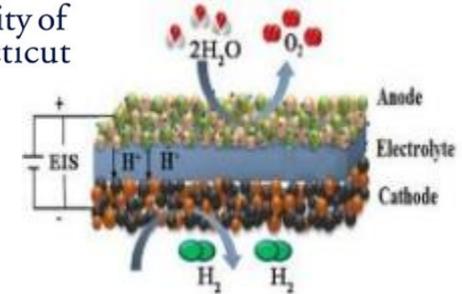
Accomplishments and Progress: University of Connecticut: Proton-Conducting Solid Oxide Electrolysis Cells for Large-scale Hydrogen Production at Intermediate Temperatures

P152

PI: Prabhakar Singh
Co-PI: Boxun Hu
Partner: PNNL



University of Connecticut



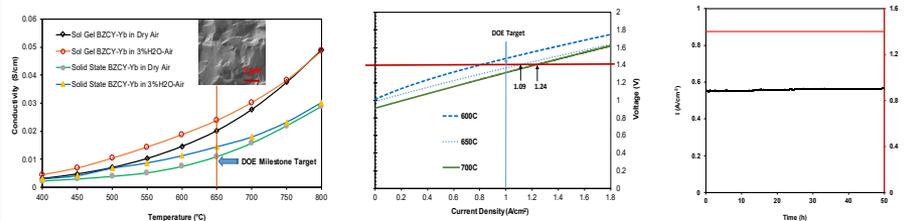
Project Goals: Identify novel materials and processing techniques to develop cost effective and efficient proton-conducting solid oxide electrolysis cells (H-SOECs) for large-scale hydrogen production at intermediate temperatures (600-800°C) to meet DOE cost (< \$2/gge H₂) and performance targets (degradation rate <4 mV/1000 h at 1A/cm²), and electrical efficiency >95%.

Approaches:

- Develop electrolyte formulations capable of densification (96-98%) below 1400°C;
- Utilize tape cast multi-layer laminated electrolyte (10-20 μM) and electrode;
- Optimize cell materials for densification, proton conductivity and structural stability;
- Develop electrode poisoning and performance degradation mitigation approaches;
- Utilize EMN core experimental and computational capabilities (INL & NREL).

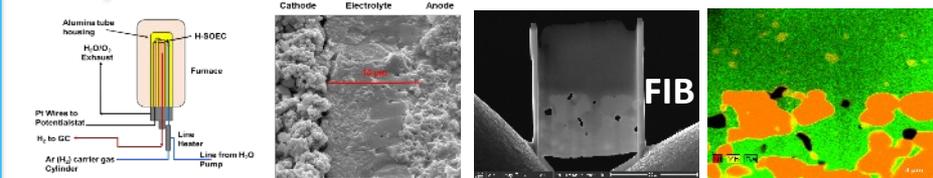
Accomplishments in BP1

- Sintered electrolyte at ≤1400°C.
- Obtained electrical conductivity of 0.01 S.cm⁻¹ at 650°C.
- Demonstrated cell performance of 1 A/cm² at 1.4 V and 650°C.
- Achieved 50 hours cell performance stability.



Focus of BP2

- Develop 50-gram batch proton-conducting electrolyte with conductivity of at least 0.02 S.cm⁻¹ at 650°C.
- Develop and test 1-inch diameter cells with stable electrolysis performance (<25 mV/1000 h) for at least 200 hours.
- Obtain current density above 1 A/cm² at ≤ 1.4 V and temperature of ≤ 650°C.





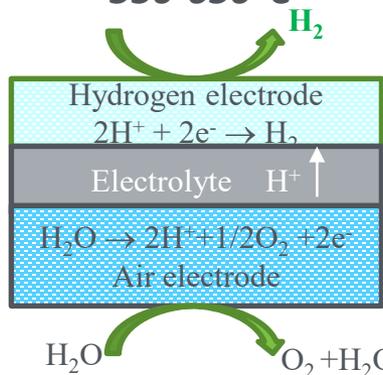
Accomplishments and Progress: United Technology Research Center Thin-Film, Metal-Supported High-Performance and Durable Proton-Solid Oxide Electrolyzer Cell

P154

Concept

Proton conducting electrolyzer

550-650 °C



Approach

Integrate low cost fabrication, material optimization and modeling

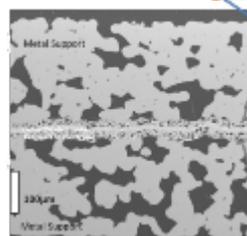
Target performance >1.0 A/cm² at 1.4 V and ≤650 °C

Plasma Spray



UTRC

Co-sintering



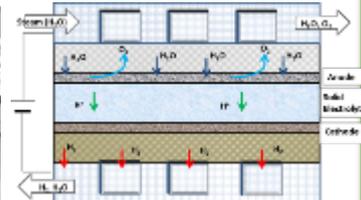
LBNL

Material optimization



INL

p-SOEC cell modeling

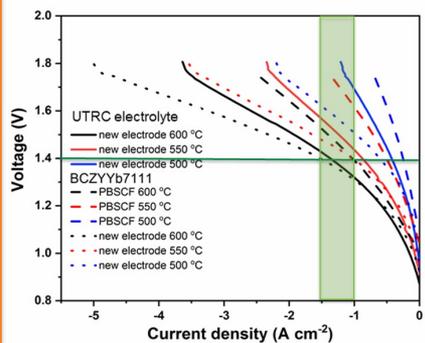


NREL

BP1 Main Accomplishments

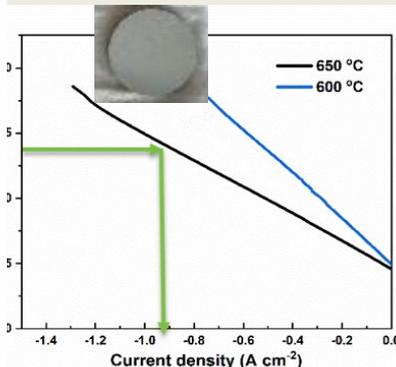
High performance material identified for p-SOEC

(>1.0 A/cm² at 1.4V & <650°C)

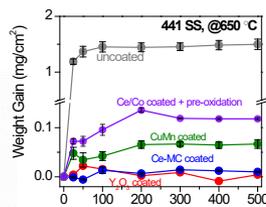


Button cell with SPS electrolyte exceeded BP1 target

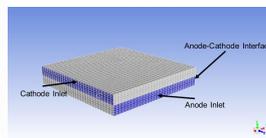
(>0.8 A/cm² at 1.4V & 650°C)



Protective coatings for SS alloys, 500h no oxidation



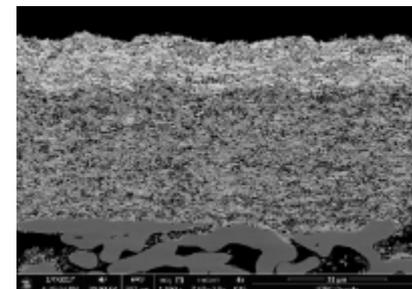
Establish cell model



BP2 Focus

- Metal cell fabrication by plasma spray
- Material optimization and durability
- Cell performance modeling

Half cell on metal support by plasma spray





Accomplishments and Progress: West Virginia University (New project, in BP 1)

Goal: IT H-SOECs for simultaneous H₂O splitting and H₂ separation with high current densities > 1.0 A/cm² at 1.4 V/cell while operating at ~700 °C (BP1).

Approach:

Electrode and full cell @ WVU

- Modeling driving
 - H₂O-splitting reaction kinetics
 - Anode structural and composition
- Candidate anodes development
- Conformal catalyst layer coating
- Cell fabrication and performance measurements

Electrocatalyst @ CSM&NREL

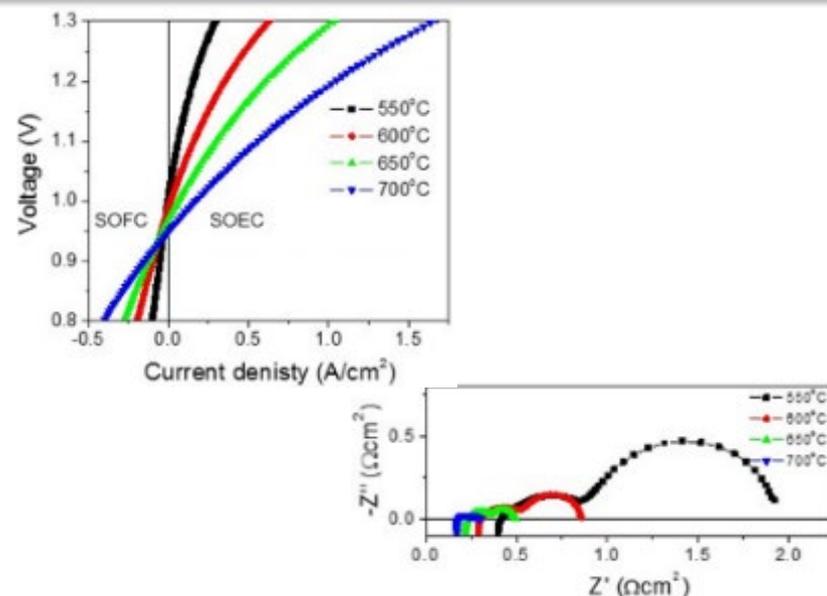
- Appropriate electrocatalyst compositions
 - High-throughput screening
- Catalysis & local surface activity
 - Operando ambient-pressure XPS

H-electrolyte @ INL:

- Advanced powder synthesis techniques
 - sol-gel and nitrate-combustion
- Post analysis
 - SEM/EDX, TEM and XPS
- Conductivity improvements

Accomplishment of BP1

- Established framework of H₂O-splitting reaction kinetics modeling
- Development of highly performing triple conducting anode, ~1A/cm² at 1.3V 700°C
- Initial testing of reference samples on Scienta-Omicron HIPP Lab System at Mines
- Reference samples are being characterized with E-XPS, high throughput screen of anode and catalyst has started
- Initiated study on improving conductivity, stability and sinterability of BCZYYb system by adjusting Zr/Ce ratio in INL





Accomplishments and Progress: Saint-Gobain (New project, in BP 1)

Project Goal

Develop oxygen electrode materials with high performance which solve the issue of electrode delamination in order to produce hydrogen below the DOE target of \$1.87/kg and to test this electrode in a stack platform that has shown degradation rates <0.2%/1000 hrs in SOFC mode.

Approach

Novel chemistries of nickelate-based materials showing **enhanced oxygen hyperstoichiometry** will be developed. These materials will be co-sintered in button cells and stacks to be tested for performance and durability.

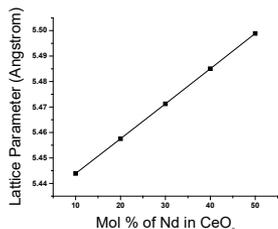
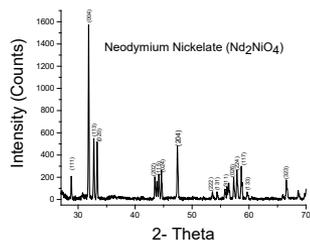
Barriers to be overcome

- | | | |
|--|---|---|
| 1) Phase stability/performance:
Identification of stability boundaries with target electrochemical properties | 2) Co-sintering with balance of cell:
Incorporate materials with stacks ensuring porosity & activity | 3) Accelerated testing:
Protocol development to probe dominant degradation mechanism |
|--|---|---|

BP1: 10/18 – 9/19

Saint-Gobain, Boston University, PNNL & INL refined testing protocol, project plan, and responsibilities

- Neodymium nickelate (NNO) and neodymium doped ceria (NDC) chosen as 1st material set to produce
- Synthesis procedures developed resulting in phase purity
- Phase pure NNO prepared
- Lattice parameter shift correlates very well with degree of Nd doping in CeO₂



Remainder of BP1: 3/19 – 9/19

- Complete synthesis of remaining target compositions
- Utilize high-temp XRD & TGA @ SNL to quantify oxygen hyperstoichiometry
- Quantify phase stability of target material systems
- Prepare button cells of baseline and new compositions
- Test baseline SOEC performance and degradation
- Down-select material compositions based on lab tests
- Test top performers in SOEC mode to show enhanced performance and durability
- Utilize microstructural analysis including CT scanning to determine degradation mode

BP2 10/19 – 9/20 Proposed Plan

- Scale testing from button cells to stacks
- Optimize stoichiometry of high performing material

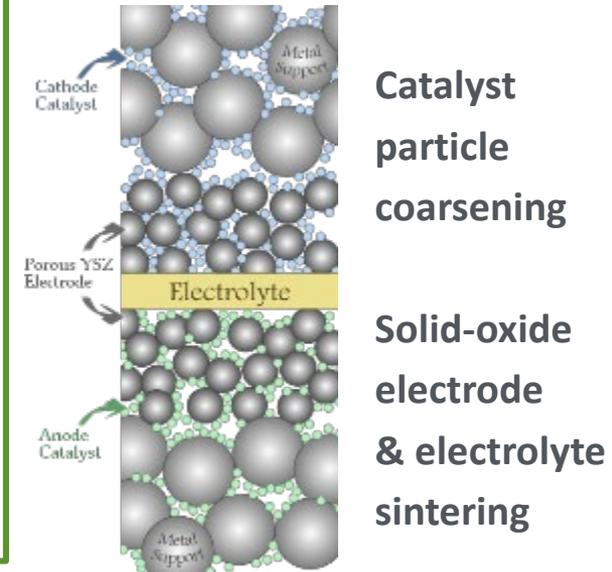


HTE Supernode : Characterization of Solid Oxide Electrode Microstructure Evolution

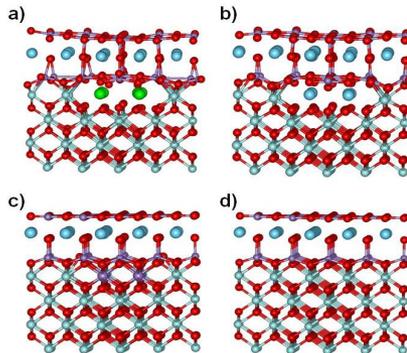
Challenge:

Degradation mechanisms in solid oxide electrolysis cells (SOECs), which are poorly understood at present, will be studied using accelerated testing at high current density, closely coupled with theory.

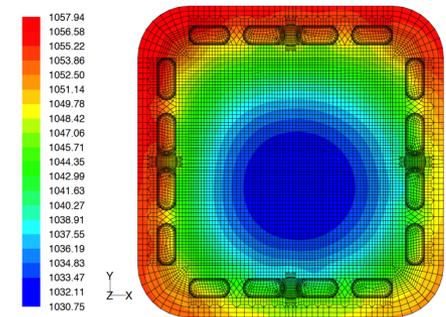
A deeper understanding of high temperature electrolysis (HTE) electrode microstructure evolution as a function of local solid-oxide composition and operating conditions is needed to develop more active, longer-life electrodes.



Electrical currents and ion forces lead to inter-crystalline structure forces



High temperatures and thermal gradients cause non-uniform grain boundary stresses



Goal: Combine HTE nodes to establish achieve a deep understanding of high temperature electrolysis (HTE) electrode microstructure evolution as a function of local solid-oxide composition and operating conditions.

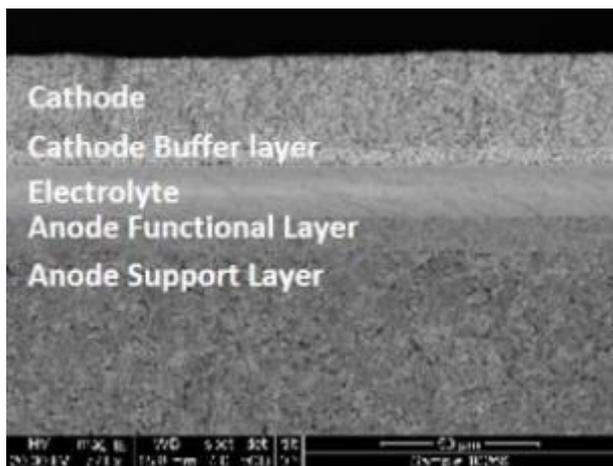


HTE Supernode : Characterization of Solid Oxide Electrode Microstructure Evolution

Accomplishments

- ✓ Supernode team established; R&D plan has been initiated
- ✓ First batch of YSZ-based SOEC fabricated using high-purity precursors.
- ✓ Sintering aid used in cathode buffer layers.
- ✓ Electrolyte thickness: 10-15 μm .

**7 nodes combined:
INL, NREL, LBNL, LLNL, Sandia**

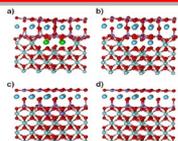




Collaboration: HTE Node Utilization

Lab	Node	Node PI	NWU	UConn	UTRC	Super-Node
NREL	Multi-Scale Thermochemical and Electrochemical Modeling for Material Scale-Up to Component and System Design	Ma, Zhiwen Martinek, Janna		✓	✓	
LBNL	Metal-Supported SOEC Cell	Tucker, Michael Wang, Ruofan	✓		✓	
INL	Advanced Materials for Elevated Temperature Water Electrolysis	Ding, Dong		✓	✓	✓
NREL	Controlled Materials Synthesis and Defect Engineering	Ginley, David Parilla, Philip Bell, Robert				✓
NREL	High-Throughput Experimental Thin Film Combinatorial Capabilities	Zakutayev, Andriy		✓		
INL	SOEC Characterization	O'Brien, James	✓			✓
NREL	Secondary Ion Mass Spectrometry (SIMS)	Harvey, Steven	✓			
SNL	Advanced Electron Microscopy	Sugar, Josh				✓
LLNL	Multi-Scale Modeling of Solid-Sate Interfaces and Microstructures in High-Temperature Water Splitting Materials	Wook, Tase Wood, Brandon Frolov, Timofey				✓

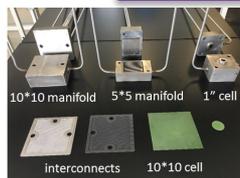
Computation



Material Synthesis



Characterization

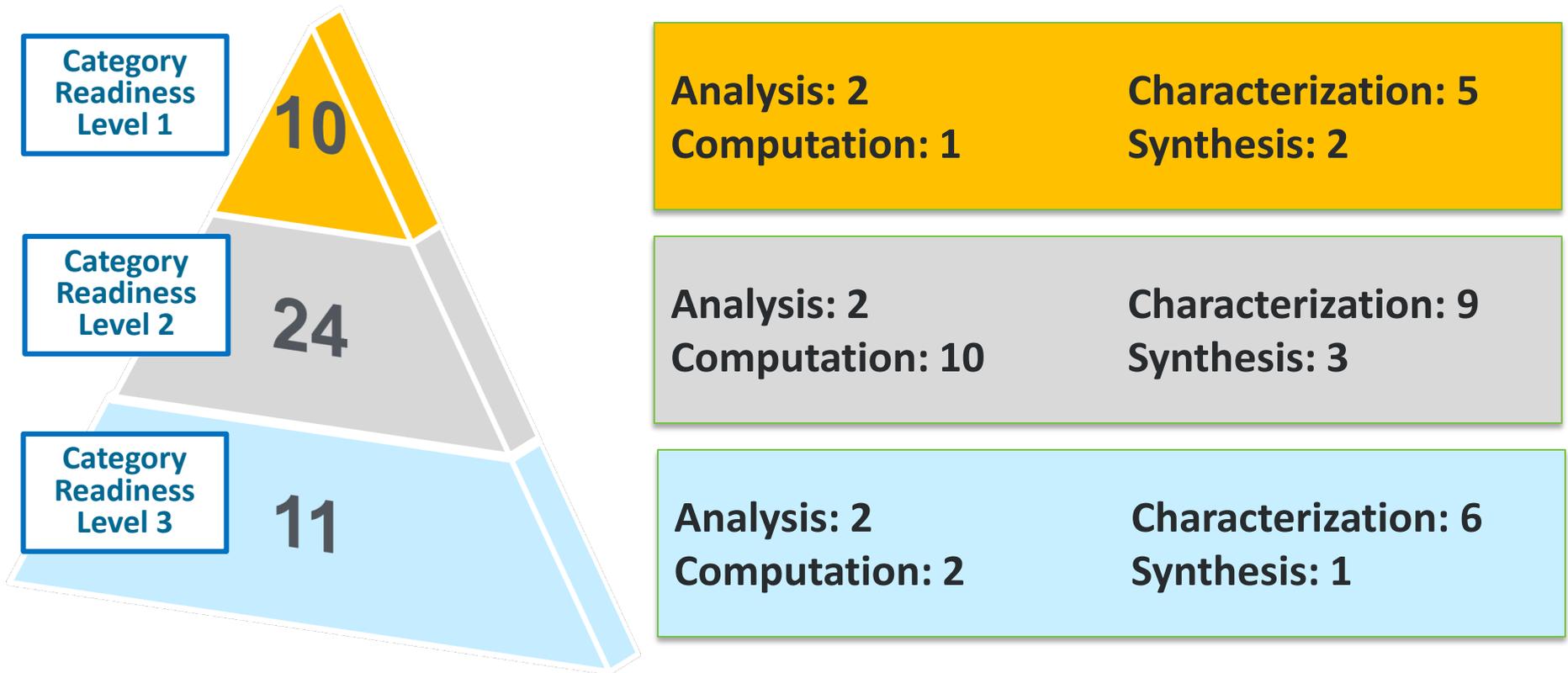


Analysis





Collaboration: 44 HTE Nodes



- Nodes comprise equipment and expertise including uniqueness
- Category refers to availability and readiness and not necessarily the expense and time commitment
- Note that many nodes span classification areas



Data Hub & Engagement with 2B Benchmarking Team (Olga Marina, PNNL – HTE Lead)

BENCHMARKING

- **Ongoing assessment of the EMN Node capabilities**
- **Collaborated with HydroGEN Benchmarking Project**
- **Leadership role in developing path forward for harmonized protocols, hardware, and materials**
- **Benchmarking working groups and annual meeting**
- **Disseminated benchmarking information**

DATA HUB

- **HTE data metadata definitions in development**
- **Several datasets uploaded to hub**



Summary - HydroGEN LTE Projects

- **HydroGEN HTE is:**
 - Supporting 5 FOA projects with 9 nodes
 - Leading a Supernode with 6 nodes
- **Projects demonstrate improvements in Proton-Conducting and Oxygen Ion Transport Electrodes / Electrolytes**
- **Working closely with the project participants and benchmarking activities to advance knowledge and utilize capabilities**



Future Work

- **Continue to enable and support research of Phase 2 Projects through lab nodes and expertise**
- **Enable new FOA awarded seedling projects (Fall 2019)**
- **Continue to develop HTE supernode to attain deeper understanding of microstructure functionality and evolution**
- **Work with the 2B team and HTE working group to establish testing protocols and benchmarks**
- **Utilize data hub for increased communication, collaboration, generalized learnings, and making digital data public**

Acknowledgements



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HydroGEN
Advanced Water Splitting Materials

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Research Teams



**United Technologies
Research Center**



University of
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SAINT-GOBAIN

 West Virginia
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Pacific Northwest
NATIONAL LABORATORY



Northwestern
University



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HydroGEN
Advanced Water Splitting Materials

HTE Supernode Team - 18 experts

