Energy Materials Network
U.S. Department of Energy

HydroGEN
Advanced Water Splitting Materials

HydroGEN: High-Temperature Electrolysis Supernode

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Presenter: Gary Groenewold, INL

Date: 5/13/2020

Venue: 2020 DOE Annual Merit Review

Project ID # P148B

NREL | BERKELEY LAB | Sandia National Laboratories | INL Idaho National Laboratory | Lawrence Livermore National Laboratory | SRNL

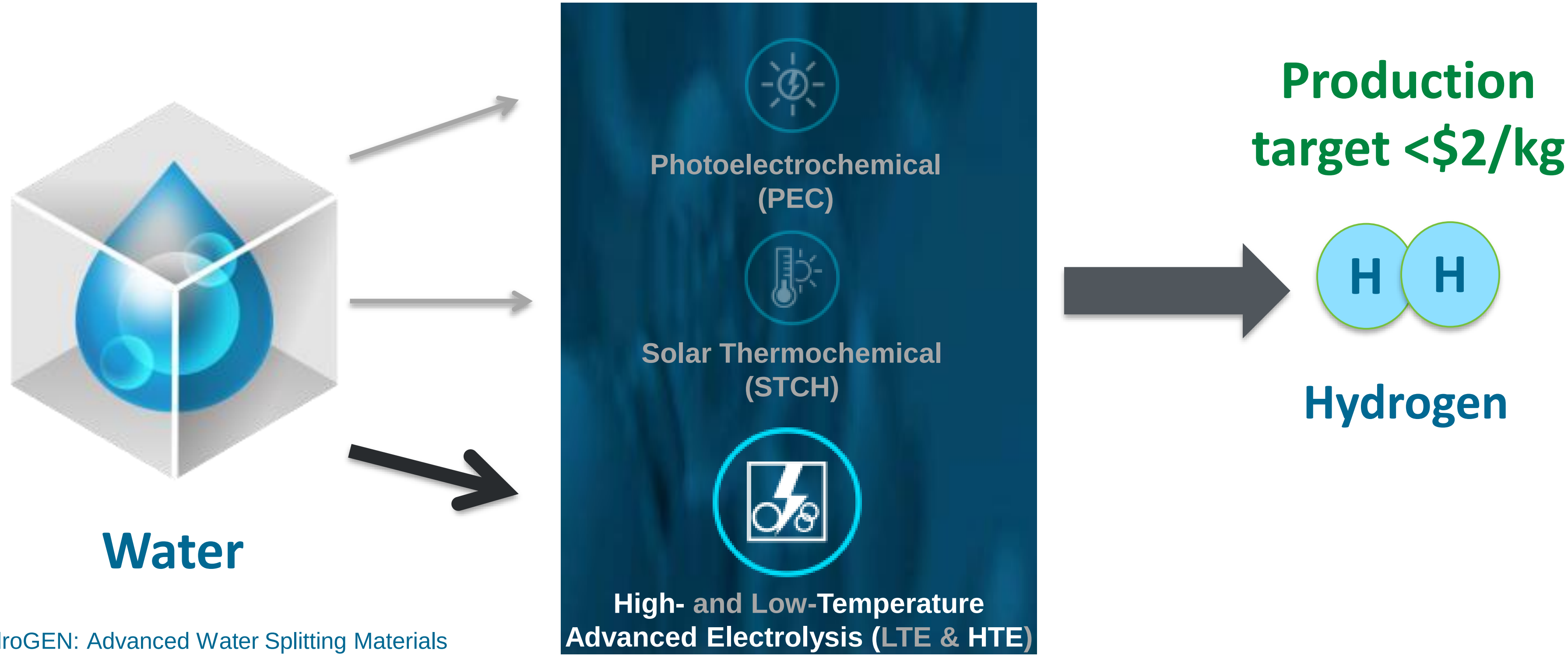
Advanced Water-Splitting Materials (AWSM)

Relevance, Overall Objective, and Impact

AWSM Consortium 6 Core Labs:

NREL (National Renewable Energy Laboratory) · Berkeley Lab · Sandia National Laboratories · INL (Idaho National Laboratory) · Lawrence Livermore National Laboratory · SRNL

<u>Accelerating R&D</u> of innovative materials critical to advanced water splitting technologies for clean, sustainable & low cost H_2 production, including:



Approach: HTE Projects & Collaboration

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

HTE Node Labs

INL Idaho National Laboratory

Sandia National Laboratories

NREL National Renewable Energy Laboratory

Berkeley Lab

Lawrence Livermore National Laboratory

Supernode

Support through:

Personnel

Equipment

Expertise

Capability

Materials

Data

HTE Projects

United Technologies Research Center

Northwestern University

REDOX Redox Power Systems, LLC

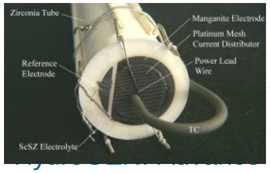
West Virginia University

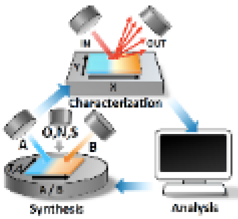
NEXCERIS where energy meets environment

Saint-Gobain

University of Connecticut

University of South Carolina





Accomplishments and Progress:
Established Nodes for Project Support

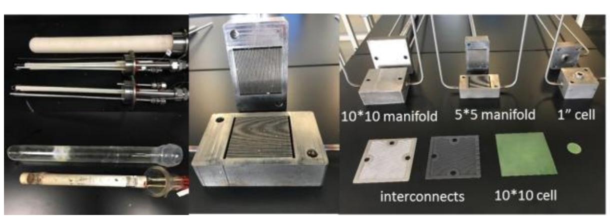
Nodes for HTE

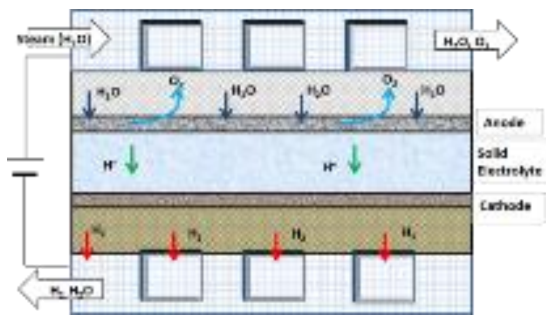
- 9 @ readiness level 1
- 22 @ readiness level 2
- 9 @ readiness level 3

10 nodes used by current HTE projects

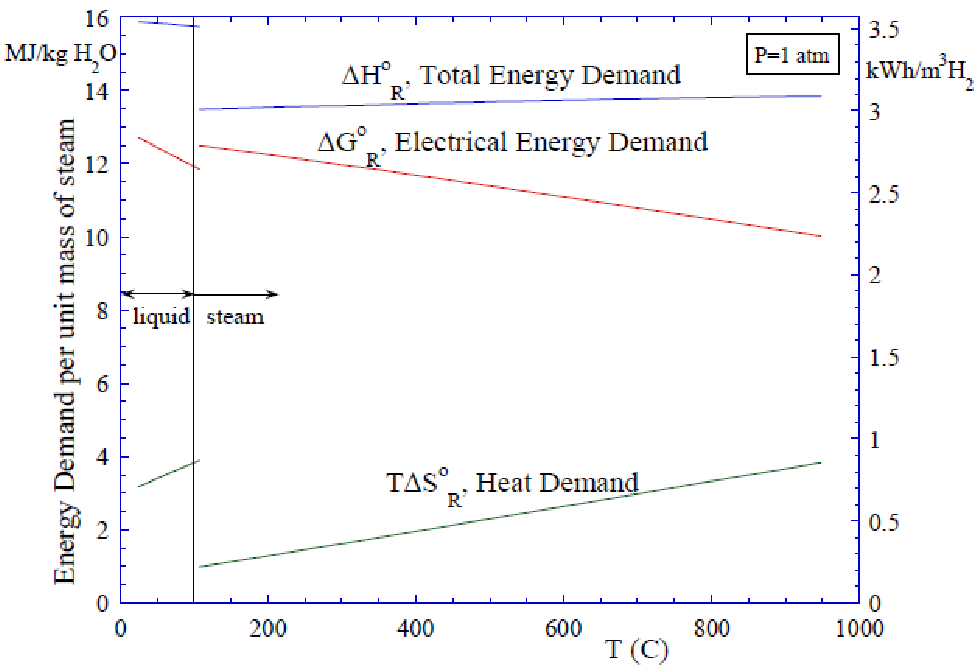
Node Classification

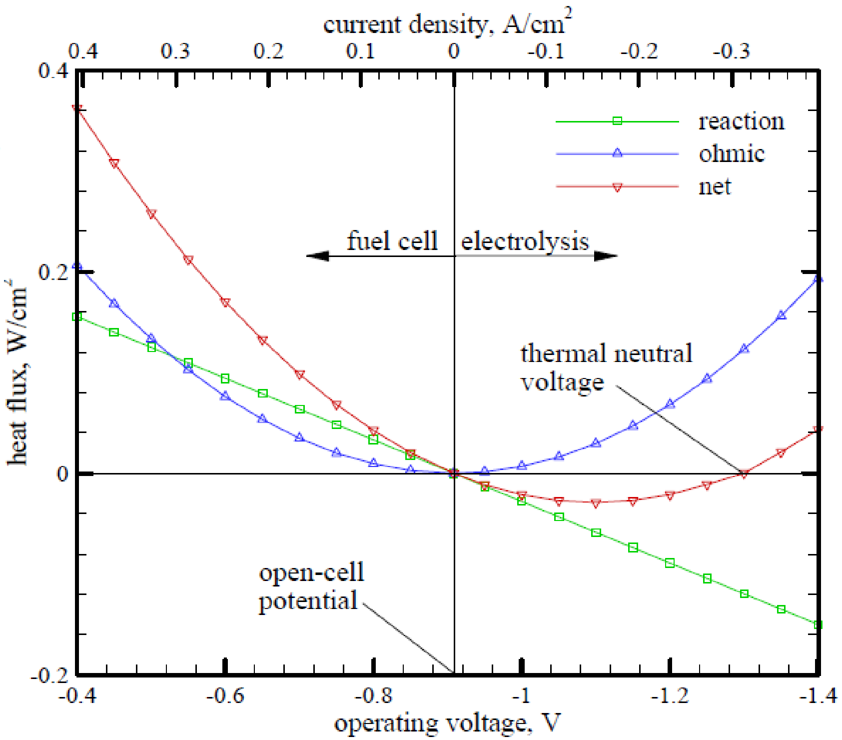
6x Analysis
6x Benchmarking
20x Characterization
13x Computation
6x Material Synthesis
5x Process and Manufacturing Scale-Up
5x System Integration





Overview: Advantages/Disadvantages of HTE



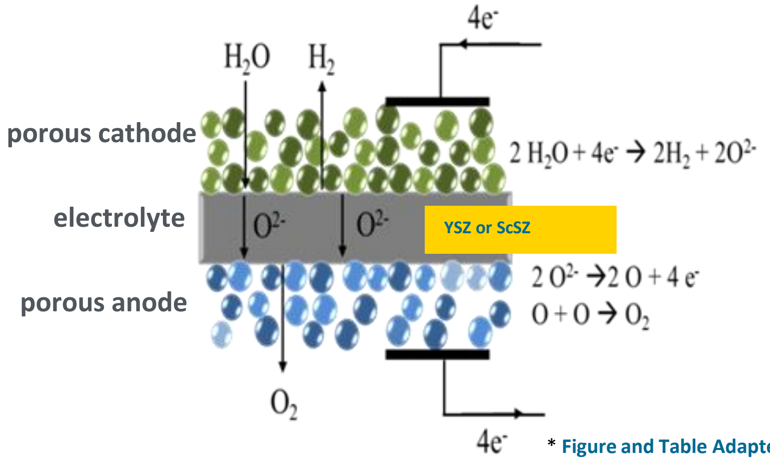


- **30-50% higher thermodynamic efficiency is possible for steam compared to water splitting (combined free energy and electricity use)**
- **Reversible operation is possible with optimal design of cells, stacks and modules**
- **Does not require highly precious metals**
- **Concerns: cell degradation,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

Overview – HTE Technology, o-SOEC, p-SOEC

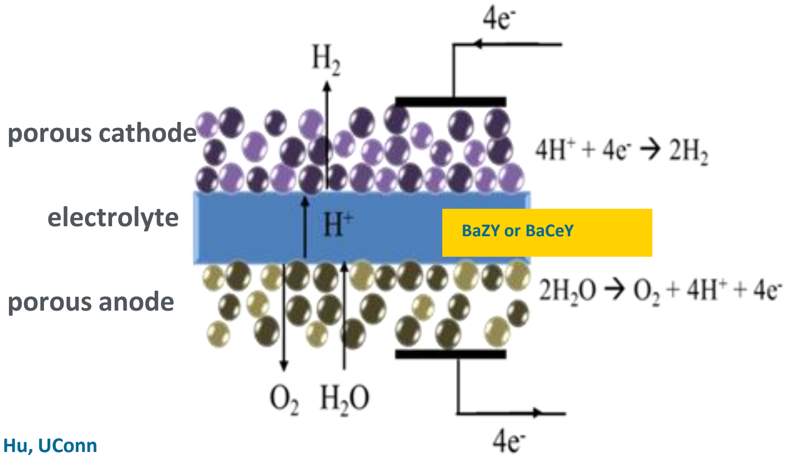
Oxygen Ion Transport Solid-Oxide Electrolysis*

(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

O^{2-}- SOEC	Attributes	H^+- SOEC
650-850°C	Operating Temperature	550-750°C
0.015 $S.cm^{-1}$ at 850°C	Electrolyte Conductivity	0.01 $S.cm^{-1}$ at 650°C
$H_2O + H_2$	Cathode Products	Pure H_2
$H_2O + O_2$	Anode Products	O_2 + sweep gas
durability decreases: microstructure evolution, Δ stresses, Cr migration	Challenges	slower kinetics, maturation of electrolyte (synthesis, densification, H^+ conduction)

HTE Supernode is focused on attacking o-SOEC issues: elemental migration, unexpected phase formation, crack and void formation, and delamination

HTE Supernode Challenges, Composition, Timeframe, and Funding

Challenges

Durability of o-SOECs

- elemental migration
- new phase formation
- void formation
- delamination and cracking

Performance of p-SOECs

- kinetics, conductivity
- maturation of the electrolyte

Composition

- INL – materials R&D, precision cell fab, electrochemical characterization
- NREL – custom materials fabrication, synchrotron characterization at SLAC
- Sandia – microscopy, materials analysis
- LLNL – multiscale modeling
- LBNL – synchrotron characterization

Timeline

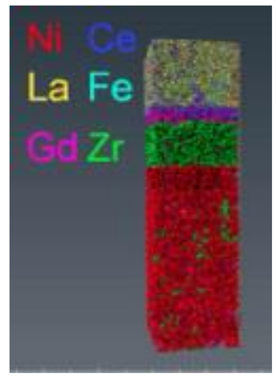
- Project start date: 2019
- Project end date: BP1 ending Sept., 2020

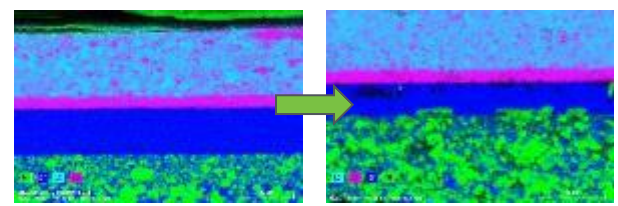
Budget

- FY20 DOE Funding: $240K
- FY20 DOE Funding: $125K
- Total DOE Funds Received to Date: $365K

HydroGEN HTE Supernode: Five Labs, Coordinated Research Capabilities

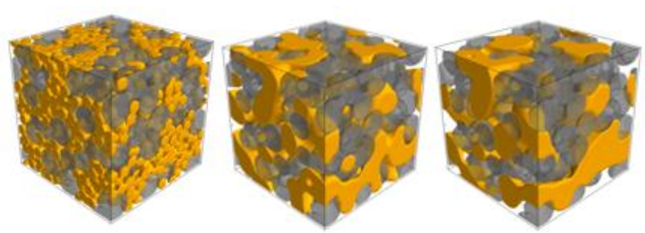
- **Coordinated R&D effort addressing o-SOEC, p-SOEC science**
- **INL**
  - Materials R&D
  - Cell Fabrication and Testing
  - Intensive Seedling Support
- **Sandia**
  - Materials micro-milling (FIB)
  - Microscopy & elemental mapping
- **NREL**
  - Custom cell design & fab
  - SLAC interface for Micro XRD, XAS, XTM
- **LLNL**
  - Multi-scale computational modeling
  - Ab initio → phase field → continuum electrochem (NWU)
- **LBNL - ALS**
  - Micro diffraction
  - Tomography





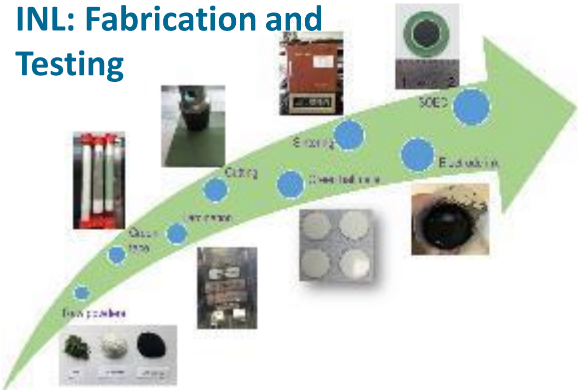
pre-operation

post-operation 750° C



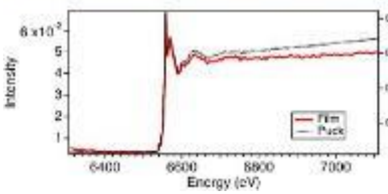
HTE Supernode Objectives – o-SOEC technology

INL: Fabrication and Testing



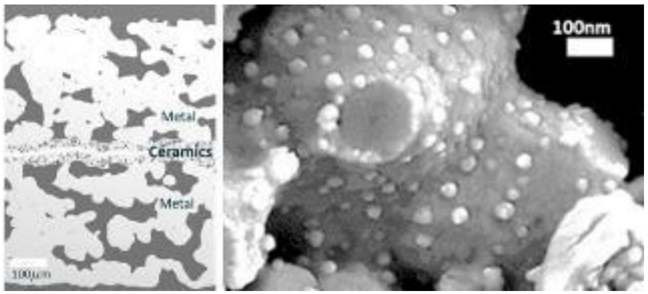
SNL: Imaging

NREL: Surface & Interface

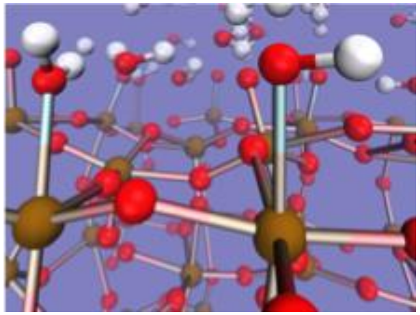


- higher efficiency, improved durability
- rationalize & predict degradation
- manipulate composition to optimize durability
- accelerate rate of technology development
- **achieve \$2/kg target for H_2 production of**

LBNL: Processing & Operando



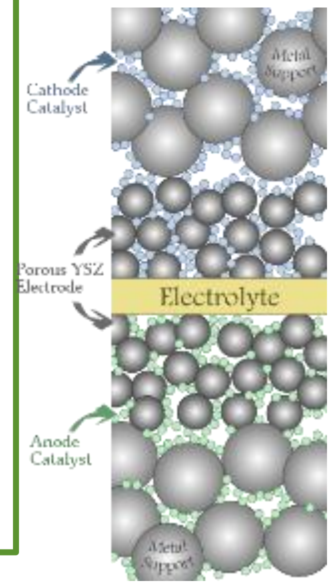
LLNL: Modeling



HTE Supernode Challenges: Characterization of Solid Oxide Electrode Microstructure Evolution

Challenges:

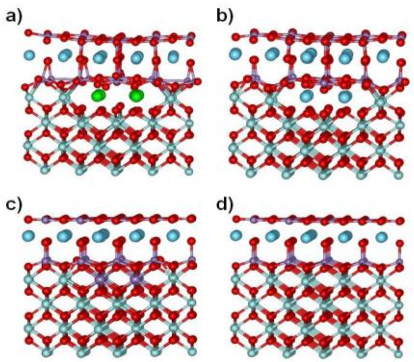
- Market acceptance of hydrogen production using high temperature electrolysis (HTE) relies on <u>cost reduction</u> and <u>durable operation</u>.
- Degradation mechanisms in oxygen-ion conducting solid oxide electrolysis cells (o-SOEC) remain elusive
  - correlation of electrode microstructure evolution to degradation under realistic operation conditions
    → not well understood



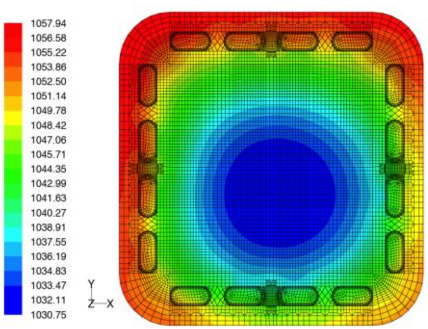
Catalyst particle coarsening

Solid-oxide electrode & electrolyte sintering

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



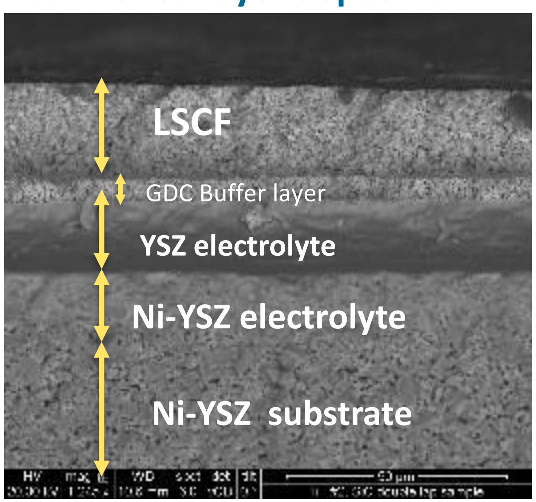
High temperatures and thermal gradients cause non-uniform grain boundary stresses



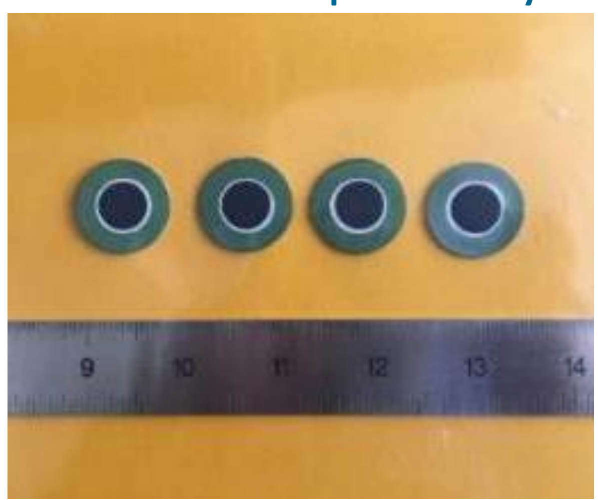
HTE Supernode Contributions: INL -- Advanced Materials for Elevated Temperature Water Electrolysis

- Capability
  - Hydrogen Lab: 30 yr. legacy of SOEC development and engineering
    - Button cell & stack testing
  - o-SOEC material research, cell fabrication
    - "button-to-large": high temp roll-to-roll, solid oxide additive manufacturing for varied cell components, configurations, electrodes
  - Electrochemical, high-throughput materials testing
- Accomplishments

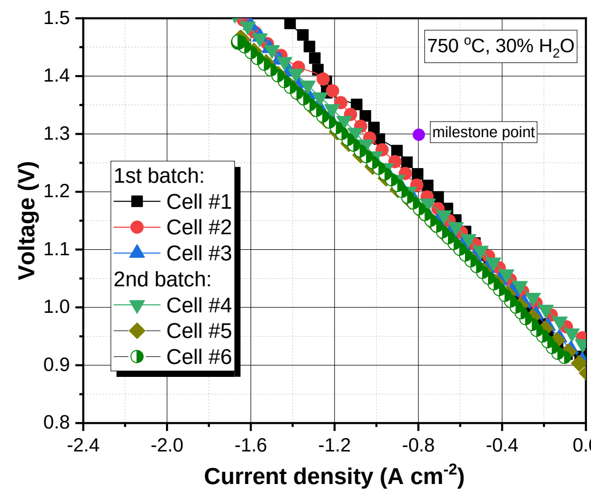
controlled layer deposition



button cell fab reproducibility



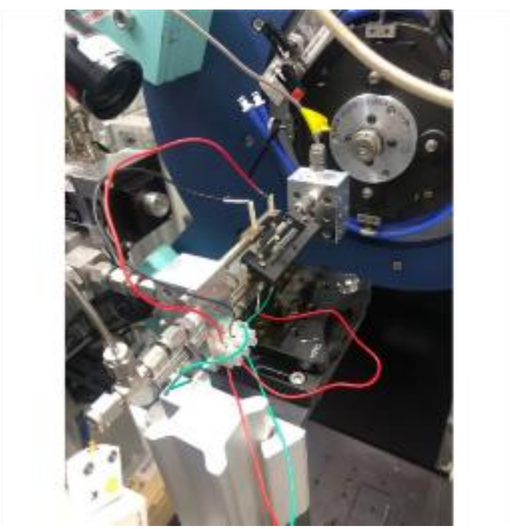
o-SOEC performance consistency

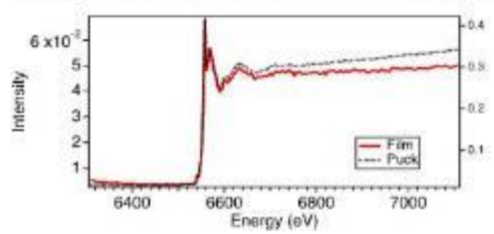


HTE Supernode Contributions: NREL -- Controlled Materials Synthesis and Defect Engineering

- Capability
  - Custom materials fabrication, targeted synthesis of representative interfaces
  - Advanced synchrotron analyses collaborating with SLAC -- ultimate goal is to use synchrotron beam-line spectroscopy (transmission or reflection mode) to resolve <u>high temperature</u> solid-oxide 3-D microstructure to depths of 20-50 µm.
  - Analyze representative HTE cell layers & interfaces with high precision at SLAC
  - Post-mortem analysis of HTE cells
    - Development of secondary phases (XRD)
    - Interdiffusion of elements (XRD, XAS, XRF)
    - Formation of voids (Tomography - TXM)
- Accomplishments

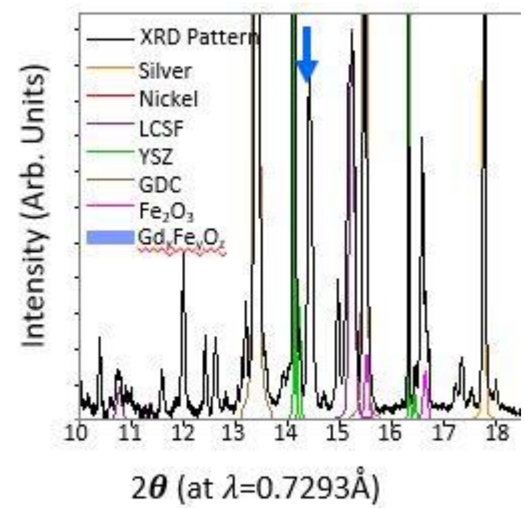
Design & fab operando stages for beam line experiments





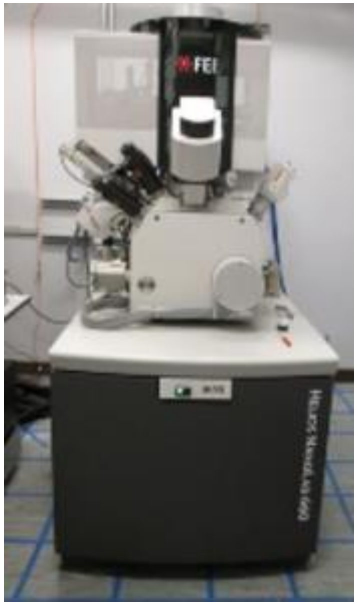
XAS measurements

$GdFeO_x$ phase identification with micro focused XRD



HTE Supernode Contributions: : Sandia – 2D and 3D Chemical and Morphological Characterization

- Capability
  - Focused ion beam sample milling
    - micro milling for vertical profiling
  - Advanced Electron Microscopy: scanning, transmission
    - high detail morphology
  - Electron Backscatter Diffraction
  - Energy Dispersive X-ray spectroscopy for High resolution elemental
    mapping (EDS)
- Accomplishments

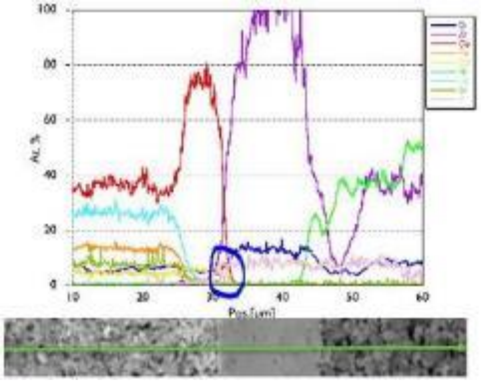


Sandia

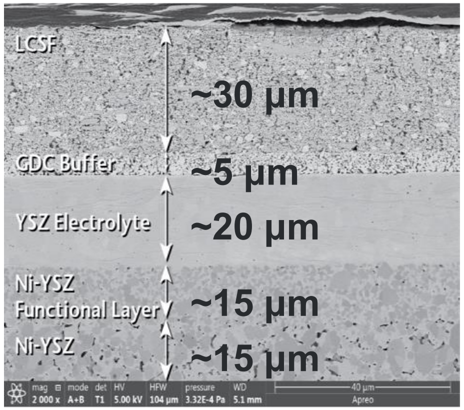
FIB/SEM/

EDS/EBSD

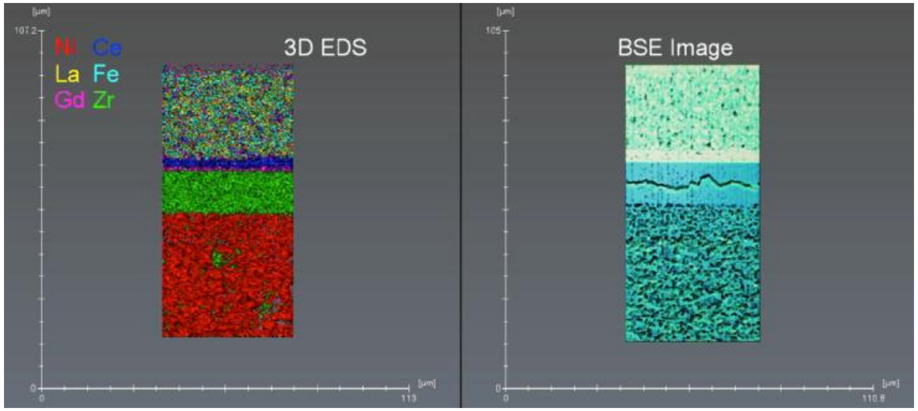
Gd migration to interface



morphology characterization



elemental mapping using EDS, and morphology using backscattered electrons – revealing a YSZ crack

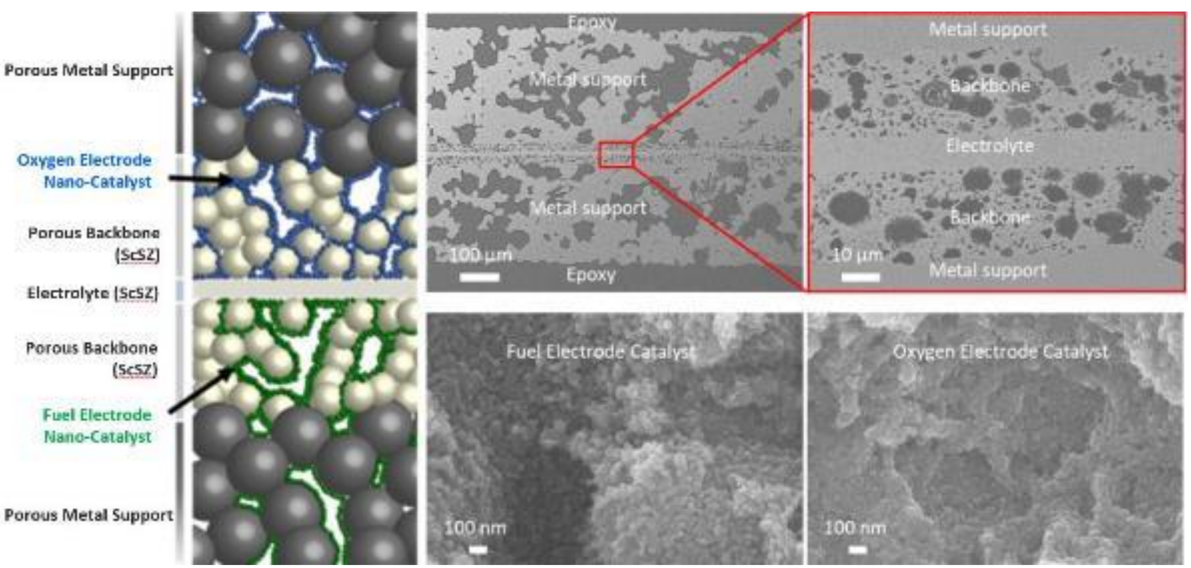


HTE Supernode Contributions: : LBNL – Processing & Operando Node

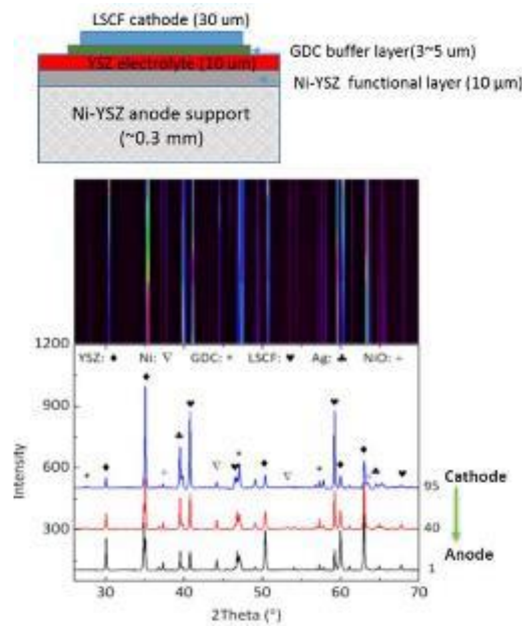
- Capability
  – Metal-Supported SOEC materials fabrication: significantly less expensive than ceramics, single high sintering step, mechanically strong, welded connections, fast temperature control, intermittent fuel/H_2 fuel tolerant
  – Characterization at ALS: tomography, non-ambient diffraction, microdiffraction

- Accomplishments

High-detail imaging enabling layer differentiation



Micro-XRD with layer-specific differentiation



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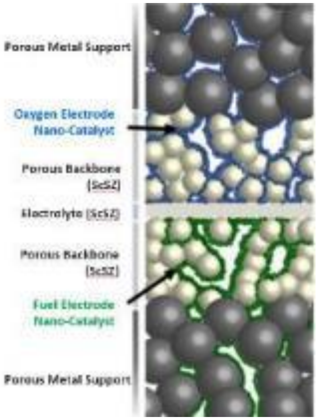
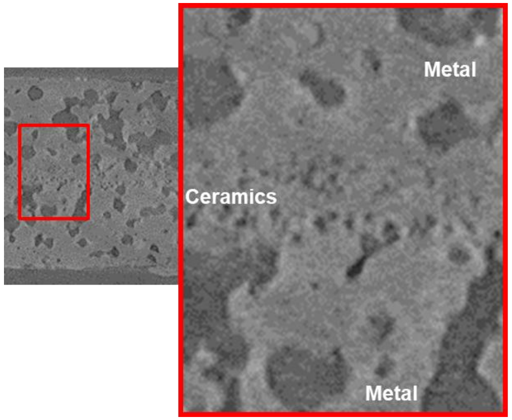
  - Characterization at ALS: tomography, non-ambient diffraction, microdiffraction
- Accomplishments

MS-SOEC

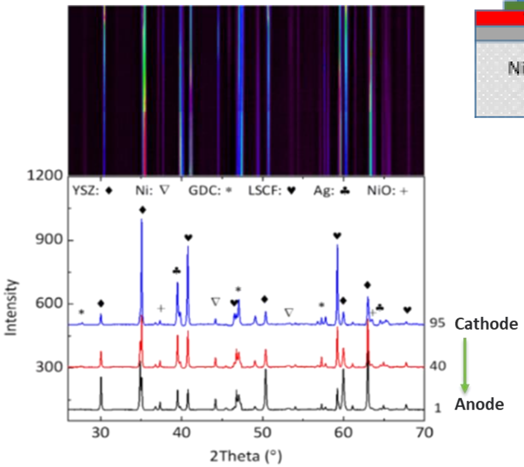


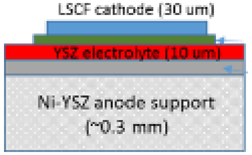
ALS

Tomography Imaging



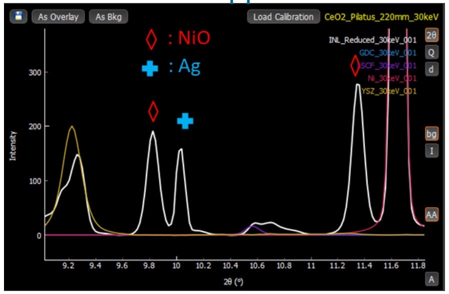
Micro-XRD with layer-specific differentiation

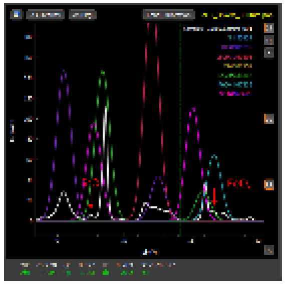




Transmission XRD Detects All Layers in Full Cell

Anode-Supported





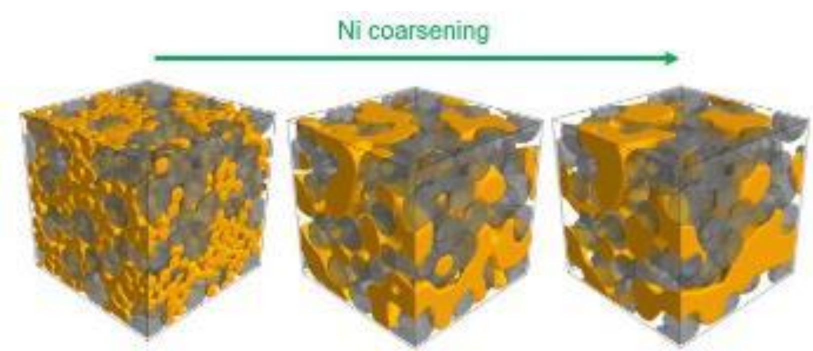
Metal-Supported

HTE Supernode Contributions: LLNL – Theory and Multi-Scale Computational Modeling

- Capability – highly complementary to experimental nodes
  - Atomistics (*ab initio*): interface & phase formation energies, atom transport, reaction kinetics
  - Computational thermodynamics (CALPHAD): phase stability v. composition and operating conditions
  - Microstructural evolution (phase field): phase transformations, coarsening kinetics, redox-driven microfracture

- Accomplishments

Advanced SOEC microstructural degradation model



Integration with quantitative CALPHAD and *ab initio* inputs

3 elements

Ni Zr O

4 phases

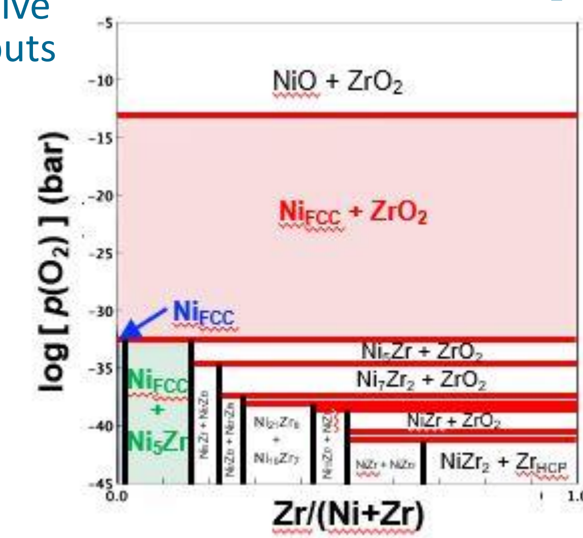
Ni_{FCC} (Zr in solution)

ZrO_2/YSZ

Ni-Zr intermetallic

Pore

Improved predictions of phase dependence on O_2



HTE Supernode Summary: Advances in Understanding o-SOEC Durability, Degradation

- Major inroads into understanding o-SOEC durability were gained through the first half of FY 2020 by the HTE Supernode
- Excellent progress in identifying phenomena that characterize failure of o-SOECs
  - Consistent, reproducible button cell platform for evaluating o-SOEC performance
  - Microscopy, elemental mapping for investigating morphology alterations and elemental migration in cell materials post-mortem
  - X-ray identification of unmodified- and new phases, and experimental strategies for higher resolution, layer specific cell interrogation of cells
  - Multiscale modeling for unraveling mechanism, key controlling factors
- Result $\rightarrow$ increased cell durability, while maintaining or improving efficiency
  - Operating strategies for current o-SOECs to increase longevity
  - Compositional options for reducing elemental migration
  - Fabrication guidance to mitigate unwanted phase formation

HTE Supernode: Future Work in o-SOEC Development, p-SOEC Evolution

- Supernode: poised to conduct the second round of experimental and theoretical studies, guided by initial results
- Capitalize on initial synchrotron X-ray, microscopy studies
  - tomographic studies using synchrotron beamline
  - stoichiometry, oxidation state using micro-XAS coupled XRF:
    - Fe segregation, localization, and secondary $Gd_xFe_yO_z$ phase formation
  - defect and void formation: Nano-TXM (30 nm resolution)
  - Initiate In-operando testing of cells & model systems: XRD, XAS, and TXM
- Model systems to isolate degradation mechanisms
  - identify and track elemental diffusion and redox in cell
  - role of sintering-aids
- Enhanced multi-scale modeling to predict migration, phase alteration, and material failure
  - thermodynamic viability using adapted DFT approaches
- Initiate p-SOEC research
  - initiate studies to identify mechanisms of electrical leakage
  - conduct durability testing to establish viability as a lower temperature alternative

HydroGEN HTE Supernode Approach: Close Interactions with Industrial, Academic Partners

- **Approach**
  - Strong communication with seedling partners collaborating with the Advanced Electrode and Solid Electrolyte Materials node at INL
    - A gateway for leveraging the microscopy, accelerator characterization and multiscale modeling capability at NREL/SLAC, Sandia, Berkeley, and Livermore

Northwestern University
SAINT-GOBAIN
REDOX
Redox Power Systems, LLC
West Virginia University
United Technologies Research Center
UCONN
UNIVERSITY OF CONNECTICUT
NEXCERIS
where energy meets environment
UNIVERSITY OF South Carolina

- **Goal**
  - Accelerate research, development, and deployment of advanced water splitting technologies for clean, sustainable hydrogen production
- **Technical Objectives**
  - Improve cell fabrication process steps, manufacturing, & cell supports
  - Improve cell performance, optimize operations parameters
  - Elucidate, eliminate, or mitigate mechanisms of degradation

Supernode connection with HTE Seedling Projects

High-level strategy: take results and approach from the HTE Supernode, apply to technology development conducted in the seedling projects

The seedlings encounter issues related to the Supernode research, only at a higher TRL

- o-SOEC development
  - **Northwestern University**: Optimize cell fabrication, testing
  - **Saint Gobain**: Thermal & chemical expansion measurement
  - **University of South Carolina**: Oxygen evolution in symmetrical, and in planar cells
  - **Nexceris**: Coupon interconnect evaluation, extending to reduced temperature evaluation in p-SOECs in BP2
- p-SOEC development
  - **UTRC**: Experimental full cell testing, focused on Faradaic efficiency measurements, and data acquisition
  - **University of Connecticut**: Cell fabrication and testing, education and training
  - **Redox**: Electrolyte stability and new compositions
  - **West Virginia University**: Electronic leakage investigations (conductivity measurement), correlated with defect chemistry and fundamental calculations

Collaboration: HydroGEN HTE Node Utilization

Lab	Node	Node PI	Nex-ceris	NWU	Redox	Saint Gobain	UConn	USC	UTRC	WVU	Super -Node
INL	Analysis and Characterization of Hydrided Material Performance	Gabriel Ilevbare, Michael Glazoff	✓								
INL	Advanced Materials for Elevated Temperature Water Electrolysis	Ding, Dong	✓	✓	✓	✓	✓	✓	✓	✓	✓
SNL	Advanced Electron Microscopy	Sugar, Josh									✓
NREL	Controlled Materials Synthesis and Defect Engineering	Ginley, David Parilla, Philip Bell, Robert									✓
NREL	Engineering of Balance of Plant (BOP) for High-Temperature Systems	Ma, Zhiwen Martinek, Janna							✓		

Collaboration: HydroGEN HTE Node Utilization

Lab	Node	Node PI	Nex-ceris	NWU	Redox	Saint Gobain	UConn	USC	UTRC	WVU	Super-Node
SNL	High-Temperature X-Ray Diffraction (HT-XRD) and Complementary Thermal Analysis	Coker, Eric				✓					
LBNL	Metal-Supported SOEC	Tucker, Michael Wang, Ruofan		✓					✓		✓
LLNL	Multi-Scale Modeling of Solid-Sate Interfaces and Microstructures in High-Temperature Water Splitting Materials	Wook, Tase Wood, Brandon Frolov, Timofey									✓
NREL	Thin Film Combinatorial Capabilities for Advanced Water Splitting Technologies	Zakutayev, Andriy					✓			✓	
INL	SOEC Characterization	O'Brien, James		✓		✓					✓

Acknowledgements

Energy Materials Network
U.S. Department of Energy

HydroGEN
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Node Experts

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David Ginley
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Eric Coker

Research Teams

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SAINT-GOBAIN
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NEXCERIS where energy meets environment
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