Energy Materials Network
U.S. Department of Energy

HydroGEN
Advanced Water Splitting Materials

HydroGEN: High-Temperature Electrolysis Supernode

G. S. Groenewold, D. Ding, R.D. Boardman

Presenter: Gary Groenewold, INL

Date: 5/13/2020

Venue: 2020 DOE Annual Merit Review

Project ID # P148D

This presentation does not contain any proprietary, confidential, or otherwise restricted information.

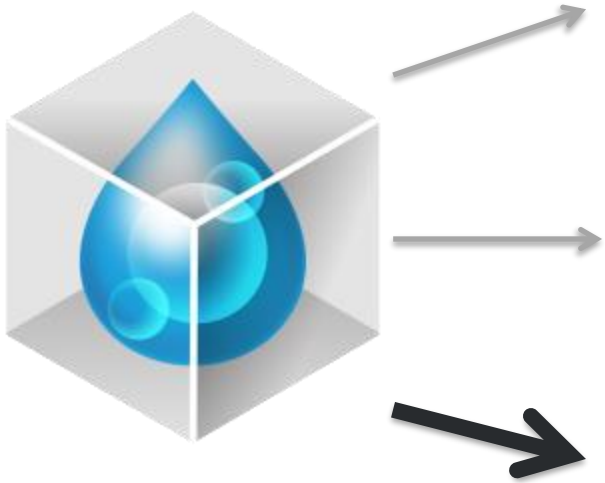
NREL · BERKELEY LAB · Sandia National Laboratories · INL Idaho National Laboratory · Lawrence Livermore National Laboratory · SRNL

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

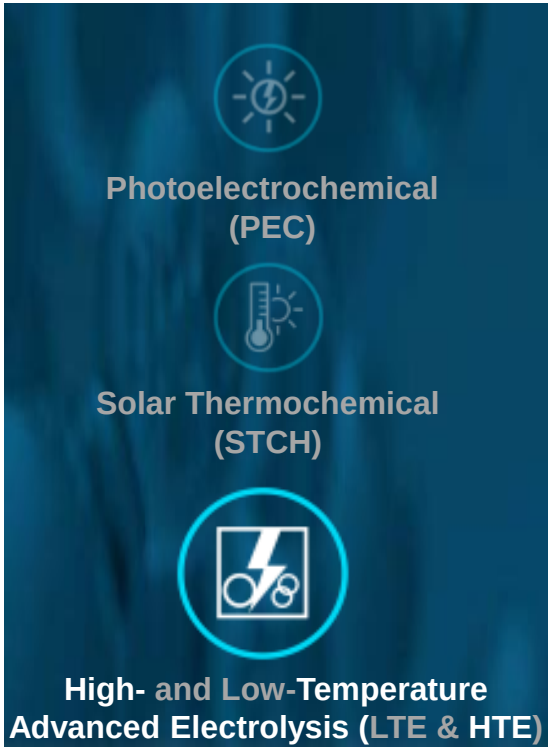
AWSM Consortium 6 Core Labs:

NREL – National Renewable Energy Laboratory
Berkeley Lab
Sandia National Laboratories
INL – Idaho National Laboratory
Lawrence Livermore National Laboratory
SRNL

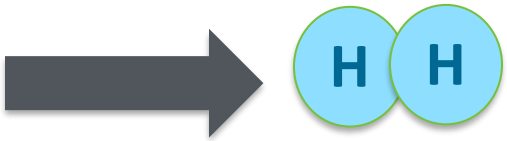
Accelerating R&D of innovative materials critical to advanced water splitting technologies for clean, sustainable & low cost H_2 production, including:



Water



Production target <$2/kg



Hydrogen

Approach: HTE Projects & Collaboration

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

HTE Node Labs

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

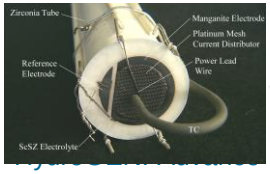
West Virginia University

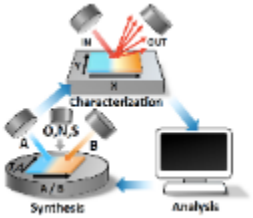
NEXCERIS where energy meets environment

SAINT-GOBAIN

University of Connecticut

UNIVERSITY OF South Carolina





Accomplishments and Progress:
Established Nodes for Project Support

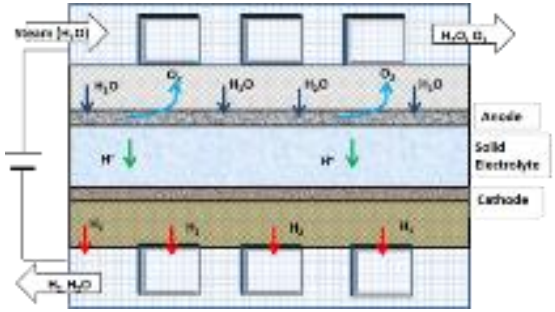
Nodes for HTE

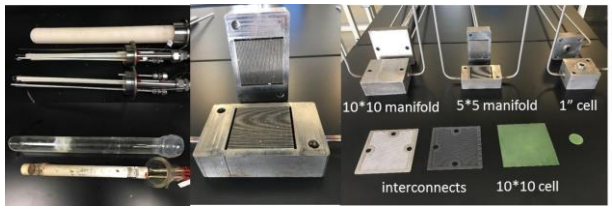
- 9 @ readiness level 1
- 22 @ readiness level 2
- 9 @ readiness level 3

10 nodes used by current HTE projects

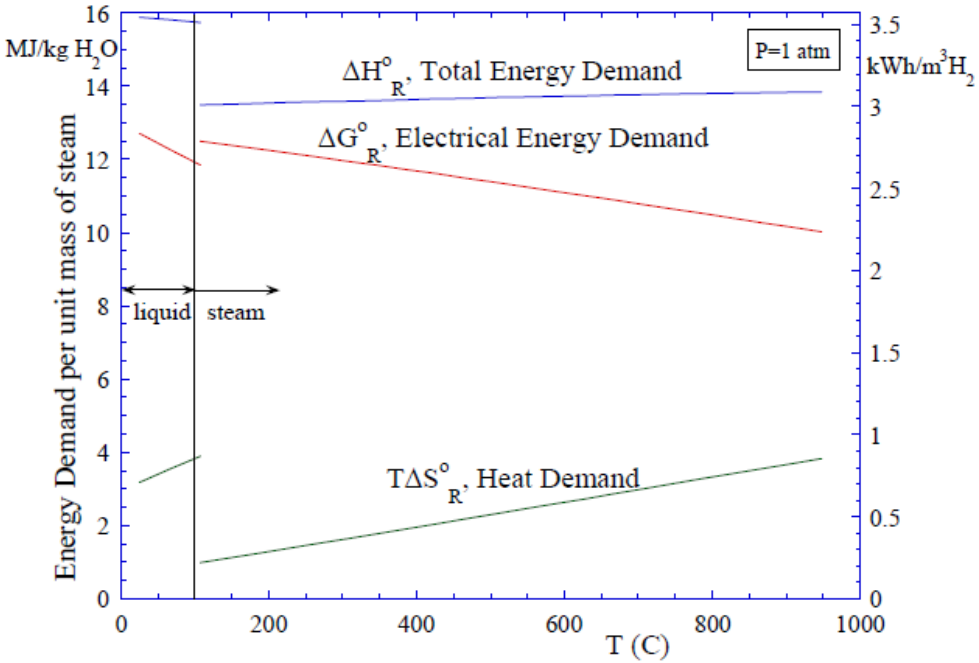
Node Classification

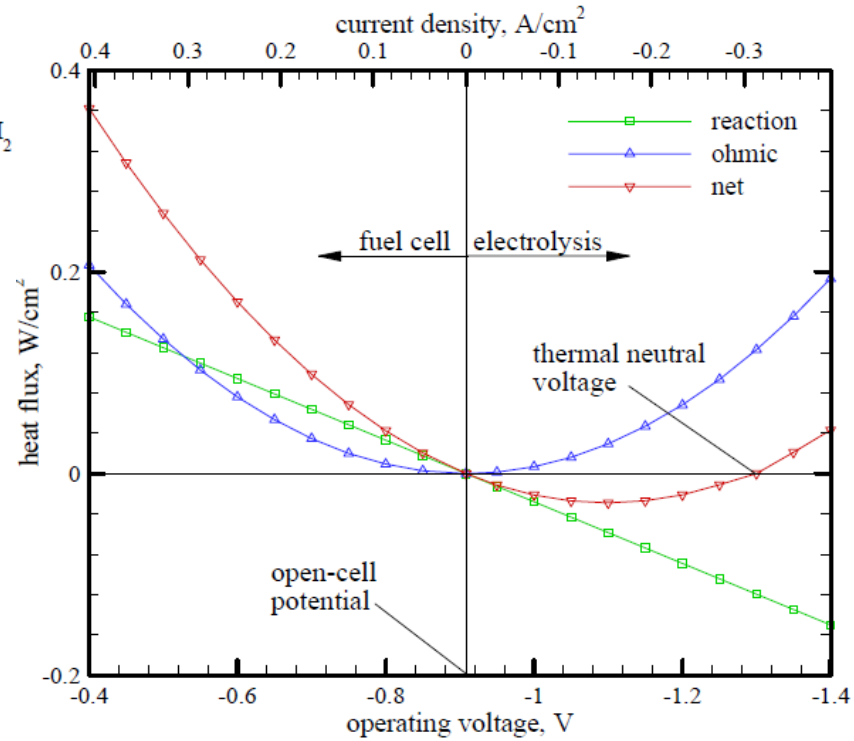
6x Analysis
6x Benchmarking
20x Characterization
13x Computation
6x Material Synthesis
5x Process and Manufacturing Scale-Up
5x System Integration





Overview: Advantages/Disadvantages of HTE



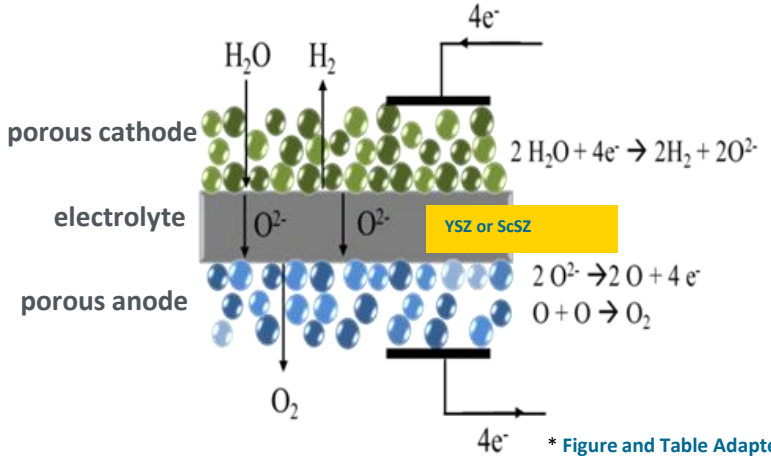


- **30-50% higher thermodynamic efficiency is possible for steam compared to water splitting (combined free energy and electricity use)**
- **Reversible operation is possible with optimal design of cells, stacks and modules**
- **Does not require highly precious metals**
- **Concerns: cell degradation,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

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

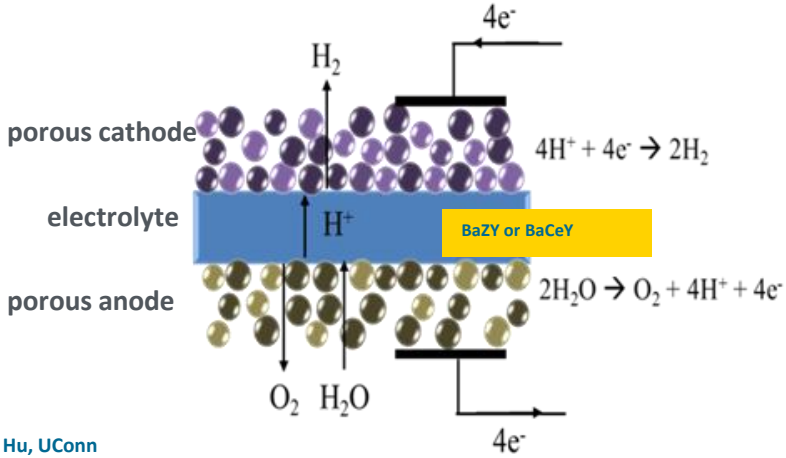
Oxygen Ion Transport Solid-Oxide Electrolysis*

(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

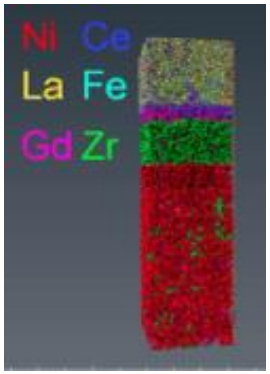
Budget

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

HydroGEN HTE Supernode: Five Labs, Coordinated Research Capabilities

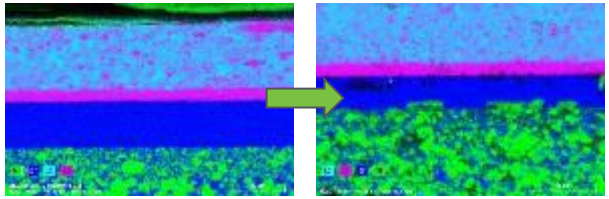
- **Coordinated R&D effort addressing o-SOEC, p-SOEC science**
- **INL**
  - Materials R&D
  - Cell Fabrication and Testing
  - Intensive Seedling Support

- **Sandia**
  - Materials micro-milling (FIB)
  - Microscopy & elemental mapping



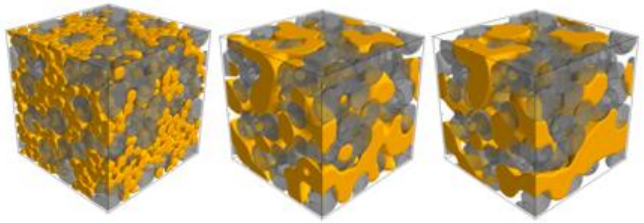
- **NREL**
  - Custom cell design & fab
  - SLAC interface for Micro XRD, XAS, XTM

- **LLNL**
  - Multi-scale computational modeling
  - Ab initio → phase field → continuum electrochem (NWU)



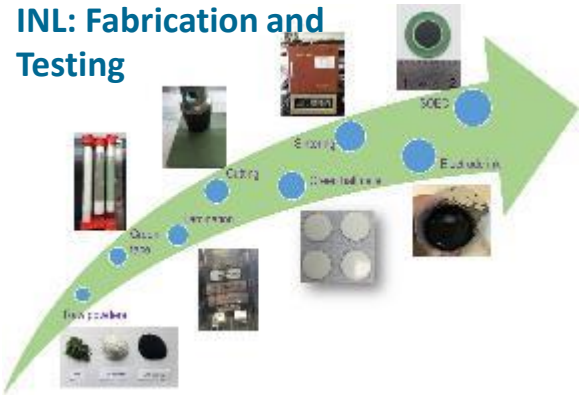
pre-operation post-operation 750° C

- **LBNL - ALS**
  - Micro diffraction
  - Tomography



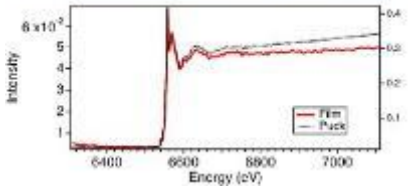
HTE Supernode Objectives – o-SOEC technology

INL: Fabrication and Testing



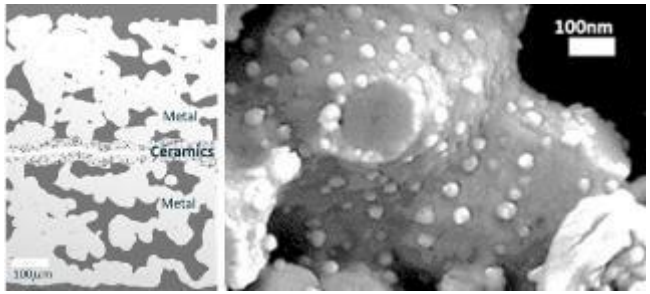
SNL: Imaging

NREL: Surface & Interface

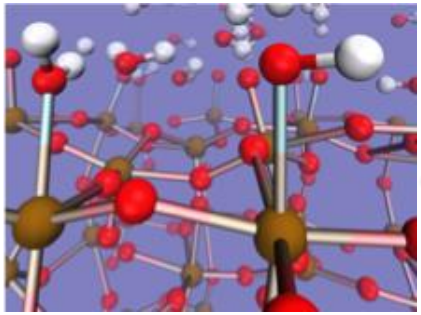


- higher efficiency, improved durability
- rationalize & predict degradation
- manipulate composition to optimize durability
- accelerate rate of technology development
- **achieve $2/kg target for H_2 production of**

LBNL: Processing & Operando



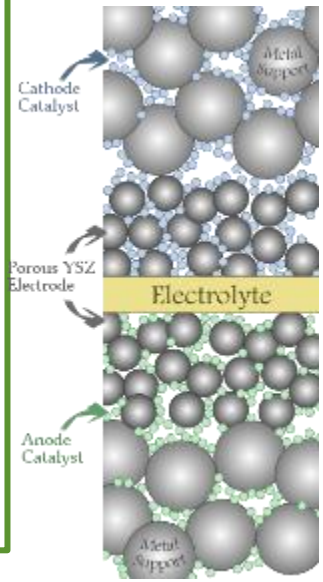
LLNL: Modeling



HTE Supernode Challenges: Characterization of Solid Oxide Electrode Microstructure Evolution

Challenges:

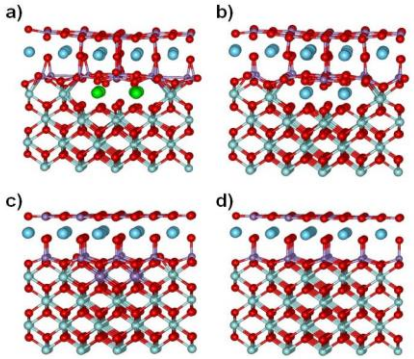
- Market acceptance of hydrogen production using high temperature electrolysis (HTE) relies on cost reduction and durable operation.
- Degradation mechanisms in oxygen-ion conducting solid oxide electrolysis cells (o-SOEC) remain elusive
  - correlation of electrode microstructure evolution to degradation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



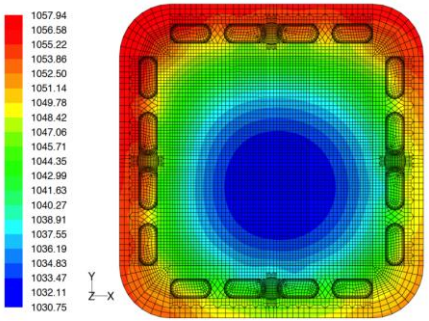
Catalyst particle coarsening

Solid-oxide electrode & electrolyte sintering

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



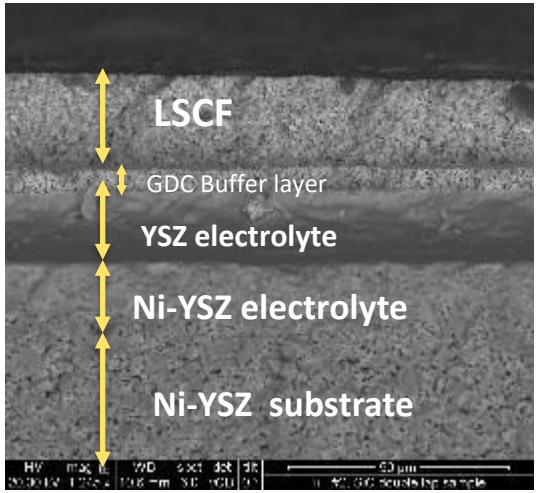
High temperatures and thermal gradients cause non-uniform grain boundary stresses



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

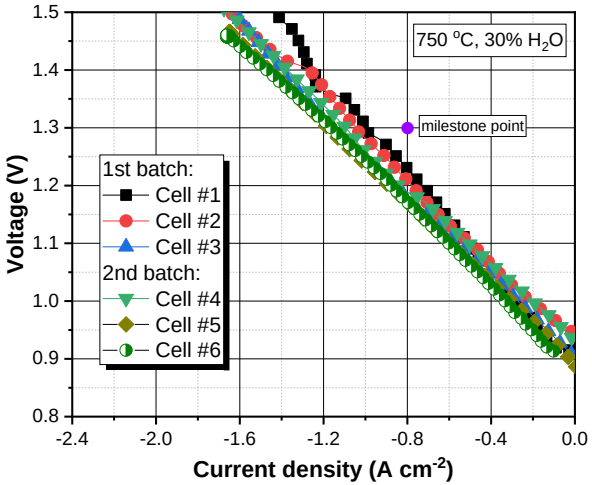
- Capability
  - Hydrogen Lab: 30 yr. legacy of SOEC development and engineering
    - Button cell & stack testing
  - o-SOEC material research, cell fabrication
    - "button-to-large": high temp roll-to-roll, solid oxide additive manufacturing for varied cell components, configurations, electrodes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

controlled layer deposition



button cell fab reproducibility

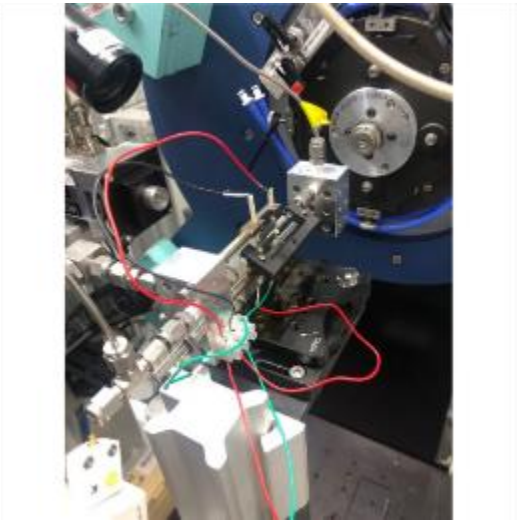
o-SOEC performance consistency

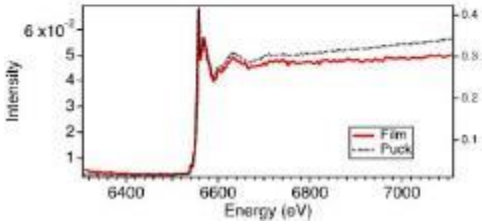


HTE Supernode Contributions: NREL -- Controlled Materials Synthesis and Defect Engineering

- 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

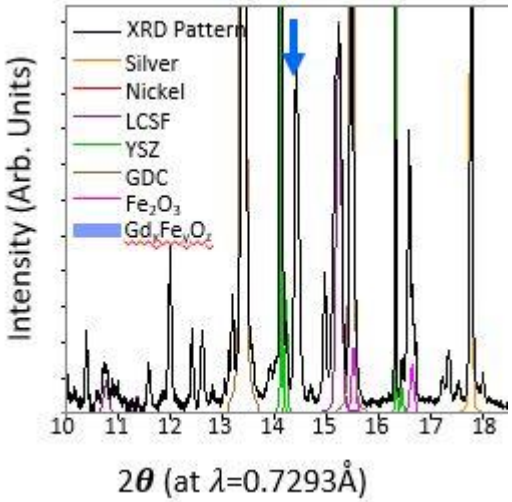
Design & fab operando stages for beam line experiments





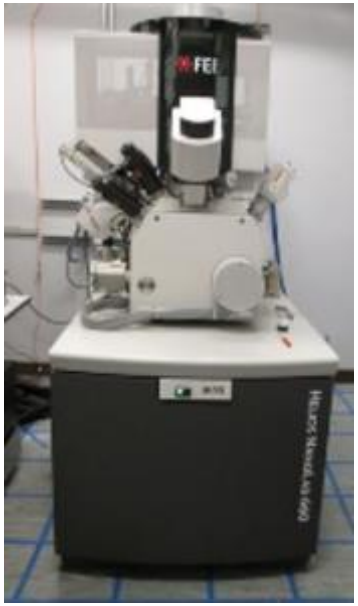
XAS measurements

$GdFeO_x$ 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

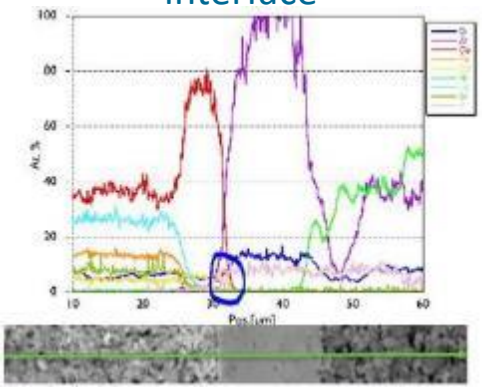


Sandia

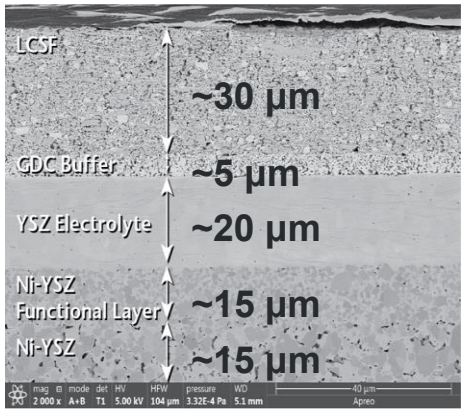
FIB/SEM/

EDS/EBSD

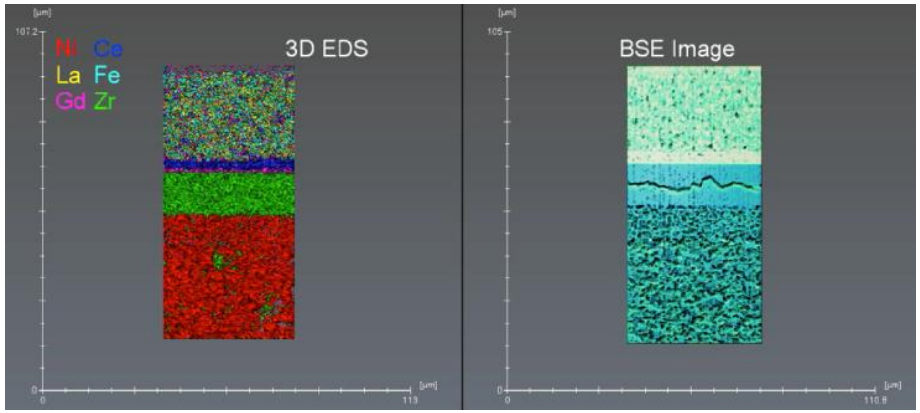
Gd migration to interface



morphology characterization



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

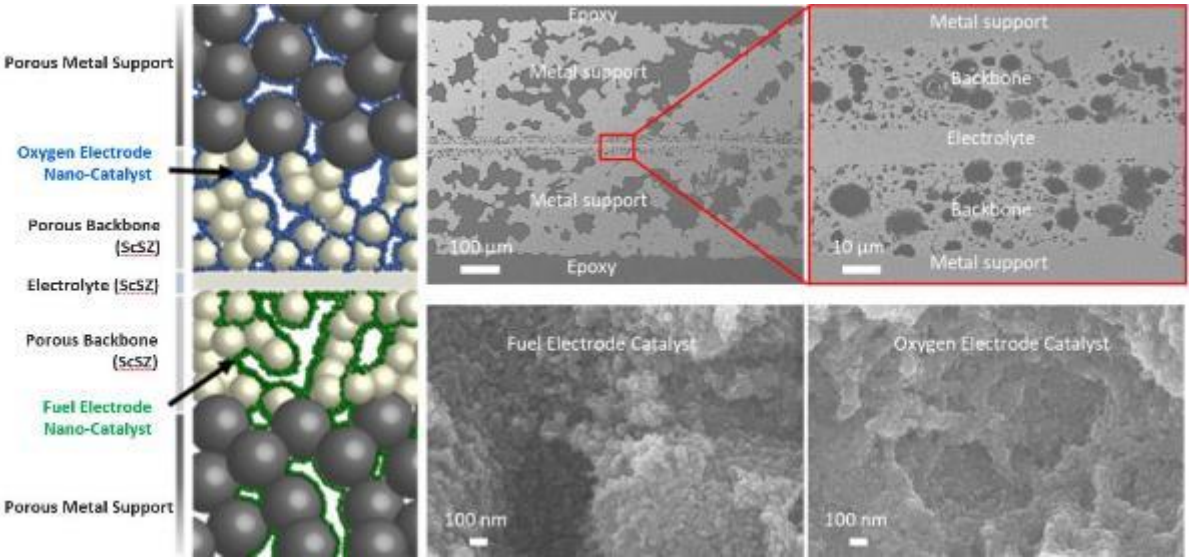


HTE Supernode Contributions: : LBNL – Processing & Operando Node

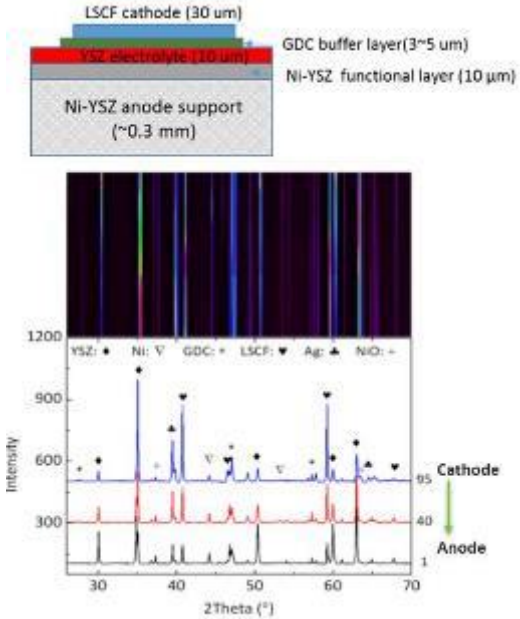
- Capability
  - Metal-Supported SOEC materials fabrication: significantly less expensive than ceramics, single high sintering step, mechanically strong, welded connections, fast temperature control, intermittent fuel/H_2 fuel tolerant
  - Characterization at ALS: tomography, non-ambient diffraction,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

- Accomplishments

High-detail imaging enabling layer differentiation



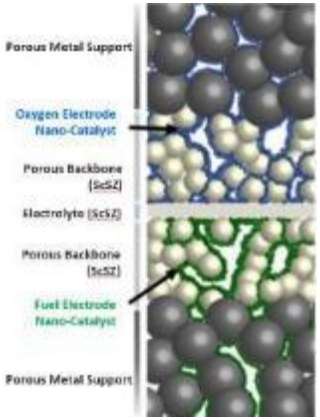
Micro-XRD with layer-specific differentiation



HTE Supernode Contributions: : LBNL – Processing & Operando Node

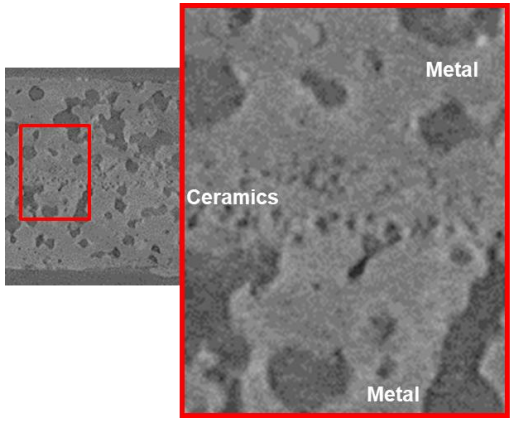
- Capability
  - Metal-Supported SOEC materials fabrication: significantly less expensive than ceramics, single high sintering step, mechanically strong, welded connections, fast temperature control, intermittent steam/H_2 fuel tolerant
  - Characterization at ALS: tomography, non-ambient diffraction,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

MS-SOEC

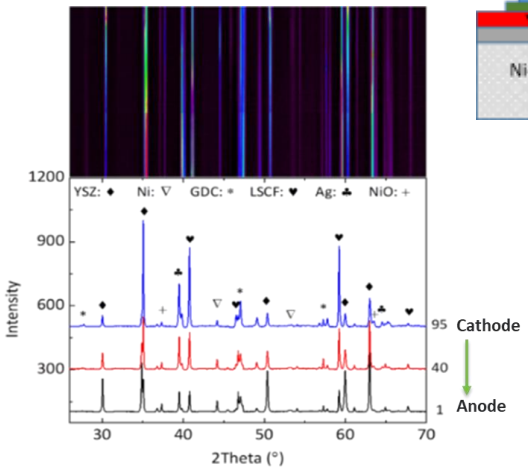


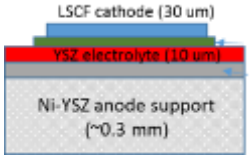
ALS

Tomography Imaging



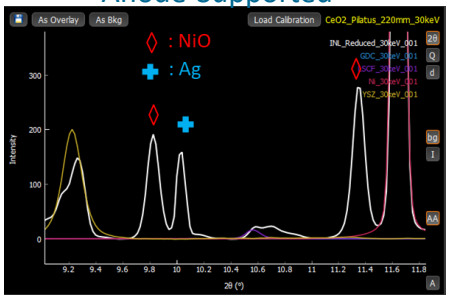
Micro-XRD with layer-specific differentiation

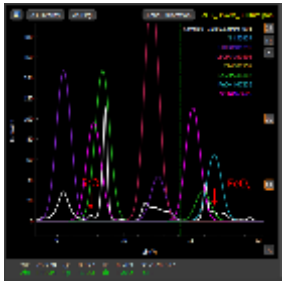




Transmission XRD Detects All Layers in Full Cell

Anode-Supported





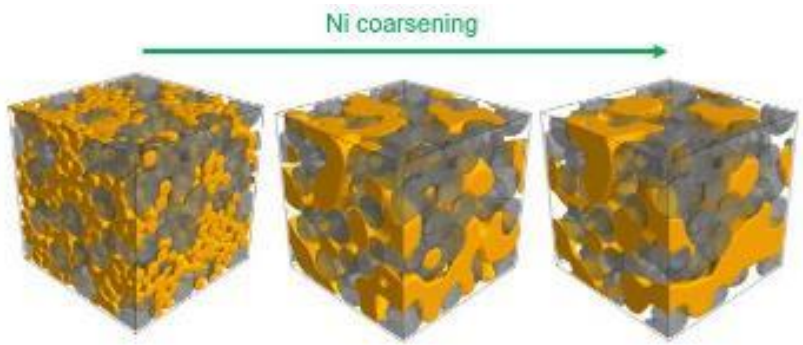
Metal-Supported

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

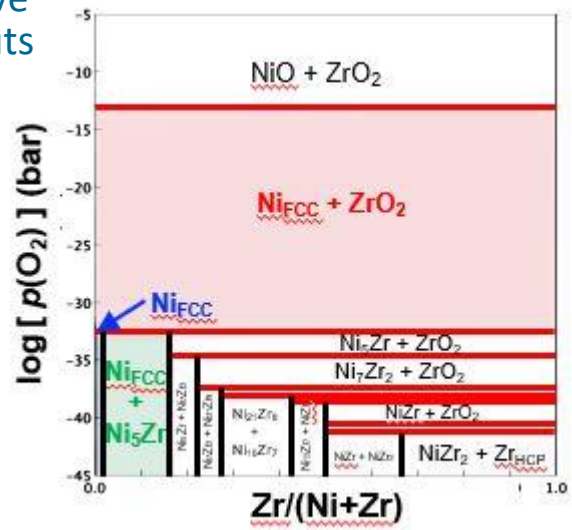
Ni_{FCC} (Zr in solution)

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 → 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	Red ox	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
Advanced Water Splitting Materials

Authors

Richard Boardman
Huyen Dinh
Michael Tucker
Dong Ding
Zhiwen Ma
James O'Brien
Adam Weber
Andriy Zakutayev

Node Experts

Dong Ding
Michael Tucker
Zhiwen Ma
James O'Brien
Andriy Zakutayev
Brandon Wood
David Ginley
Josh Sugar
Eric Coker

HTE Project Leads

Scott Barnet, NU
Tianli Zhu, UTRC
Prabhakar Singh, UConn
John Pietras, Saint Gobain
Xingo Liu
Olga Marina, PNNL

Research Teams

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

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

Acknowledgements

Energy Materials Network
U.S. Department of Energy

HydroGEN
Advanced Water Splitting Materials

HTE Supernode Team - 18 experts

Scott Barnett
Peter Voorhees

Northwestern University

Richard Boardman
Dong Ding
James O'Brien
Hanping Ding
Gary Groenewold

Idaho National Laboratory

Huyen Dinh
David Ginley
Zhiwen Ma
Robert Bell
Philip Parilla

NREL
NATIONAL RENEWABLE ENERGY LABORATORY

Sandia National Laboratories

Josh Sugar

Mike Tucker
Fengyu (Kris) Shen
Dula Parkinson
Nobumichi Tamura
Matin Kunz

BERKELEY LAB
Lawrence Berkeley National Laboratory

Lawrence Livermore National Laboratory

Brandon Wood
Joel Berry
Penghao Xiao

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