Solid-State Electrochemical Cell
R&D Progress at NETL

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Team Lead, NETL SSEC R&D

22nd Annual Solid Oxide Fuel Cell Project Review Meeting
November 17, 2021
Outline

• Introduction

• Recent Progress Summary
  • Cell and Stack Degradation Modeling and Simulation
  • Electrode Design and Engineering
  • Strategic Systems Analysis and Engineering

• Summary of Other Efforts

• Wrap-Up
NETL SSEC R&D Team

**NETL (Federal Staff)**
- Gregory Hackett, Team Lead (NETL)
- Harry Abernathy (NETL)
- Travis Shultz (NETL)
- Ron Breault (NETL)
- Rich Pineault (NETL)
- Yves Mantz (NETL)
- Yuhua Duan (NETL)
- Slava Romanov (NETL)
- Youhai Wen (NETL)
- Randy Gemmen (NETL)

**West Virginia University**
- Harry Finklea (Chemistry Emeritus)
- Ismail Celik (MAE Emeritus)
- David Mebane (MAE)
- Ed Sabolsky (MAE)
- Xueyan Song (MAE)
- Xingbo Liu (MAE)
- Yun Chen (WV Research Corp.)
- Bo Guan (WV Research Corp.)
- Jose Bohorquez (MAE, Student)
- Joshua Tenney (MAE, Student)

**NETL (Site Support Team)**
- Tom Kalapos (LRST)
- Billy Epting (LRST)
- Arun Iyengar (KeyLogic)
- Lynn Fan (LRST)
- Rick Addis (USSE2)
- Tianle Cheng (LRST)
- Youngseok Jee (LRST)
- Jian (Jay) Liu (LRST)
- Yueh-Lin Lee (LRST)
- Tao Yang (LRST)
- Yinkai Lei (LRST)
- Giuseppe Brunello (LRST)
- Hunter Mason (LRST)
- Yoosuf Picard (LRST)
- Kyle Buchheit (KeyLogic)
- Fei Xue (LRST)
- TBD Experimentalist (LRST)

**Carnegie Mellon University**
- Paul Salvador (MSE)
- Shawn Litster (MechE)
- Tony Rollett (MSE)
- Liz Holm (MSE)
- Hokon Kim (MSE, Grad Student)
- William Kent (MSE, Grad Student)

**Clemson University**
- Kyle Brinkman (MSE - Chair)
- Jack Duffy (MSE)

**Georgia Southern University**
- Hayri Sezer (Engineering)

**Penn State University**
- Long-Qing Chen (MSE)
- Yanzhou Ji (MSE, Student)

**University of Wisconsin-Madison**
- Dane Morgan (MSE)
- Ryan Jacobs (MSE)

**Wake Forest University**
- Michael Gross (Chemistry)
- Sixbert Muhoza (Post-Doc)

**TARGETED FOCUS:**
- Collaboration
- Technology Transfer
- Open-source tool development

Currently 50+ SOFC Team Members
Cell and Stack Degradation
Modeling and Simulation
Integrated Cell Degradation Model

3D Reconstruction of SOFC Electrodes

Degradation Models

Microstructural Analysis

Multiphysics Performance Model

Degradation of Cell Performance

Cost of Electricity or Hydrogen

Simulations run on thousands of unique SOFC electrode microstructures
Coupling Advanced Techniques

Microstructure Evolution in Ni-YSZ Electrodes under Operating Conditions

- Coupling the phase field model, microstructure analysis toolset, and multiphysics model for modeling the microstructure evolution

- Ni bulk diffusion, Ni(OH)$_2$ formation and diffusion through the pore phase and Ni-YSZ wettability change are incorporated in the model as the driving forces of the microstructure evolution

Adapting Capability to R-SOC/SOEC Mode

Multiphysics, ECR Characterization, Performance, Infiltration Modeling
Incorporation of Additional Degradation Mode

Simulation of Mechanical Degradation Considering Microstructures

Expanded modeling capability of simulating crack growth considering SOC microstructures

Microstructure

2 μm

Interphase cracking and through cracking under thermal and redox cycling, respectively

Thermal Cycling

Redox Cycling

Crack Length vs # of Cycles

Crack

Intact

YSZ

Pore

Ni
Convolutional Neural Networks

### Super-resolution

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<td>TPB</td>
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<tr>
<td>+ tortuosity, SA, etc</td>
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</table>

### Microstructure Generation

<table>
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<th>Value</th>
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<td>TPB</td>
<td>4.1</td>
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<tr>
<td>+ tortuosity, SA, etc</td>
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</tbody>
</table>

Tim Hsu et al., "Microstructure generation via generative adversarial network for heterogeneous, topologically complex 3d materials." *JOM* v73 pg 90 (2021)
Machine Learning Results of Analysis

Cathode (Air Electrode) Feature Importance Ranking

Each cathode feature’s impact on lifetime energy produced at 400 mA/cm²

- LSM/YSZ
- Porosity
- D-LSM
- D-YSZ
- D-pore
- HF-YSZ
- σ-YSZ
- σ-pore
- σ-LSM
- HF-pore
- HF-LSM

Low LSM/YSZ ratio, low porosity, and small solid particles are beneficial
Lifetime Energy as Figure of Merit

Voltage decay is important but misses whether electrode was a poor performer to begin with.

Need a single figure-of-merit that captures both initial performance and stability.

Lifetime energy production – at a given current density, up to a given time.

Proxy for $/kWh, which is what a plant operator would care about.

Area = lifetime energy produced [Wh/cm²]

Power [W/cm²] vs. Time [h]

Proxy for $/kWh, which is what a plant operator would care about.
Cathode Feature Importance Ranking

Impact on voltage decay [%/khr]
Lower is better

Impact on lifetime energy [Wh/cm²]
Higher is better

Small LSM particle sizes are bad for voltage decay, but net good for lifetime performance.

Lower LSM/YSZ ratio is good for both metrics.
Too Much Progress is a Good Problem to Have…

Large Cell Simulations
- DREAM SOFC Full 3D Planar
- Hydrogen/Hydrocarbon Fuels
- Contaminant Poisoning
- Impedance Analysis
- Electrolysis/r-SOC Operation

Defect Chemistry
- Provides electronic and energetic insights
- Parameters integrated with phase field or reaction models

EC Reaction Analysis
- Developed ERMINE framework for MOOSE
- Direct simulation of SOC physics in 3D microstructures
- Deeper look at heterogeneity, reaction distribution

TPB Boundary Activity Map
Performance Enhancement & Degradation Mitigation
SOC Electrode Design and Engineering
SOC Electrode Design and Engineering

Designing, Developing, and Deploying Advanced Electrode Eng. Techniques

Objectives
- Enhancement of performance and longevity
- Materials engineering
- Microstructure engineering

Benefits
- Cell/stack cost reduction
- Cell overpotential reduction
- Thermo-chemical / thermo-mechanical stability increase
- Reduced cost-of-electricity and/or cost of hydrogen produced

Approach
- **DESIGN** of materials and nanostructures
- **DEVELOPMENT** through tailored electrode construction
- **DEPLOYMENT** in commercial SOC systems
• **Hybrid Materials**: Metal and organic components mixed at atomic level
• **Sintering in inert atmosphere**: carbon template forms *in-situ* and remains during sintering; carbon is subsequently burned out at 700°C.
Infiltration of Nano-Structured Catalysts

- In-situ carbon templating method expanded to larger set of SOC materials
  - Reductions in $R_p$ and $R_\Omega$ possible by adding nano-size ionic conductors to backbone.
  - Protonic conducting oxide ($\text{BaCe}_{0.2}\text{Zr}_{0.7}\text{Y}_{0.1}\text{O}_{3-\delta}$) also formulated for IT-SOC applications

**Previous results:** Power curves of infiltrated LSM/YSZ baseline cells

**New Results:** Decrease in polarization resistance of LSCF/SDC baseline cells when infiltrated with nano-SDC (nSDC)
**Additive Manufacturing of SOC**

**Functionally Graded Electrodes to Mitigate Degradation, Boost Performance**

- Built automated layer-by-layer dip-coating and aerosol spray deposition systems to create 3D functionally graded electrode structures
  - Can vary **composition**, **particle size**, and **porosity** of composite electrode components
- Aerosol system has six inlet tubes (2 cleaning solutions, 4 electrode compositions)
  - Can change nozzle to change the width of deposited stripe
- Systems will be used to create optimized electrodes designed through simulations

**YSZ backbone porosity varied in z-direction on YSZ substrate**

Electrode composition varied from inlet to outlet on 5×5 cm² substrate (YSZ used for cost considerations during system development phase)
Advanced Characterization of Cation Interdiffusion across Interfaces

500-hour SOFC operation test: 0.75 A/cm², 800°C

“Burn-in” 5.4% / 1000 h performance loss

Change in cation ratios during LSM burn-in period different from longer term operation, showing impact of different driving forces

APT studies done in collaboration with J. D. Poplawsky at ORNL
Strategic Systems Analysis and Engineering
Pulling It All Together
Systems Analysis Recent Progress

Solid Oxide Cell and Stack Cost Production Study Expansion

Rationale

- A robust cell and stack production cost tool was developed previously.
- In response to the SOFC Program's investment in SOEC technology, this tool will be expanded to include SOEC production and additional cell geometries.

Approach

- Cell and stack cost production spreadsheet tool will include all necessary cost inputs:
  - Raw materials, equipment, energy, etc.
- Tool will allow sensitivity studies to be conducted on SOFC and SOEC:
  - Total production, materials costs, etc.
- Detailed guidance document/instructions being prepared to accompany tool.

Outcome

- Spreadsheet tool and guidance document in preparation.
- Scheduled completion March 2022.
In response to DOE interest, the SOFC Program has expanded its portfolio to include high-temperature solid-state electrolysis technology. A detailed understanding of the merits/demerits of coupling SOFC/SOEC technology versus a single reversible SOC unit is needed as a basis for future analysis.

The analysis will consider the incorporation of reversible SOC and SOFC/SOEC paired equipment and details on the effects of integrated equipment in a hybridized energy system. For example, capital cost of reversible SOC vs stability of separate SOFC/SOEC units will be a critical consideration.

The analysis will provide critical information to serve as a foundation to inform the SOFC Program on targeted R&D needed for integrated energy systems with SOC technology(ies). Targeted guidance for future analysis scope and scheduled completion March 2022.

APPENDIX E – AREA OF INTEREST 5: SOLID OXIDE ELECTROLYSIS CELL (SOEC) TECHNOLOGY DEVELOPMENT FOR HYDROGEN PRODUCTION

AOI Issue Date 01/15/2021
Submission Deadline for Full Applications 03/01/2021
DOE Share ($K) – 80% 1,000
Cost Share ($k) – 20% 250
Anticipated No. of Awards 8
Maximum Period of Performance 24 months (Single Phase/Single Budget Period)
Wrap-Up
NETL Capability Overview

High Temperature Electrochemical Systems (SOFC / r-SOC / SOEC)

FECM-SOFC Program
Goals and Objectives

NETL Unique Capability and Achievements

- Only team capable of modeling from atoms-to-COE
- Published high-resolution cell reconstruction datasets
- World leader in characterizing and simulating heterogeneity
- Advanced interface characterization - Atom Probe Tomography
- First using machine learning to create 40,000+ synthetic microstructures
- Able to provide targeted cell development feedback to industry
- In-situ high temp optical fiber sensor development (temp / gas comp)
- Experimental testing/electrode engineering/infiltration successes
- Extensive capability in strategic systems analysis and engineerings

Modeling from the Atomistic Scale to the System Scale (w/ PNNL)

Electrode Sub-volumes

Single Cell

Multi-Cell Stack
PNNL SOFC-MP

IGFC System

7/14/2021
Additional Efforts

Support of Other DOE/FECM/SOFC Program/HFTO Projects

Aris Energy Solutions, LLC

- System operational and producing power for the NETL site!
- Please see presentation later today!
- Project: DE-FE31978

UNDEERC

- Applying NETL capability to syngas fueled, tubular SOC
- Please see presentation from November 16
- Project: DE-FE31977

H₂NEW Laboratory Consortium (EERE/HFTO)

- Contributing expert guidance on modeling and characterizing SOC performance for new HFTO program
Final Announcement

• This will be Greg’s final presentation as NETL Team Lead for SSEC R&D at these meetings

• NETL is in the process of transitioning the Team Lead role to Dr. Harry Abernathy

• Greg will continue to be involved in NETL Systems Analysis efforts and will continue to participate in these meetings in that role
THANK YOU!

VISIT US AT: www.NETL.DOE.gov

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