

# NETL R&D: SOFC Materials Development and Degradation Modeling



21<sup>ST</sup> Annual Solid Oxide Fuel Cells Project Review Meeting

Gregory A. Hackett, Ph.D. – Team Lead

NETL Research and Innovation Center

July 10, 2020

Harry Abernathy, Ph.D.

Shiwoo Lee, Ph.D.

Tom Kalapos, Ph.D.



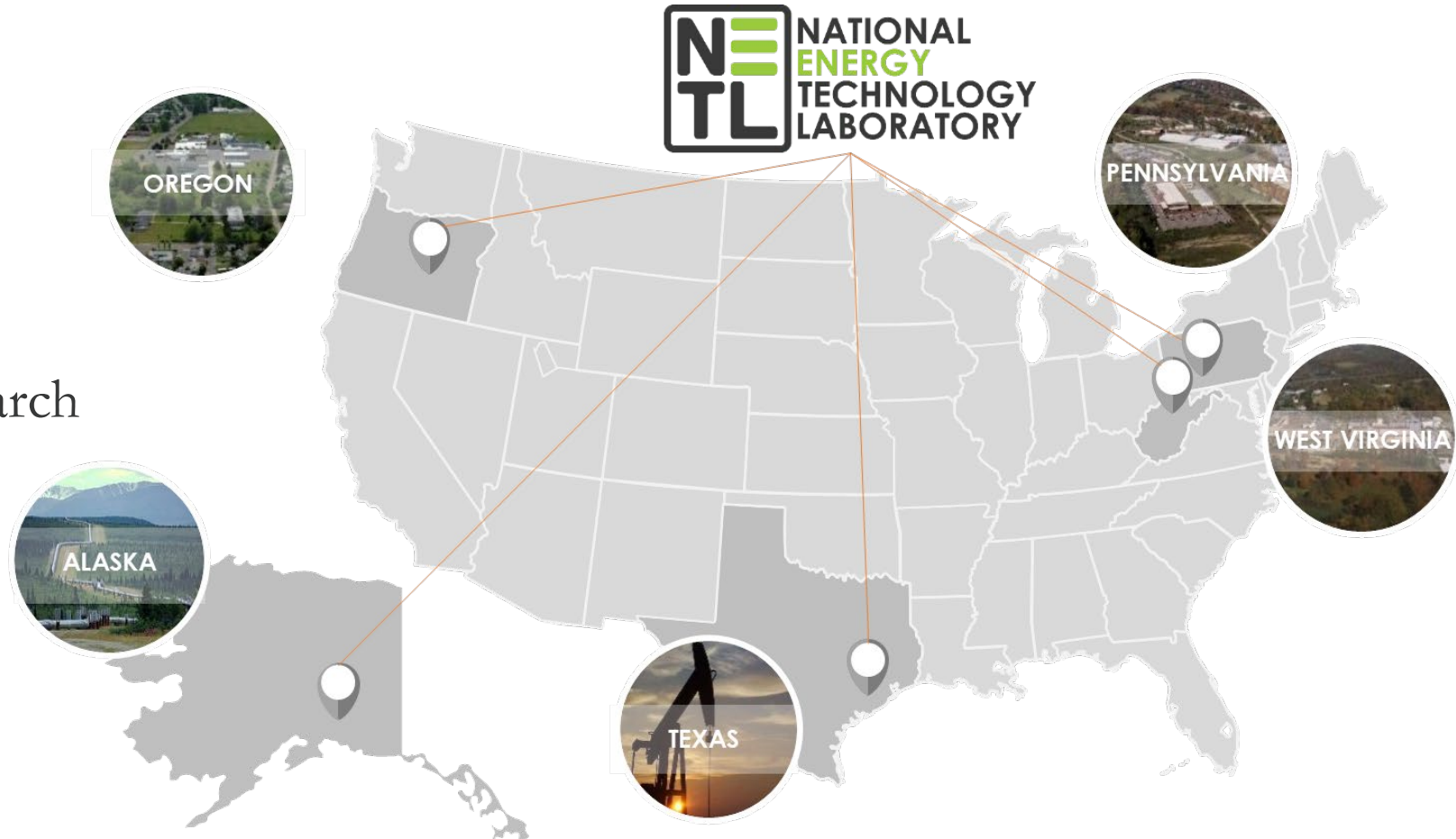
Solutions for Today | Options for Tomorrow





# Outline

- **NETL SOFC Research Team (EY20)**
- **NETL SOFC Research Portfolio**
  - Electrode Engineering Research and Development Progress
  - Cell and Stack Degradation Evaluation and Modeling Progress
  - Systems Engineering and Analysis Progress



# NETL SOFC Research Team



## NETL (Federal Staff)

- Gregory Hackett, Team Lead (NETL)
- Travis Shultz (NETL)
- Rich Pineault (NETL)
- Yves Mantz (NETL)
- Yuhua Duan (NETL)
- Slava Romanov (NETL)
- Youhai Wen (NETL)
- Dustin McIntyre (NETL)
- Jonathan Lekse (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 Corporation)
- Bo Guan (WV Research Corporation)
- Jose Bohorquez (MAE, Student)

## NETL (Site Support Team)

- Tom Kalapos (LRST)
- Harry Abernathy (LRST)
- Shiwoo Lee (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)
- Billy Epting (LRST)
- Hunter Mason (LRST)
- Yoosuf Picard (LRST)

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

## Carnegie Mellon University

- Paul Salvador (MSE)
- Shawn Litster (MechE)
- Tony Rollett (MSE)
- Tim Hsu (MSE)
- Hokon Kim (MSE, Grad. Student)
- Randall Doane (MSE, Grad Student)
- Elizabeth Holm (MSE)

## Clemson University

- Kyle Brinkman (MSE - Chair)
- Jack Duffey (MSE)

## 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)

## Western Carolina University

- Hayri Sezer (Engineering)

Currently 50+ SOFC Team Members

# NETL SOFC Field Work Proposal Overview

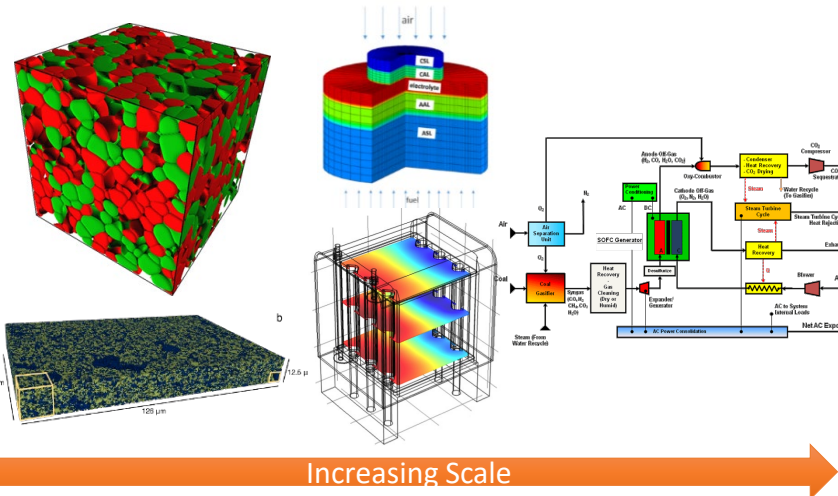
Enabling SOFC Technology through Research and Development at NETL

**CHALLENGE:** SOFC technology is cost prohibitive due to long-term performance degradation

**APPROACH:** Develop detailed degradation modeling tools and improve performance / longevity of SOFC

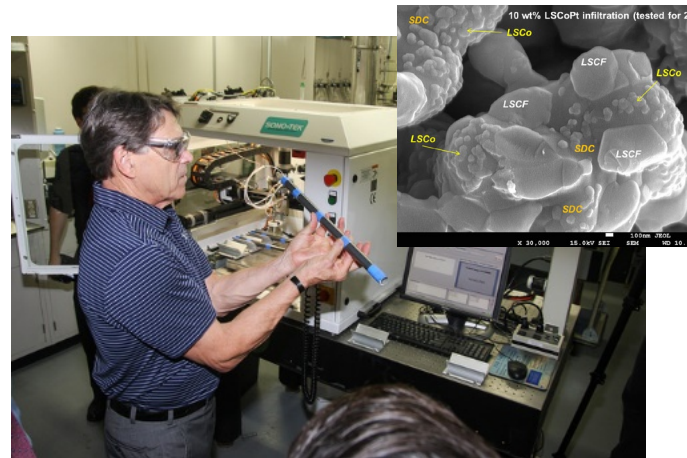
## Cell and Stack Degradation Modeling

- Degradation Prediction Tools
- Atoms-to-System Scale Bridging
- Experimental Validation
- HT Fiber-Optic Sensors



## Electrode Engineering

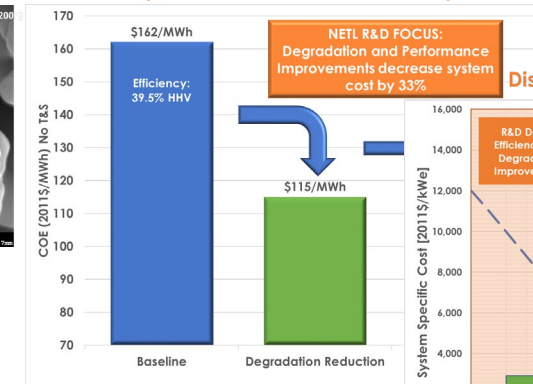
- Degradation Mitigation
- Microstructure Optimization
- Technology Transfer to Industry
- System Demonstrations



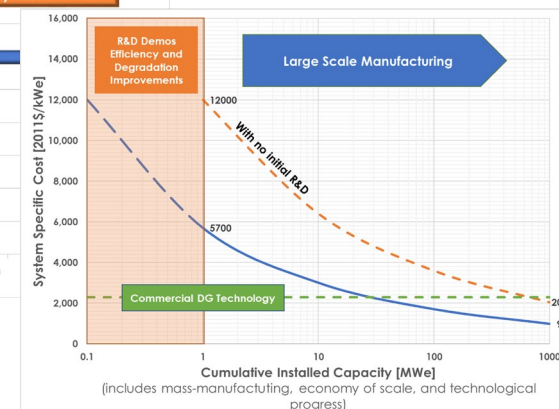
## Systems Engineering and Analysis

- Techno-Economic Analysis
- Hybrid Configuration Assessment
- R&D Goals Evaluation

### Utility Scale Techno-Economic Analysis

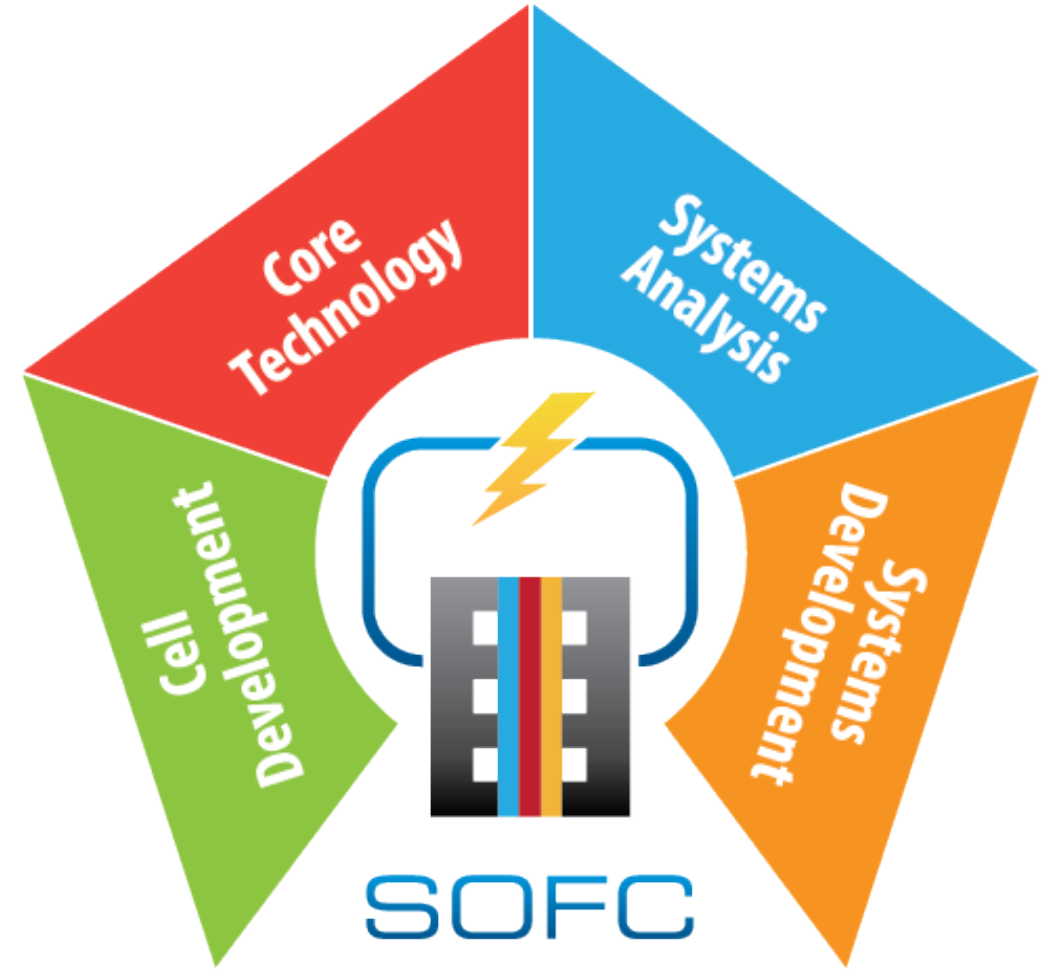


### Distributed Generation Market Analysis



# Performance Enhancement & Degradation Mitigation

SOFC Electrode Engineering



# SOFC Electrode Design and Engineering

Designing, Developing, and Deploying Advanced Electrode Engineering Techniques

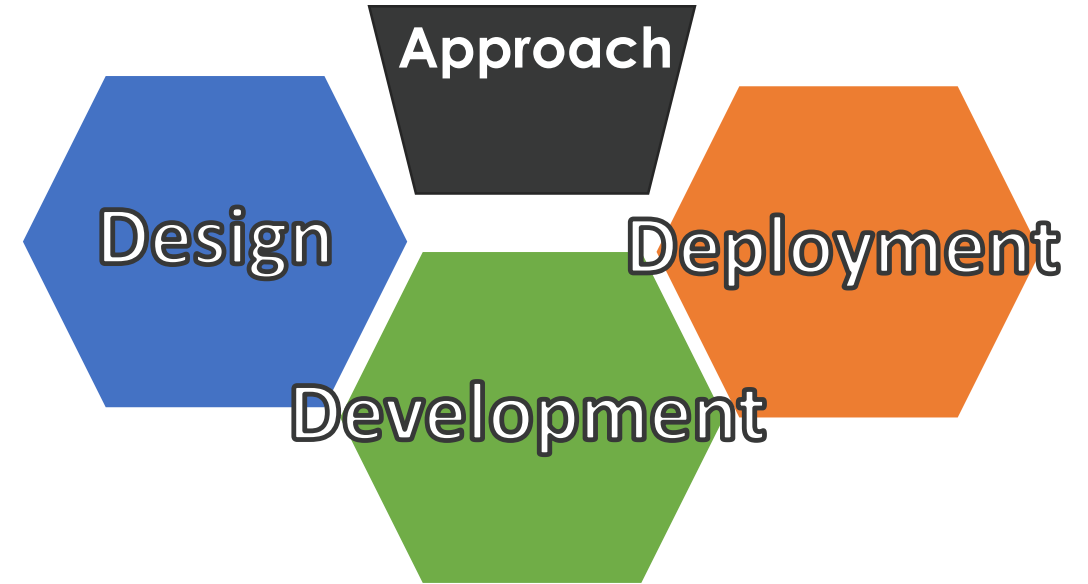


- **Objectives**

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

- **Benefits**

- Stack cost reduction
- Cell overpotential reduction
- Thermo-chemical / thermo-mechanical stability increase



**DESIGN** of materials and nanostructures

**DEVELOPMENT** through tailored electrode construction

**DEPLOYMENT** in commercial SOFC systems



# Enabling SOFC Technology through R&D at NETL

Electrode Engineering – Enhances Performance and Increases Reliability

## Concept to Market Readiness

### COMMERCIALIZATION

Technology available for implementation in SOFC production line

### DEMONSTRATION

Technology implemented and tested at SOFC stack (kW) scale

### SYSTEM TESTING

Evaluate technology on several commercial developer cells

### DEVELOPMENT

Patents obtained

### DISCOVERY

Proof of Concept

2019-20

Licensing to SOFC commercial developer

TRL 7-8

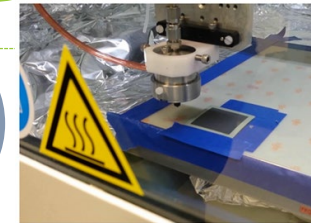
Atrex Cells



2017-18

Direct collaboration with Atrex Energy to scale up technology

TRL 6



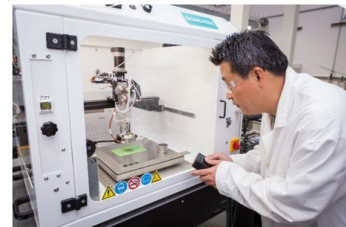
Demonstration at Lab Scale

2012-16

Demonstration on commercially relevant scale

TRL 4-5

Sonotek Sonic Spray Coater used for technology scale-up

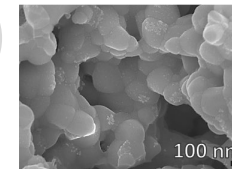


2009-12

Technology validated on SOFC button cells (several W) scale

TRL 2-3

Electrode infiltration technique evaluated



Infiltrated Cathode

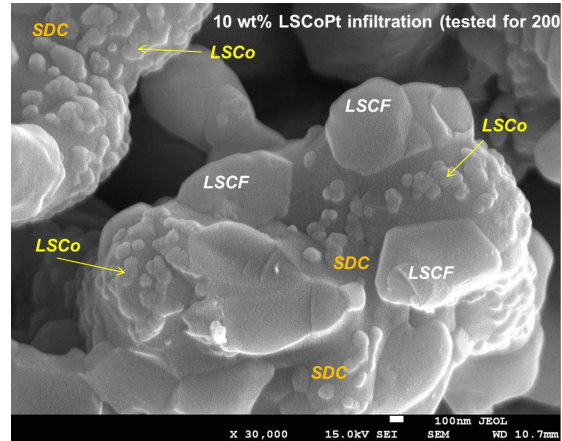
2009

# Electrode Design and Engineering

Electrode Infiltration Capabilities

## Industrial Scale Electrode Infiltration Technology

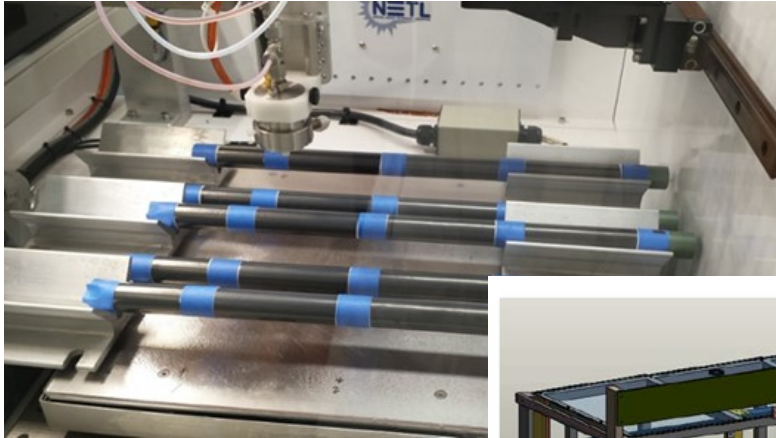
- NETL has developed and patented a single-step cathode infiltration technique that can be utilized by commercial SOFC manufacturers to improve their cell performance and durability
  - Proven performance gains of
    - 10% peak power increase
    - 33% reduction in degradation rate
    - 200% lifetime increase
  - Low-cost (\$0.006/cm<sup>2</sup>)
  - Scalable
  - Ready for technology transfer
    - Collaboration with industry
    - NDAs executed
  - Ready for any cell geometry



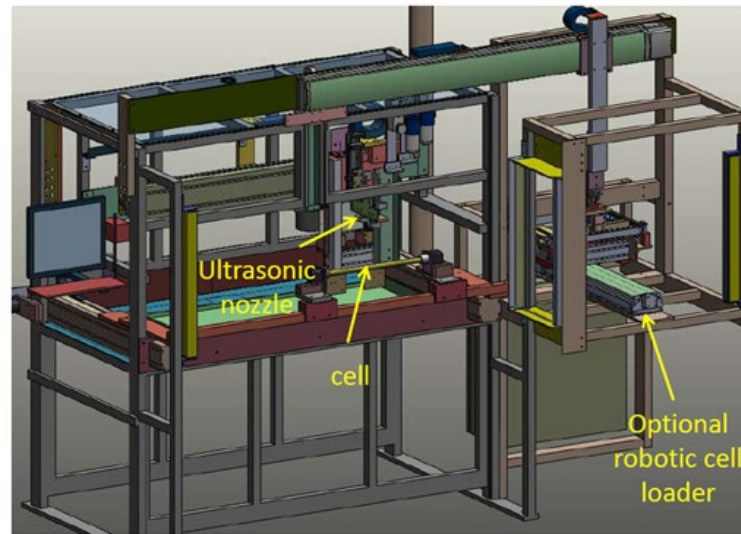
Secretary Perry inserting an SOFC (Atrex Energy) into the Sono-Tek Spray Coater



## Demonstration on Commercial Developer SOFC Stack



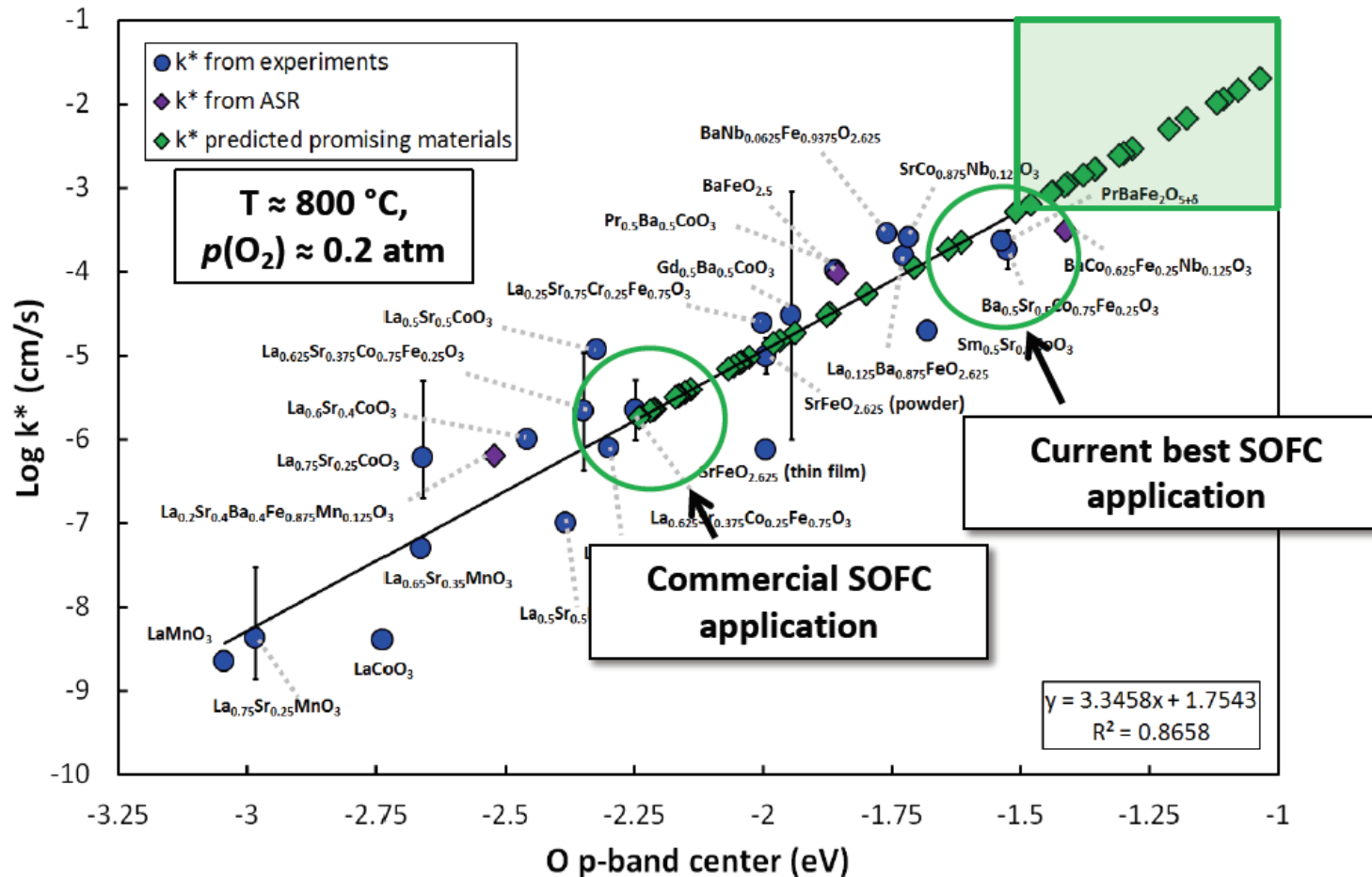
Spray infiltration  
process at  
NETL



- Results showed the infiltration process applied to Atrex Energy tubular cells **reduced the processing time** required for cathode infiltration to one day.
- Atrex Energy constructed a **factory-scale automatic spraying infiltration system** based on the NETL's technology.
- A **1.5 kW stack was tested** utilizing the infiltration process. The process improved the Atrex fuel cell stack performance without noticeable degradation for 2000 hours.

Factory-scale automatic spray infiltration system installed at Atrex Energy

## Computational Design of Materials

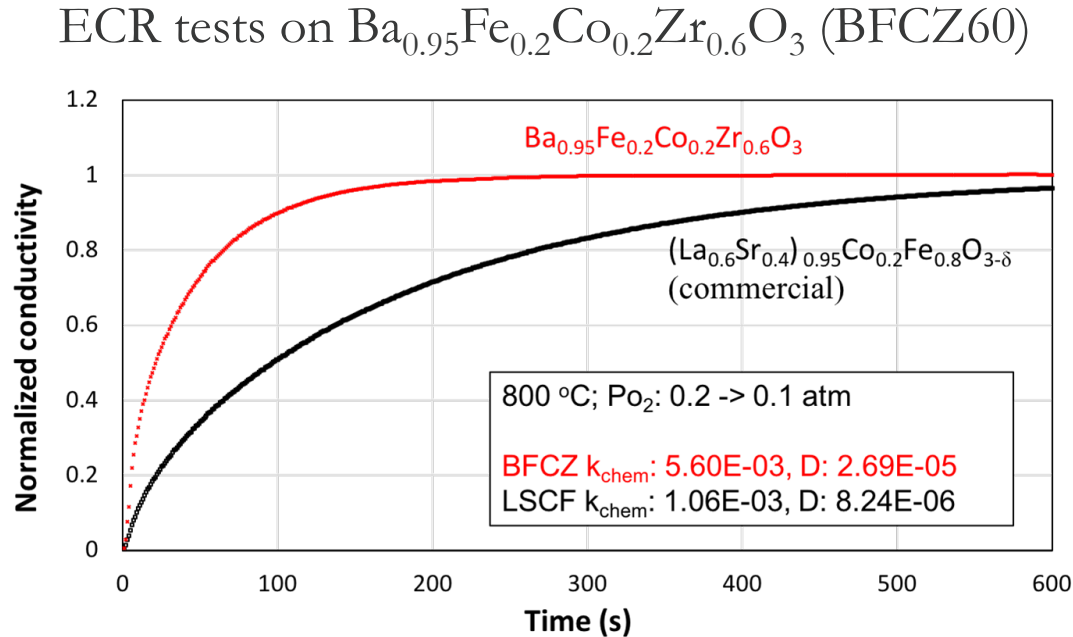


- Utilizing computational chemistry techniques such as density functional theory, we can theorize a more active electrode material
- Theory shows that there are several materials with the potential to be exceptionally active compared to today's materials.

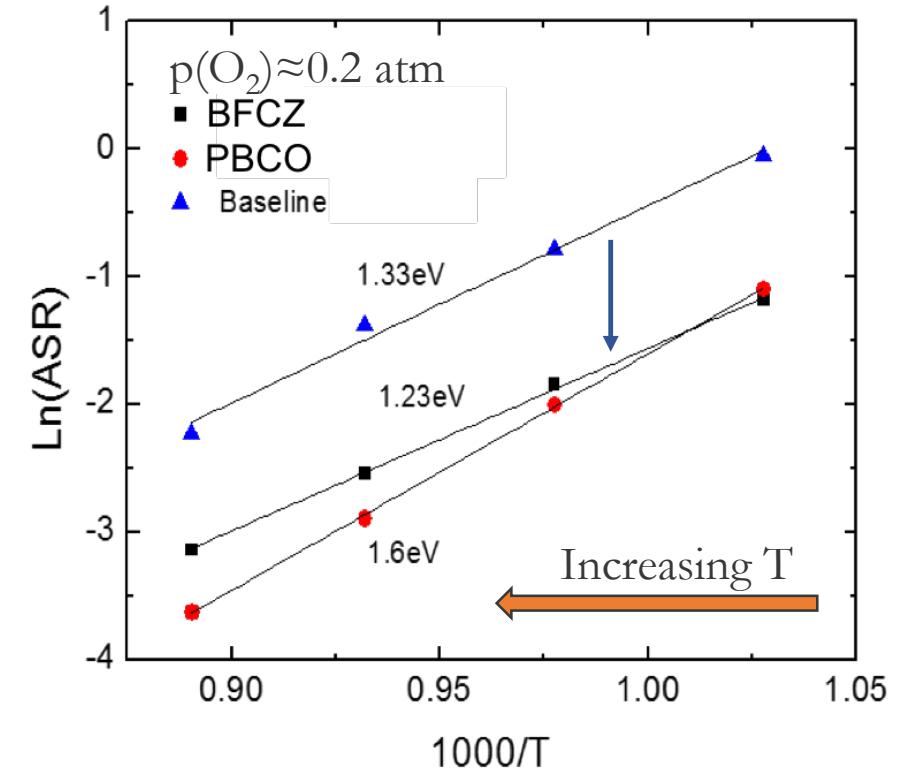
Linear correlation of  $k^*$  and O p-band center [1]



## Experimental Verification



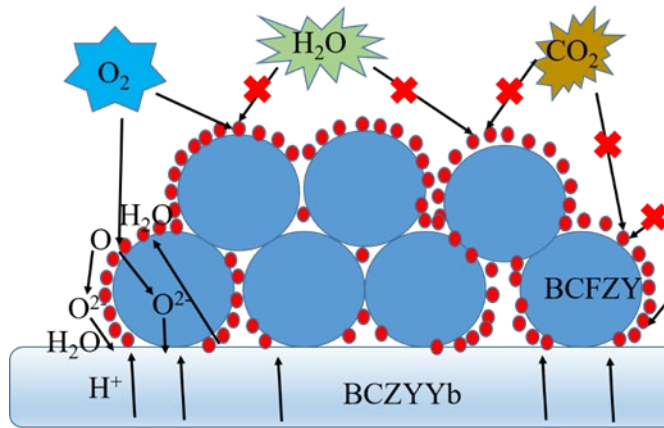
- ECR measurements on BFCZ containing 60% Zr resulted in **5× higher  $k_{\text{chem}}$**  and **3× higher  $D_{\text{chem}}$**  than LSCF



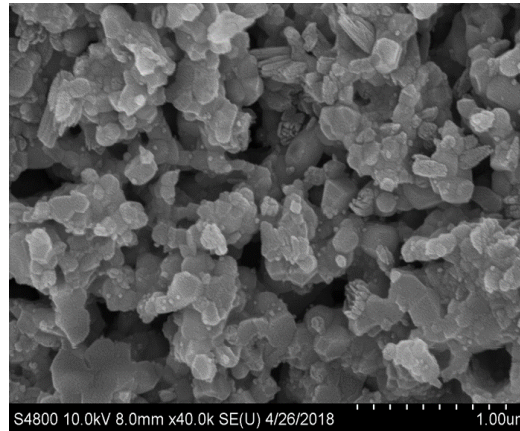
- Infiltration of LSM cathode with BFCZ containing 75% Zr resulted in **reduced ASR by about 10×**, and comparable performance to  $(\text{PrBa})_2\text{Co}_2\text{O}_{5+x}$

## Proton Conducting SOFC Electrodes

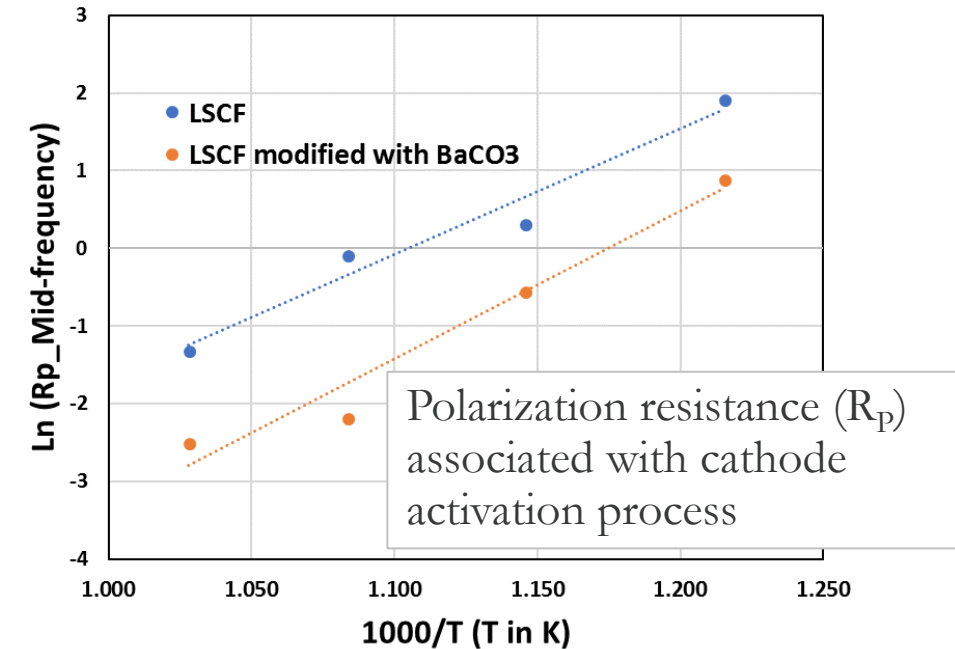
- Electrolyte: BCZYYb, Cathode: BCFZY or LSCF
- Electrocatalyst:  $\text{BaCO}_3$ , nano-BCFZY, etc.**



Cathode infiltration in Proton SOFCs



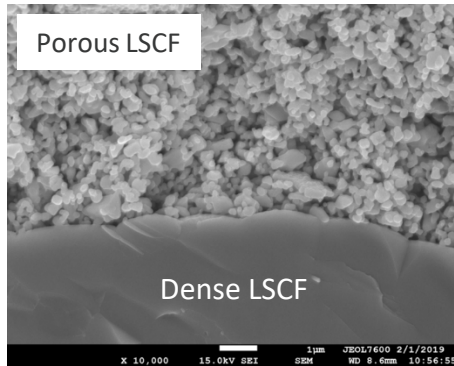
LSCF electrode infiltrated with  $\text{BaCO}_3$



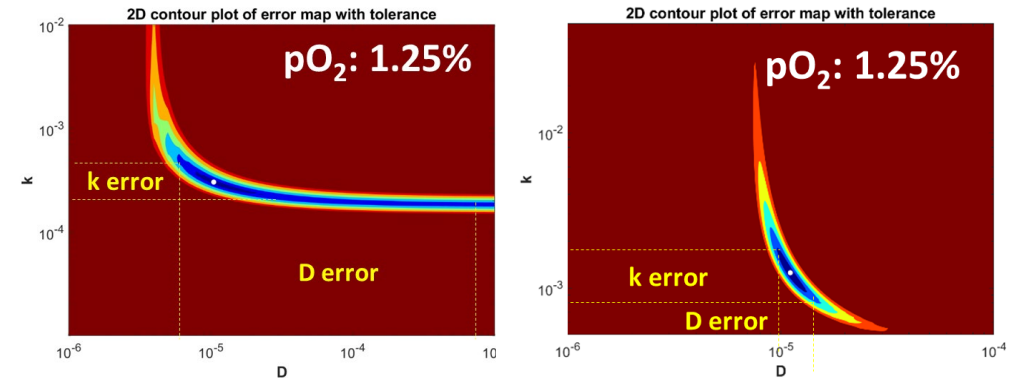
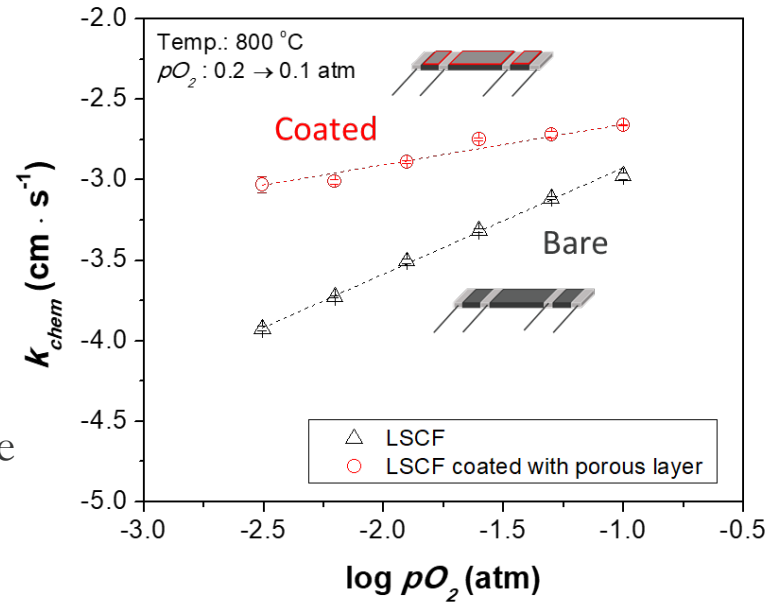
- The ASR of the  **$\text{BaCO}_3$ -infiltrated LSCF cathode** ( $0.08 \, \Omega \cdot \text{cm}^2$ ) is significantly less than that of the pure LSCF cathode ( $0.27 \, \Omega \cdot \text{cm}^2$ ) at  $700^\circ\text{C}$



## Modified ECR (Electrical Conductivity Relaxation)



Porous LSCF layer on dense LSCF ECR sample



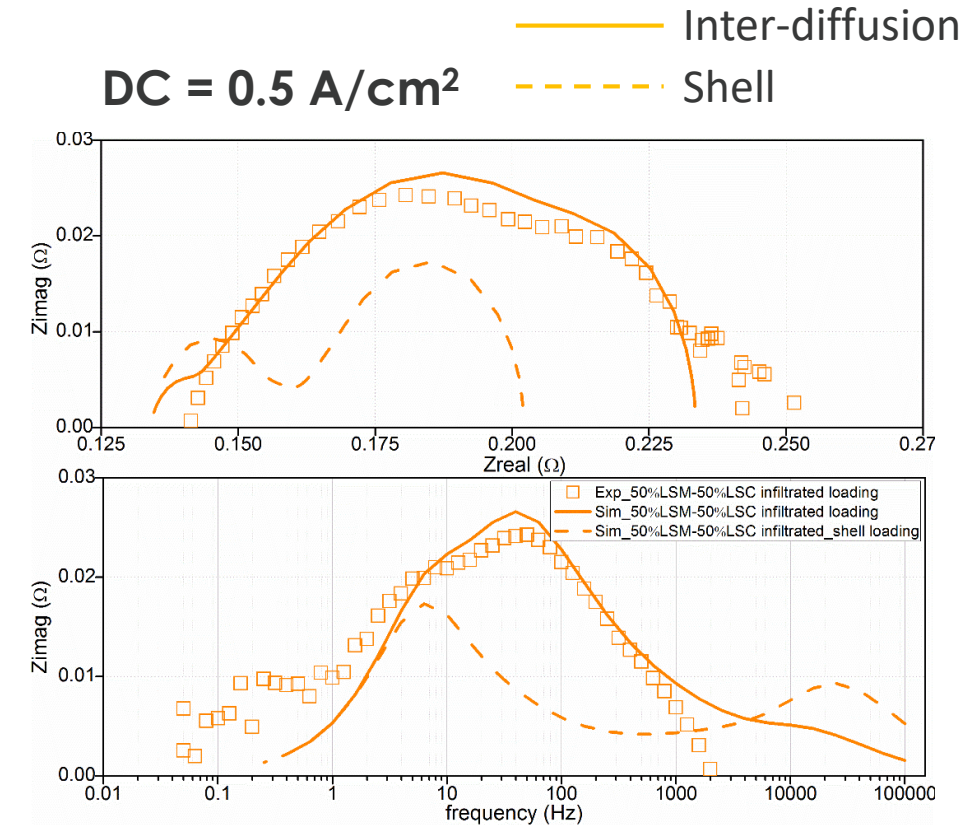
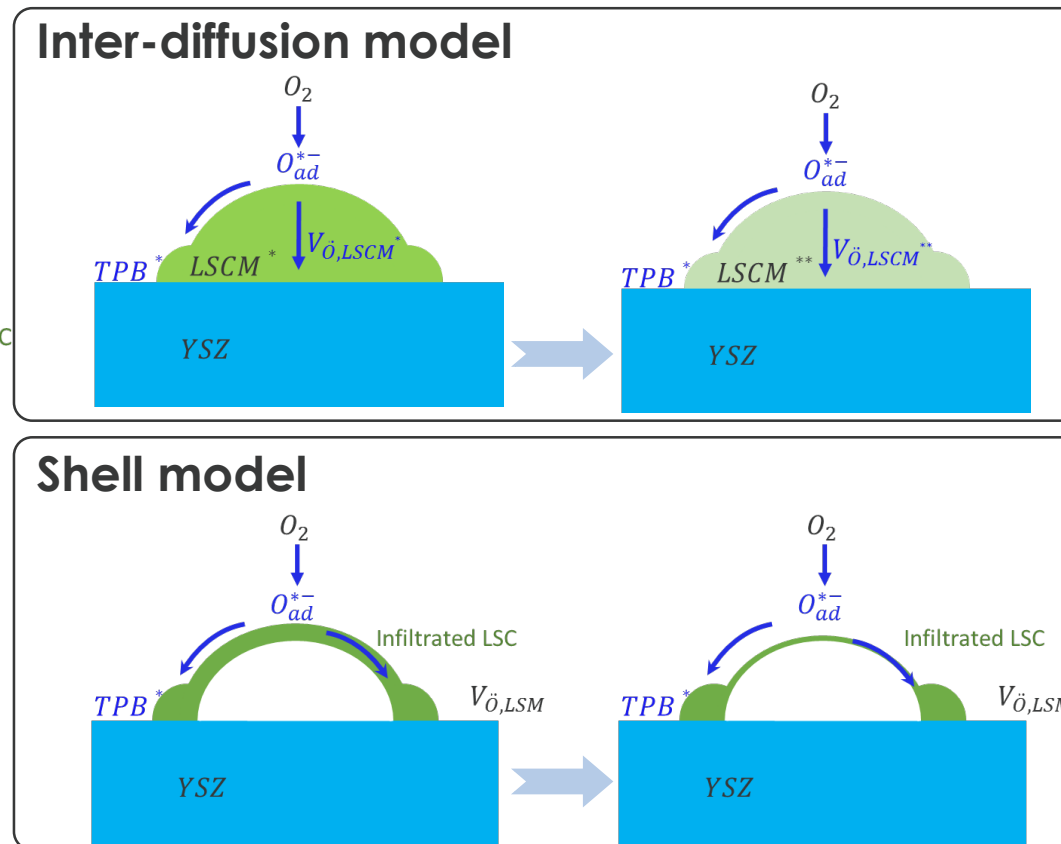
Error map for the calculated  $k_{chem}$  and  $D_{chem}$  at  $pO_2 = 1.25\%$  (a) bare LSCF, (b) LSCF coated with porous layer.

- A novel approach of determining bulk diffusion coefficient ( $D_{chem}$ ) using the electrical conductivity relaxation (ECR) was developed.
- Coating the surfaces of bar samples with porous, in-kind particles (e.g. porous LSCF on dense LSCF bar sample) enabled reduction in the characteristic thickness ( $L_c$ ) and determination of  $D_{chem}$  values with minimal error, which couldn't be achieved by conventional methods.

# Modeling of Infiltrated Electrode

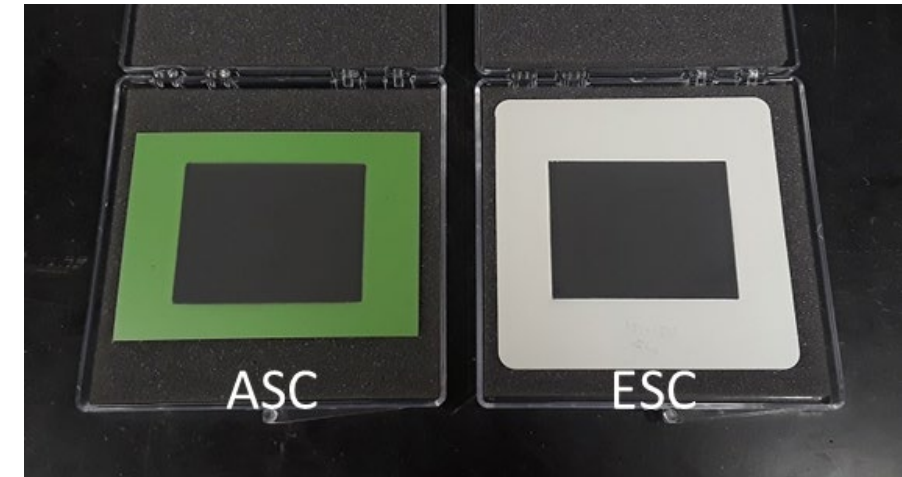
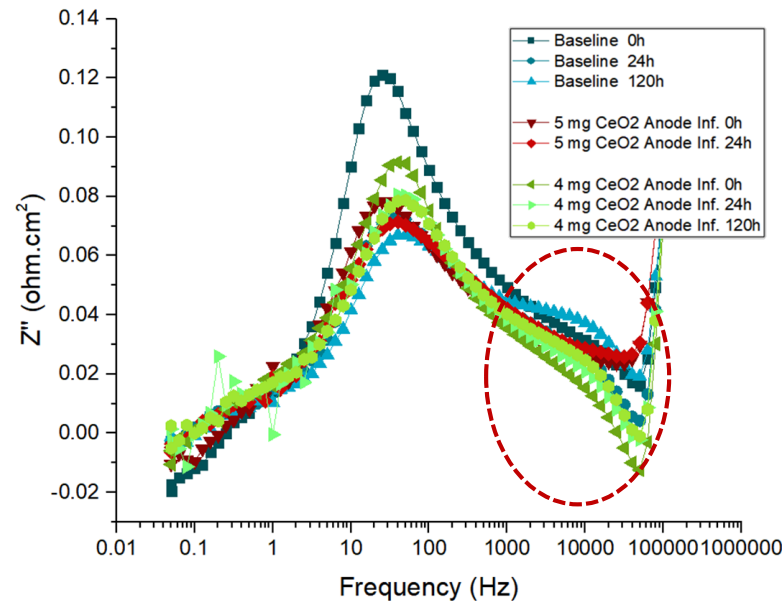
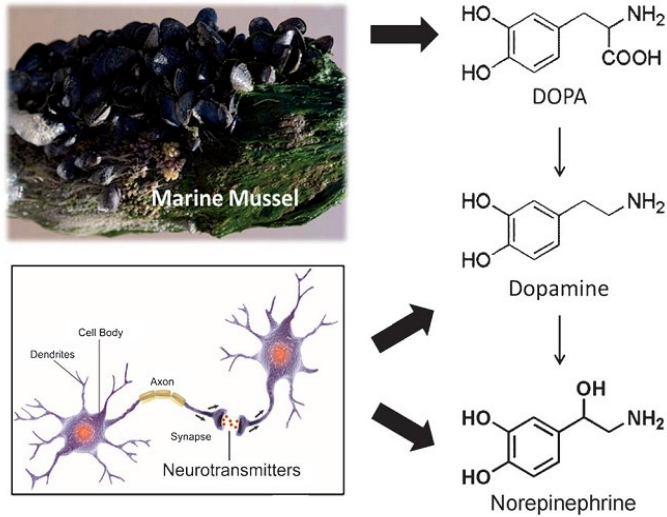
Performance of infiltrated LSM/YSZ composite cathodes is investigated via multi-physics simulations with a multistep charge transfer oxygen reduction reaction (ORR) mechanism.

## LSC infiltrated LSM/YSZ cathode: Inter-diffusion Model vs. Shell Model





## Bio-Surfactant Assisted PRE-REDUCED SOFC Anode Infiltration



Electrocatalyst-infiltrated planar cells

- pNE offers smoother and more uniform coating

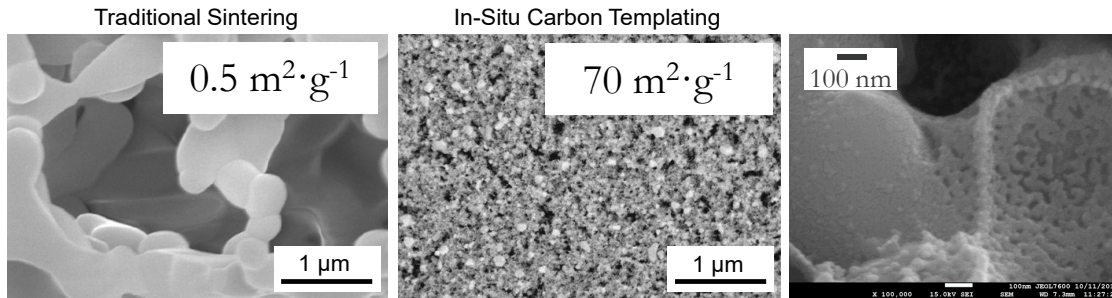
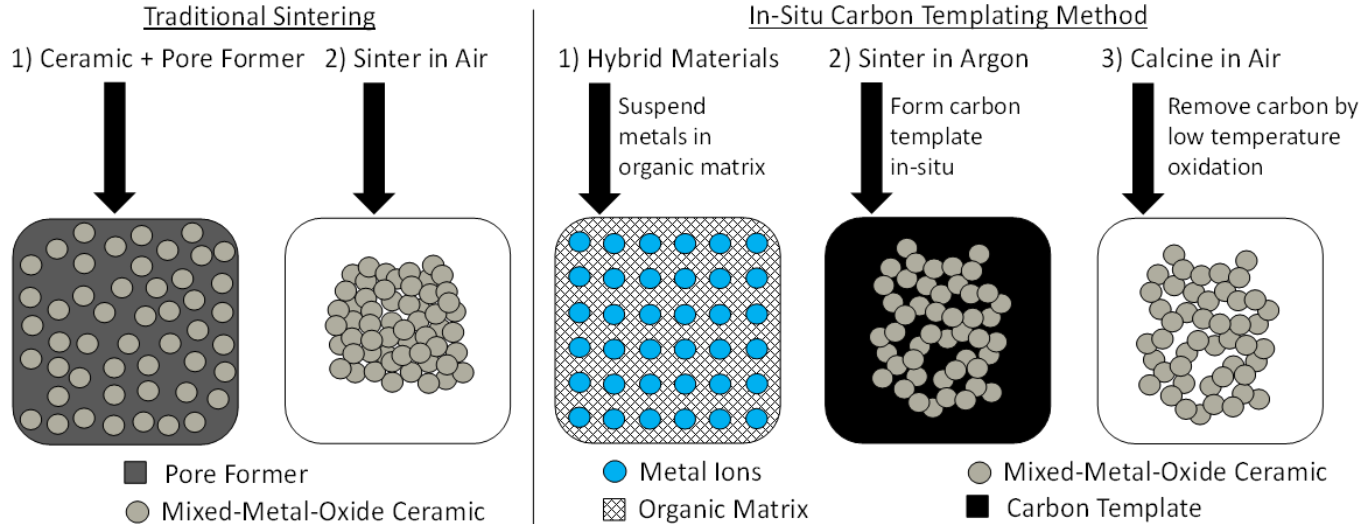
H. Lee, et al., *Angew. Chem. Int.*, (2013) 9187

- Anode resistance of industry cells decreased by bio-surfactant assisted infiltration

- The bio-surfactant assisted infiltration protocol was **verified on industrial planar fuel cells.**

# High Surface-Area Nanostructured Cathodes

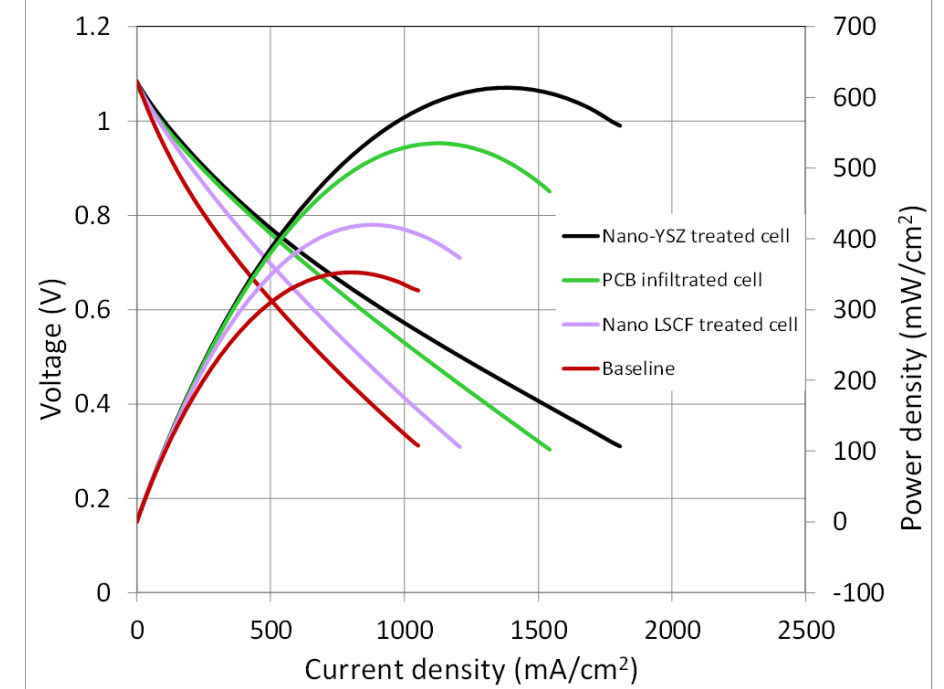
## Hybrid Materials-Assisted Templating



Traditional  
sintering

In-situ carbon  
templating

Nano-YSZ  
infiltrated  
LSM-YSZ



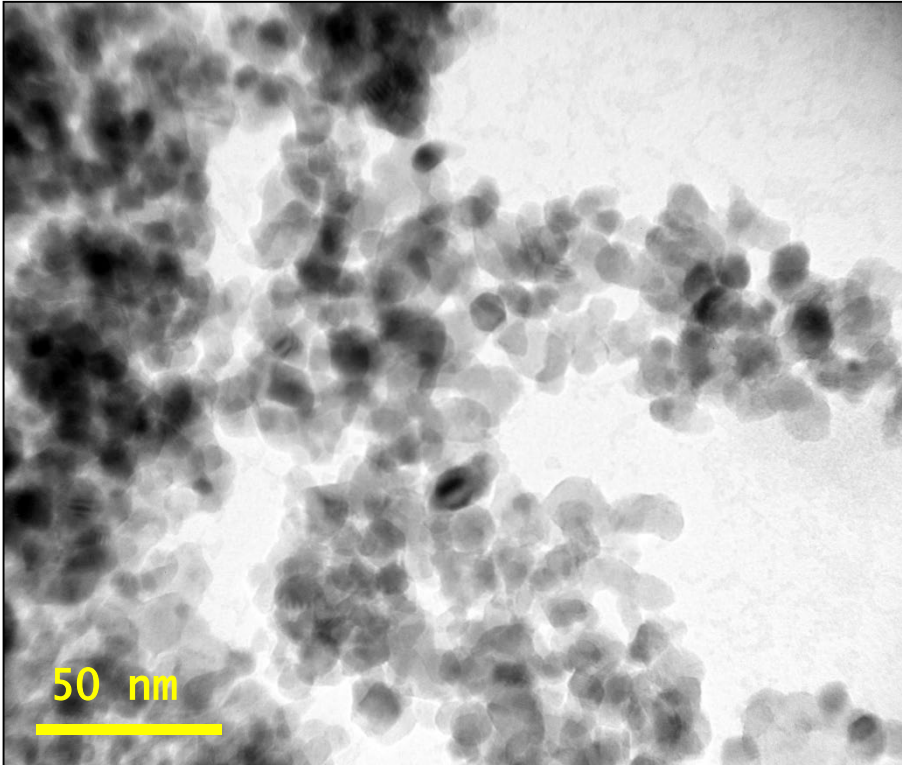
**Nano-YSZ infiltrated LSM-YSZ cathode showed stable performance:**

- Nano-YSZ infiltrated: **0.67% over 200 h**
- (PrBa)CoO<sub>x</sub> infiltrated: **1.86% over 200 h**

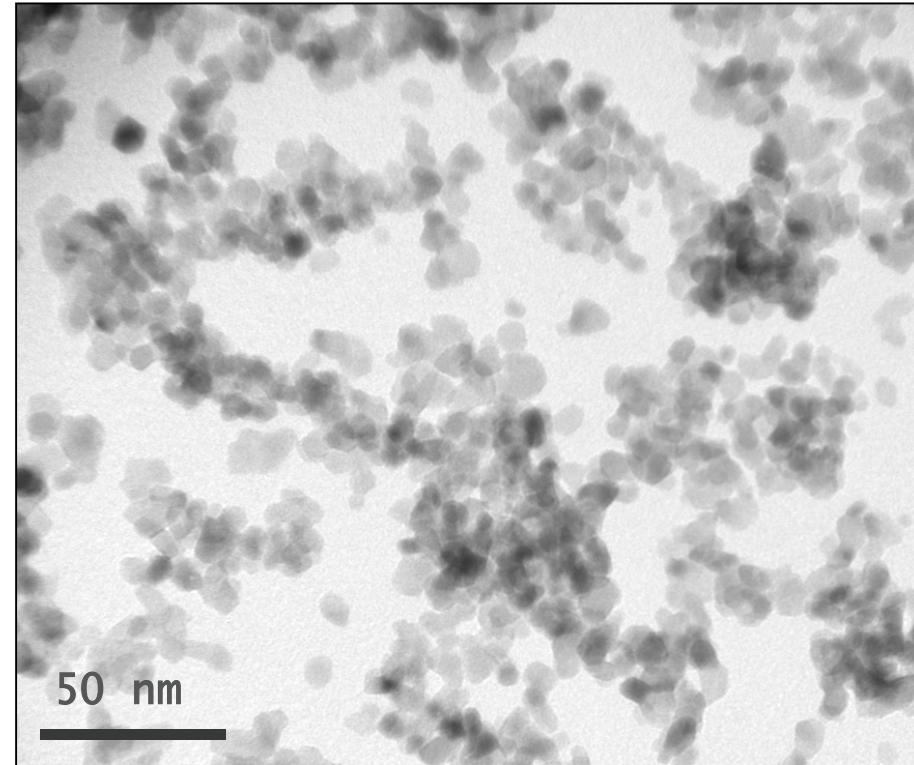


# Hybrid Materials-Assisted Templating

YSZ - 850°C in N<sub>2</sub>, 700° C in Air



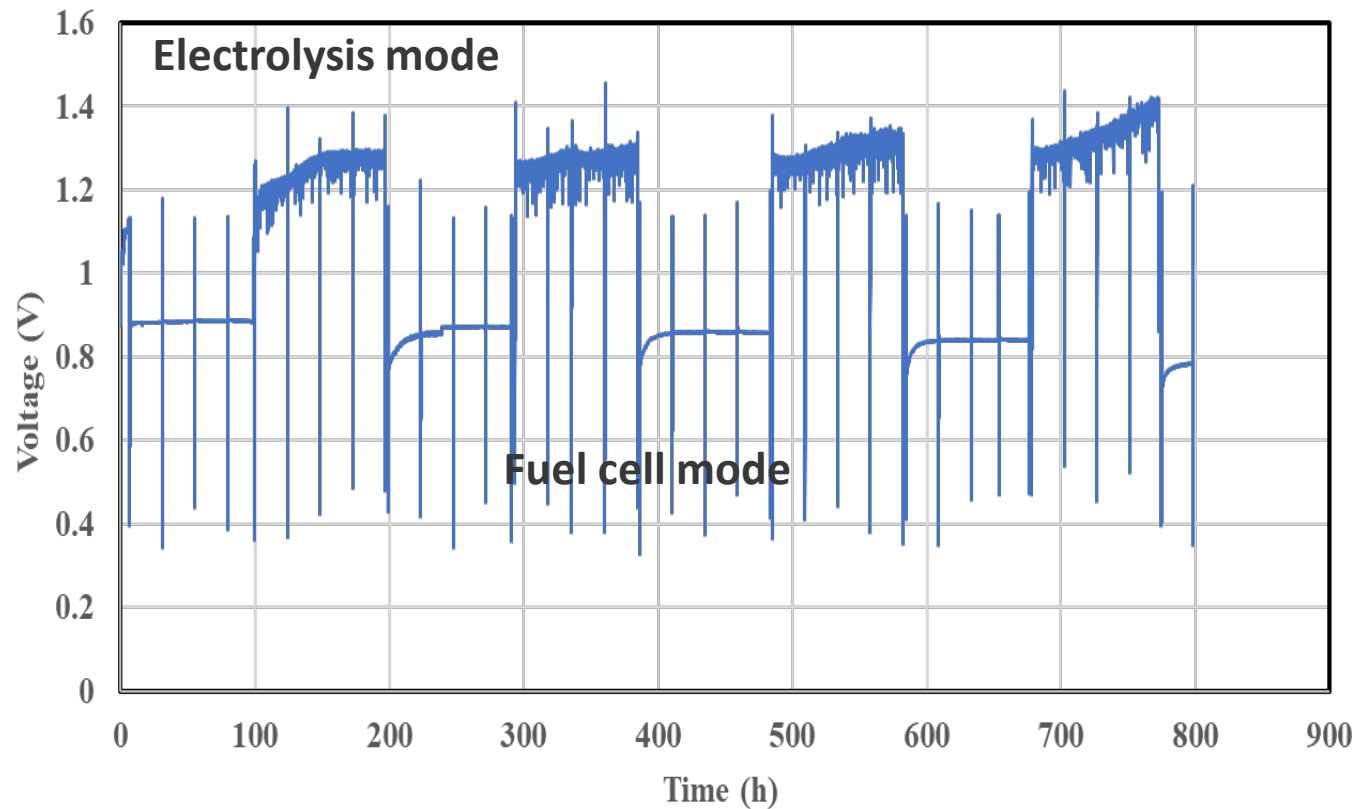
YSZ - 1250°C in N<sub>2</sub>, 700° C in Air



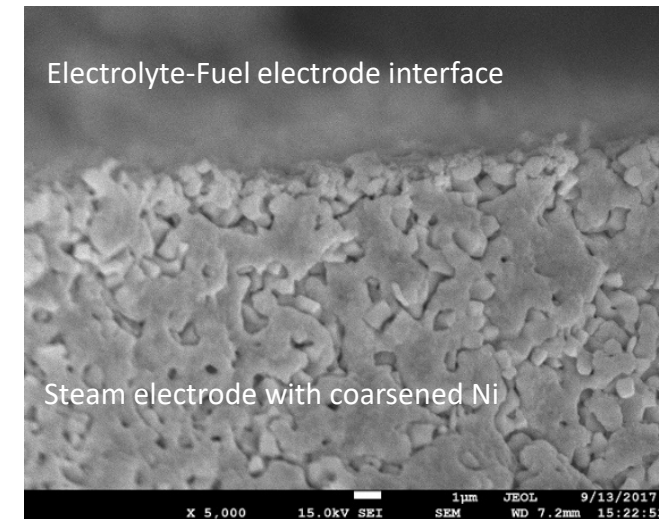
The properties of the nanoparticles are controlled by varying the processing conditions

# Reversible Mode Operation

## Recent Effort – Reversible Mode Operation



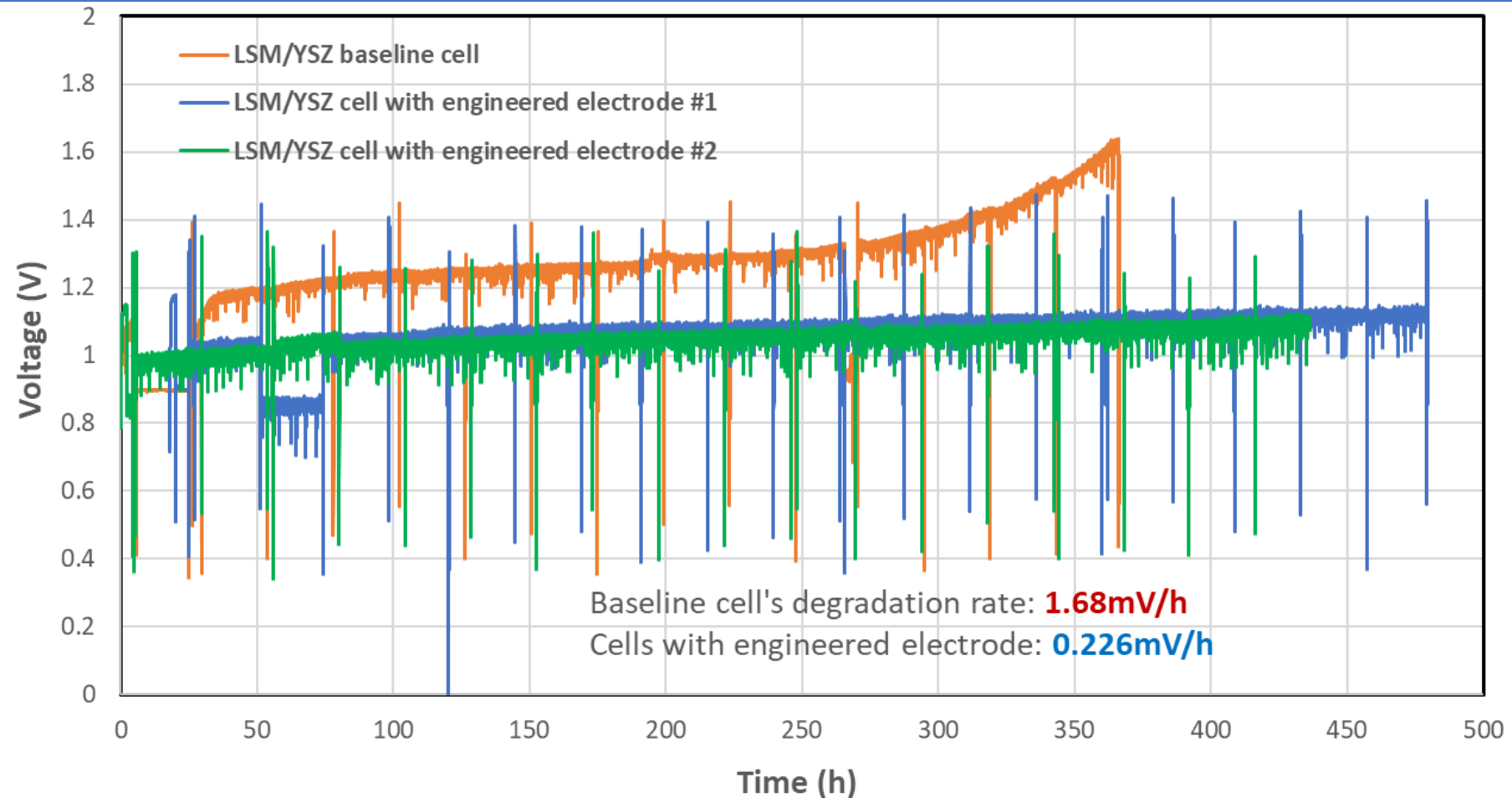
- Anode-supported commercial cell (**LSM/YSZ cathode**)
- Temperature: **800°C**
- Electrolysis (cathode): **60% H<sub>2</sub>O** – 10% H<sub>2</sub> – 30% N<sub>2</sub>
- Fuel Cell (anode): **25% H<sub>2</sub> – 75% N<sub>2</sub>**



Delamination and Ni phase coarsening were evident from the cell tested under high steam conditions

## Solid Oxide Electrolysis Cell with Engineered Electrode

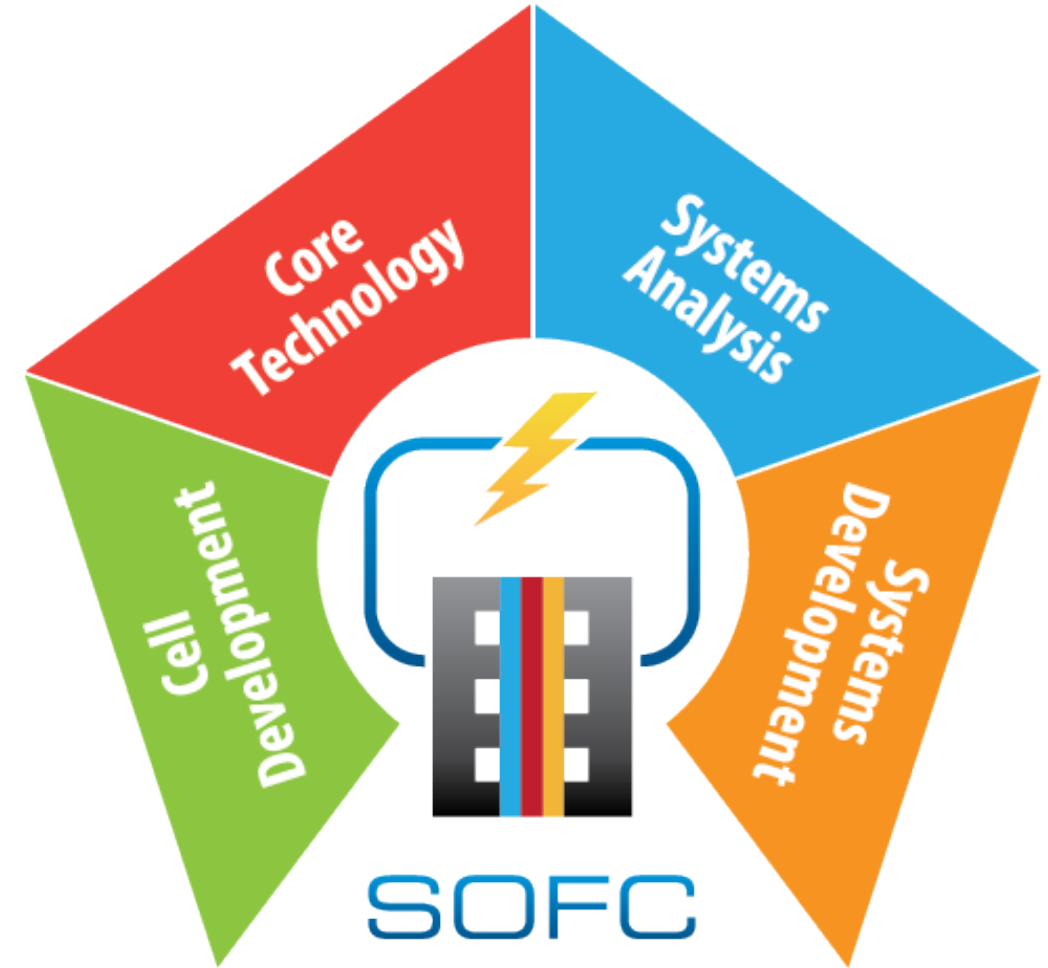
- An **LSM/YSZ** cell with engineered electrodes
- Temperature: **800°C**
- Electrolysis (cathode):  
**60% H<sub>2</sub>O** – 10% H<sub>2</sub> – 30% N<sub>2</sub>
- Fuel Cell (anode): **25% H<sub>2</sub>** – 75% N<sub>2</sub>





# Cell and Stack Degradation

Technologies and Toolsets Under Development

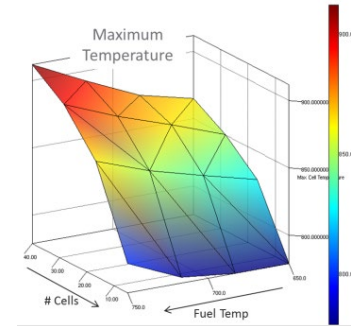


# Task 2 Background

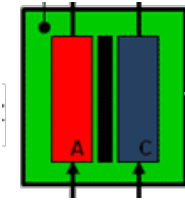
NETL/PNNL Collaboration to Complete Scaling Process

Need design and engineering at several scales to facilitate wide-scale SOFC commercialization

Link NETL and PNNL models at different scales to inform system level and life cycle analyses

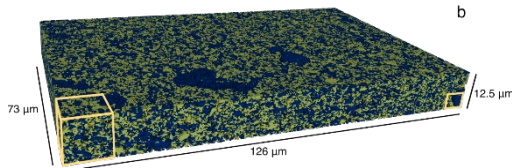
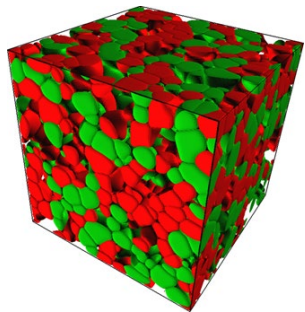


Response Surface Analysis

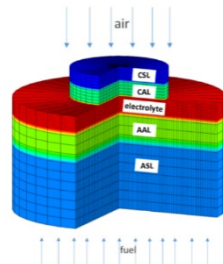


Reduced Order Model (ROM)

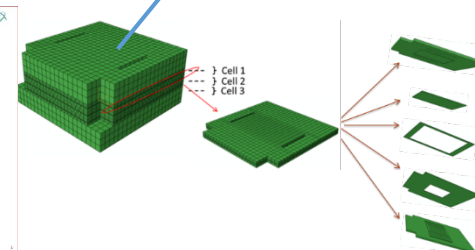
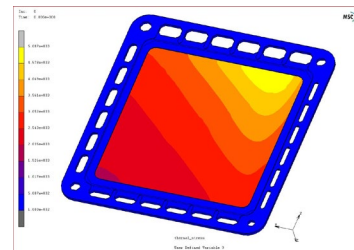
Increasing Scale



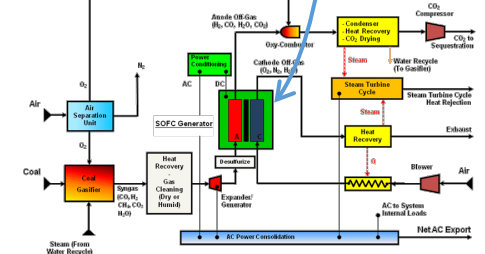
Electrode Microstructure



Single Cell



Multi-Cell Stack



IGFC System Model

NETL

PNNL

NETL

# Enabling SOFC Technology through R&D at NETL

## Predictive Modeling – Reduction of Cost for SOFC Systems

### TOOL RELEASE

Release of SOFC Predictive Modeling Tool into public domain

### DEMONSTRATION

Fully integrate all degradation models into SOFC operation model

### MATURATION

Demonstration of degradation models integration into SOFC operation model

### DEVELOPMENT

Critical SOFC degradation modes identified, expansion of SOFC operation model

### DISCOVERY

Proof of Concept

### Concept to Market Readiness

2020

Demonstrate how microstructure and operating conditions affect plant-level cost-of-electricity

2018-19

Scale-bridge from microscale to cell to stack/system level (collaborate with PNNL)

2015-18

Use of plasma-FIB to create world's largest reconstruction commercial developer cells

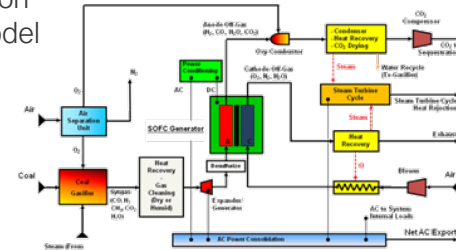
2013-15

Use of focused-ion beam (FIB) to reconstruct electrodes, evaluate operationally-relevant properties

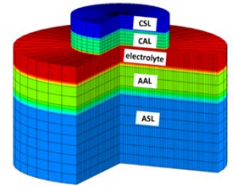
2012

Concept of Predictive "Hurricane" Model for SOFC

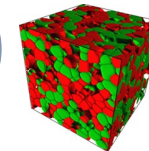
Integrated Gasification Fuel Cell System Model



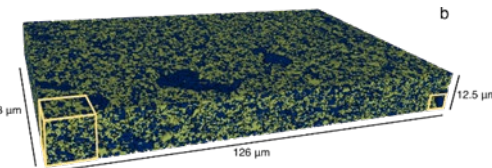
SOFC Operation "Multi-physics" Model



Particle coarsening degradation model



Cathode microstructure reconstruction

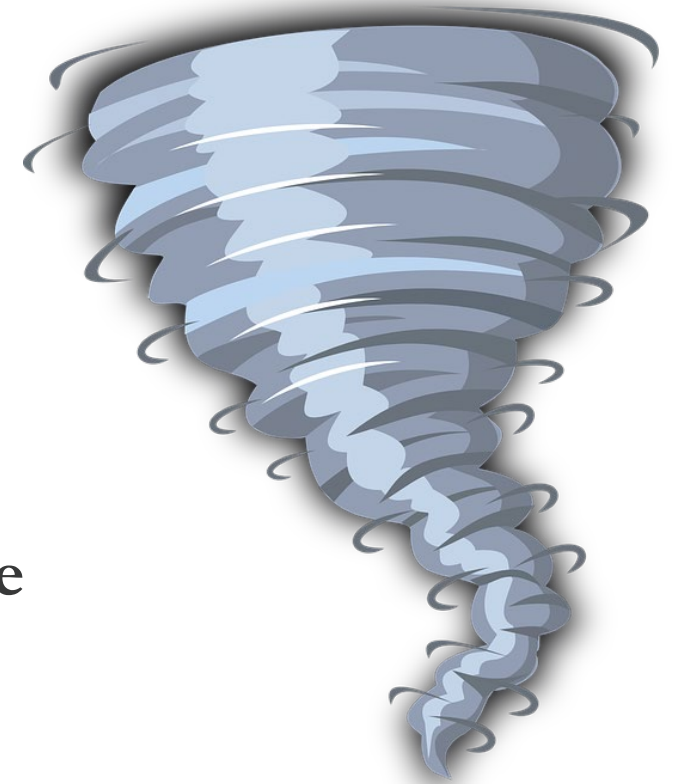


Hurricane prediction concept



# Recent Progress

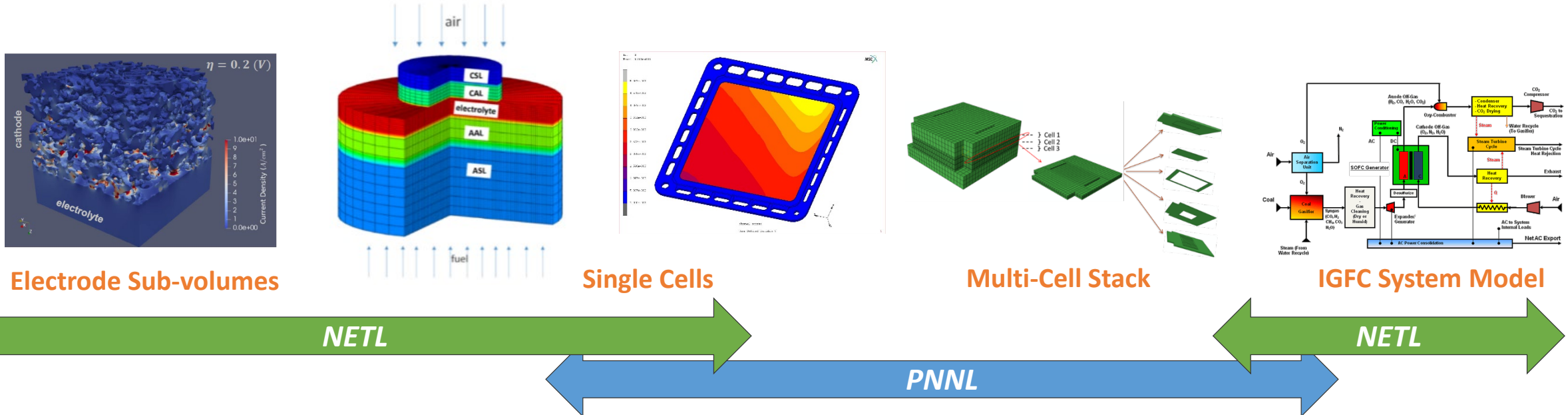
- **Converting modeling tools to open-source platform(s)**
- **Integrated multiple degradation modes into predictive framework**
  - Particle coarsening, secondary phase formation, contaminant interactions, etc.
  - Utilizing principle component analysis and machine learning to understand complex model parameter interactions
- **Working with SOFC commercial developer to demonstrate high-temperature fiber optic sensors via NDA**
  - Temperature and gas composition measurement
- **Predictive modeling tools scheduled for initial release by March 2021**



# World Leading SOFC Research

Modeling from Atoms to Cost-of-Electricity

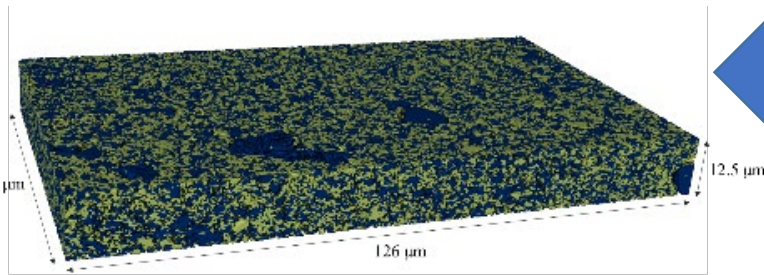
- The SOFC group at NETL is the only solid oxide fuel cell research team capable of modeling from the atomistic scale to the system scale
  - Atoms to cost-of-electricity



# World Leading SOFC Research

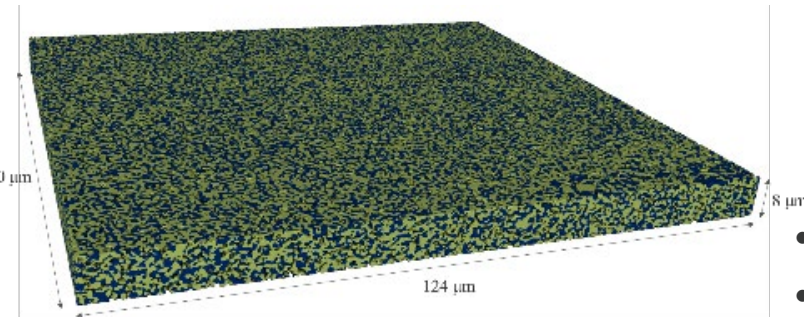
Production of High-resolution 3-D SOFC Microstructure Reconstructions

- The SOFC group at NETL is the only solid oxide cell research team that has published high resolution electrode reconstruction datasets
  - Crucial for accurate characterization of electrode heterogeneity



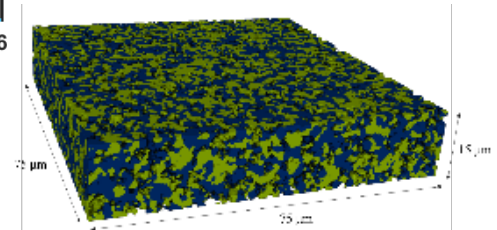
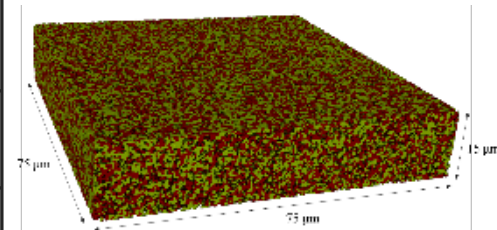
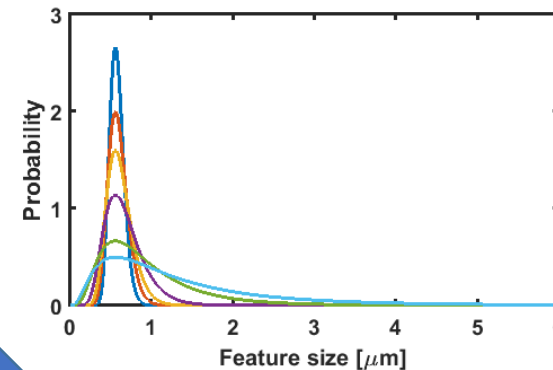
## Real Microstructures

- Predict behavior of industrial cells
- Baseline for improving particular cell



## Synthetic Microstructures

- Explore more features
- More efficient than creating/imaging 1000s of real cells

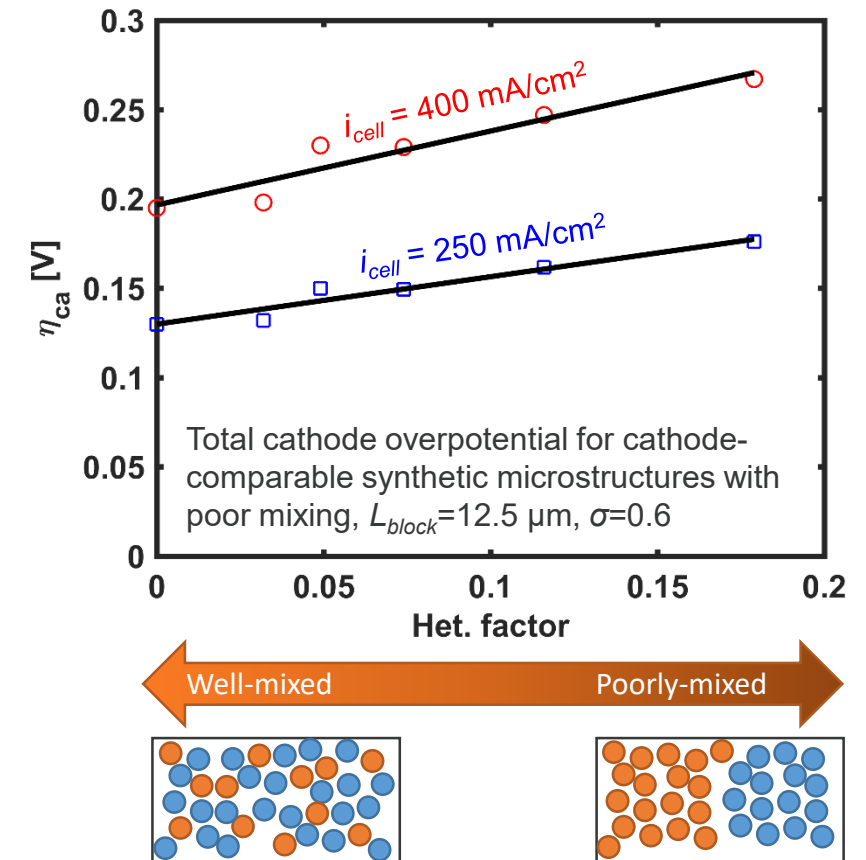
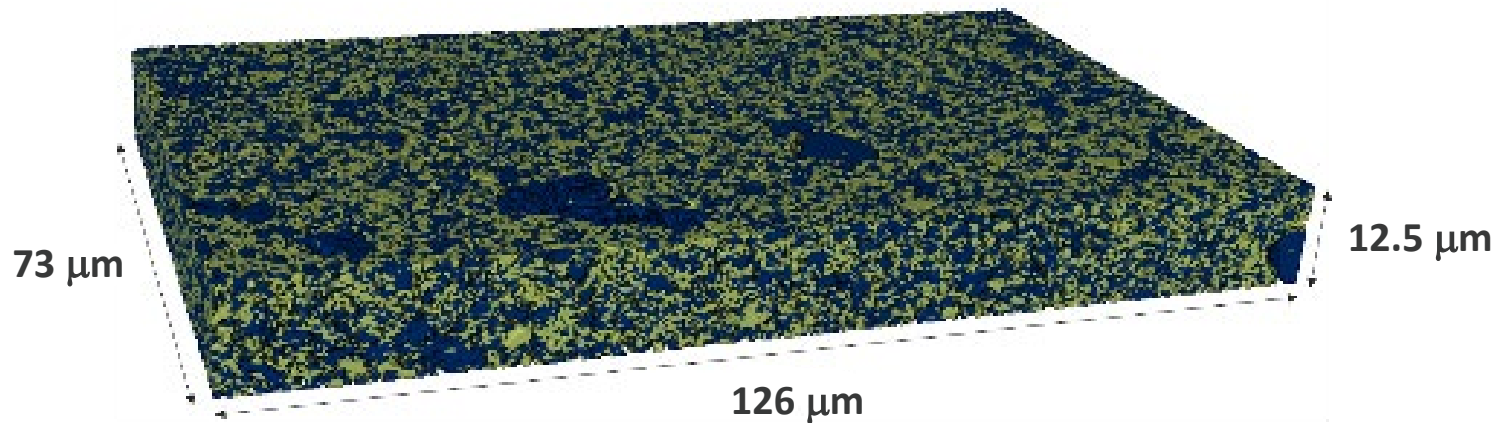




# World Leading SOFC Research

Microstructural Heterogeneity Characterization and Simulation

- The SOFC Group at NETL is the world leader in characterizing and simulating heterogeneity in porous electrodes
  - **First** in using machine learning to create synthetic microstructures that more accurately capture heterogeneity in real electrodes

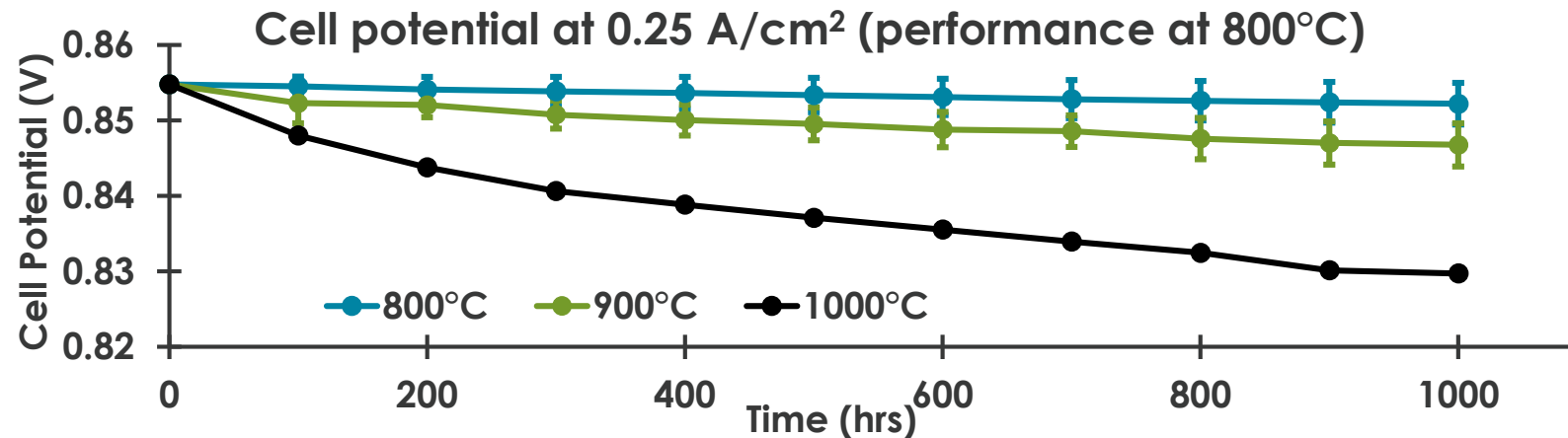


Multiphysics modelling by Dr. Hunter Mason

# World Leading SOFC Research

Synthetic Microstructures Explored to Date

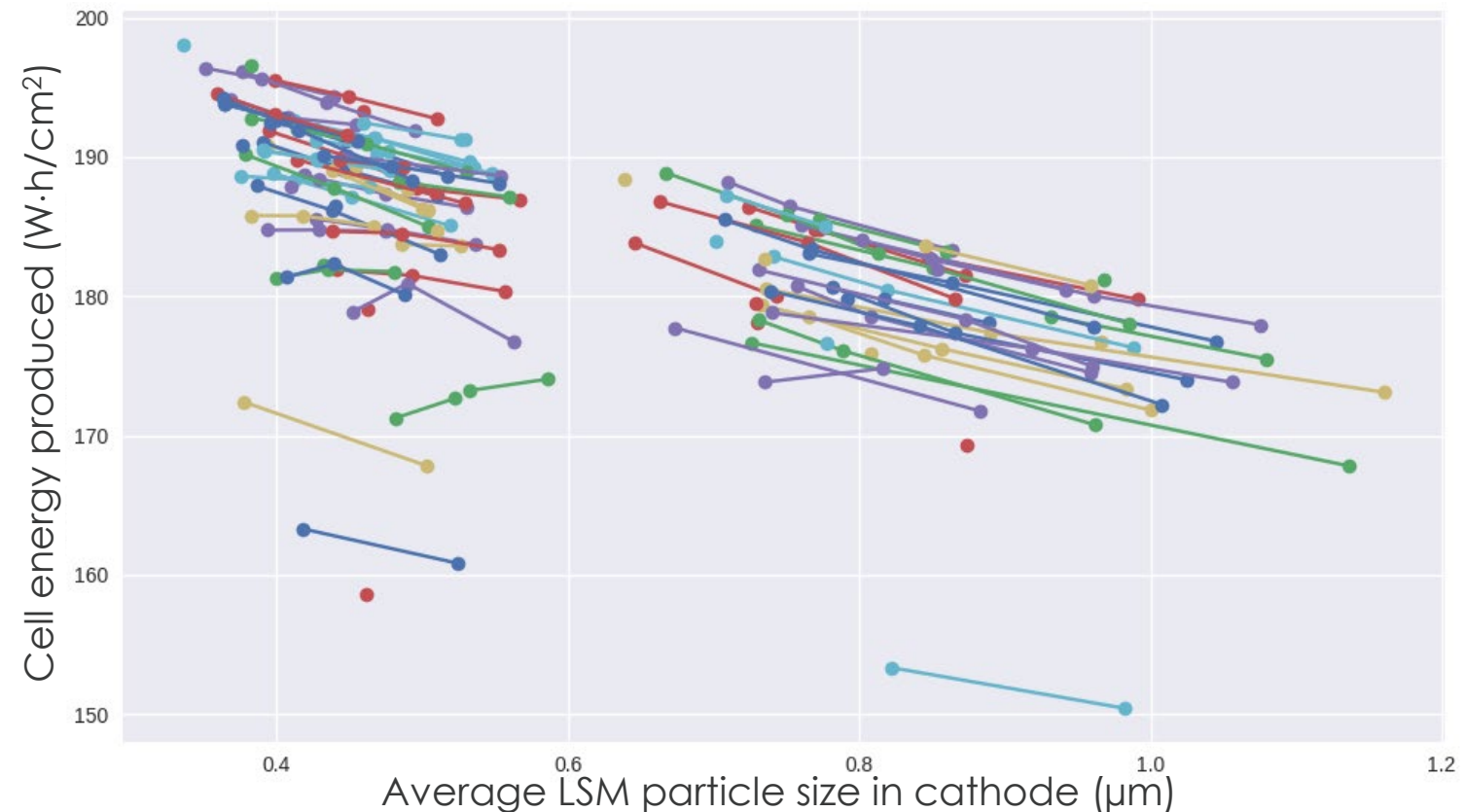
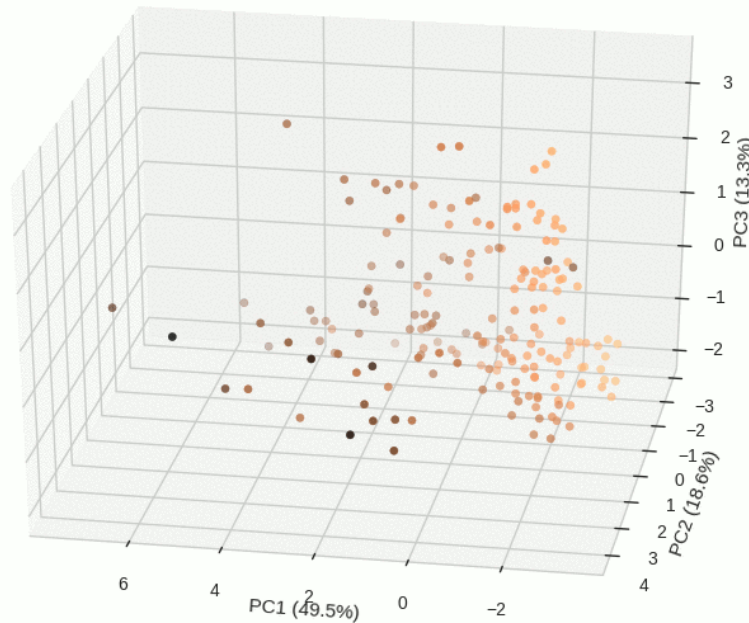
- The largest and broadest bank of unique electrode microstructures (45,000) has been generated by NETL
  - JOULE 2.0
  - Varied phase fractions, phase fraction distributions of three phases (COMPOSITION)
  - Varied particle sizes, particle size distributions (MICROSTRUCTURE)
- Simulated particle coarsening of 500+ unique cathode microstructures



# World Leading SOFC Research

Connecting Microstructure to Cost-of-Electricity

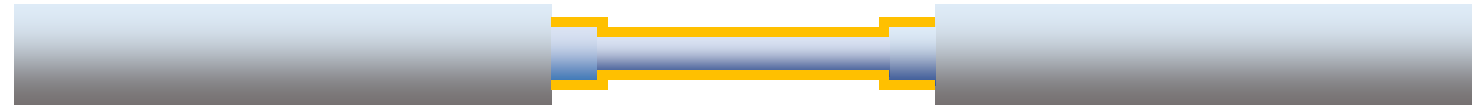
- NETL has developed a sound methodology to connect microstructural changes to the cost-of-electricity
  - Figure of Merit:  $\text{W}\cdot\text{h}/\text{cm}^2$
  - Trends discovered via principal component analysis





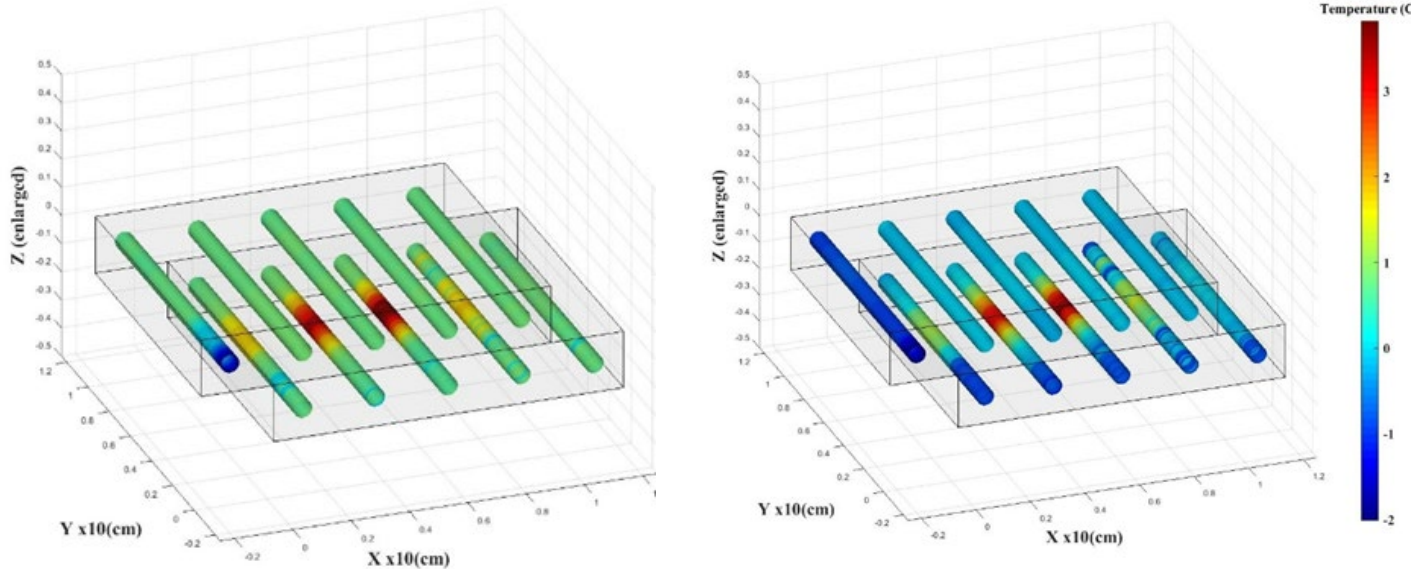
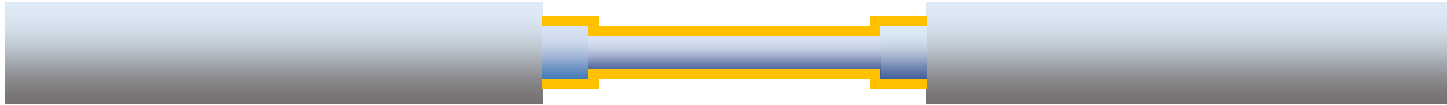
# High Temperature Optical Fiber Sensor

- Multi-application technology under development for high temperature sensing
  - Demonstrated in SOFC environment
- In-situ sensing of
  - Temperature distribution
  - Gas composition
    - CO, CH<sub>4</sub>, H<sub>2</sub>, CO<sub>2</sub>
- Novel coatings for optimal selectivity
- Of interest to several SOFC commercial developers

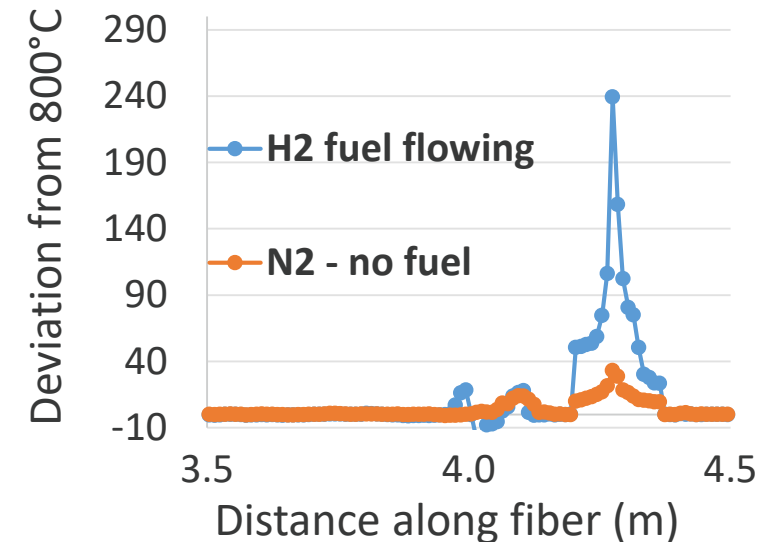


# High Temperature Optical Fiber Sensor

## Distributed In-situ Temperature and Gas Composition Sensing



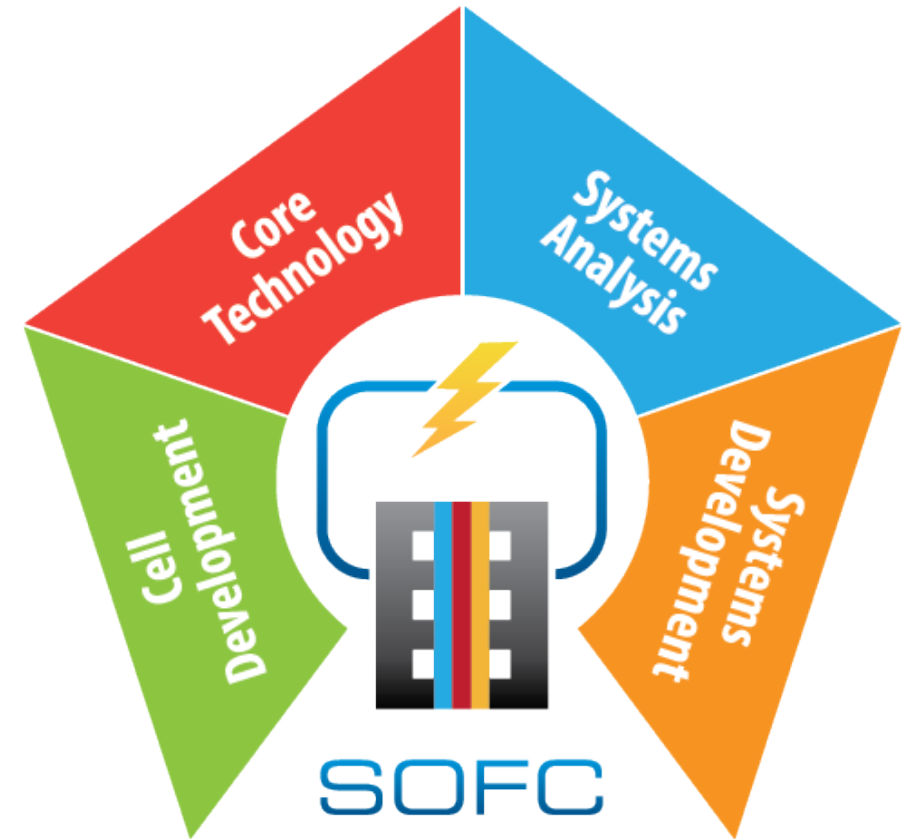
Thermal transients at 30 and 90 s from 5×5 cm<sup>2</sup> ASC at 750°C with H<sub>2</sub> fuel after 2A load



Failure detection: Temperature spike from cracked cell at 800°C

# Systems Engineering & Analysis

Pulling It All Together



U.S. DEPARTMENT OF  
**ENERGY**



## Techno-Economic Analysis of Integrated Gasification Fuel Cell (IGFC) Systems

**Motivation:** Techno-Economic Analyses (TEA) of SOFC systems are used to provide DOE-FE and the public with cost and performance information for SOFC technologies

**Objective:** This study updates performance, cost, and pathway information for IGFC systems to aid in the development of targeted R&D approaches for SOFC fueled by gasified coal

### Study Details

**Pathway cases are developed to demonstrate incremental progress from state-of-the-art to advanced SOFC performance**

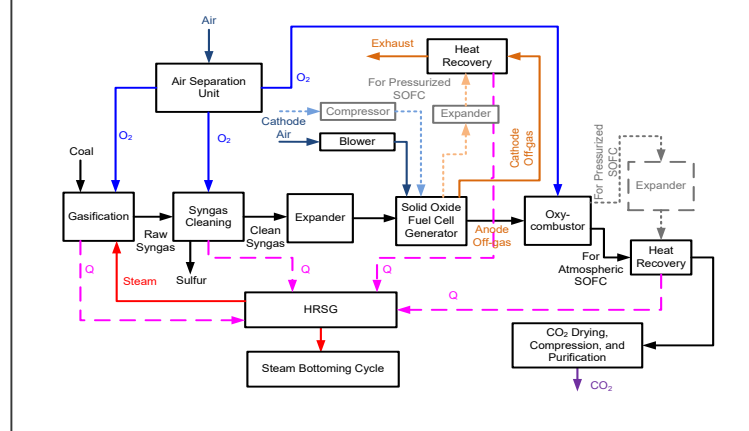
- Includes technology updates of other critical system components such as carbon capture, gasification units, etc.

**Study updates a previously released report (2013). Updates include:**

- Year dollar update to 2018\$, performance data generated by PNNL stack model ROM, SOTA vs Advanced SOFC, vent gas recirculation concept, capture and non-capture cases, and updated gasification costs

**Report to be Released June 2020**

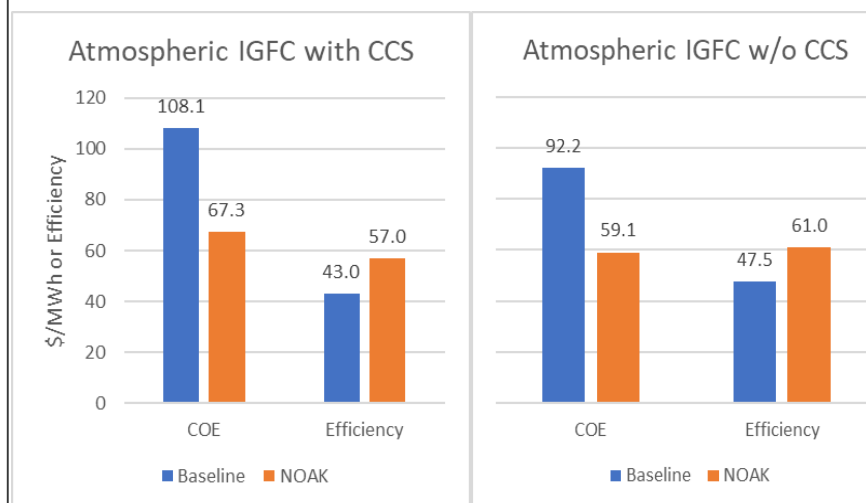
### IGFC Configuration



### Principal Investigator

Gregory A. Hackett

### Preliminary Results



Report includes detailed expansion of results for pressurized cases and more

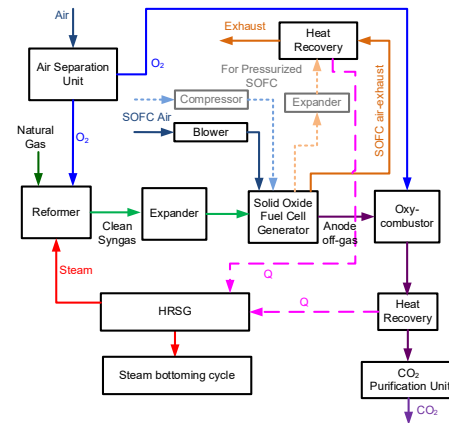
**Motivation:** Techno-Economic Analyses (TEA) of SOFC systems are used to provide DOE-FE and the public with cost and performance information for SOFC technologies

## Study Details

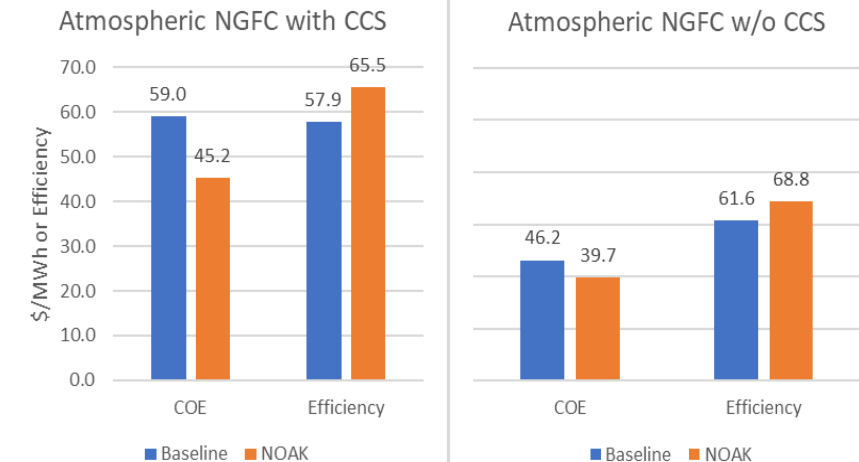
- Includes technology updates of other critical system components such as carbon capture, external reformers, etc.

- Year dollar update to 2018\$, performance data generated by PNNL stack model ROM, SOTA vs Advanced SOFC, vent gas recirculation concept, capture and non-capture cases, and on cell reforming percentage sensitivities

## NGFC Configuration

Principal Investigator

## Preliminary Results



U.S. DEPARTMENT OF  
**ENERGY**

## Distributed Generation (DG) as a Potential Market for SOFC

**Motivation:** DOE-FE's development plan for SOFC technology includes demonstration of commercial units at the DG scale ( $\approx 1$  MW)

**Objective:** A market study is performed to describe how SOFC technology fits into a competitive DG market and projects cost reductions associated with demonstration of multiple units

### Study Details

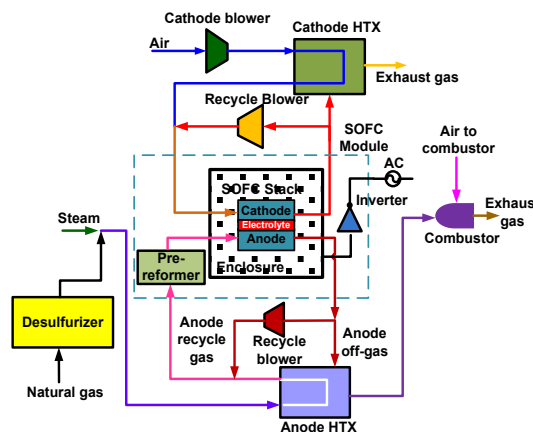
**Document describes the current distributed generation market and the potential for SOFC technology within it**

- Study analyzes several market studies, detailing the capacity potential for SOFC technology in the DG market
- Study projects how many DG demonstration units at 1-MWe are needed to reach the \$900-\$1000/kW cost target
- Sensitivities (such as natural gas price) applied for SOFC and other DG scale technologies for comparison

**Incorporates anticipated penetration for other DG technologies including wind, solar, reciprocating engine, microturbines, etc.**

[Report Available Online \(link below\)](#)

### SOFC-DG Configuration



### Principal Investigator

Gregory A. Hackett

### Key Study Results

Parameter	Nth of a Kind SOFC DG Performance
Net AC Power [kWe]	1000
Operating Pressure [atm]	1.0
Operating Temp. [°C (°F)]	750 (1382)
Cell Voltage [V]	0.830
Current Density [mA/cm <sup>2</sup> ]	400
Net AC Efficiency [HHV]	61.3
Module Cost [2011\$/kWe]	452
BOP Cost [2011\$/kWe]	531
Total System [\$/kWe]	983

Study predicts 25-90 1-MWe units will be needed to reach this cost per kW



## SOFC Cell and Stack Production Cost Study

**Motivation:** DOE-FE's cost targets for SOFC require multiple (25+) demonstrations at the distributed generation scale ( $\approx 1$  MW) for viability

**Objective:** Develop a comprehensive tool to assist SOFC commercial developers understand the costs associated with large scale production of solid oxide fuel cells and stacks

### Tool Details

**The SOFC cell and stack production cost tool is an Excel-based tool intended for public release**

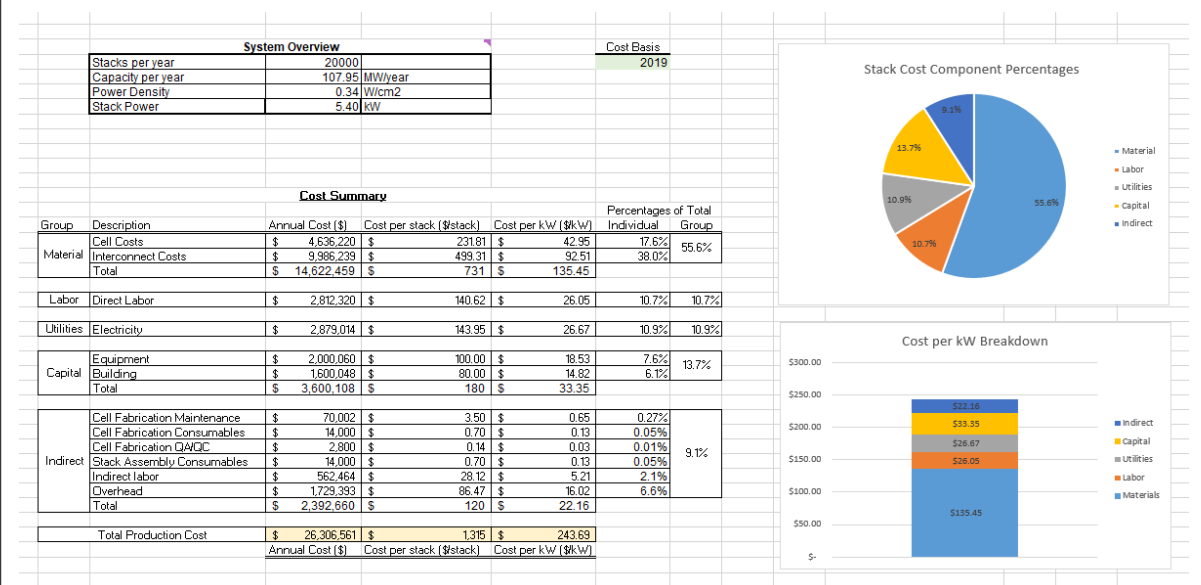
- Tool includes all of the necessary cost inputs including raw materials, equipment costs, labor costs, etc.
- Tool will allow for sensitivities to be conducted on parameters such as total production scale, materials costs, electricity costs, etc.
- Default values provided will serve as an example case study

**Tool will be accompanied by a detailed user manual with instructions and a worked examples**

- Can be easily modified to include the necessary materials for hydrogen producing SOEC

**Tool scheduled for completion July/August 2020**

### Spreadsheet Tool Example



# Solid Oxide Fuel Cells

## Hydrogen Production Cost Analysis from Solid Oxide Electrolysis Systems

**Motivation:** DOE-FE SOFC Program has recently adjusted their objectives to include relevant SOFC technologies for hydrogen production, when operated in electrolysis mode

**Objective:** Conduct an independent analysis of hydrogen production costs from high temperature electrolysis cells and compare the results to those described by EERE in their 2016 report

### Study Details

DOE-FE has requested that NETL conduct a study similar to the 2016 EERE report on hydrogen production with a basis of 50,000 kg per day

- NETL will conduct an independent assessment of hydrogen production costs, with updated information, as available
- Sensitivity analyses will be conducted similar to those conducted in the original report

A summary report and slide presentation will be developed to describe the similarities and differences between NETL's and EERE's results

**Anticipated Study Completion TBD**  
**(Likely July/August 2020)**

### EERE Report Information

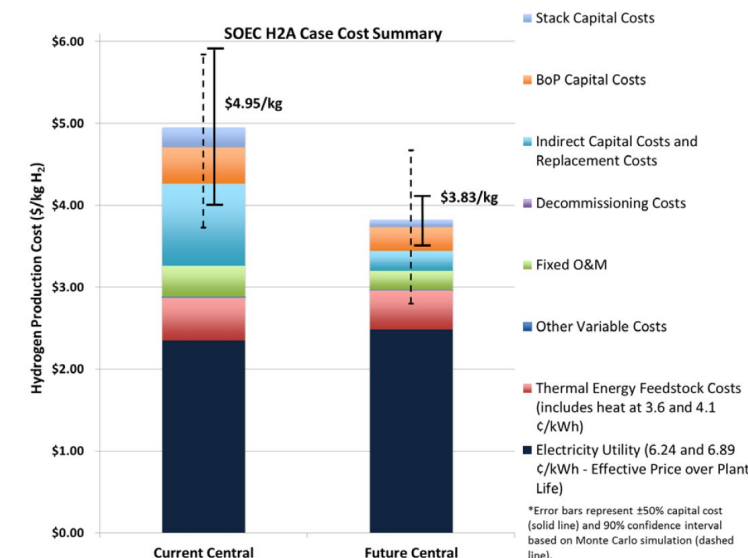
DOE Hydrogen and Fuel Cells Program Record	
Record #: 16014	Date: 2/17/16
Title: Hydrogen Production Cost from Solid Oxide Electrolysis	
Originators: David Peterson, Eric Miller	
Peer Reviewed by Industry Representatives: Annabelle Brisse, Joseph Hartvigsen, Randy Petri, and Greg Tao	
Approved by: Sunita Satyapal	Date: 5/31/16

This study made use of the NREL Hydrogen Analysis Model, which is available online

### Principal Investigator

Gregory A. Hackett

### Original Study Key Results



# Solid Oxide Fuel Cell R&D Progress Review



## Cell and Stack Degradation Modeling

- Formalized partnership with developer to integrate high temperature fiber optic sensors into SOFC stack
- Demonstrated ability to predict SOFC performance degradation from multiple modes
  - Progress toward public release of predictive toolsets by March 2021
- Added to simulations:
  - Infiltrated materials
  - Reversible SOFC operation

## Electrode Engineering

- Tested a commercial SOFC in reversible mode for 2400 h, cycling between fuel cell and electrolysis mode every 100 h
  - Infiltrated commercial SOFC showed significant reduction in degradation when operating under electrolysis mode
- Developing novel approach for increased electrode surface area via carbon templating
- Novel materials discovery and fabrication

## Systems Engineering and Analysis

- Robust cell and stack production cost model under development
  - Intended for public release this summer
- IGFC and NGFC techno-economic analyses under final NETL review
  - Intended for public release this summer
- Scoping study completed on hybrid carbon conversion technologies with SOFC component



# THANK YOU!

---

VISIT US AT: [www.NETL.DOE.gov](http://www.NETL.DOE.gov)

**Gregory A. Hackett, Ph.D.**

Research Team Lead, Solid Oxide Fuel Cells

National Energy Technology Laboratory

U. S. Department of Energy

304-285-5279

[Gregory.Hackett@netl.doe.gov](mailto:Gregory.Hackett@netl.doe.gov)



U.S. DEPARTMENT OF  
**ENERGY**