

# **A Multifunctional Isostructural Bilayer Oxygen Evolution Electrode for Durable Intermediate-Temperature Electrochemical Water Splitting**

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May 30, 2020

Project ID P190

# Project Overview

## Timeline

- Start date: 10/1/2019
- End date: 9/30/2022
- Actual starting date: 2/10/2020

## Barriers

- High degradation rate
- Oxygen/electrolyte interfacial delamination
- Cr-poisoning

## Budget

- Total Project Budget: \$1.25M
- Total Recipient Share: \$0.25M
- Total Federal Share: \$1.0M
- Total DOE Funds Spent: \$17K as of March 31, 2020

## Partners

- Prof. Xinfang Jin, University of Massachusetts at Lowell
- INL for testing nodes usage
- NREL for modeling nodes usage

# Relevance

- Objectives:

To develop a highly active and Cr-resistant oxygen electrode for high-durability, high-efficiency and high-rate hydrogen production via high temperature solid oxide electrolysis cells (HT-SOECs). HT-SOEC is currently an integrated part of HydroGEN Consortium R&D program

- Key Milestones:

Metric (in stack)	State of the Art	Expected Advance
Button cell @ $j = 0.9 \text{ A/cm}^2$ , $700^\circ\text{C}$	$\eta_{\text{OE}} > 0.20 \text{ V}$	$\eta_{\text{OE}} < 0.15 \text{ V}$
Planar 2-cells @ $j = 0.8 \text{ A/cm}^2$ and $700^\circ\text{C}$	$\eta_{\text{OE}} > 0.25 \text{ V}$ $> 15 \text{ mV/kh}$ $1000 \text{ h}$	$\eta_{\text{OE}} < 0.15 \text{ V}$ $< 10 \text{ mV/kh}$ $1000 \text{ h}$
Tubular cells @ $j = 0.9 \text{ A/cm}^2$ and $700^\circ\text{C}$	$\eta_{\text{OE}} > 0.20 \text{ V}$ $> 10 \text{ mV/kh}$ $5000 \text{ h}$	$\eta_{\text{OE}} < 0.15 \text{ V}$ $< 8 \text{ mV/kh}$ $5000 \text{ h}$

- Impact

Accelerating commercialization of SOEC technology for low-cost and high-efficiency hydrogen production

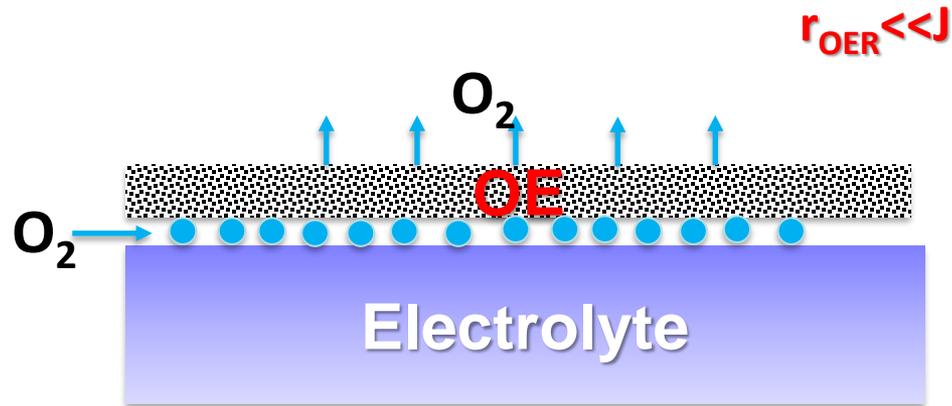
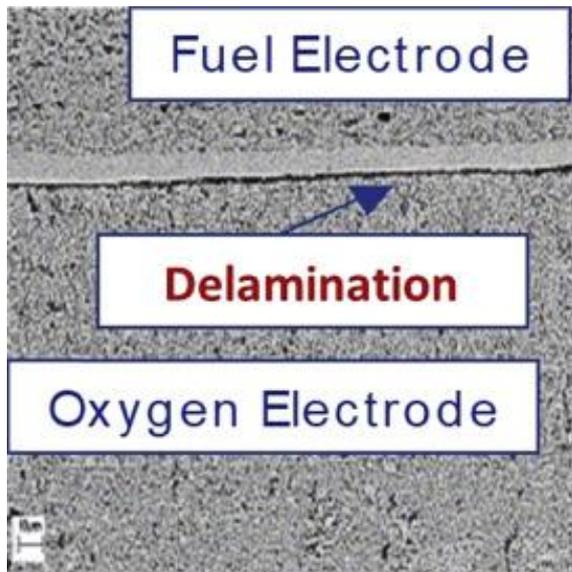
# Approach

- Huang group at UofSC
  - Process optimization of isostructural bilayer OE for scale-up fabrication
  - Fast screening of the bilayer OE and performance evaluations at button-cell scale
  - Cell performance demonstrations at bench-scale and stack level
- Jin group at UMass Lowell
  - Development of a combined chemical and mechanical model
  - Identification of failure modes
  - Establishment of mitigation strategies

# Milestone Status (FY19-20)

<b>Date</b>	<b>Milestones</b>	<b>Completion</b>
5/31/2020	Finalize bilayer OE loading	60%
8/31/2020	Finalize bilayer OE calcination condition	60%
11/30/2020	Finalize the design and fabrication of STEHC testing fixture	10%

# Project Background- Barrier #1

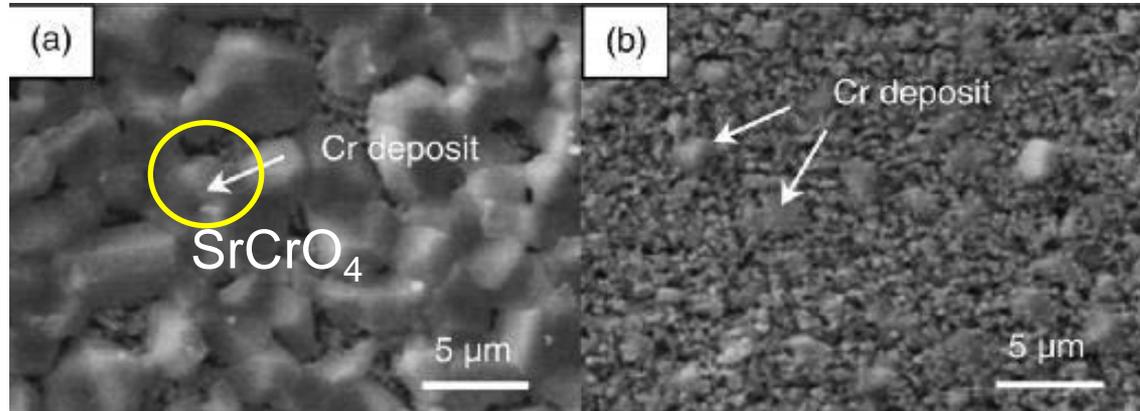


A. Virkar, International journal of hydrogen energy 35 (2010) 9527-9543

Coralli, et al. Science and Engineering of Hydrogen-Based Energy Technologies,  
Hydrogen Production and Practical Applications in Energy Generation,  
2019, p39-122.

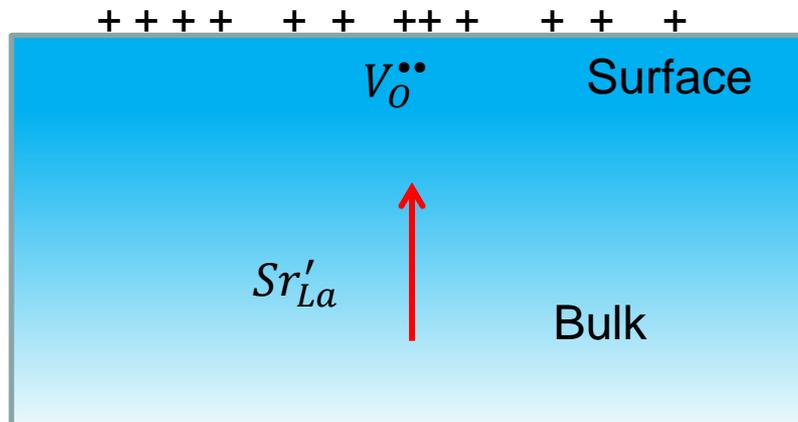
# Project Background- Barrier #2

Cr-poisoning (LSCF at 900°C for 50h with Fe-Cr alloy)



Solid State Ionics 179 (2008) 1459–1464

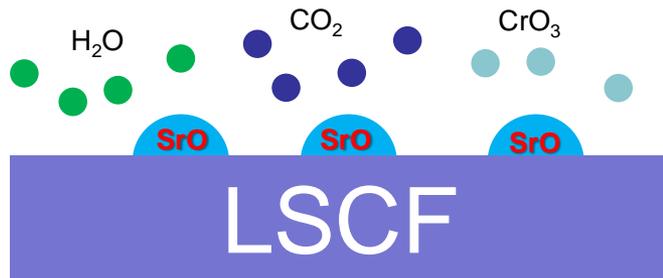
Electrostatic interaction:  $V_{O^{\bullet\bullet}} - 2Sr'_{La}$



**Sr as an acceptor dopant is the culprit for Sr-segregation**

# Surface Sr-Segregation: Consequences

*LSCF electrode under practical conditions*



- H<sub>2</sub>O and CO<sub>2</sub> can compete with O<sub>2</sub> for adsorption on oxygen vacancy sites
- SrO + H<sub>2</sub>O → Sr(OH)<sub>2</sub>
- SrO + CO<sub>2</sub> → SrCO<sub>3</sub>
- SrO + CrO<sub>3</sub> → SrCrO<sub>4</sub>



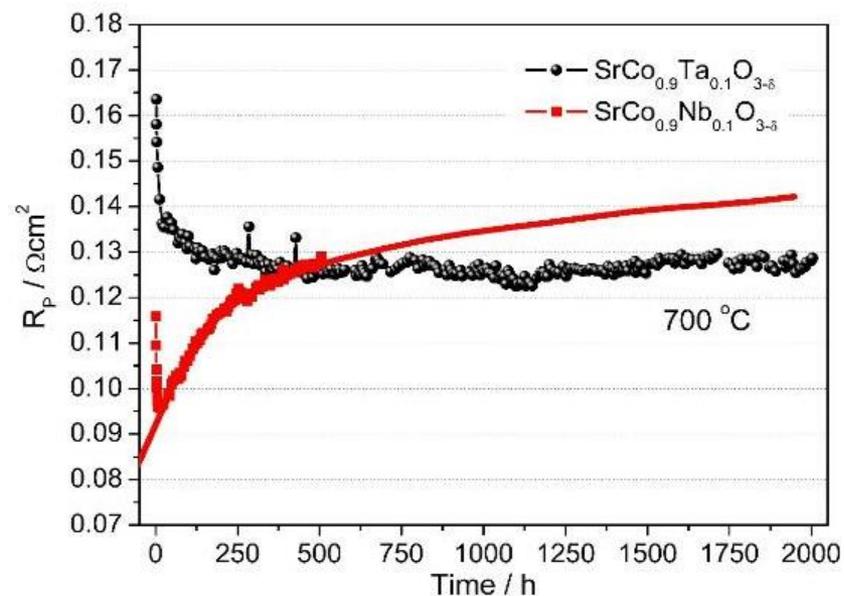
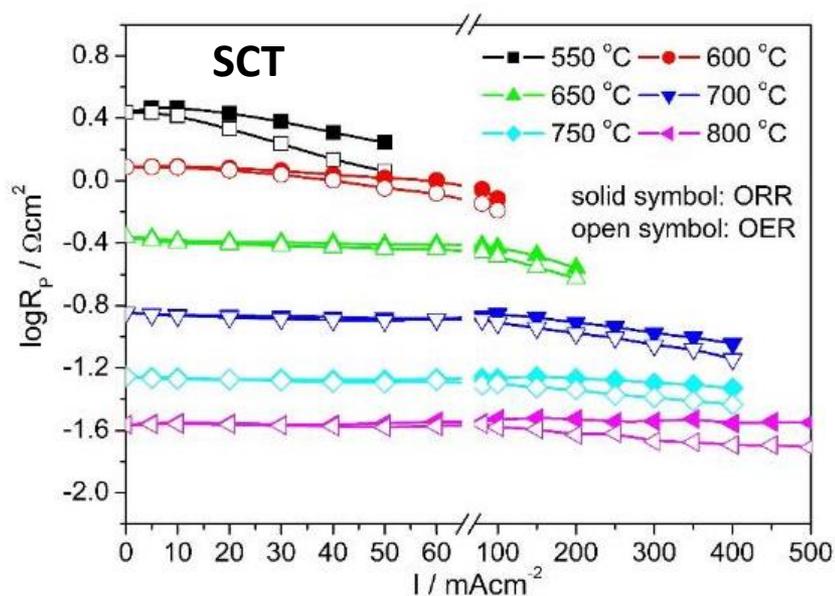
# A New Bilayer OE Concept



**Core:** LSCF-GDC

**Shell:**  $\text{SrCo}_{0.9}\text{Ta}_{0.1}\text{O}_{3-\delta}$  (SCT)

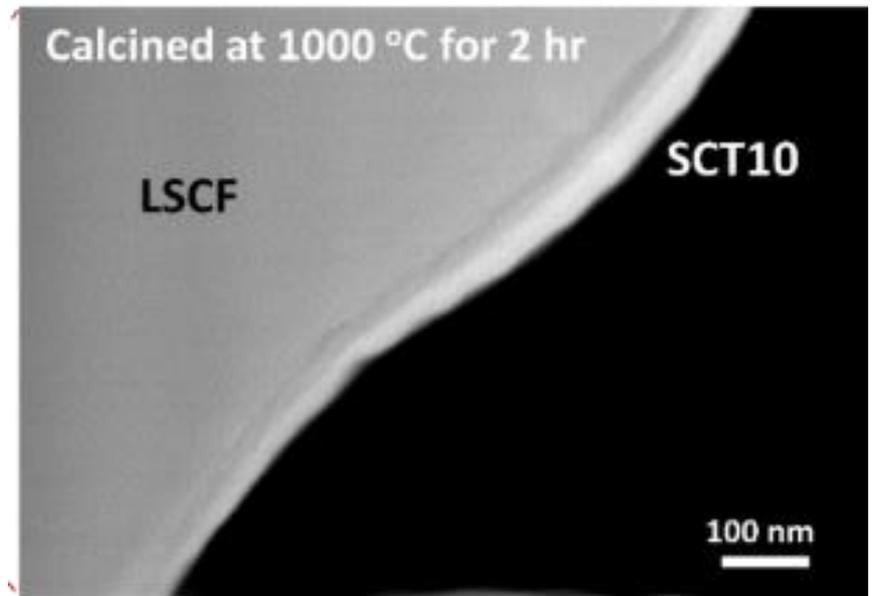
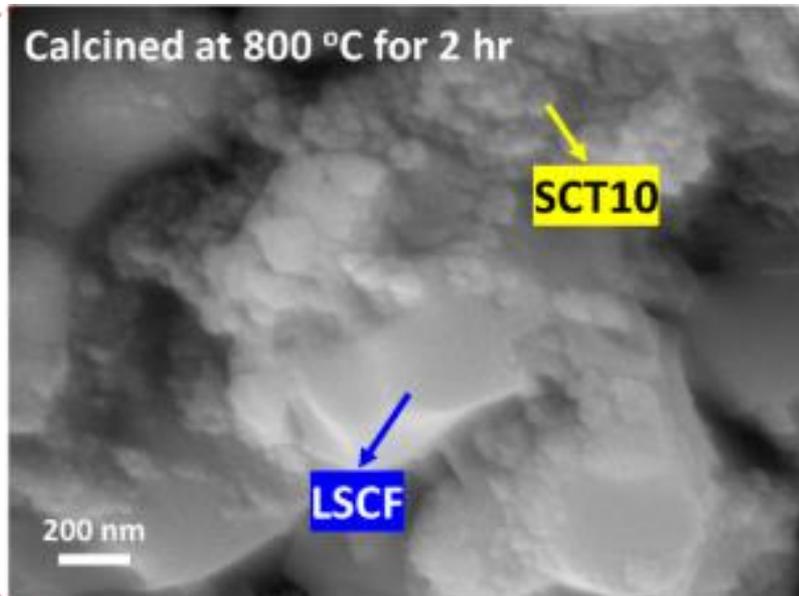
# Prior Work on the Shell Material



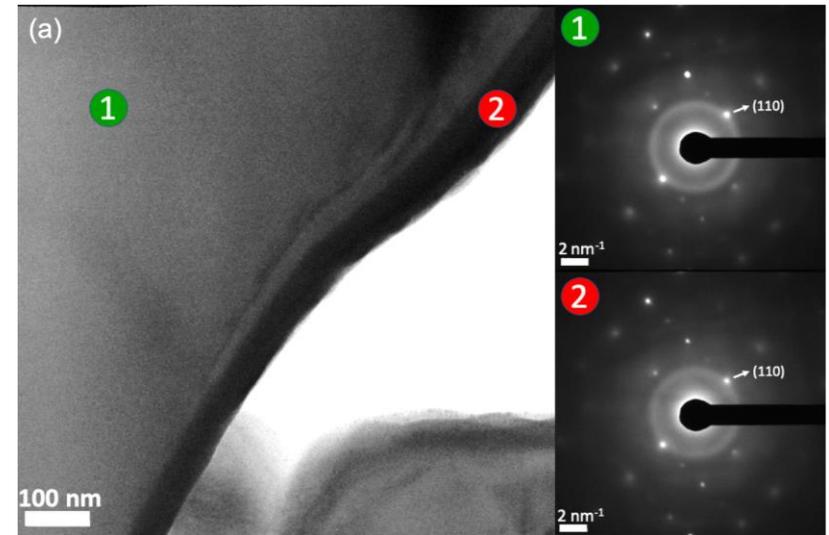
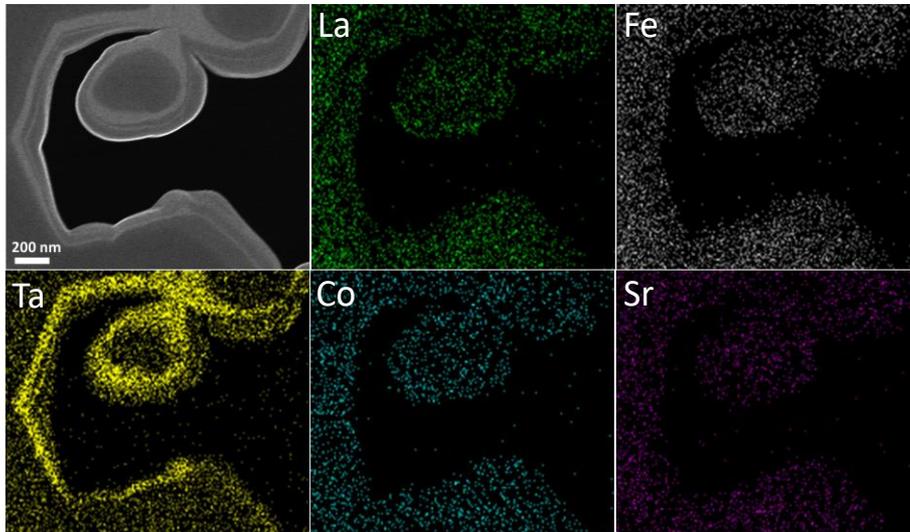
Wang and Huang et al., *Journal of Materials Chemistry A*, 2017, 5, 8989-9002.

# The Bilayer Structure

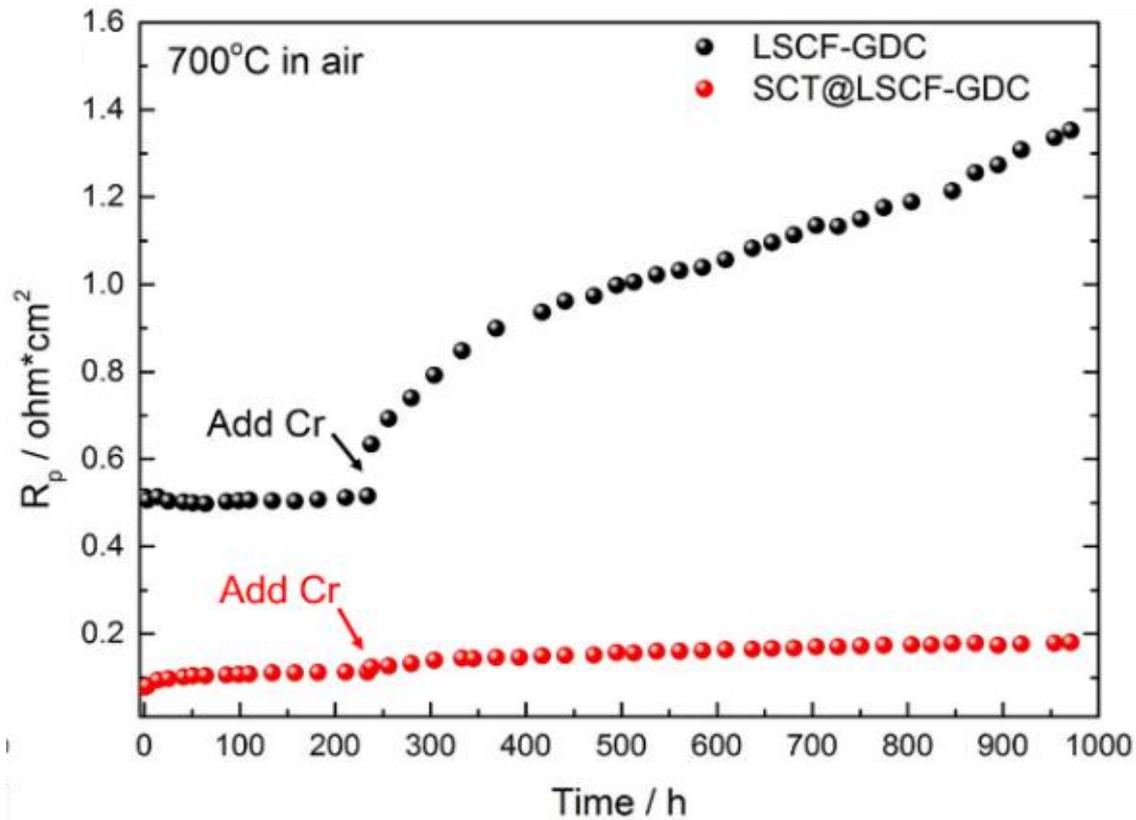
The calcination temperature effect



# The Isostructural Bilayer Structure

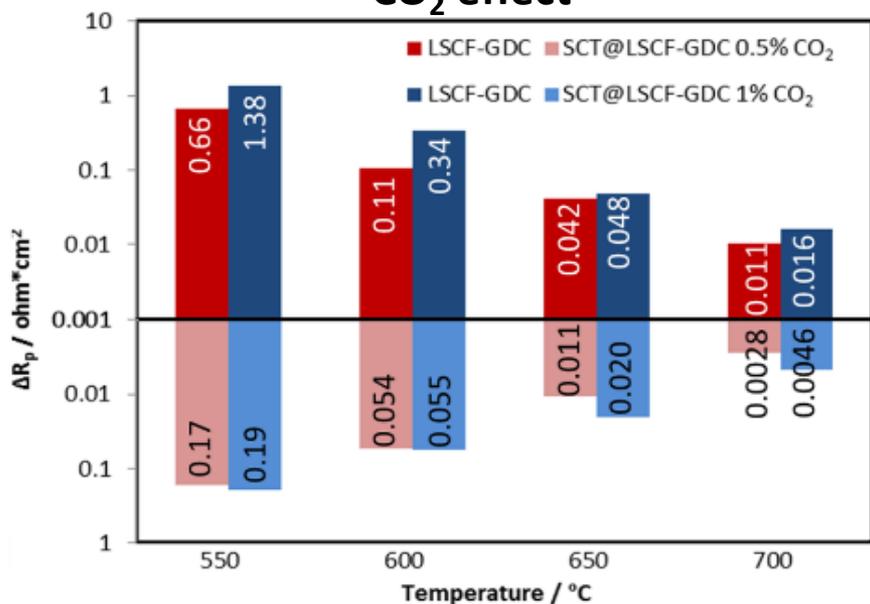


# The Cr-Tolerance

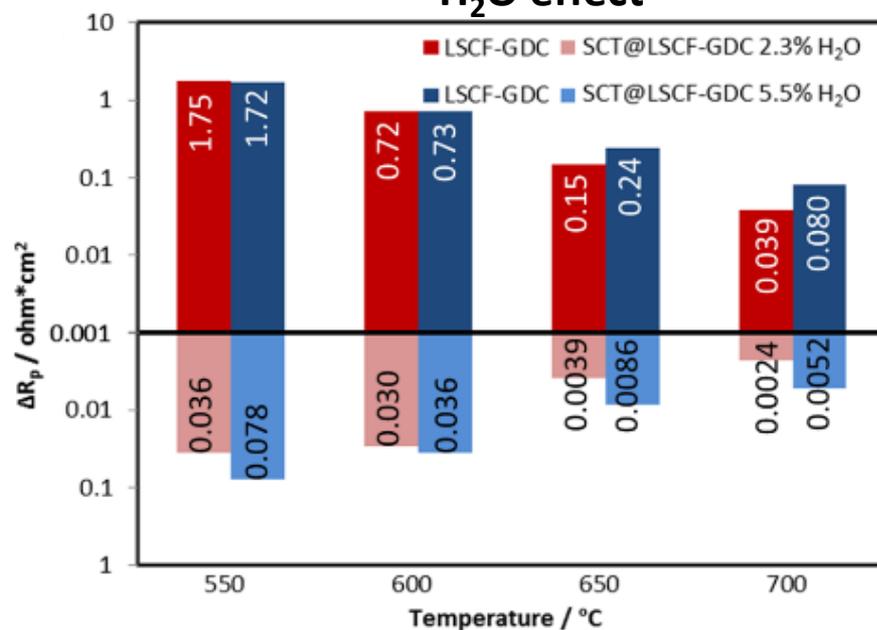


# The CO<sub>2</sub> and H<sub>2</sub>O Tolerance

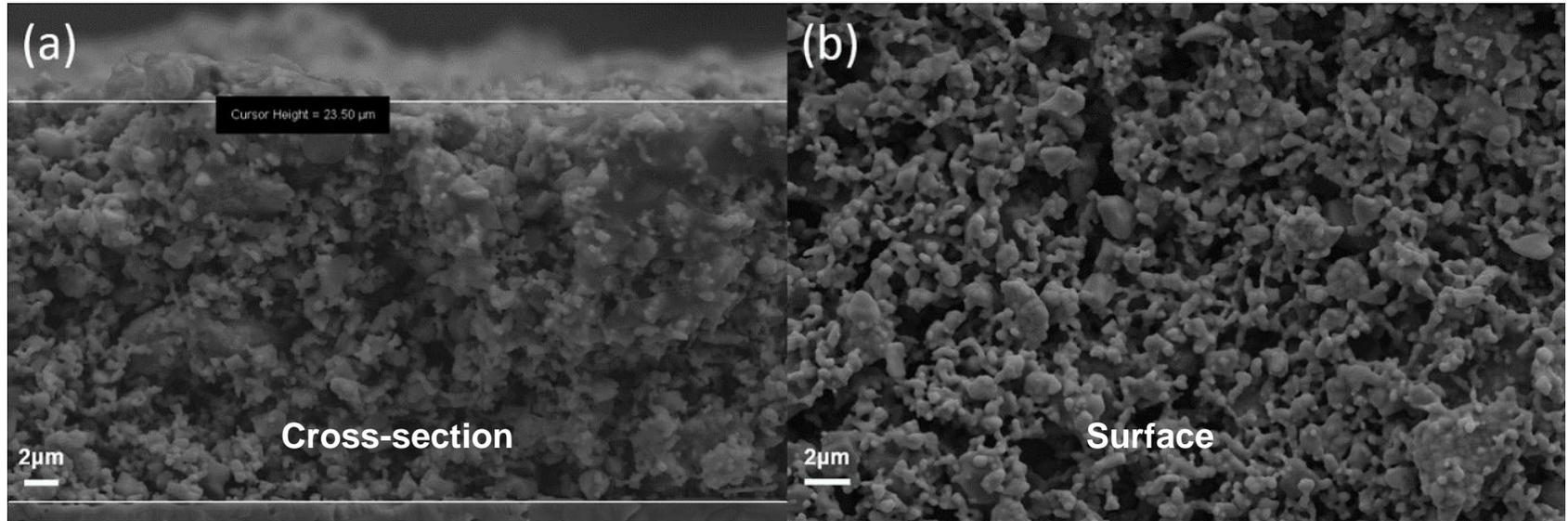
## CO<sub>2</sub> effect



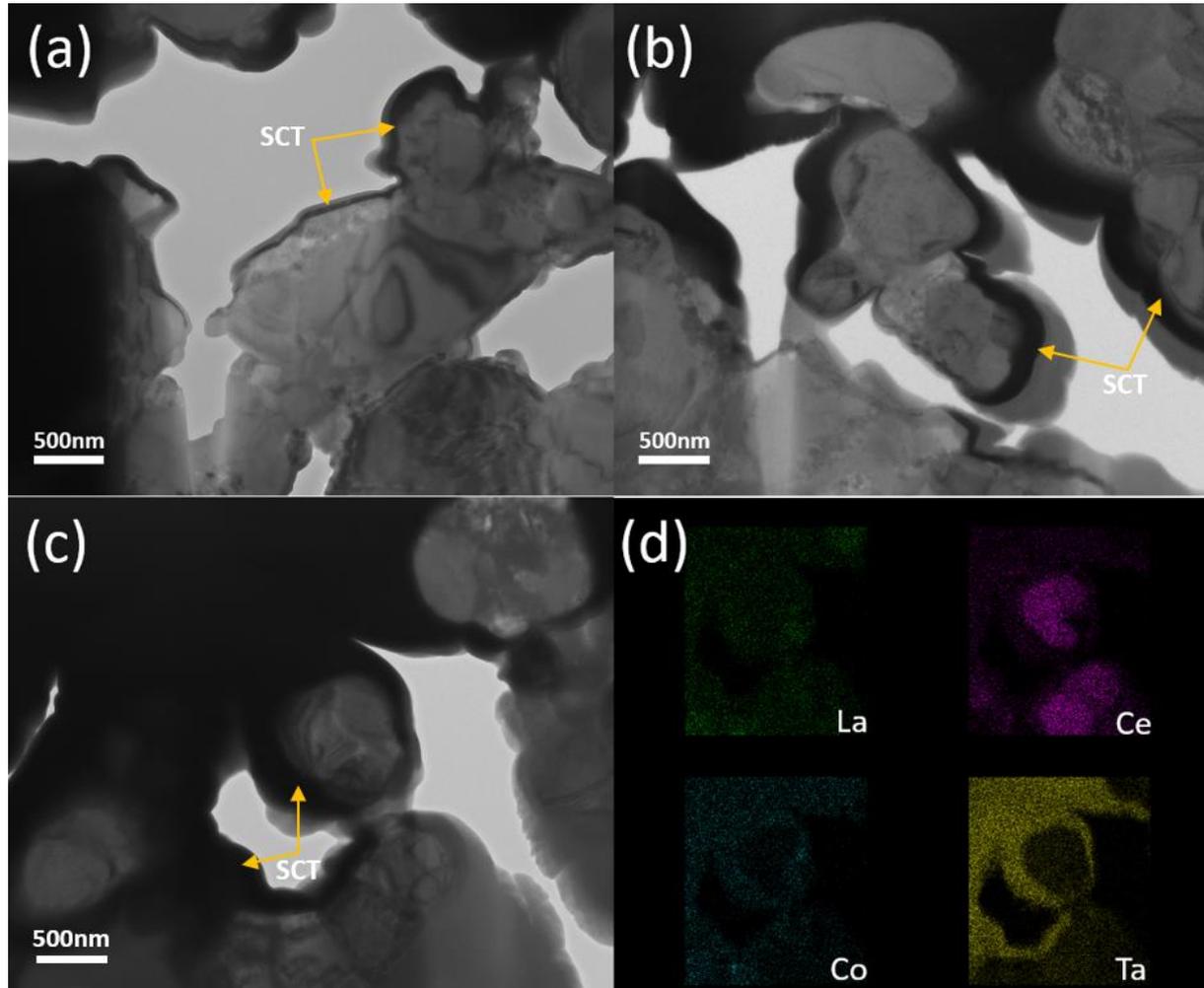
## H<sub>2</sub>O effect



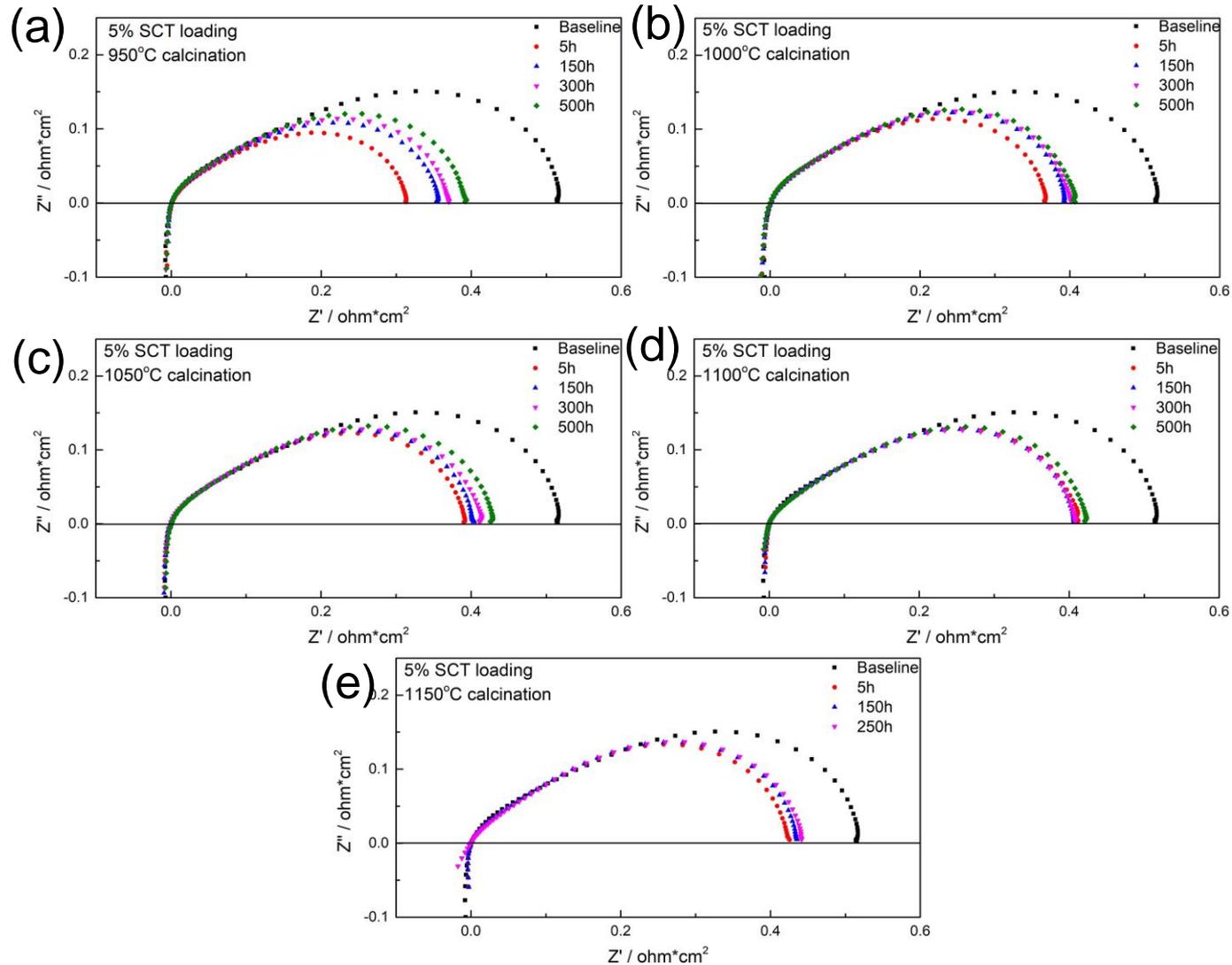
# Accomplishment and Progress: LSCF-GDC Skeleton



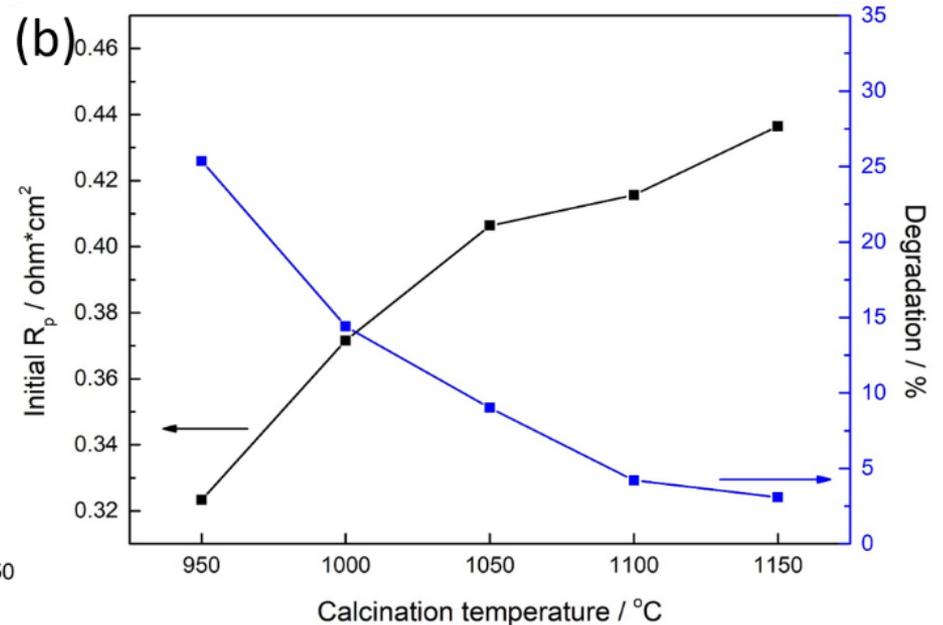
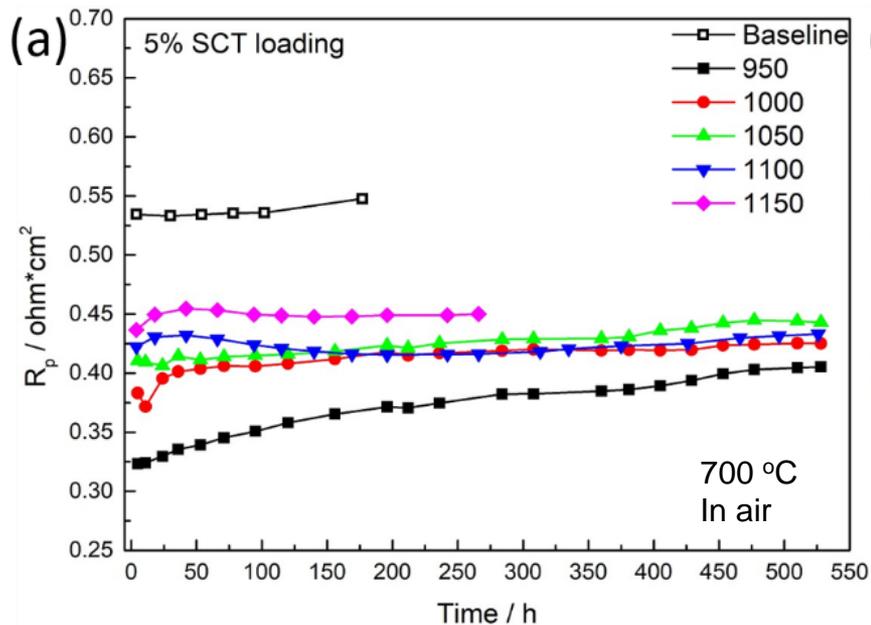
# SCT@LSCF-GDC



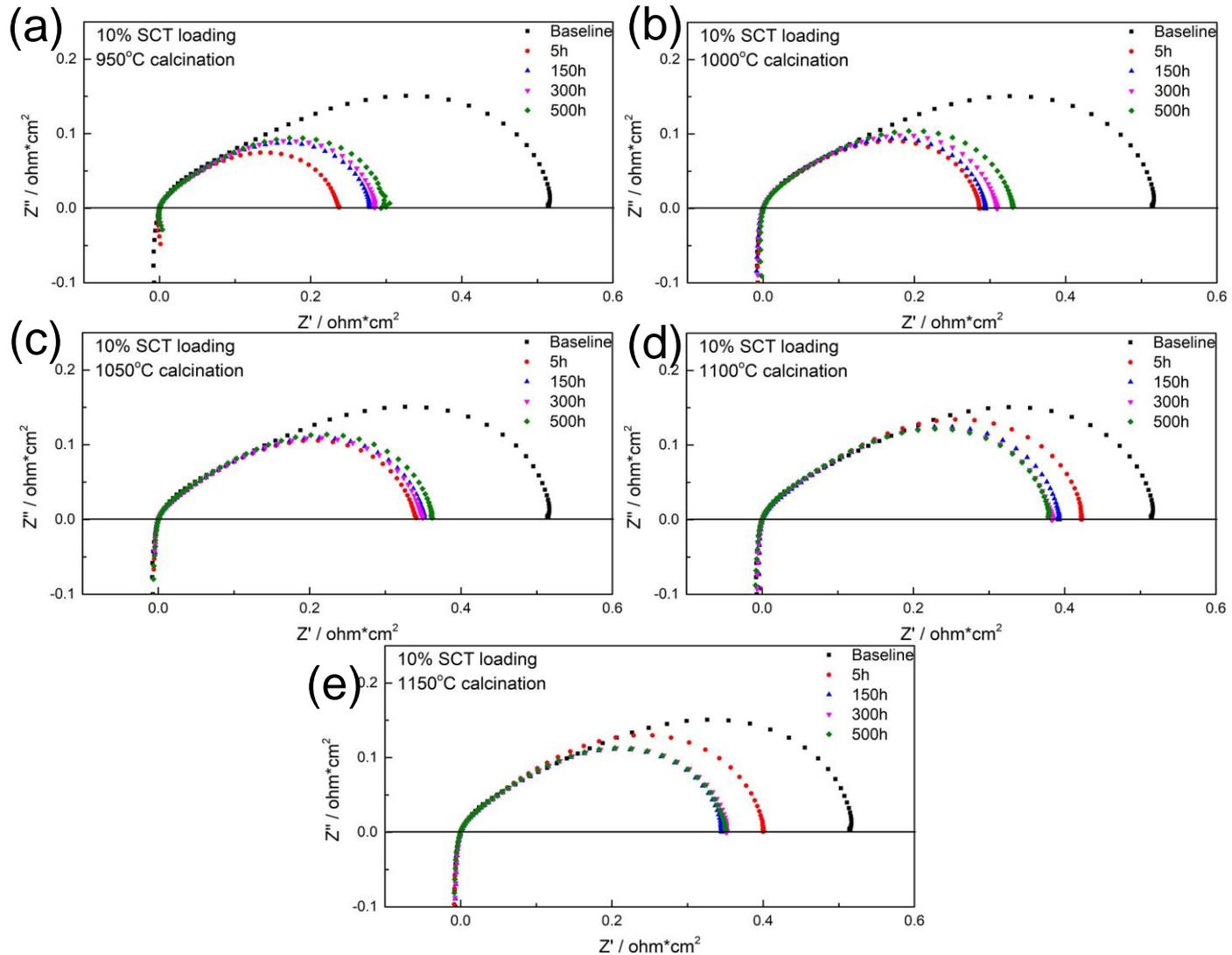
# EIS Spectra of 5%SCT@LSCF-GDC



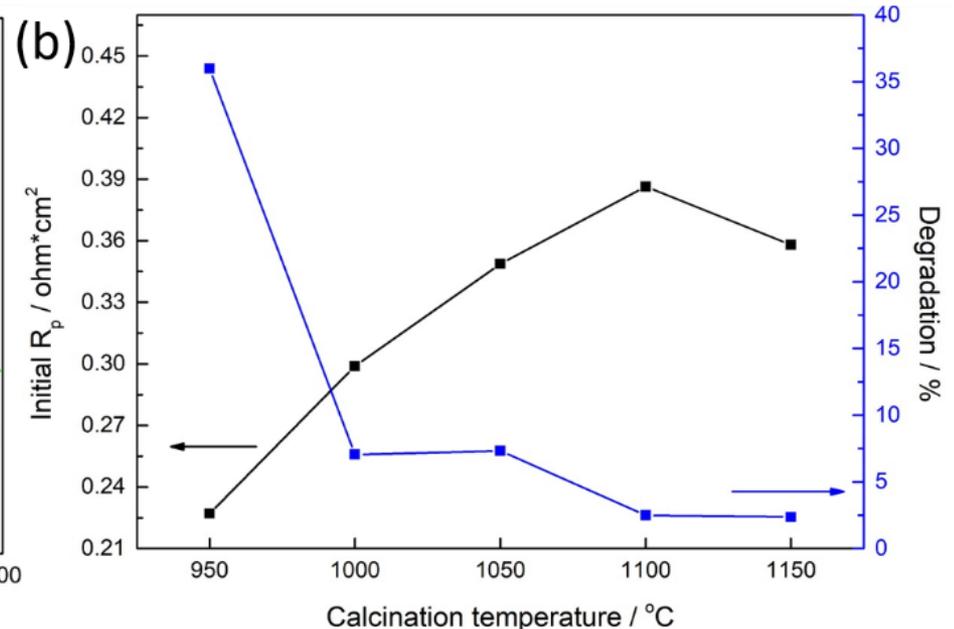
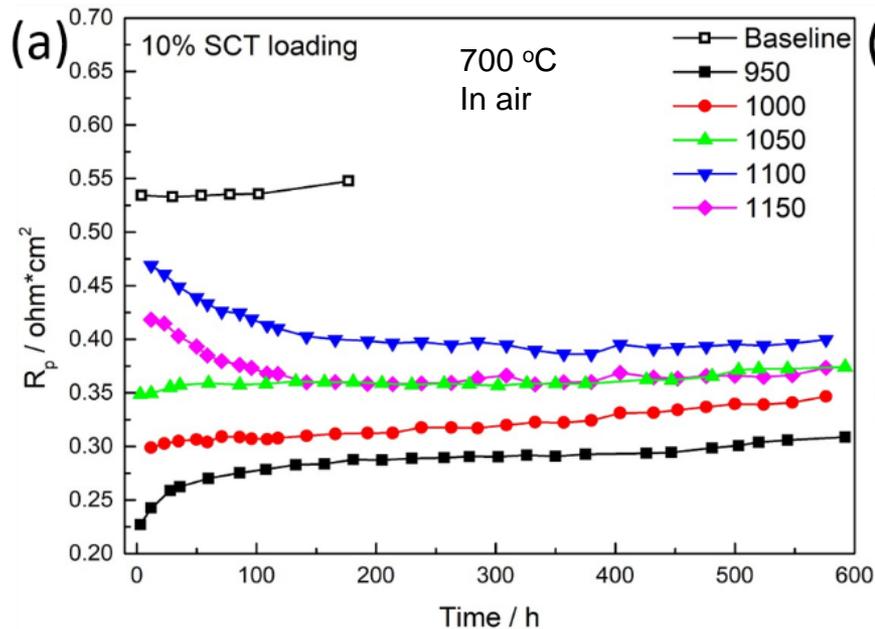
# Long-term stability of 5%SCT@LSCF-GDC



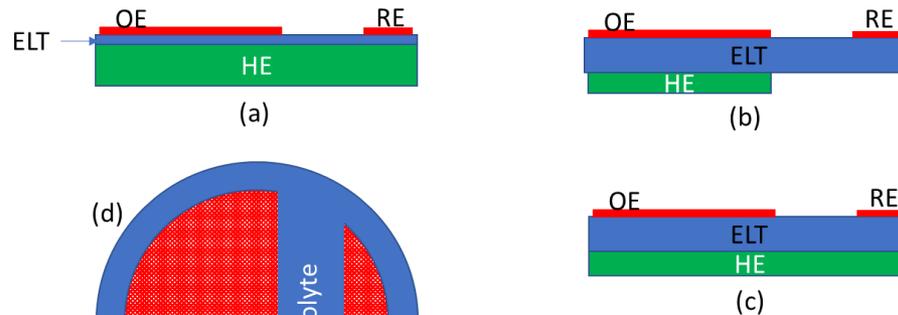
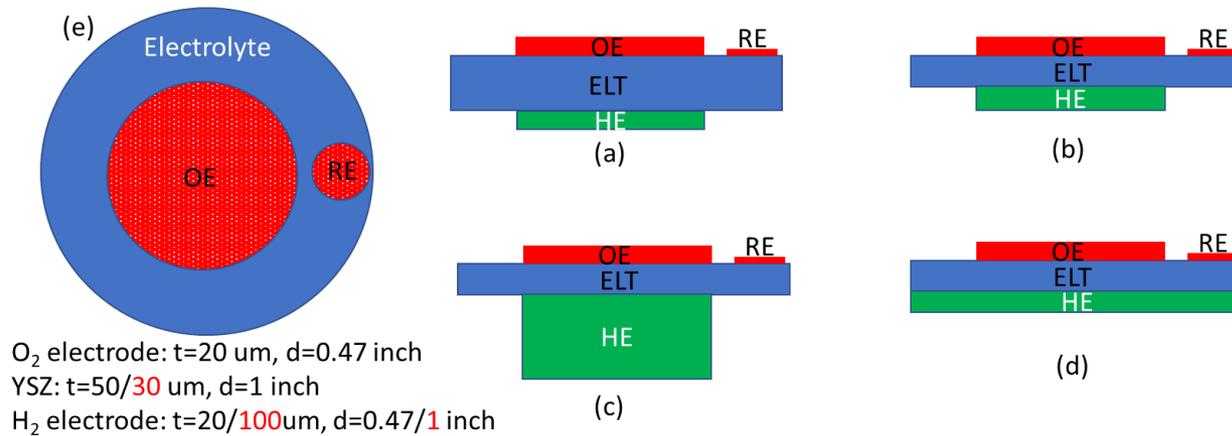
# EIS Spectra of 10%SCT@LSCF-GDC



# Long-Term Stability of 10%SCT@LSCF-GDC

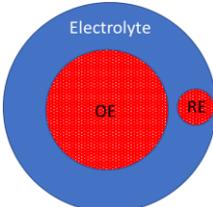
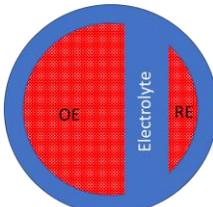


# Theoretical Analysis on 3-electrode Configuration



O<sub>2</sub> electrode: t=20 μm, d=0.79 inch  
 YSZ: t=30/50 μm, d=1 inch  
 H<sub>2</sub> electrode: t=500/20 μm, d=0.79/1 inch

# Modeling Results of Three-Electrode Overpotential Measurements

Top View	Cross-Section	Potential @ RE	Potential @ middle of ELT @ 0.4A/cm <sup>2</sup>	Validity for 3-Electrode Measurement
		-0.310V	-0.310V	Valid
		-0.307V	-0.307V	Valid
		-0.240V	-0.240V	Valid
		0V	-0.307V	Invalid
		0V	-0.250V	Invalid
		-0.307V	-0.307V	Valid
		-0.270V	-0.307V	Error

# Collaboration and Coordination

- University of Massachusetts at Lowell
  - subcontractor, focusing on multiphysics modeling
- Idaho National lab
  - Nodes usage, to reproduce and validate bilayer OE at a larger scale
- National Renewable Energy Lab
  - Nodes usage, to develop thermochemical and electrochemical multiphysics models

# Remaining Challenges and Barriers (FY20-21)

- Demonstration of bilayer OE milestone performance at button-cell level
- Demonstration of bilayer OE milestone performance larger-cell level.
- Establishing multiphysics models for degradation understanding and mitigation.

# Proposed Future Work (FY20-21)

Milestone	Date	Description	Status
2.2	02/28/21	Meet button-cell performance: $\leq 0.15$ V OER overpotential at 1 A/cm <sup>2</sup> for 1 kh @ 700 °C	Not started
4.1	02/28/21	Establish a Multiphysics model to describe OER degradation mechanisms	Not started
4.2	02/28/21	Demonstrate a combined thermal/electrochemical model	Not started
Go/No-Go	02/28/21	Demonstrate the best OER performance specified in Milestone 2.2	Not started
3.1	05/31/21	Successfully transfer the bilayer OE fabrication process to INL for scale-up fabrication	Not started

# Technology Transfer Activities

- Officially filed a US utility patent on February 27, 2020, “Method to Make Isostructural Bilayer Oxygen Electrode”, Serial No. 16/802,608.

# Summary- *Progress and Accomplishments*

- Completed the loading-calcination-morphology- $R_p$  relationship study for 5% and 10% SCT added LSCF-GDC OEs. A clear trending is observed.
- Built a new test rig (including steam generator, high temperature reactor, on-line MS gas analysis system) for button and tubular cells.
- Started anode-supported thin film electrolyte button cell fabrication
- Computationally studied seven 3-electrode configurations for possible overpotential measurement of the OE.