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

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Project ID P190

Project Overview

Timeline	Barriers
• Start date: 10/1/2019 • End date: 9/30/2022 • Actual starting date: 2/10/2020	• High degradation rate • Oxygen/electrolyte interfacial delamination • Cr-poisoning
Budget	**Partners**
• 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	• 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= 0.9A/cm^2, 700^{o}C	$\eta_{OE}>0.20V$	$\eta_{OE}<0.15V$
Planar 2-cells @ j= 0.8A/cm^2 and 700^{o}C	$\eta_{OE}>0.25V$ >15 mV/kh 1000h	$\eta_{OE}<0.15V$ <10 mV/kh 1000h
Tubular cells @ j= 0.9A/cm^2 and 700^{o}C	$\eta_{OE}>0.20V$ >10 mV/kh 5000h	$\eta_{OE}<0.15V$ <8 mV/kh 5000h

- Impact

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

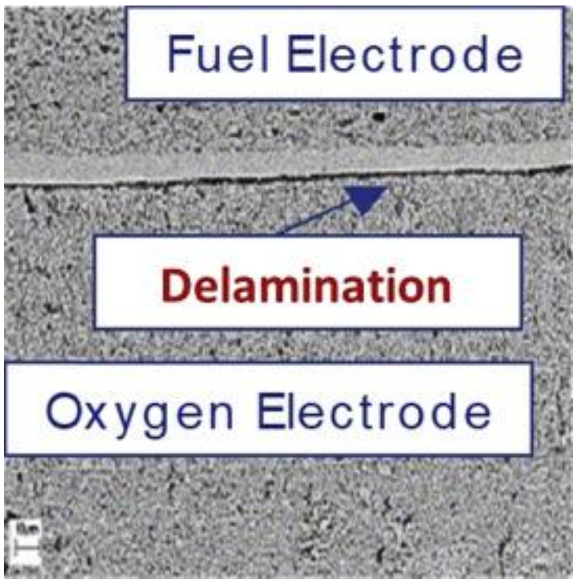
Approach

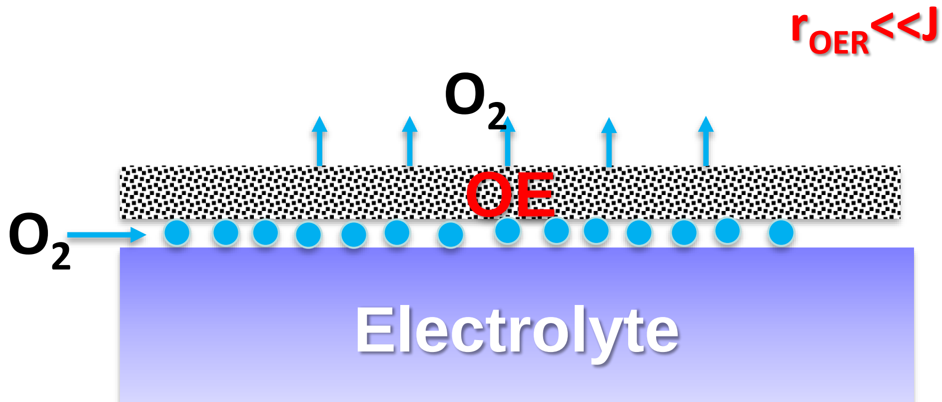
- Huang group at UofSC
  - Process optimization of isostructural bilayer OE for scale-up fabrication
  - Fast screening of the bilayer OE and performance evaluations at button-cell scale
  - Cell performance demonstrations at bench-scale and stack level
- Jin group at UMass Lowell
  - Development of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

Milestone Status (FY19-20)

Date	Milestones	Completion
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



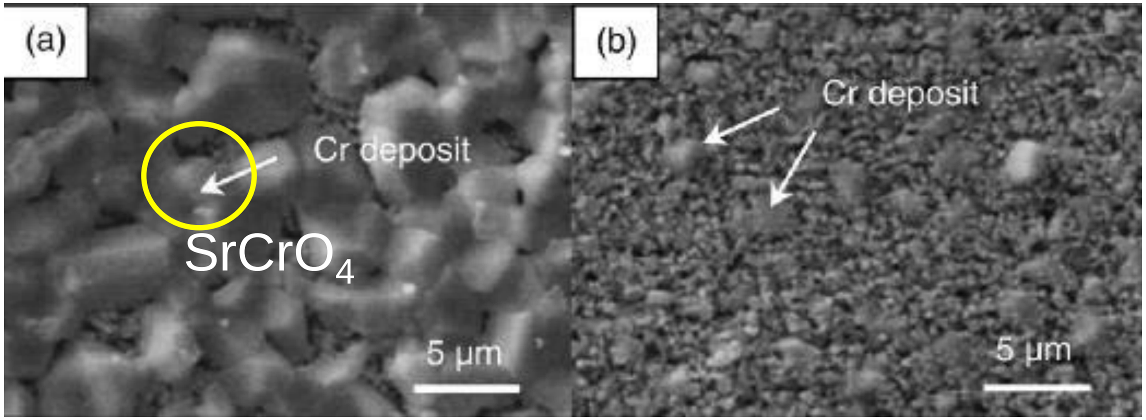


A. Virkar, International journal of hydrogen energy 35 (2010) 9527-9543

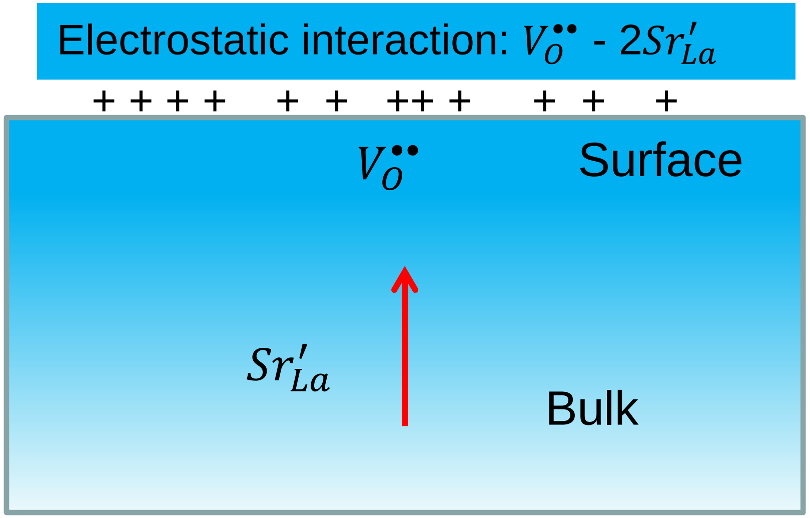
Coralli, et al. Science and Engineering of Hydrogen-Based Energy Technologies, Hydrogen Production and Practical Applications in Energy Generation, 2019, p39-122.

Project Background- Barrier #2

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



Solid State Ionics 179 (2008) 1459–1464

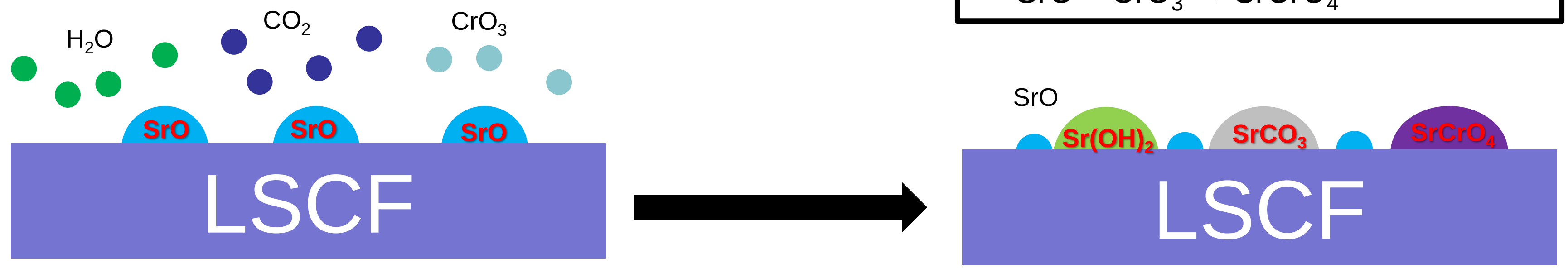


Sr as an acceptor dopant is the culprit for Sr-segregation

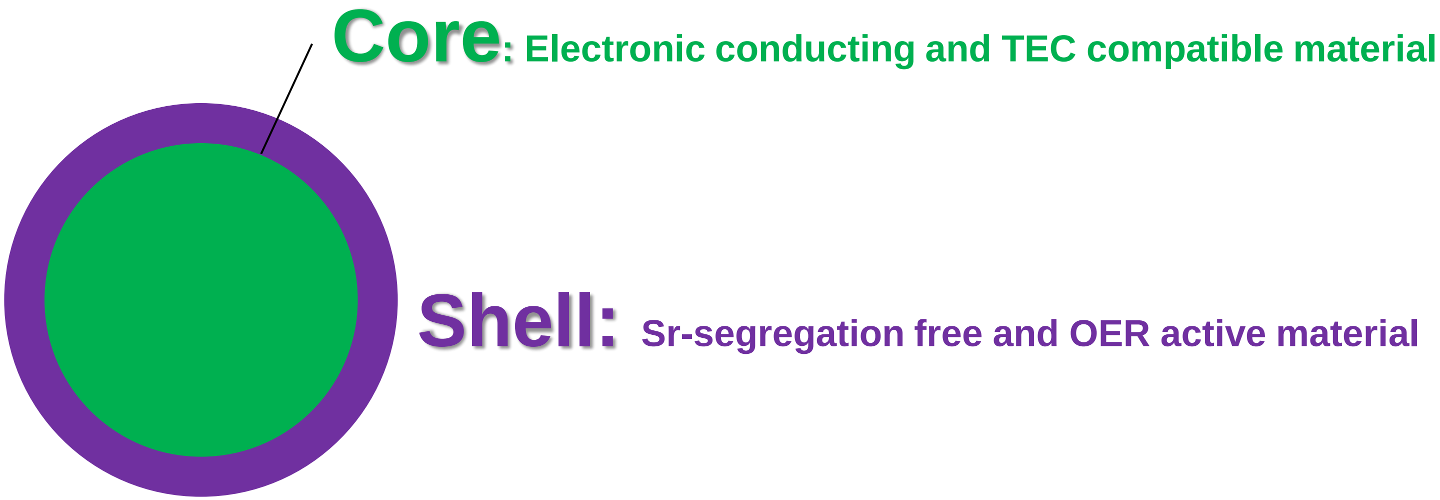
Surface Sr-Segregation: Consequences

- H_2O and CO_2 can compete with O_2 for adsorption on oxygen vacancy sites
- $SrO + H_2O \rightarrow Sr(OH)_2$
- $SrO + CO_2 \rightarrow SrCO_3$
- $SrO + CrO_3 \rightarrow SrCrO_4$

LSCF electrode under practical conditions



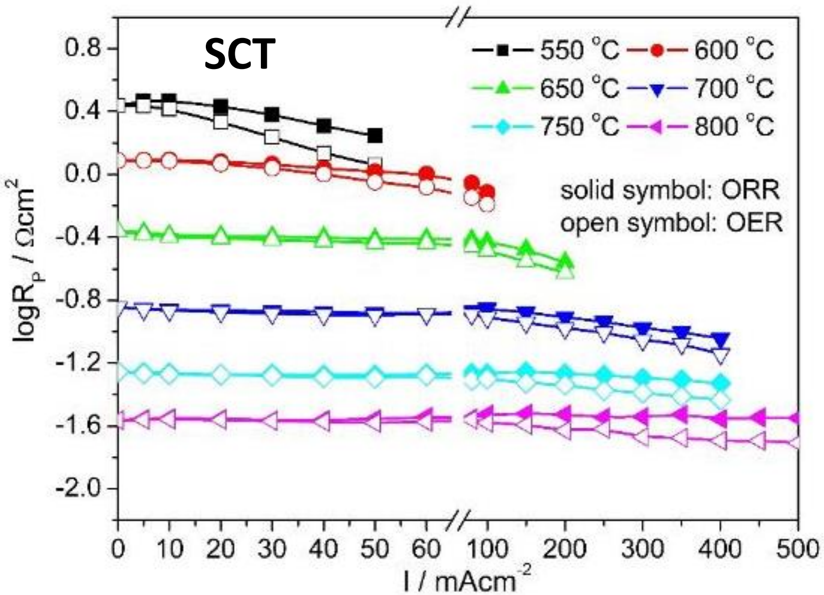
A New Bilayer OE Concept

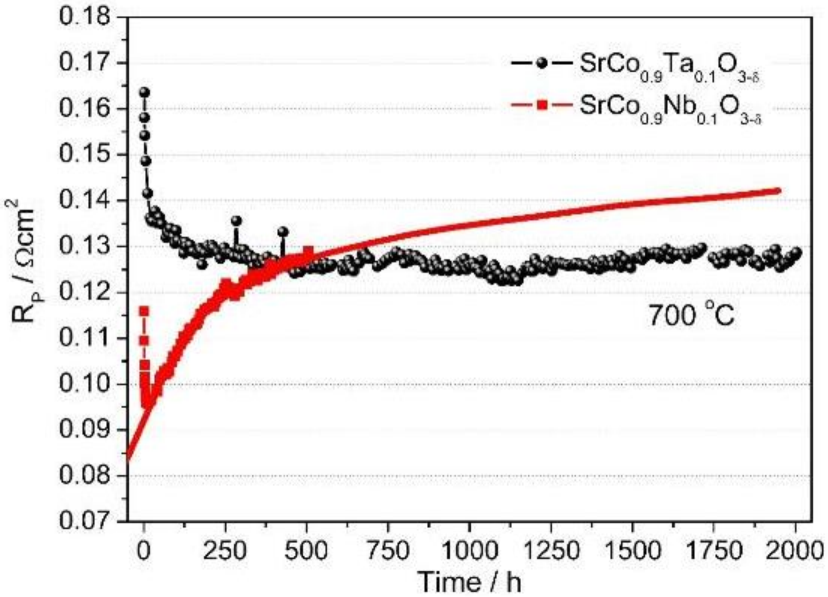


Core: LSCF-GDC

Shell: $SrCo_{0.9}Ta_{0.1}O_{3-\delta}$ (SCT)

Prior Work on the Shell Material

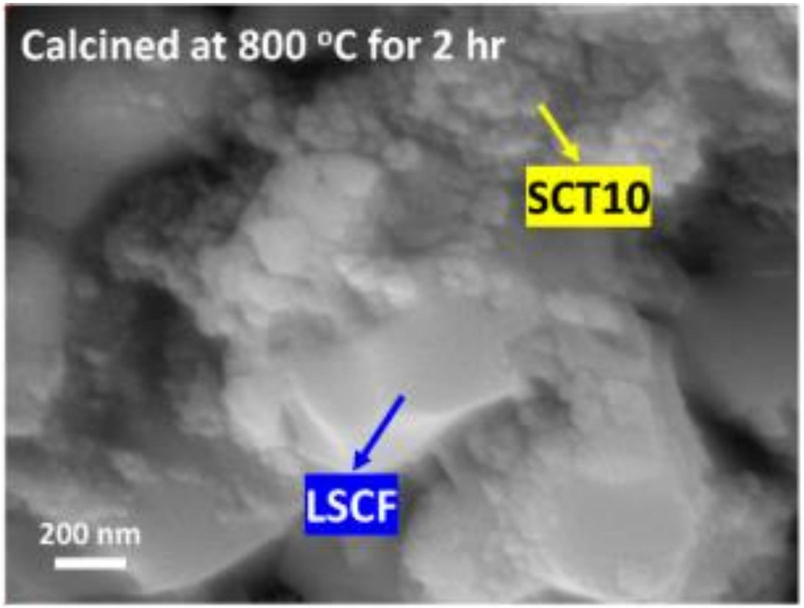


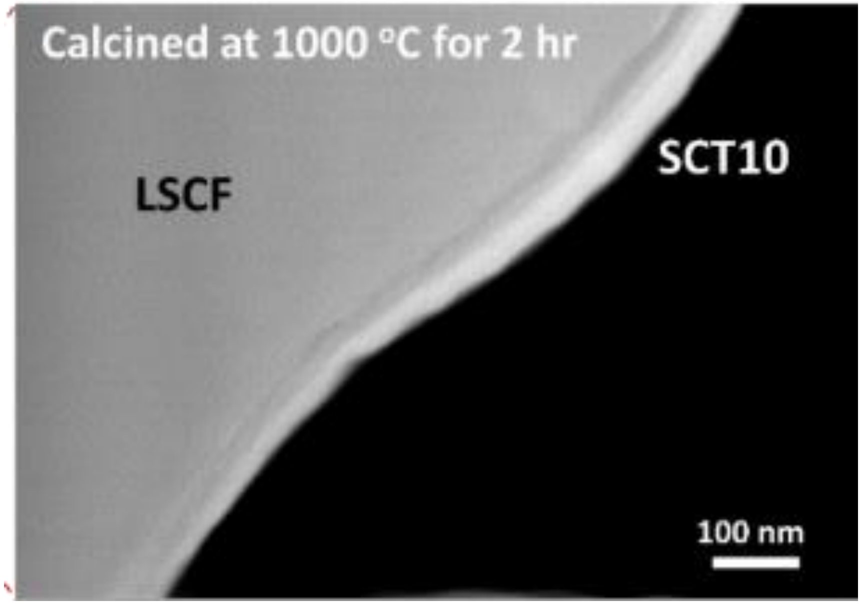


Wang and Huang et al., **Journal of Materials Chemistry A**, 2017, **5**, 8989-9002.

The Bilayer Structure

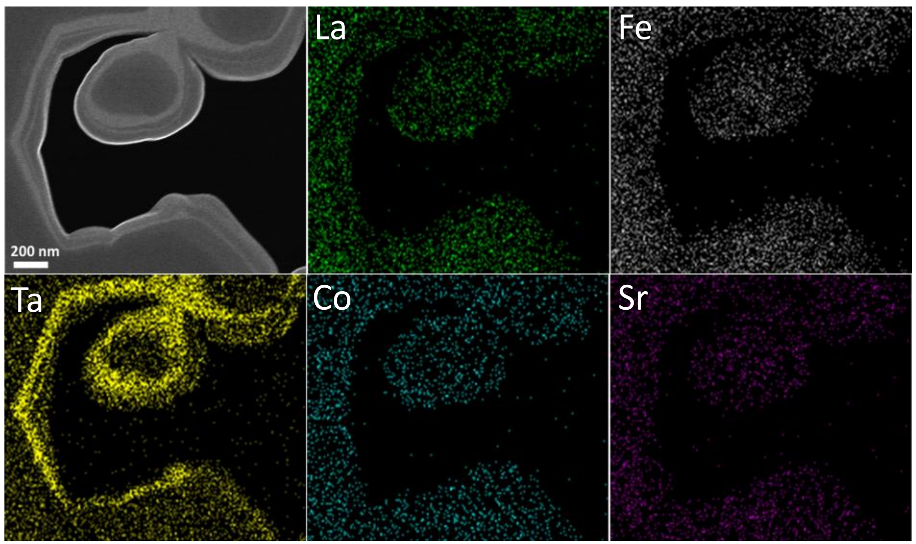
The calcination temperature effect

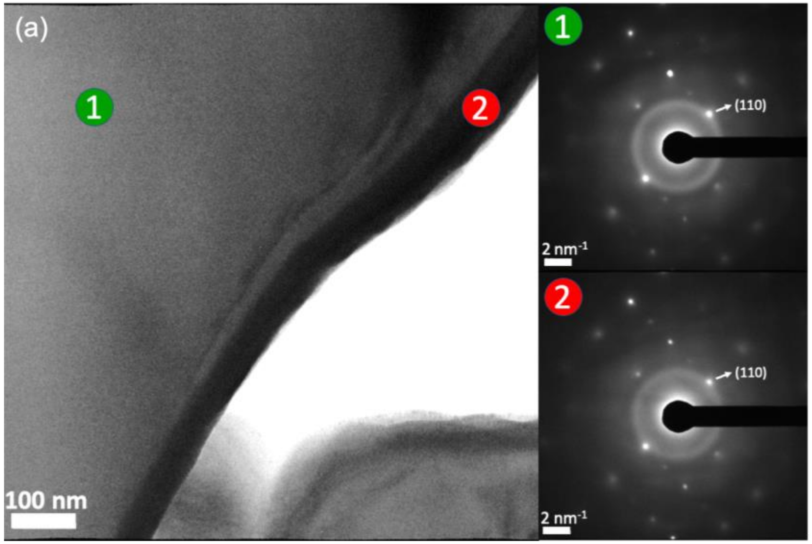




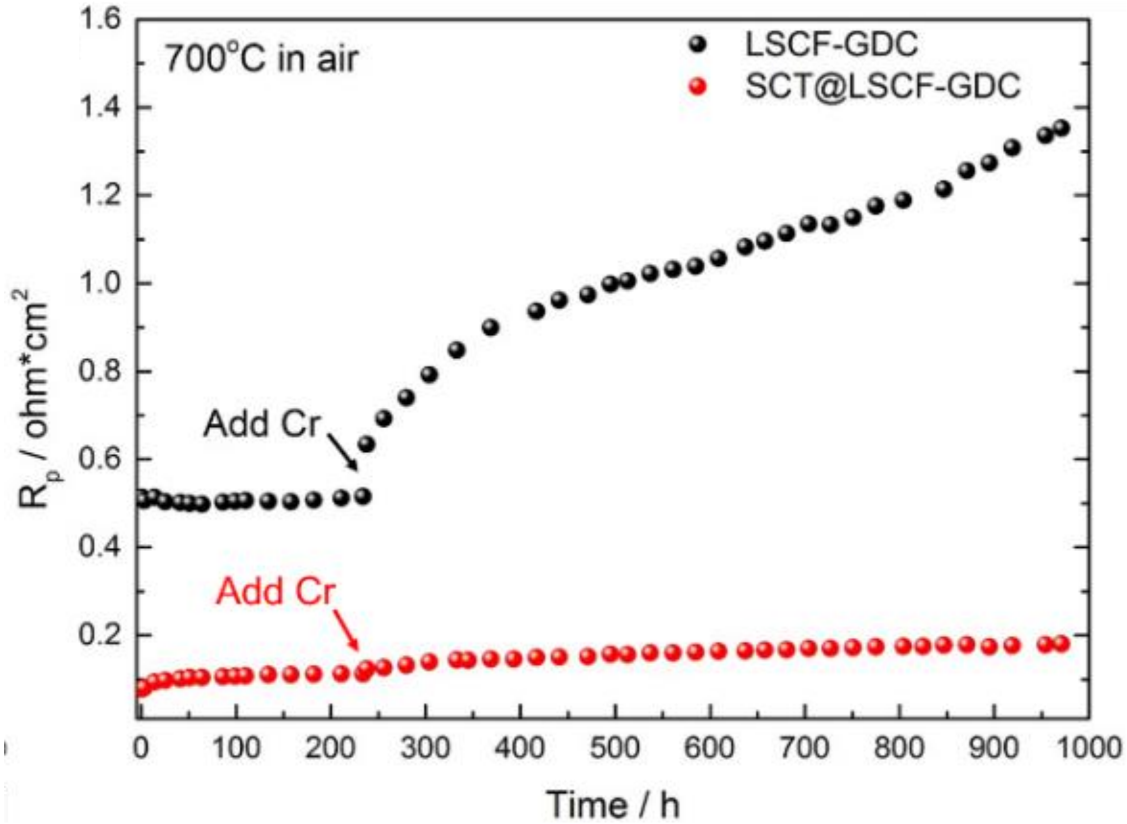
Yang and Huang et al. **Journal of Materials Chemistry A**, 2020, 8, 82-86

The Isostructural Bilayer Structure

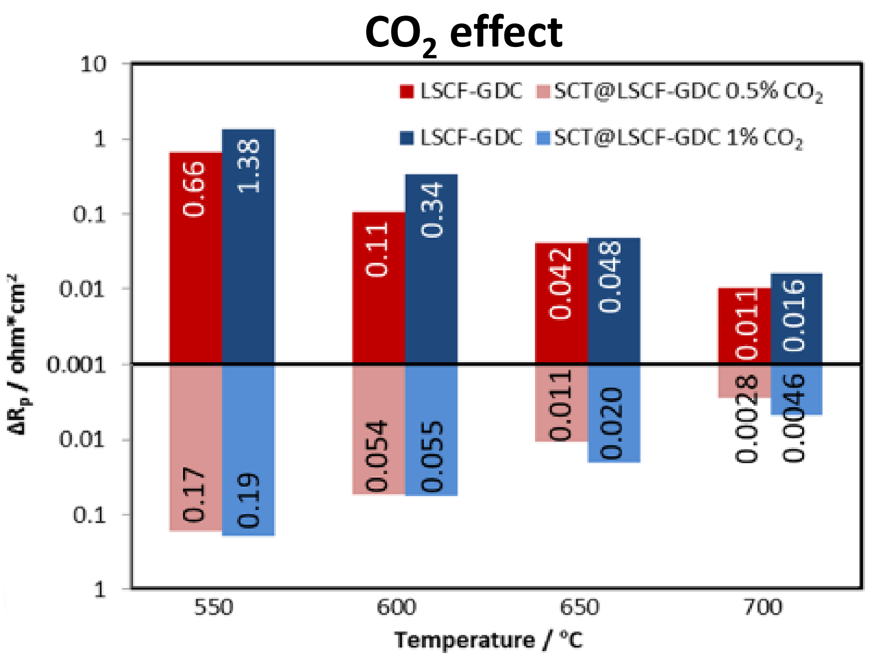


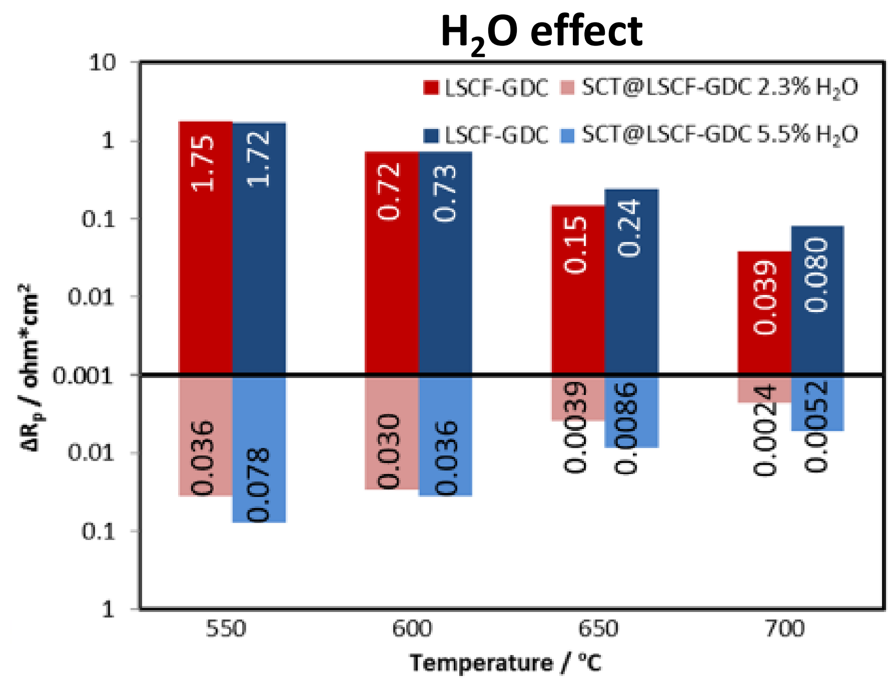


The Cr-Tolerance

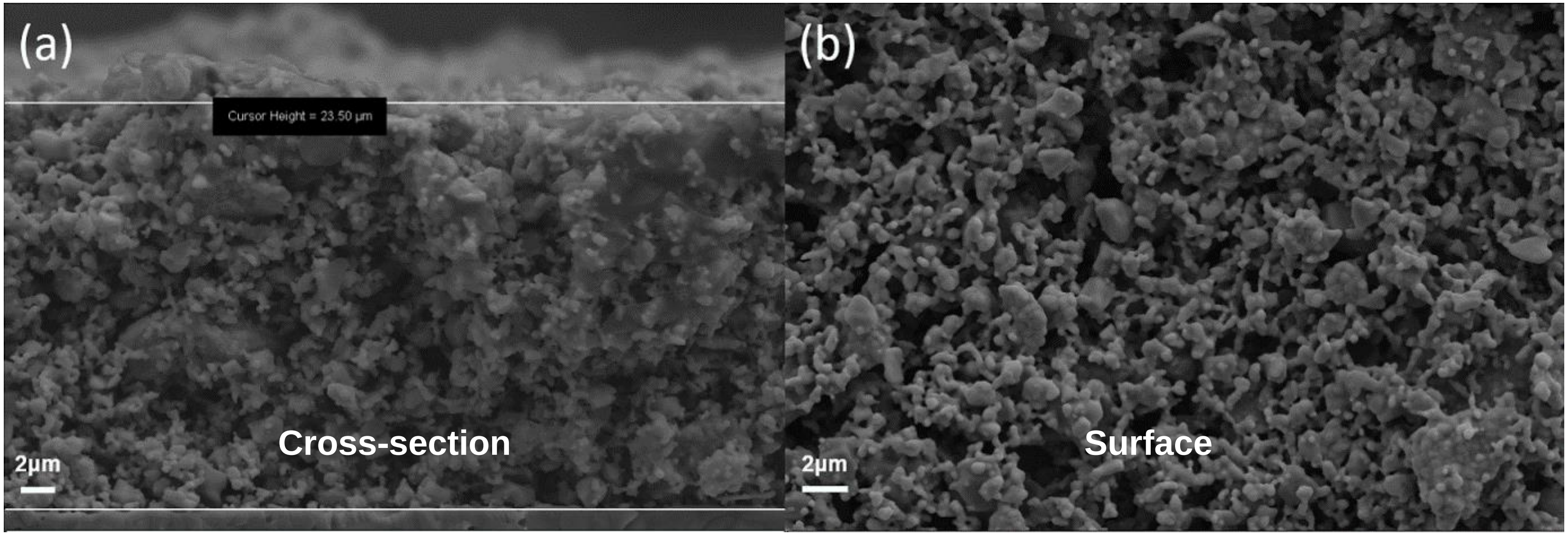


The CO_2 and H_2O Tolerance

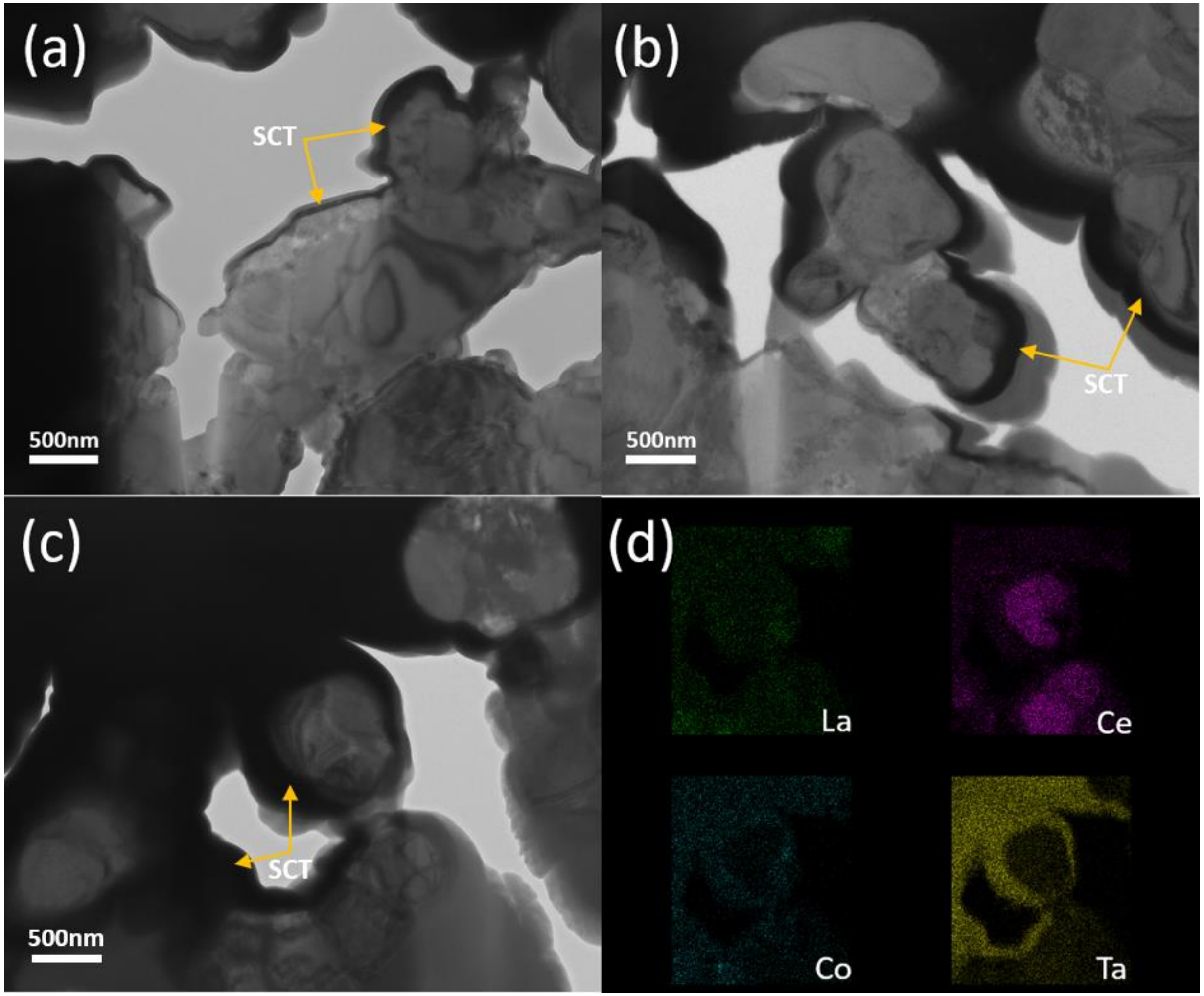




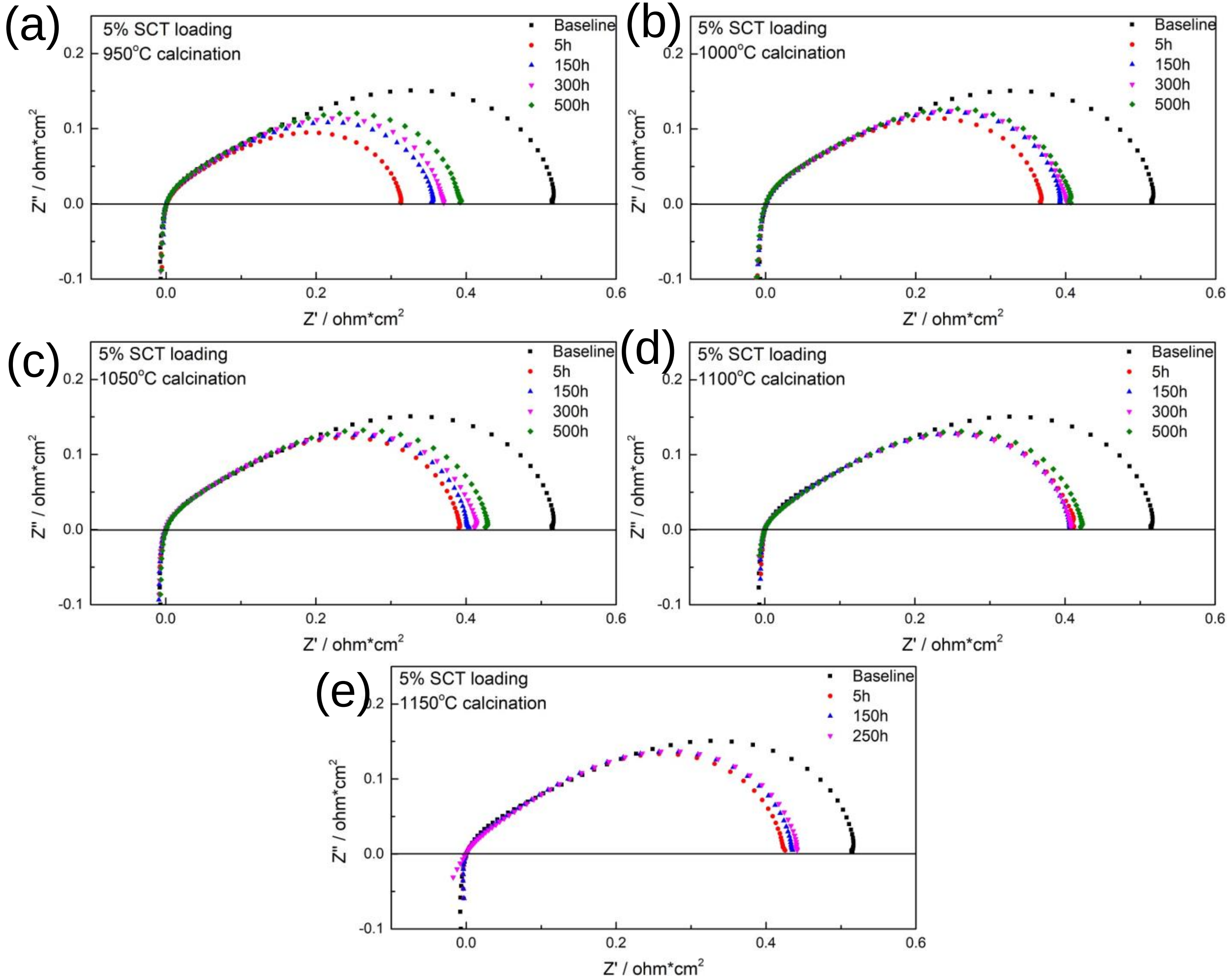
Accomplishment and Progress: LSCF-GDC Skeleton



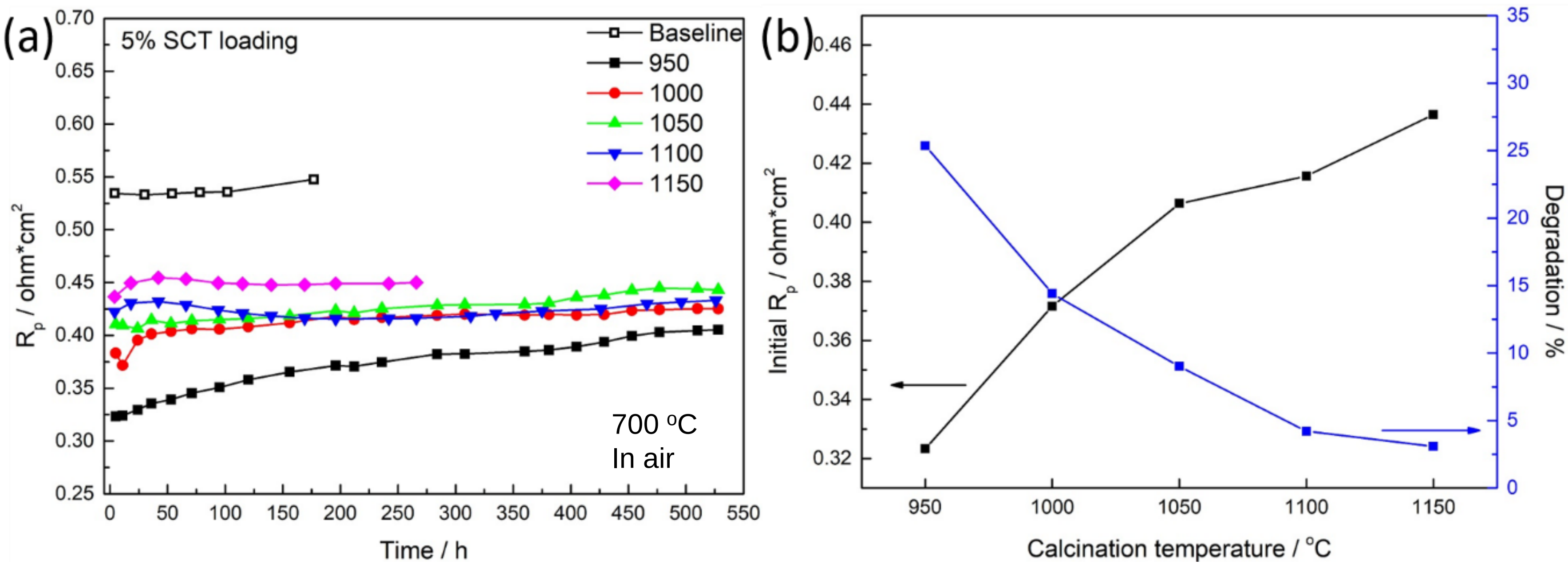
SCT@LSCF-GDC



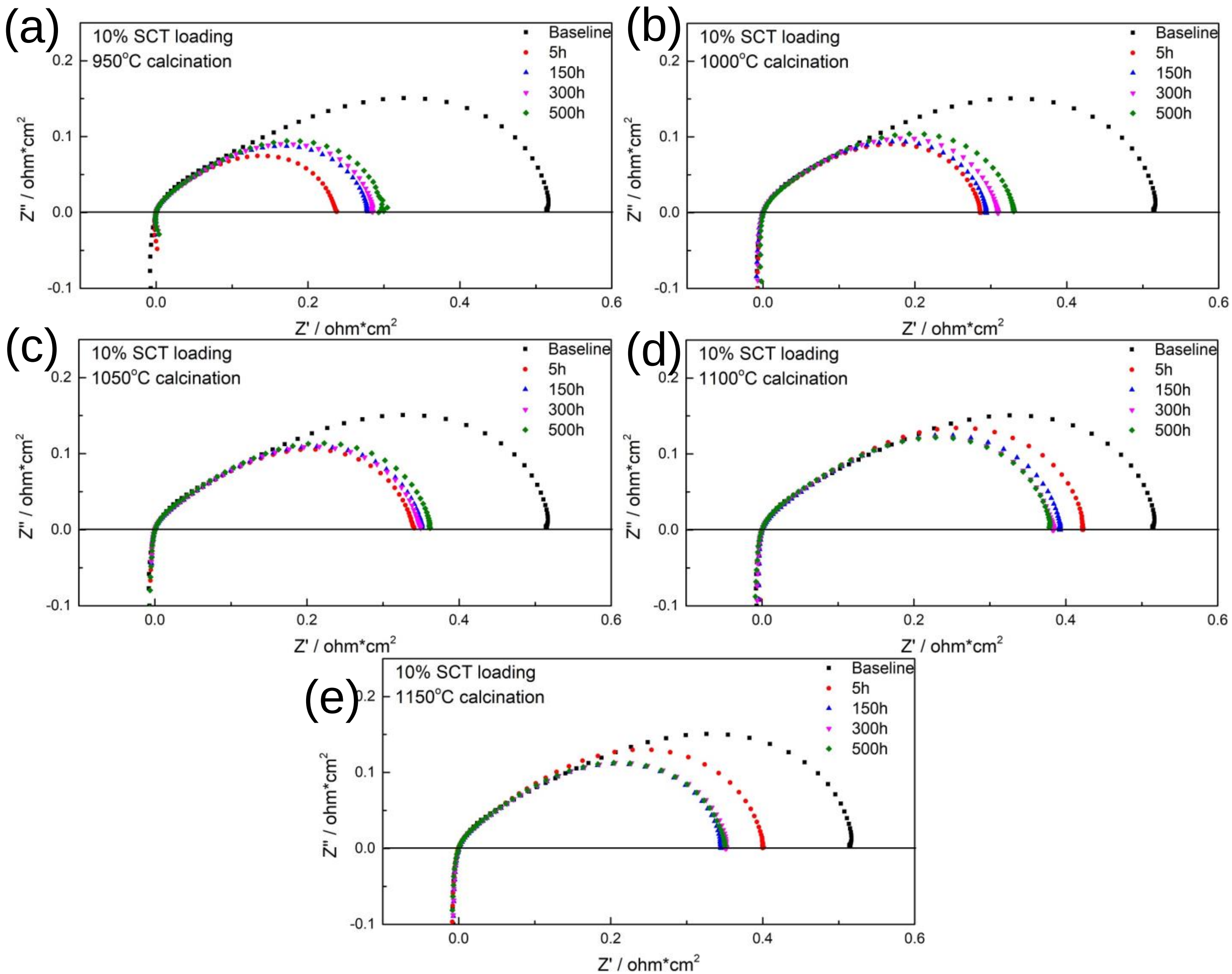
EIS Spectra of 5%SCT@LSCF-GDC



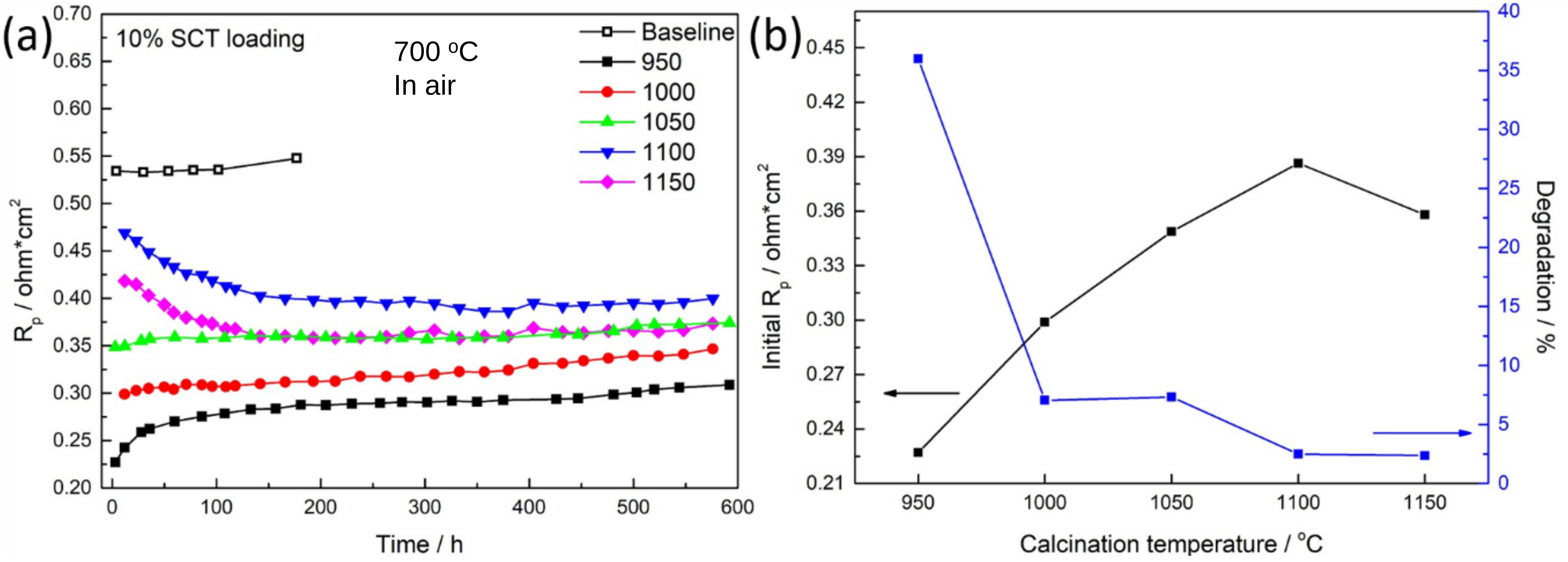
Long-term stability of 5%SCT@LSCF-GDC



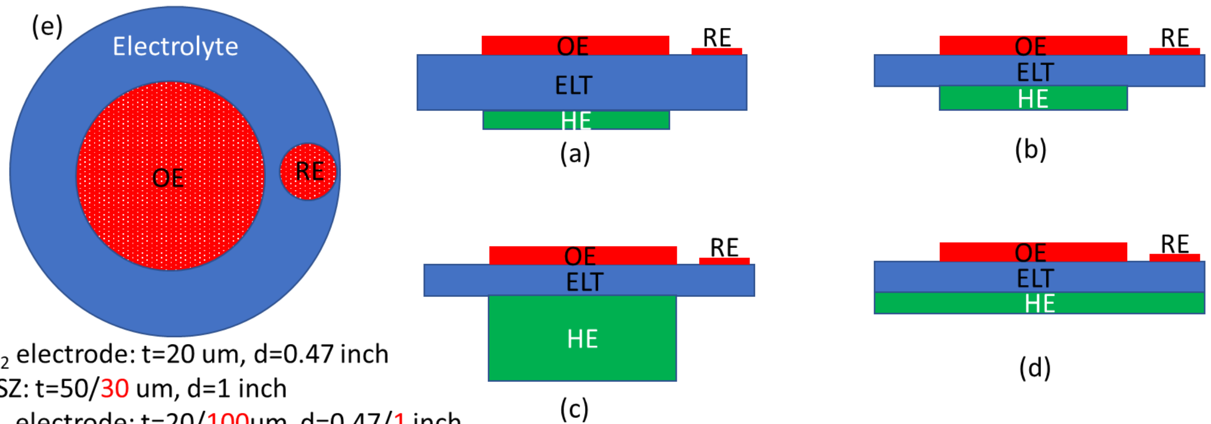
EIS Spectra of 10%SCT@LSCF-GDC



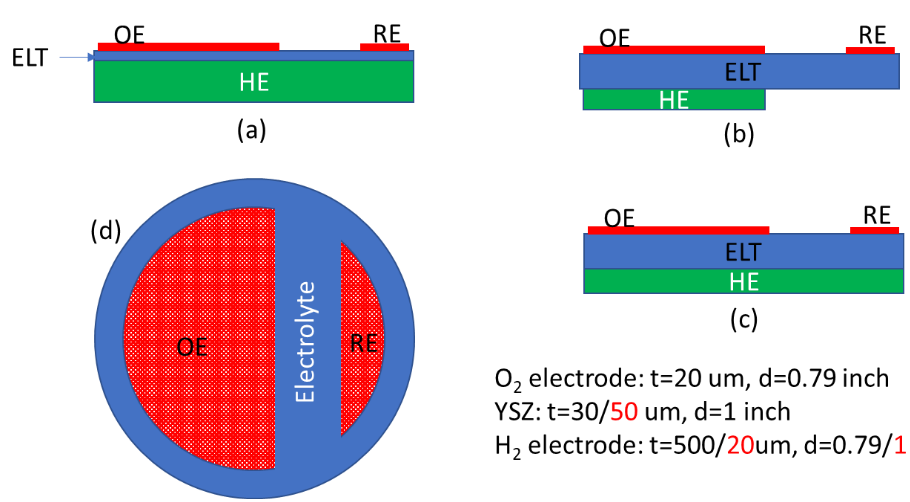
Long-Term Stability of 10%SCT@LSCF-GDC



Theoretical Analysis on 3-electrode Configuration

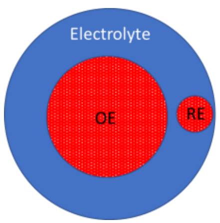
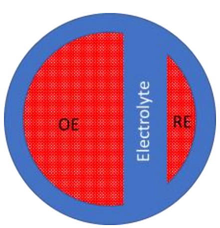


O_2 electrode: t=20 um, d=0.47 inch
YSZ: t=50/30 um, d=1 inch
H_2 electrode: t=20/100um, d=0.47/1 inch



O_2 electrode: t=20 um, d=0.79 inch
YSZ: t=30/50 um, d=1 inch
H_2 electrode: t=500/20um, d=0.79/1 inch

Modeling Results of Three-Electrode Overpotential Measurements

Top View	Cross-Section	Potential @ RE	Potential @ middle of ELT@0.4A/cm²	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: ≤0.15 V OER overpotential at 1 A/cm^2 for 1 kh @ 700 ^oC	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.