DE-FE0031972

Reversible SOFC-SOEC Stacks Based on Stable Rare-Earth Nickelate Oxygen Electrodes

Dr. John Pietras[1], Dr. Srikanth Gopalan[2], Dr. Yu Zhong[3]
Dr. Wenyuan Li[4], Dr. Whitney Colella[5]

1 Saint-Gobain
2 Boston University
3 Worcester Polytechnic Institute
4 West Virginia University
5 Gaia Energy Research Institute

26 October 2022

SAINT-GOBAIN GROUP

Commitment to achieve **carbon neutrality in 2050**

World or European leader in most of our businesses

Founded over
350
years ago

Locations in
76
countries

Saint-Gobain Research
8
cross-business R&D centers

More than
166,000
employees

Approximately
800
manufacturing facilities

Around
3,500
sales outlets

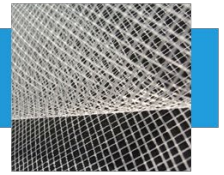
HIGH PERFORMANCE SOLUTIONS


ABRASIVES & COMPOSITE SYSTEMS


CERAMICS


LIFE SCIENCES


MOBILITY


CONSTRUCTION INDUSTRY

REGIONAL BUSINESS


CERTAINTEED BUILDING MATERIALS


SAGE ELECTROCHROMIC GLASS

SAINT-GOBAIN

CERAMICS – KEY FIGURES

SEFPRO It's a matter of trust. · ZirPro · Valoref · SAINT-GOBAIN

Over **5,500 employees**

Manufacturing sites in **10 countries**

50 plants

Global sales operations

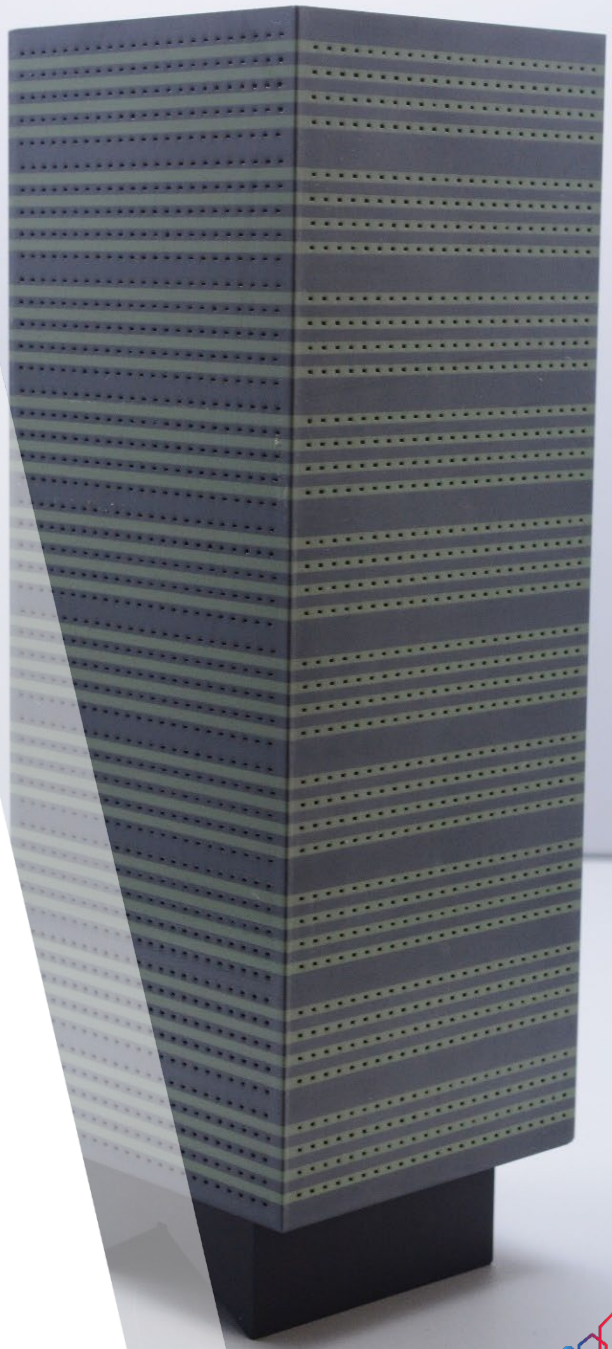
DESIGN → **MANUFACTURE** → **DISTRIBUTE** → **RECYCLE**

Strong expertise in materials science leading to solutions co-developed with our customers

State-of-the-art materials for critical industrial applications

In-depth customer knowledge for a personalized digital experience

A progressive increase in the proportion of recycled content in our products



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REVERSIBLE SOFC-SOEC STACKS BASED ON STABLE RARE-EARTH NICKELATE OXYGEN ELECTRODES: DE-FE0031972

Objectives to be reached during this project

1. Establish state-of-the-art oxygen electrode materials
2. Stabilize Ni-YSZ hydrogen electrode against Ni migration - utilize infiltration
3. Quantify the effect of cell & stack design on durability – then improve it
4. Develop and quantify cost-effective and scalable manufacturing

BOSTON UNIVERSITY

WPI

WestVirginiaUniversity.

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Gaia Energy Research Institute LLC (Gaia)
Energy, Environmental, and Engineering Research

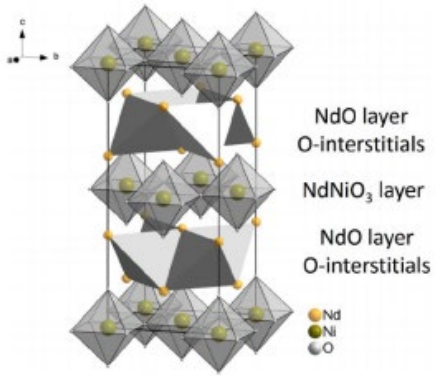
Entering Budget Period 2

Grant Program Activity	Budget Federal	Non-Federal	Total
Budget Period 1	$796,976	$203,754	$1,000,730
Budget Period 2	$798,961	$197,689	$996,650
Budget Period 3	$794,730	$196,226	$990,956
Totals	**$2,390,667**	**$597,669**	**$2,988,336**

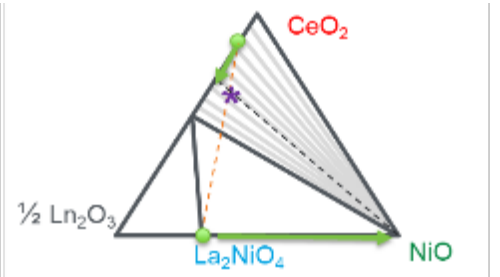
Acknowledgement

NATIONAL ENERGY TECHNOLOGY LABORATORY

Ln_2NiO_4 Air Electrode



Performance Potential



Stability Challenge

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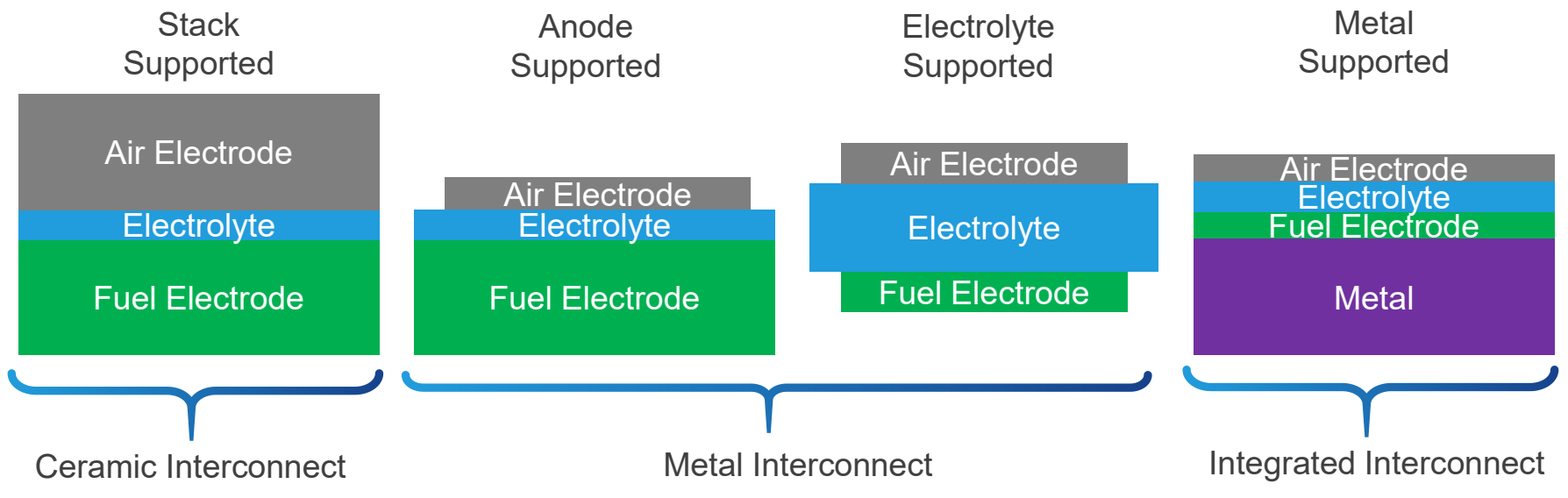
WIDESPREAD ADOPTION OF HIGH TEMPERATURE ELECTROLYSIS RELIES ON HIGH VOLUMES – FOCUS ON TRANSFERABLE RESULTS

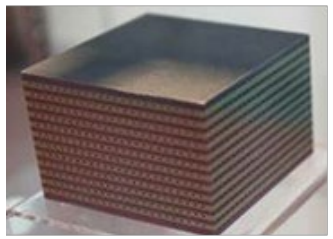
Designs can be grouped by the thickest layer of an individual cell and how they are connected into a stack

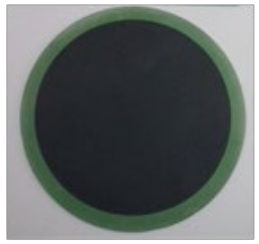


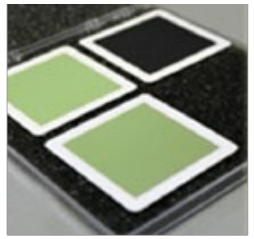
Commonalities

- Air electrode activity vs. temp.
- High humidity @ fuel electrode
- Interconnects Metal or Ceramic

Stack Developers







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FOCUS ON STACK AND REVERSIBLE OPERATION MODE ISSUES

SOLUTIONS AT EACH LEVEL DESIGNED TO BE PORTABLE TO MANY SYSTEM CONFIGURATIONS

Cell Level

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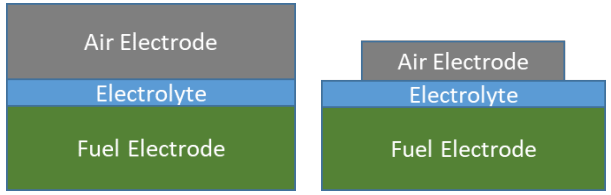
Oxygen Electrode (Nickelates)



Stack Supported | Anode Supported

Fuel Electrode (Nickel migration)

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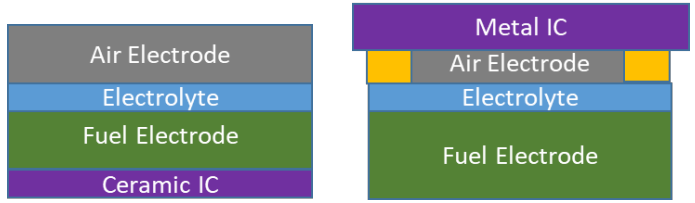
Stack Level

WPI · West Virginia University

Nickelate – Cr interconnect interactions



Stack Supported | Anode Supported

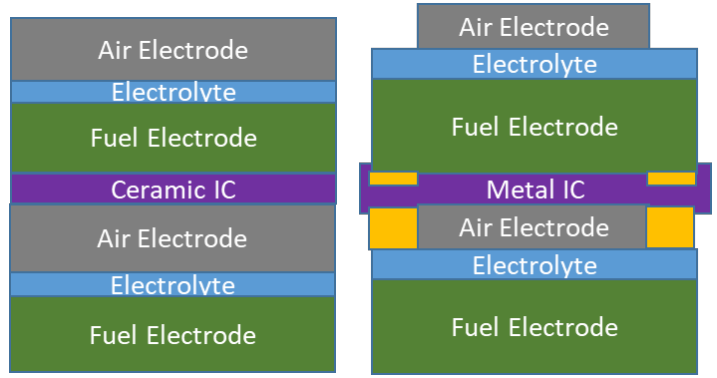
Ceramic IC humidity interactions

West Virginia University · SAINT-GOBAIN

Scale Up

SAINT-GOBAIN · West Virginia University

Solution integration and stack testing



Stack Supported | Anode Supported

Techno-economic analysis

Gaia Energy Research Institute LLC (Gaia)
Energy, Environmental, and Engineering Research

WPI · SAINT-GOBAIN

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CELL LEVEL MODELING

DETERMINE AND MODEL THE OXYGEN DIFFUSION MECHANISM IN NICKELATE STRUCTURE

Cell Level

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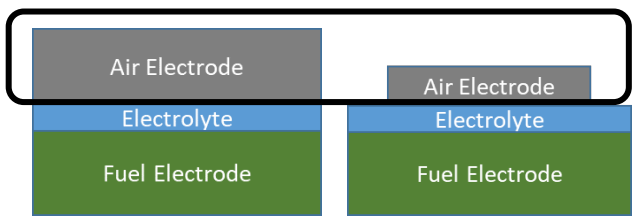
Oxygen Electrode Investigations (**Nickelates**)



Stack Supported

Anode Supported

Fuel Electrode Investigations (**Nickel migration**)

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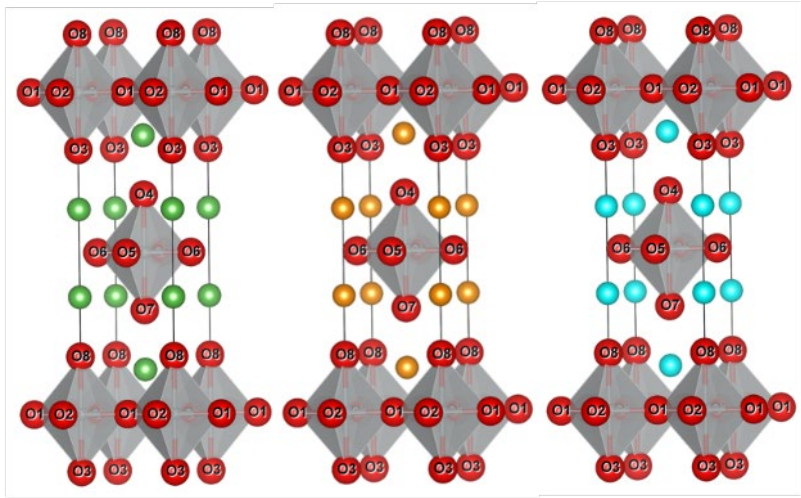
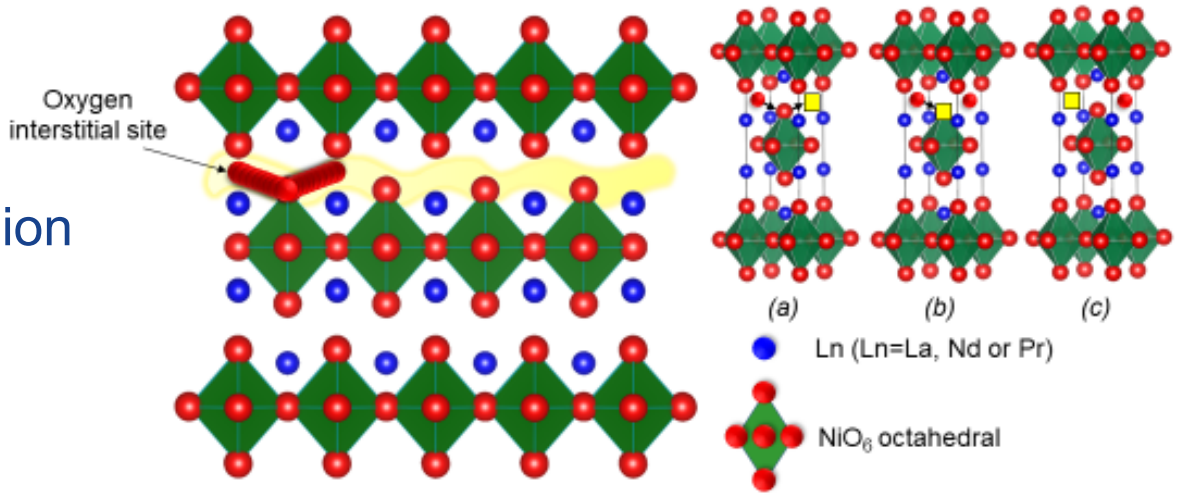
Vacancy Diffusion

vs

Interstitial Diffusion



$La_4Ni_2O_8$ $Nd_4Ni_2O_8$ $Pr_4Ni_2O_8$



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CELL LEVEL MODELING

VACANCY DIFFUSION HAS VERY HIGH ACTIVATION ENERGY, MOST LIKELY NOT ACTIVE

Cell Level

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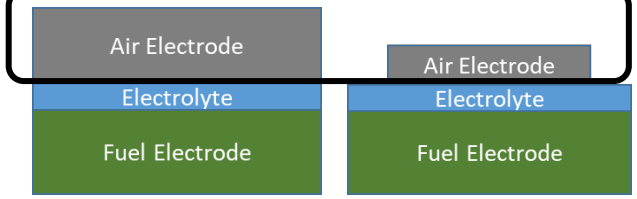
Oxygen Electrode Investigations (**Nickelates**)



Stack Supported

Anode Supported

Fuel Electrode Investigations (**Nickel migration**)

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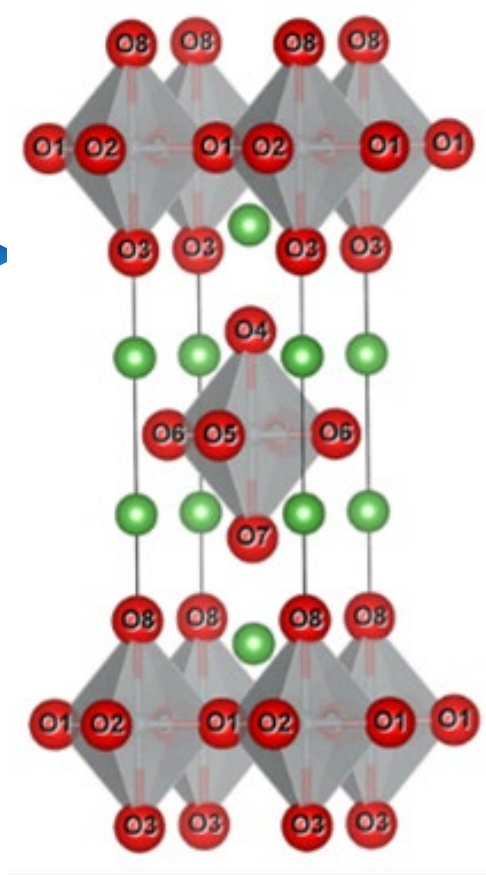
Eight oxygen sites

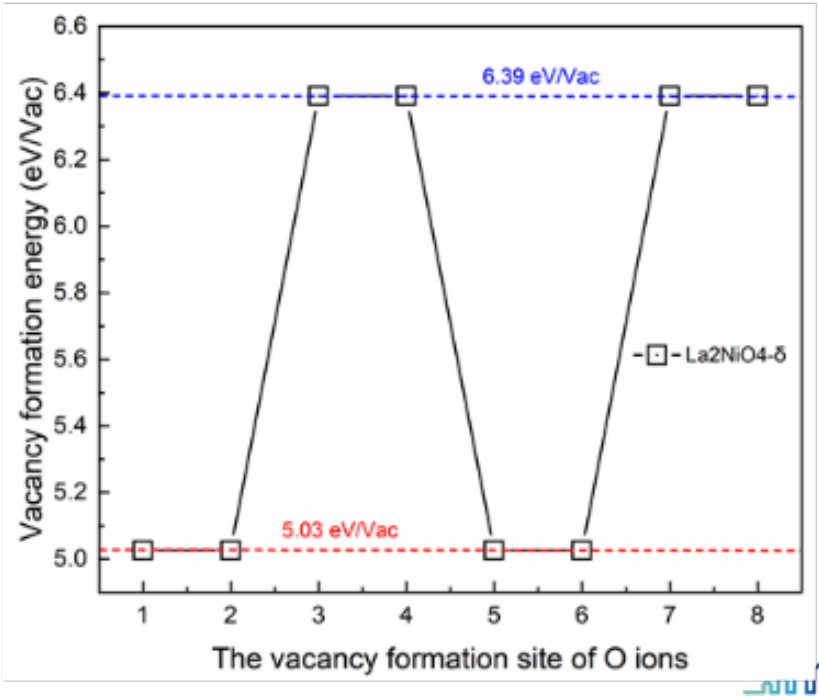


Vacancy Creation

$La_4Ni_2O_8$ $Nd_4Ni_2O_8$ $Pr_4Ni_2O_8$

$$Ln_4Ni_2O_8 \xrightarrow{-O} Ln_4Ni_2O_7$$

$$\Delta E_{Vf} = E^{ab\ initio}_{Ln4Ni2O7}(vac) - E^{ab\ initio}_{Ln4Ni2O8} + \frac{1}{2}\left(E^{ab\ initio}_{O2} + \Delta h^{0}_{O2}\right)$$



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CELL LEVEL MODELING

THREE PATHS FOR OXYGEN DIFFUSION THROUGH INTERSTITIAL MOVEMENT

Cell Level

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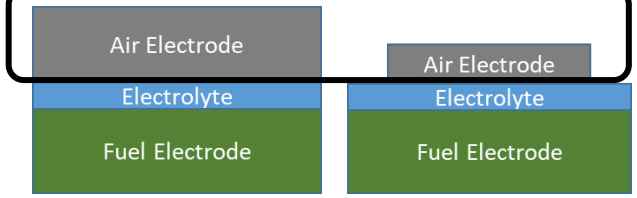
Oxygen Electrode Investigations (**Nickelates**)



Stack Supported

Anode Supported

Fuel Electrode Investigations (**Nickel migration**)

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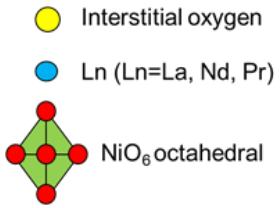
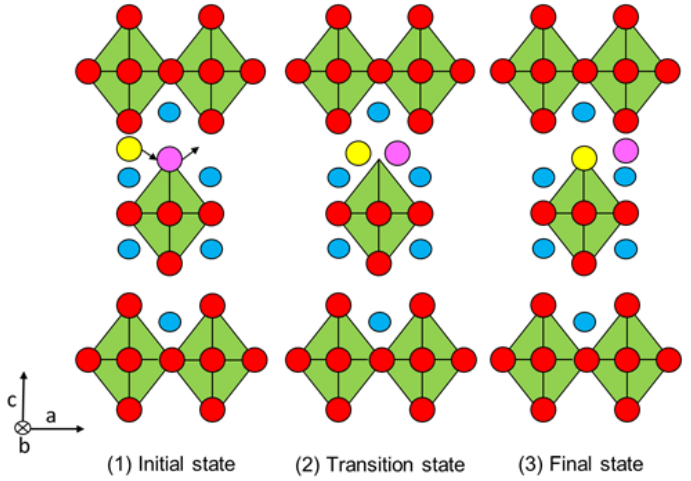
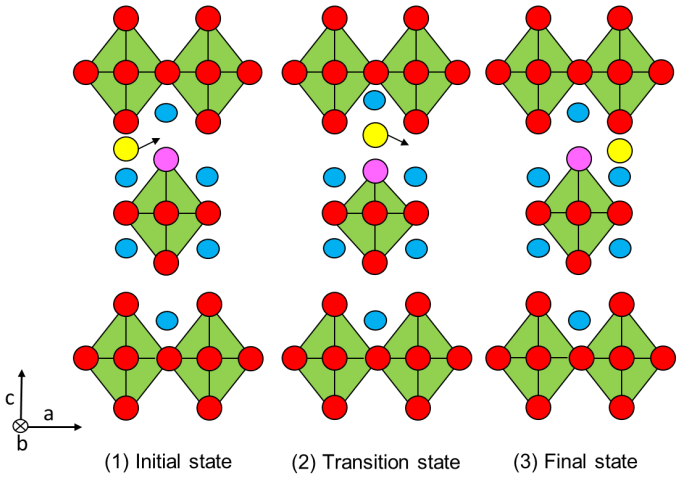


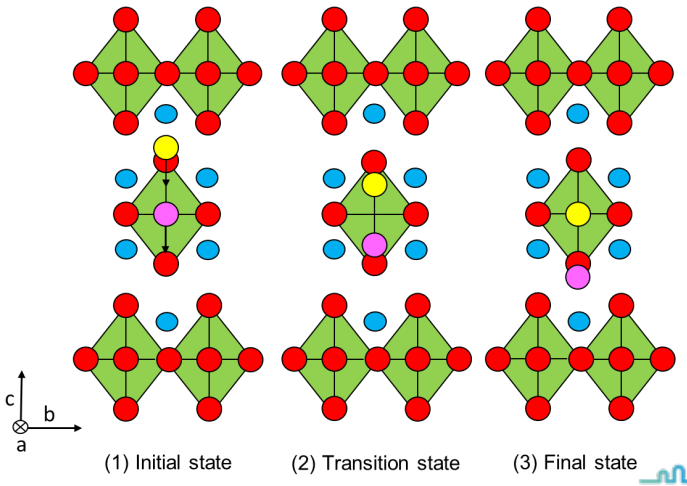
Path 1: bumping along a-b plane



Path 2: squeezing through a-b plane



Path 3: bumping along a-c plane



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CELL LEVEL MODELING

CALCULATED IONIC CONDUCTIVITY IN NICKELATE SYSTEM

Cell Level

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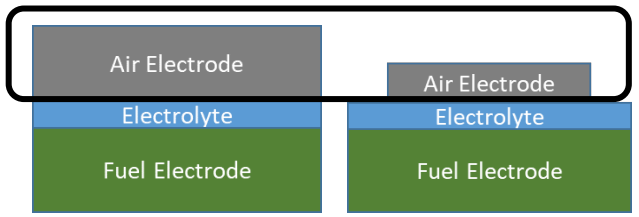
Oxygen Electrode Investigations (**Nickelates**)



Stack Supported

Anode Supported

Fuel Electrode Investigations (**Nickel migration**)

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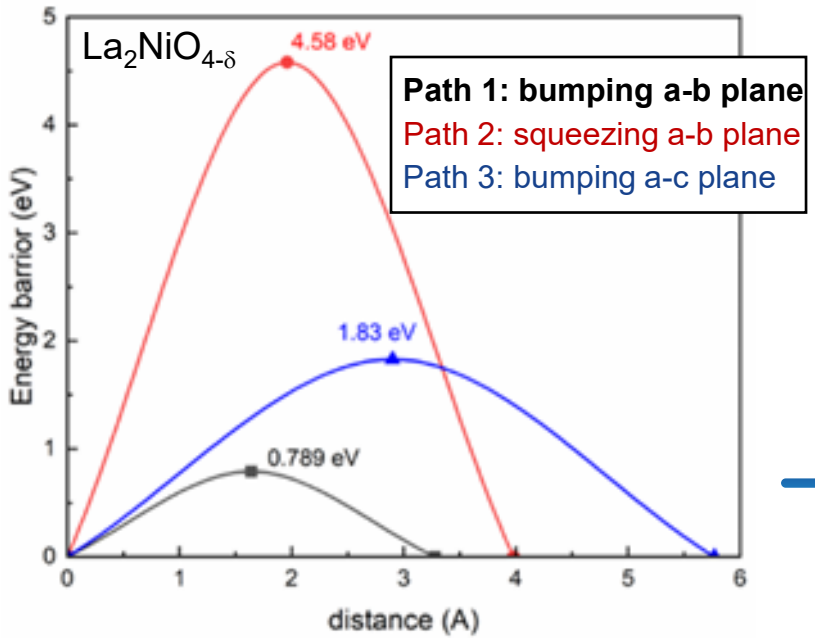
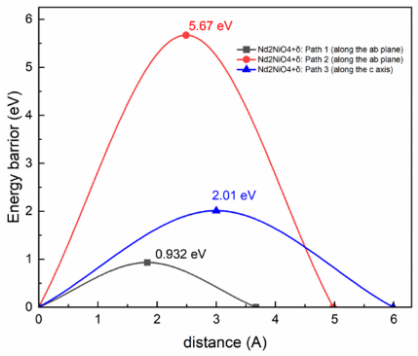
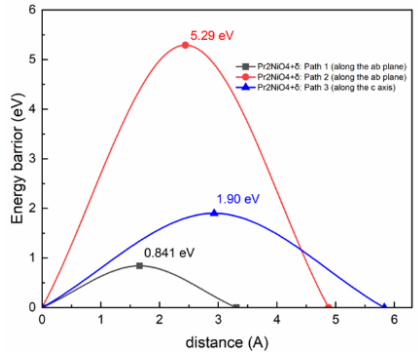
Energy barrier in $Ln_2NiO_{4-\delta}$



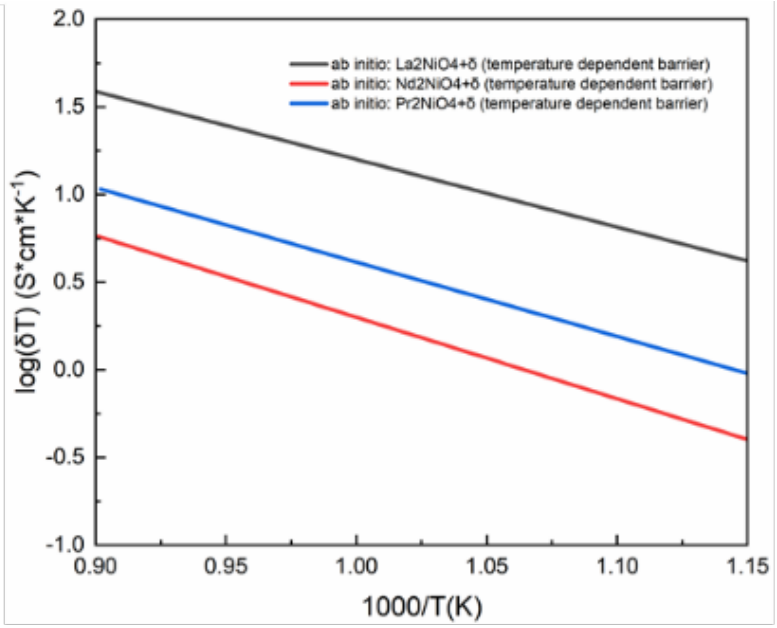
$Nd_2NiO_{4-\delta}$



$Pr_2NiO_{4-\delta}$



Conductivity in $Ln_2NiO_{4-\delta}$ La > Pr > Nd



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CELL LEVEL OPTIMIZATION STUDY

INVESTIGATE COMPOSITION/DESIGN PERFORMANCE-STABILITY RELATIONSHIP

Cell Level

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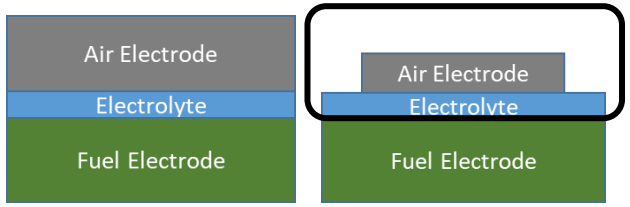
Oxygen Electrode Investigations (Nickelates)



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Fuel Electrode Investigations (Nickel migration)

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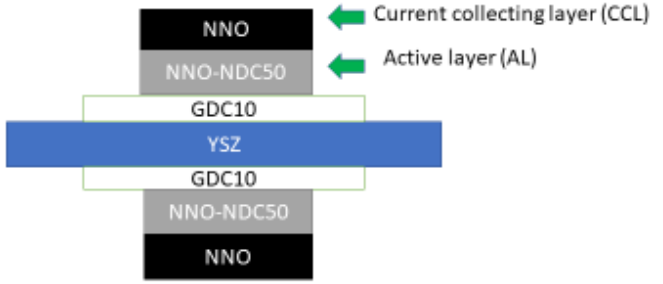
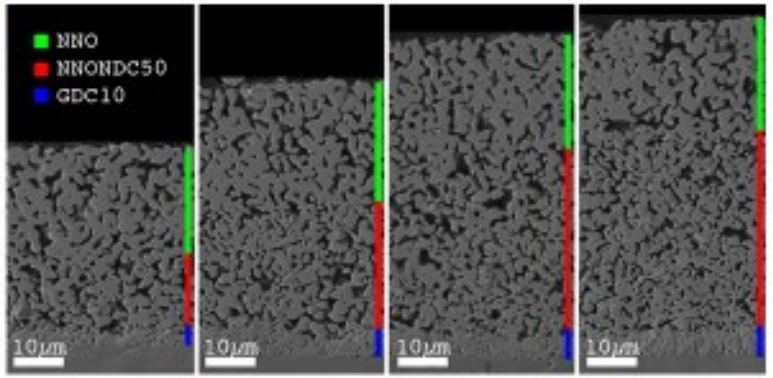
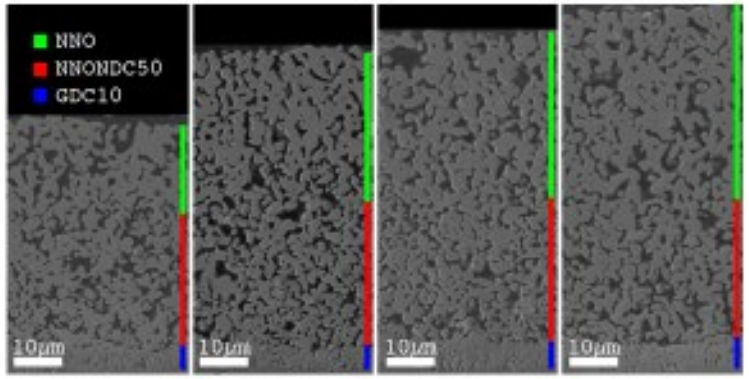
Architecture Optimization Using Symmetrical Cells

- Active layer (NNO-NDC50) thickness
- Current collecting layer (NNO) thickness
- Measurement of ohmic and polarization resistance of the symmetrical cells at 800 and 700°C in air





AL thickness variation



CCL thickness variation

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CELL LEVEL DEVELOPMENTAL WORK

STACK AGNOSTIC SOLUTIONS FOR MODE SWITCHING ON THE AIR ELECTRODE

Cell Level

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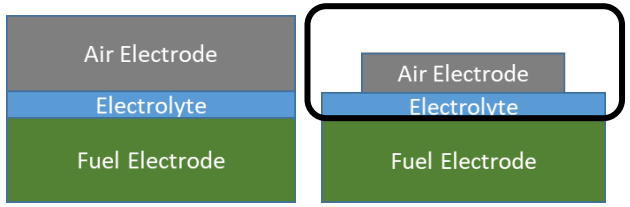
Oxygen Electrode Investigations (Nickelates)



Stack Supported | Anode Supported

Fuel Electrode Investigations (Nickel migration)

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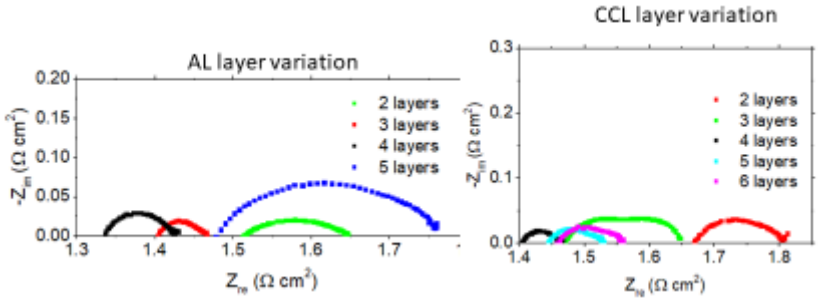
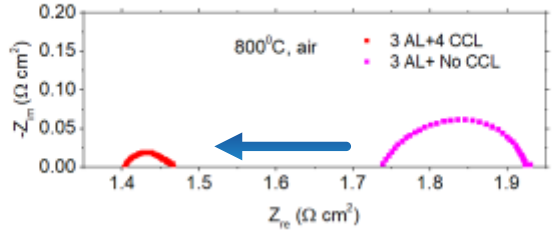
Optimizing increase of active sites vs diffusion distance to the electrolyte

- Optimized thicknesses for AL and CCL in symmetrical cell: 3 AL (27 µm) and 4 CCL (23 µm)

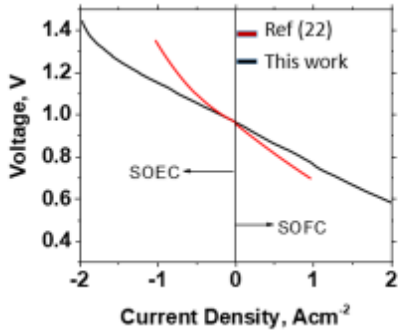


Cell with combined optimized layer thicknesses

- 24% improvement in the ohmic resistance and 170% improvement in the polarization resistance due to the CCL addition.



Improved cell performance



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CELL LEVEL DEVELOPMENTAL WORK

STACK AGNOSTIC SOLUTIONS FOR MODE SWITCHING ON THE AIR ELECTRODE

Cell Level

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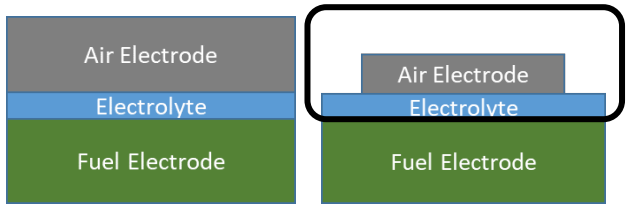
Oxygen Electrode Investigations (Nickelates)



Stack Supported

Anode Supported

Fuel Electrode Investigations (Nickel migration)

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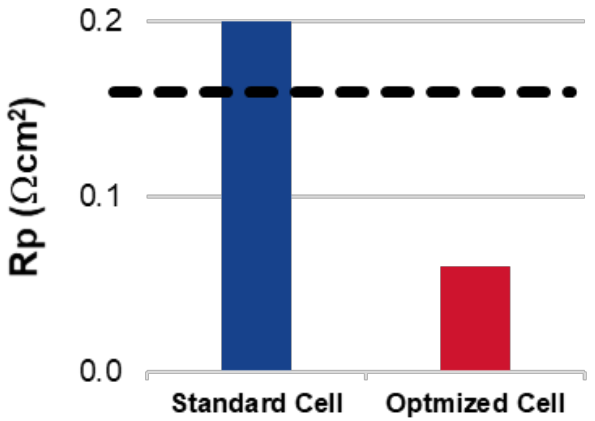
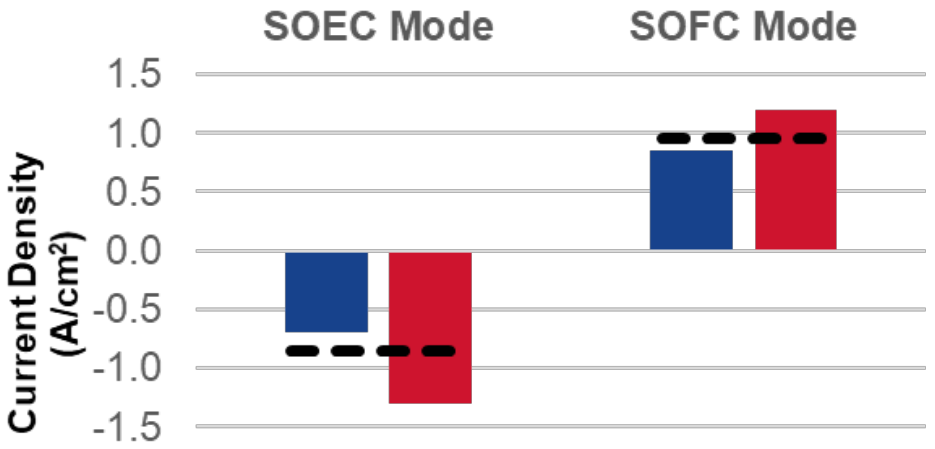
Passed Budget Period 1 Performance Milestones

Reduce polarization by 15%



Increase performance by 10%



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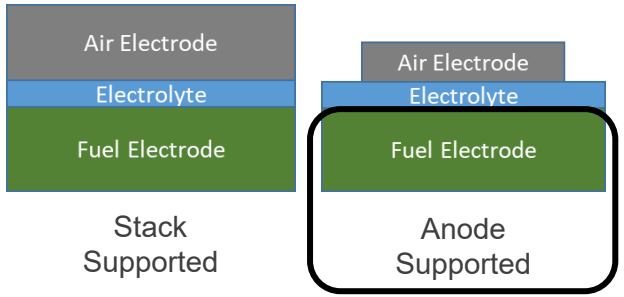
CELL LEVEL DEVELOPMENTAL WORK

NICKEL MIGRATION AS A FUNCTION OF CURRENT AND HUMIDITY

Cell Level

Oxygen Electrode Investigations (**Nickelates**)

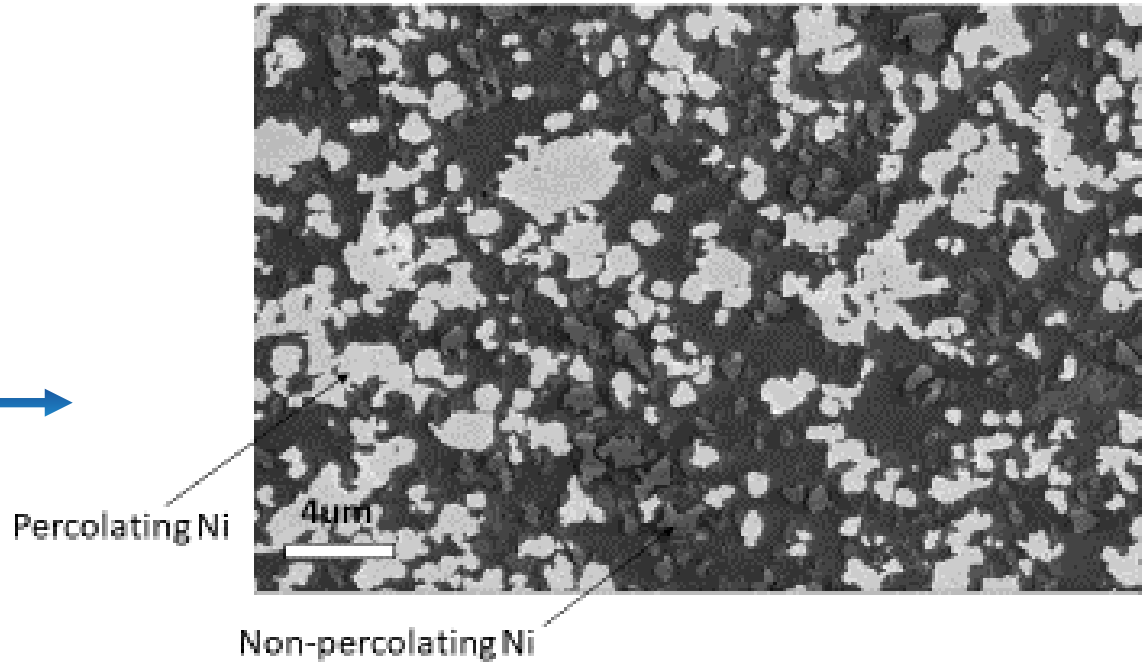


Fuel Electrode Investigations (**Nickel migration**)

Mitigation of Ni migration

- Focus solutions on MIEC anode side infiltration
- Dynamic SOFC-SOEC mode switching
- A range of simulated fuel compositions & temperatures
- Microstructure and compositional evolution probed through SEM, TEM and SEM-FIB analysis
- Studies to be guided by CALPHAD

Visualization of percolating Ni through low voltage SEM



CELL LEVEL DEVELOPMENTAL WORK

NICKEL MIGRATION AS A FUNCTION OF CURRENT AND HUMIDITY

Cell Level

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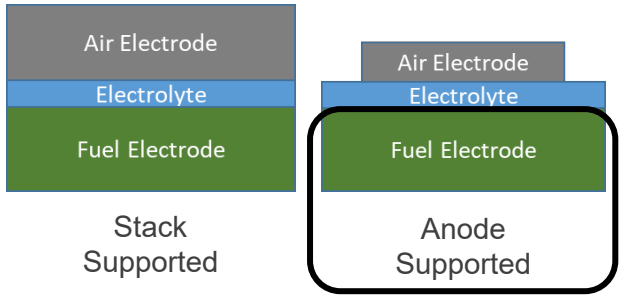
Oxygen Electrode Investigations (**Nickelates**)



Fuel Electrode Investigations (**Nickel migration**)

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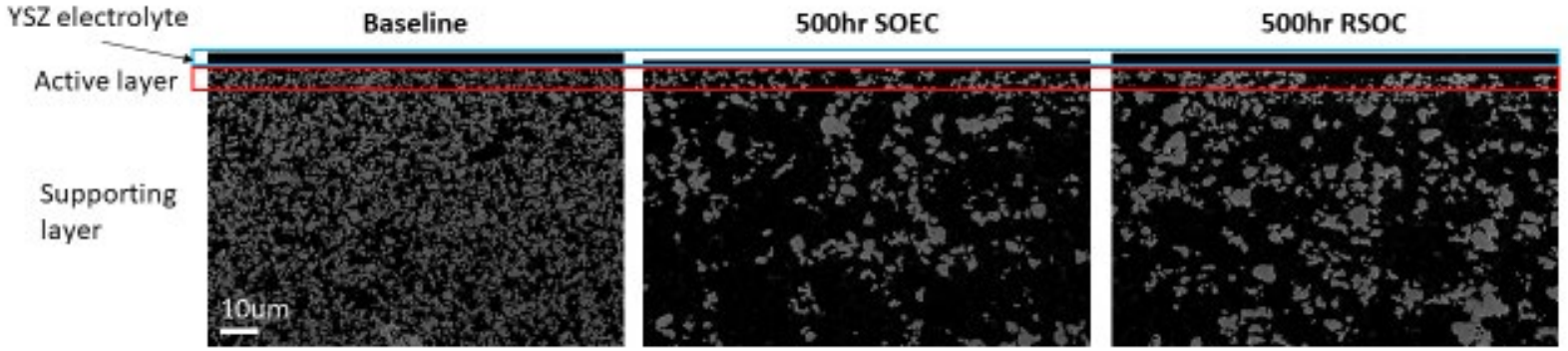
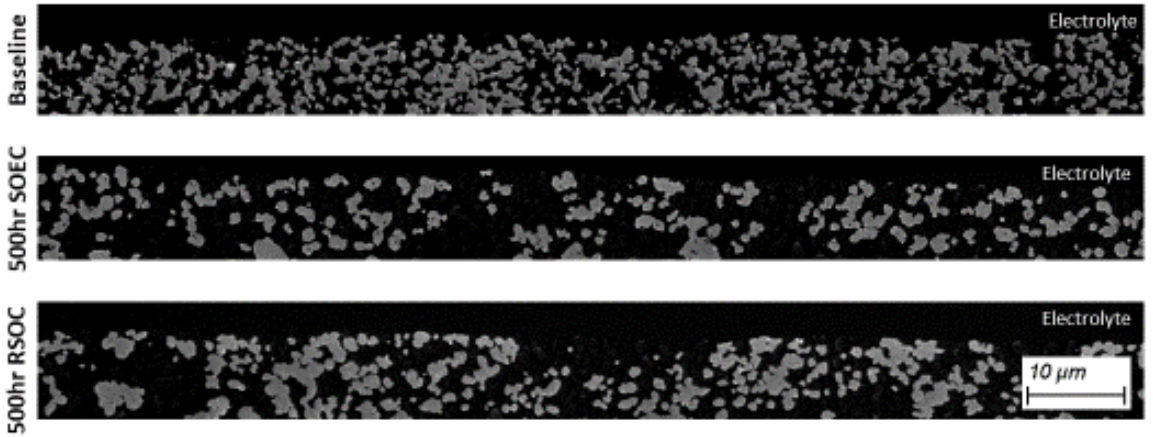
Imaging percolating nickel in the hydrogen electrode



Quantification of change as a function of operation mode



Percolated Nickel	
Volume Fraction	PSD (μm)
0.41	1.24
0.23	1.41
0.27	1.55

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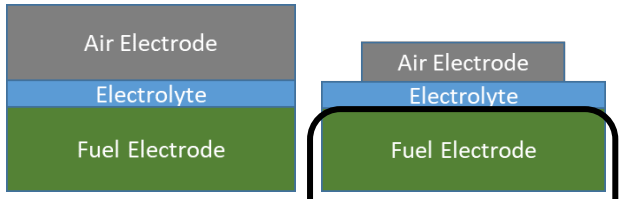
CELL LEVEL DEVELOPMENTAL WORK

DURABILITY MEASUREMENTS IN SOEC MODE

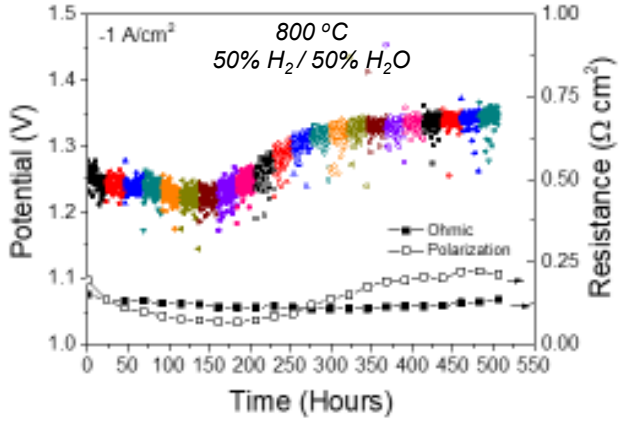
Cell Level

Oxygen Electrode Investigations (**Nickelates**)

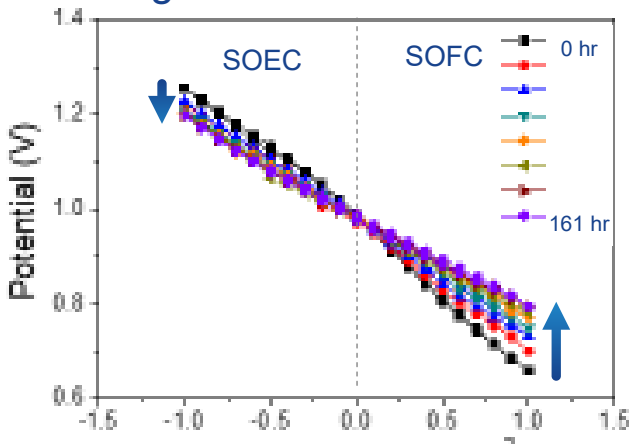


Fuel Electrode Investigations (**Nickel migration**)

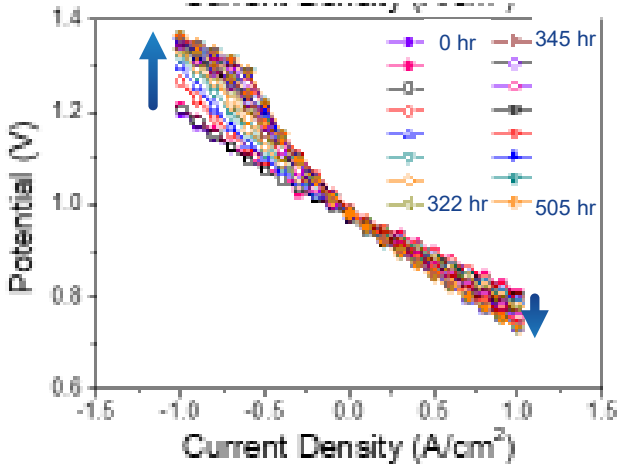
Performance shows improvement over first 150 hrs followed by degradation which slows over time



Improvement more significant in SOFC mode



Degradation more significant in SOEC mode

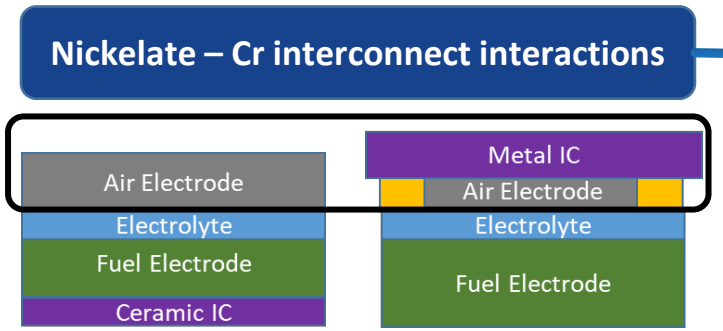


CHALLENGES OF CELL TO CELL CONNECTIONS WITHIN A STACK

DEVELOPMENT OF ACCELERATED CHROME DEGRADATION TEST

Stack Level

Nickelate – Cr interconnect interactions

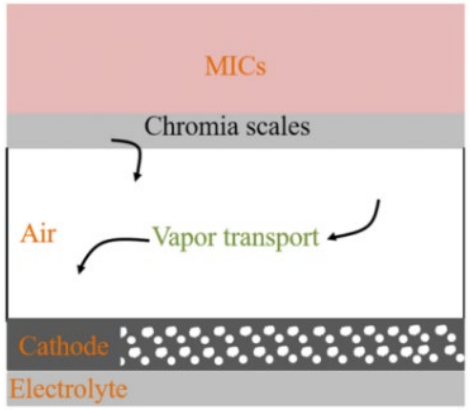


Stack Supported

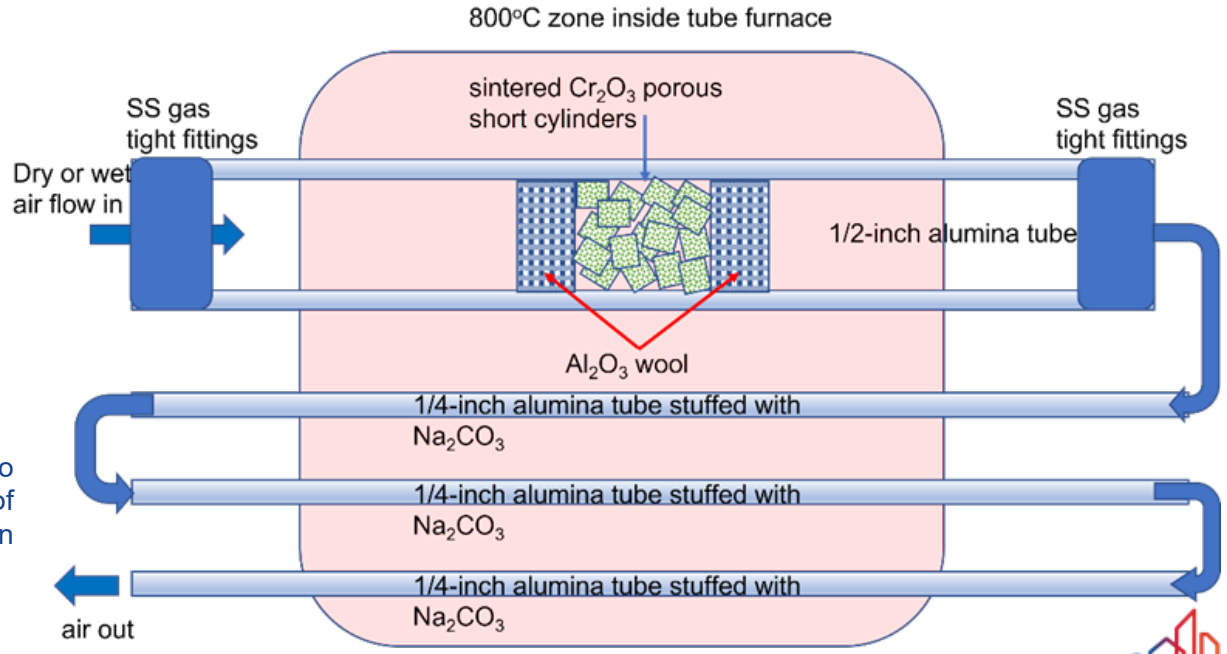
Anode Supported

Ceramic IC humidity interactions

Schematic of chrome transport within a stack



Schematic of chrome testing infrastructure



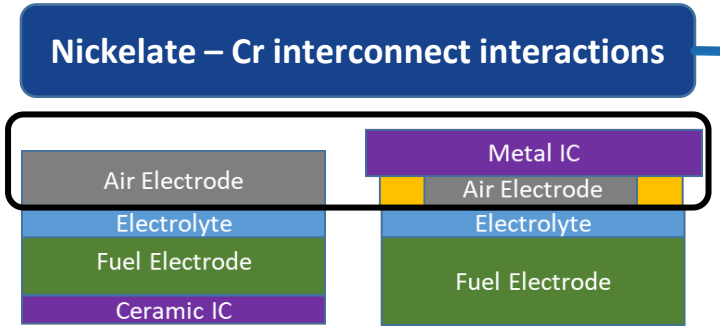
Na_2CO_3 collected to measure Cr content of condition

CHALLENGES OF CELL TO CELL CONNECTIONS WITHIN A STACK

INVESTIGATING EFFECT OF METALLIC INTERCONNECTS ON NICKELATE AIR ELECTRODES

Stack Level

Nickelate – Cr interconnect interactions

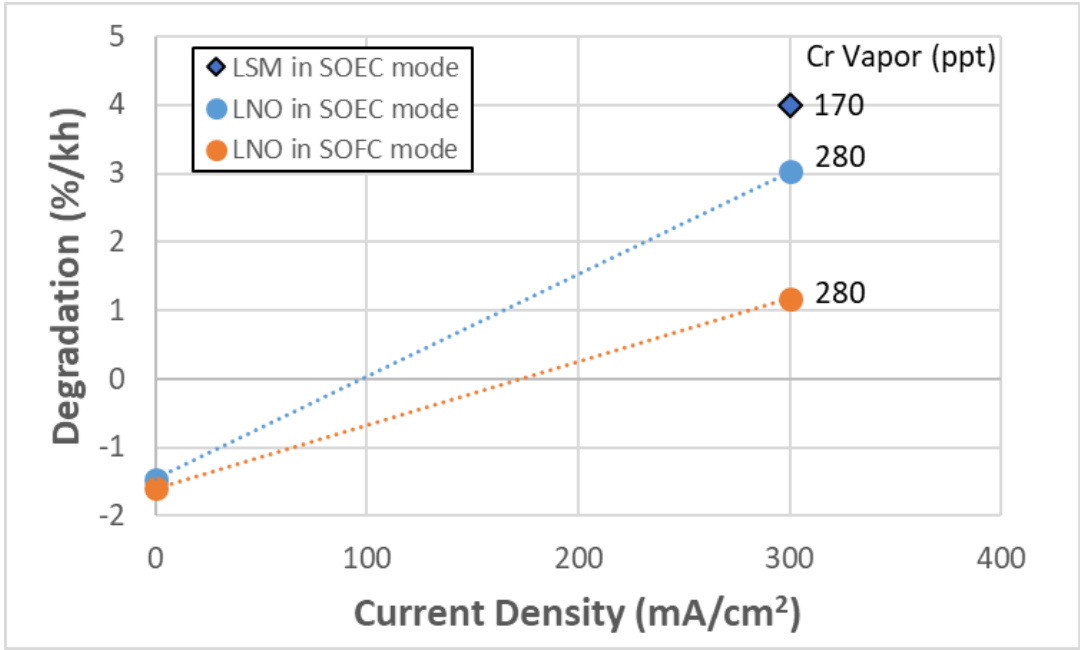


Stack Supported

Anode Supported

Ceramic IC humidity interactions

Nickelate electrode shows lower degradation rate under greater chrome vapor level compared to baseline LSM



800 °C, 60% H_2 / 40% H_2O, dry air

ENABLING STACK AGNOSTIC VOLUME PRODUCTION

LOW COST, HIGH VOLUME POWDER PRODUCTION CRITICAL FOR SOEC/SOFC ADOPTION

Reproduce solid-state synthesis method
Confirmed with XRD

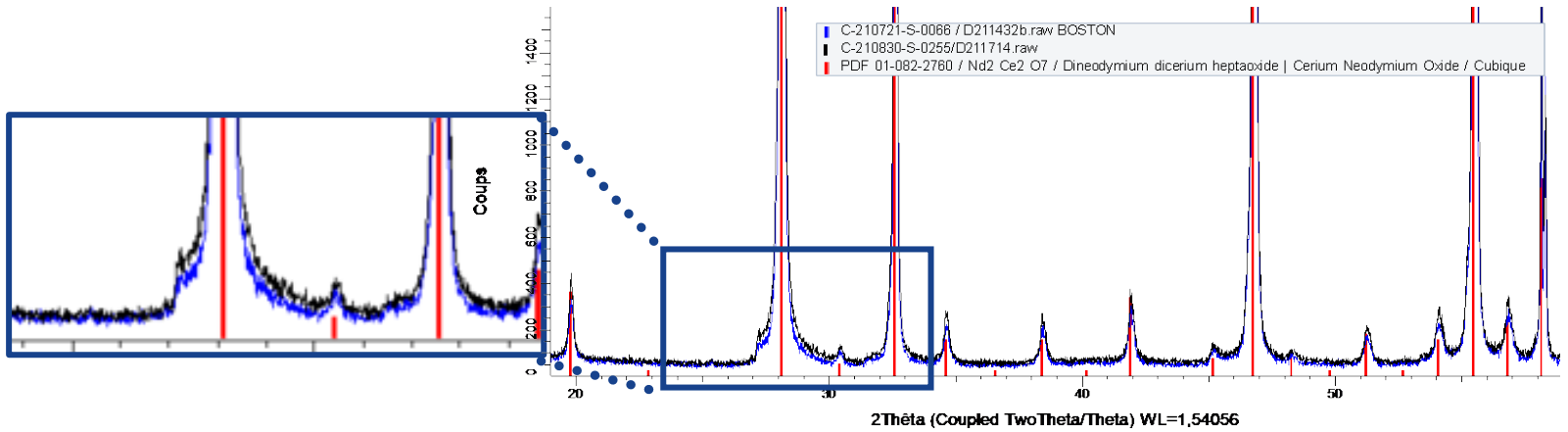
Synthesis by wet-chemistry method
- Confirmed with XRD
- Optimized calcine temp

Synthesis by electrofusion or induction furnace
- Confirmed with XRD

Develop milling procedure
Confirmed in-situ and with dilatometry

Reliable, scalable production of NDC-50

Powder Production
- Multiple commercial paths for power production
- Reproducible phase purity
- Particle size control



Scale Up

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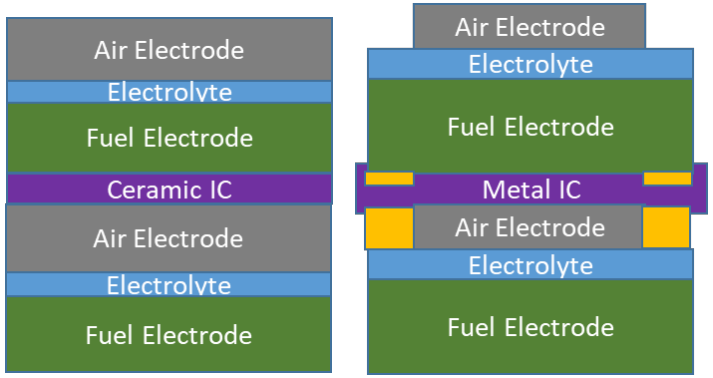
Solution integration and stack testing



Techno-economic analysis

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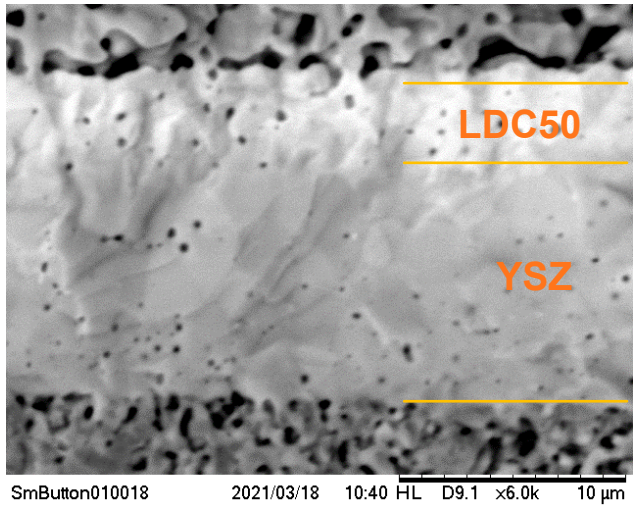
ENABLING STACK AGNOSTIC VOLUME PRODUCTION

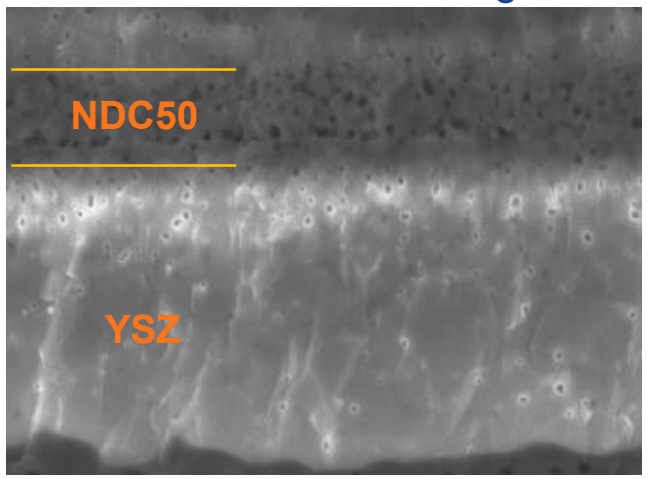
SUBCOMPONENT FABRICATION OF CONSISTENT, THIN LAYERS

Simultaneous Multi-layer Roll-to-Roll Casting

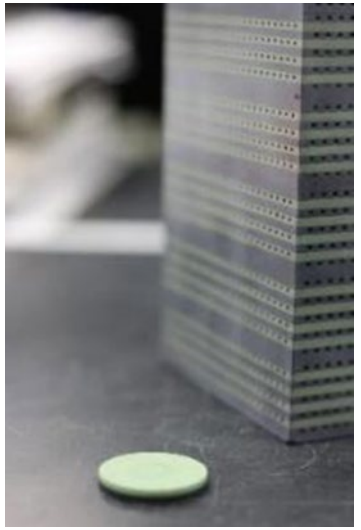


~5um barrier layer

Consecutive Layer Roll-to-Roll Casting



Compatible with standard stack production techniques



Scale Up

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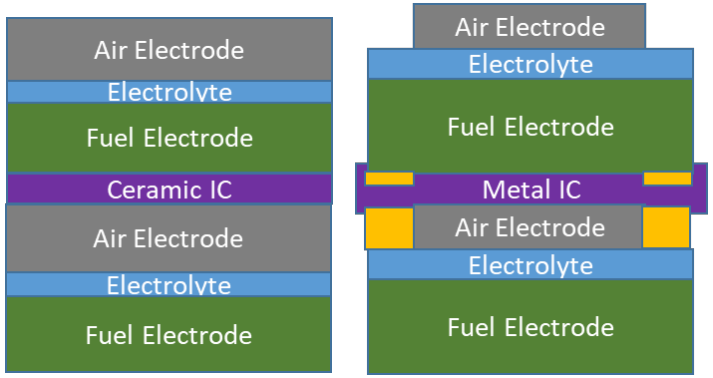
Solution integration and stack testing



Techno-economic analysis

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SUMMARY

Achievements

- Developed a robust, first principals model of Ln_2NiO_4
- Optimized the oxygen electrode geometry resulting in improved performance
- Developed a simple technique to quantify microstructural changes due to Ni migration
- Created a test stand and protocol to measure degradation due to chrome vapor
- Scaled powder production process for critical materials within the air electrode
- Developed a simultaneous multi-layer roll to roll process enabling thin and low-cost production

Next Steps

- Use model to predict behavior of complex stoichiometries
- Produce powders and test cells with advanced air electrode compositions
- Develop infiltration techniques to mitigate Ni migration in high water vapor conditions
- Investigate operational conditions, compositions which minimize degradation behavior
- Scale solutions to short stack level testing
- Utilize performance data for system level performance and cost modeling