
# 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*

18 April 2023

# 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

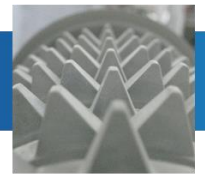
**More than**
**166,000**
employees

**Approximately**
**800**
manufacturing facilities

**Around**
**3,500**
sales outlets

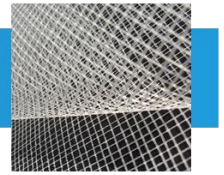
**HIGH PERFORMANCE SOLUTIONS**


**ABRASIVES & COMPOSITE SYSTEMS**


**CERAMICS**


**LIFE SCIENCES**


**MOBILITY**


**CONSTRUCTION INDUSTRY**

**REGIONAL BUSINESS**


**CERTAINTEED BUILDING MATERIALS**


**SAGE ELECTROCHROMIC GLASS**

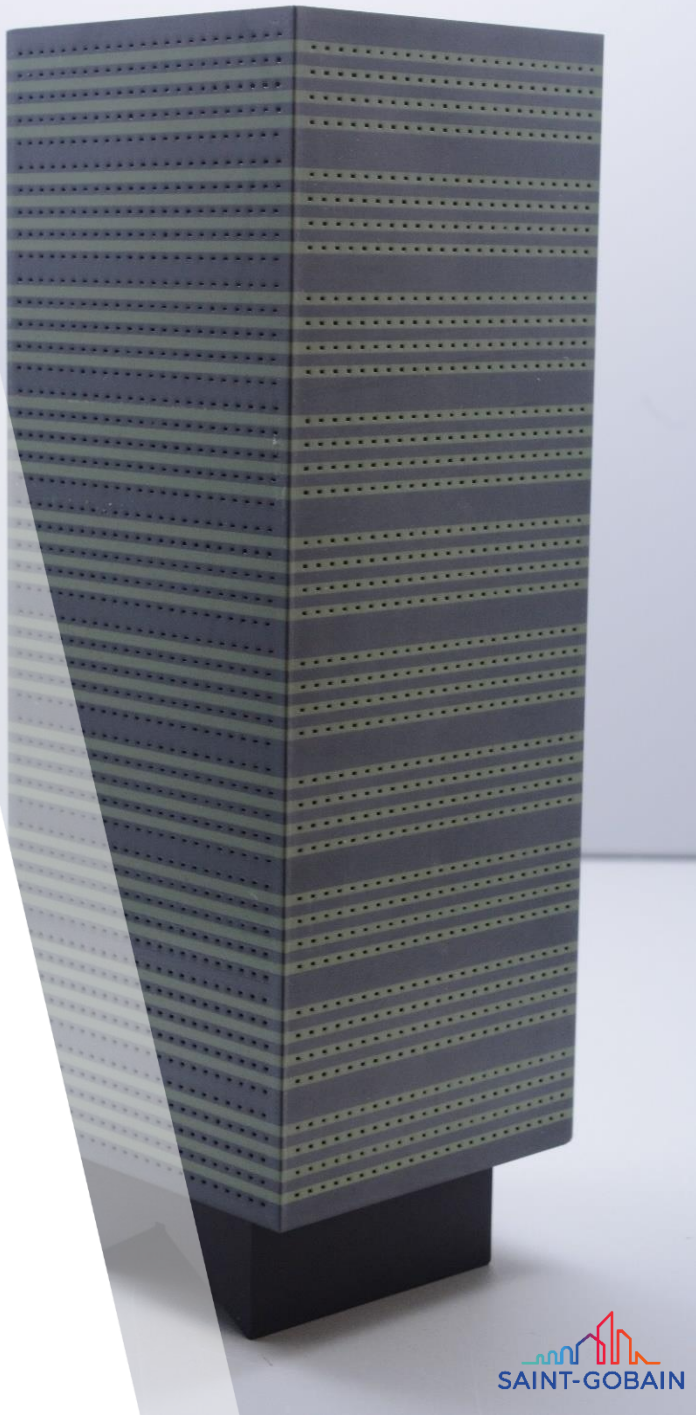
SAINT-GOBAIN

# 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** |



SAINT-GOBAIN

# 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.

SAINT-GOBAIN

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

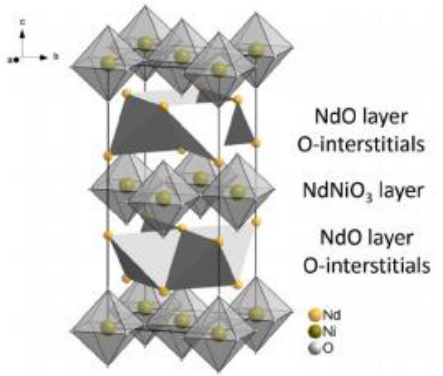
## Completing Budget Period 2

| 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** |

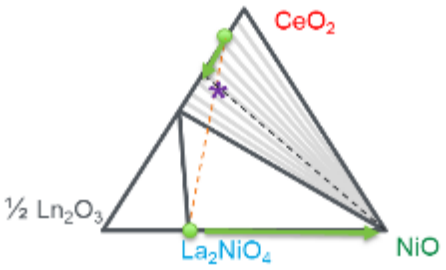
**Acknowledgement**

NATIONAL ENERGY TECHNOLOGY LABORATORY

$Ln_2NiO_4$ Air Electrode




Performance Potential




Stability Challenge

SAINT-GOBAIN

# TASKS AND DELIVERABLES

| Task | Description | Budget Period 1 | Budget Period 2 | Budget Period 3 |
|---|---|---|---|---|
| 1 | **Project management and planning** | • Project Management Plan<br>• Technology Maturation Plan<br>• Reports as specified | • Reports as specified | • Technology Maturation Plan<br>• Reports as specified |
| 2 | **Oxygen electrode development** | | • Report detailing reductions in polarization resistance | |
| 3 | **Hydrogen electrode reversible durability** | • Report detailing improvement in Ni migration | | • Report detailing impact of infiltrants on impurity tolerance |
| 4 | **Thermodynamic analysis** | | • Report summarizing compositions to minimize interconnect reactions | • Report describing optimum performance in both SOEC and SOFC modes |
| 5 | **Impact of stack architecture on cell stability** | | | • Library of Cr-poisoning to oxygen electrode performance in degradation |
| 6 | **Scale up to stack production** | • Button cells delivered | • Short stack delivered | |
| 7 | **Button cell performance and durability testing** | | • Report detailing improvement in current density of cells<br>• Report quantifying compositional and microstructural changes | • Report summarizing milestones 7.3 and 7.4 on improved degradation rate of cells testing in mode switching |
| 8 | **Stack testing** | | | • Report quantifying microstructural changes in first gen. stack after mode switching operation<br>• Report quantifying improvements over first-generation stack after mode switching |
| 9 | **Techno-economic analysis** | | | • Report on effect of improvements made on the cost and operation of stack/system |

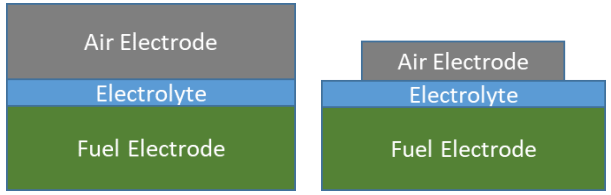
# FOCUS ON STACK AND REVERSIBLE OPERATION MODE ISSUES

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

### Cell Level

BOSTON UNIVERSITY · SAINT-GOBAIN · WPI

**Oxygen Electrode (Nickelates)**




Stack Supported | Anode Supported

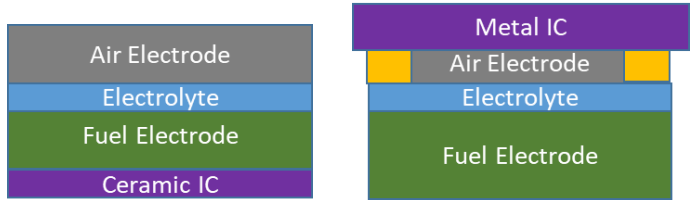
**Fuel Electrode (Nickel migration)**

BOSTON UNIVERSITY · SAINT-GOBAIN · WPI

### Stack Level

WPI · West Virginia University

**Nickelate – Cr interconnect interactions**




Stack Supported | Anode Supported

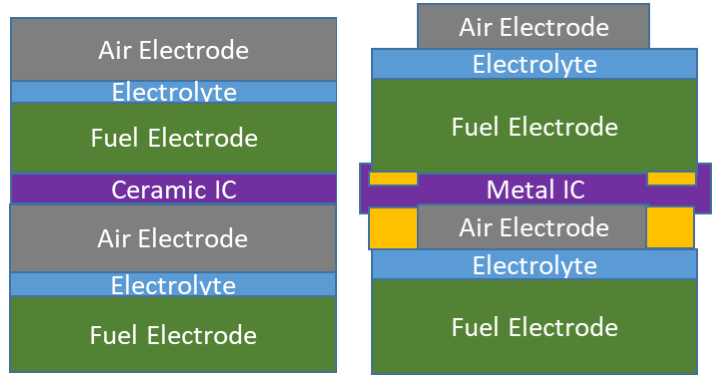
**Ceramic IC humidity interactions**

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

SAINT-GOBAIN

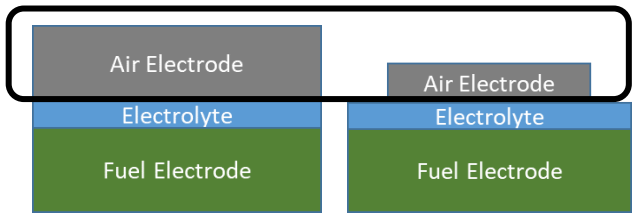
# CELL LEVEL MODELING

## DETERMINED LOW ENERGY PATH FOR OXYGEN DIFFUSION BY INTERSTITIAL MOVEMENT

Cell Level

BOSTON UNIVERSITY SAINT-GOBAIN WPI

**Oxygen Electrode Investigations (Nickelates)**

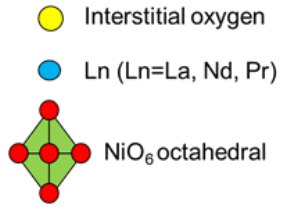



Stack Supported

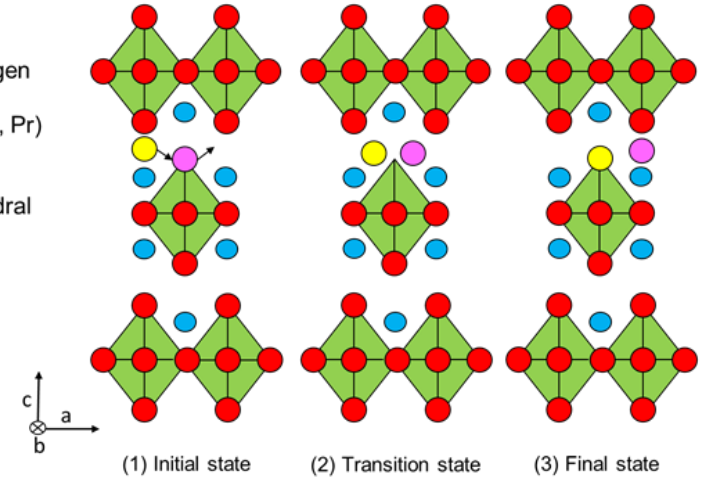
Anode Supported

**Fuel Electrode Investigations (Nickel migration)**

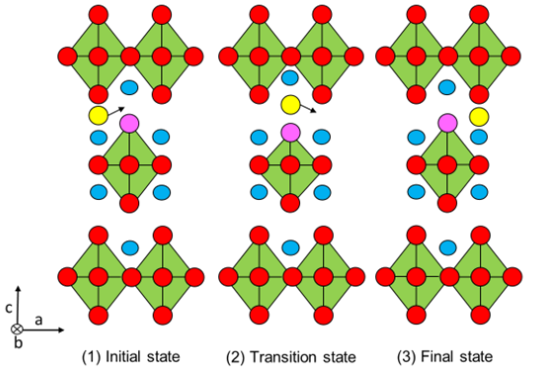
BOSTON UNIVERSITY SAINT-GOBAIN WPI

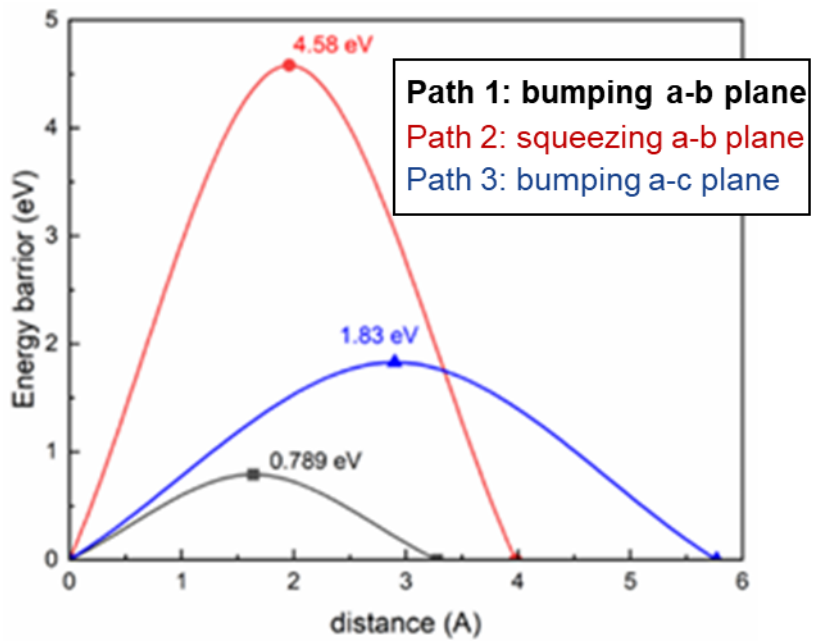



Path 1: bumping along a-b plane

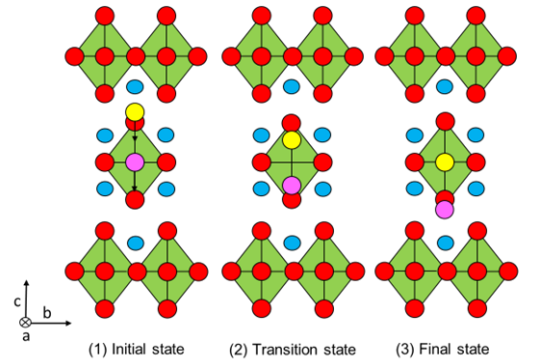



Path 2: squeezing through a-b plane




Path 3: bumping along a-c plane




SAINT-GOBAIN

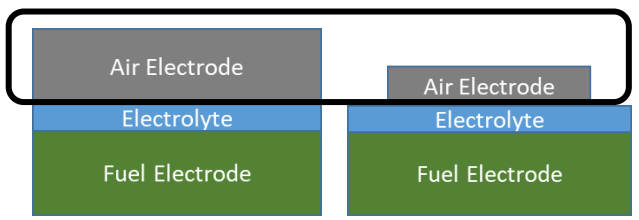
# CELL LEVEL MODELING

## CALCULATED TEMPERATURE DEPENDENT PROPERTIES OF THE NEODYMIUM NICKELATE SYSTEM

Cell Level

BOSTON UNIVERSITY · SAINT-GOBAIN · WPI

**Oxygen Electrode Investigations (Nickelates)**




Stack Supported

Anode Supported

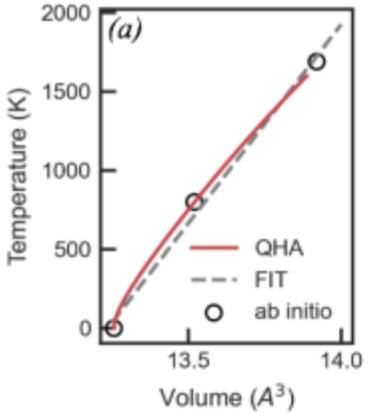
**Fuel Electrode Investigations (Nickel migration)**

BOSTON UNIVERSITY · SAINT-GOBAIN · WPI

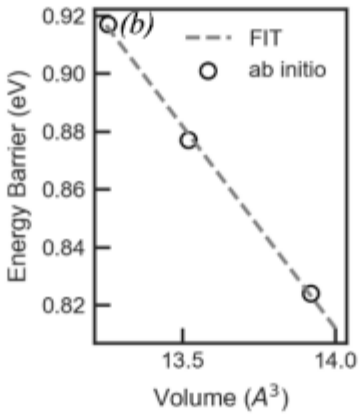
**Model refinement taking into consideration lattice expansion with increasing temperature**

$Nd_2NiO_{4-\delta}$

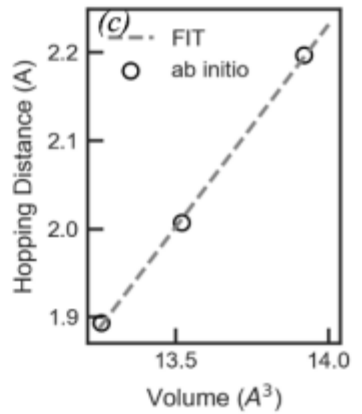
Thermal expansion



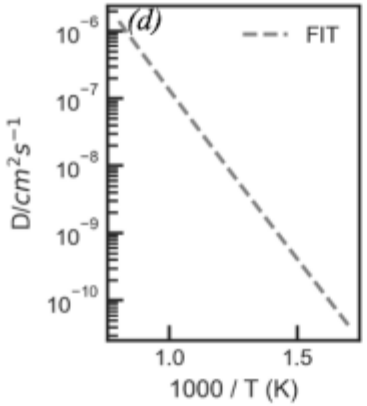

Energy Barrier



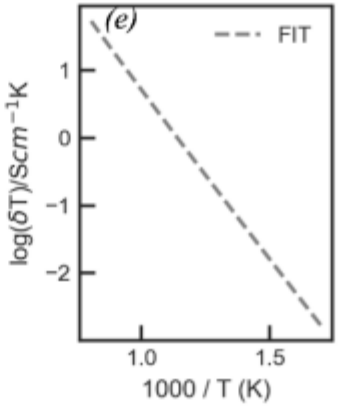

Hopping distance




Diffusivity




Ionic conductivity




SAINT-GOBAIN

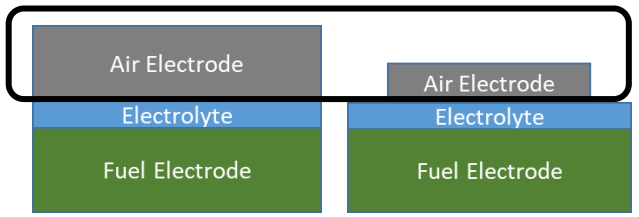
# CELL LEVEL MODELING

## IONIC CONDUCTIVITY RANKING COMPLETED

Cell Level

BOSTON UNIVERSITY SAINT-GOBAIN WPI

Oxygen Electrode Investigations (**Nickelates**)



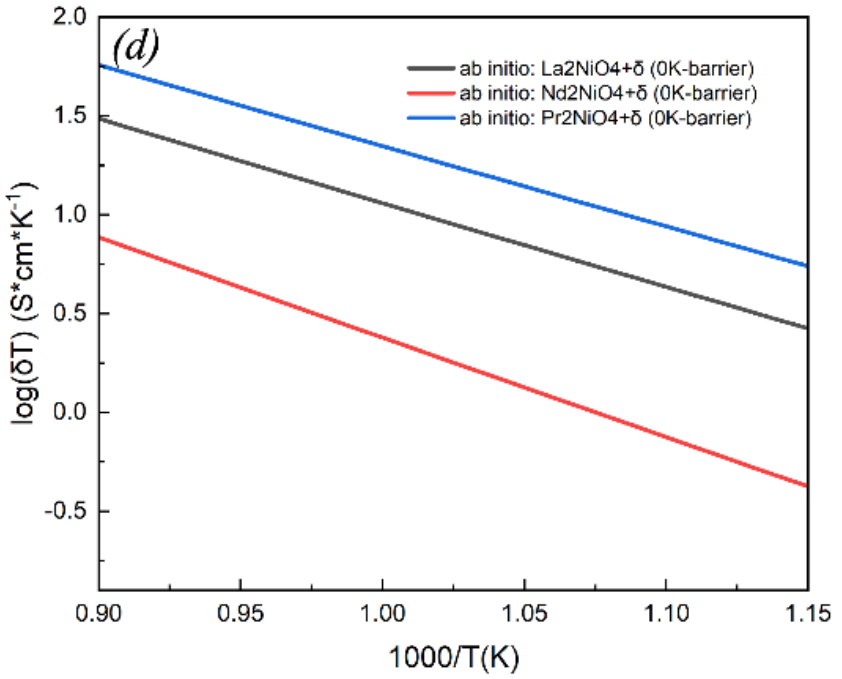

Stack Supported

Anode Supported

Fuel Electrode Investigations (**Nickel migration**)

BOSTON UNIVERSITY SAINT-GOBAIN WPI

Major result:
Ionic Conductivity in $Ln_2NiO_{4-\delta}$
Pr > La > Nd




SAINT-GOBAIN

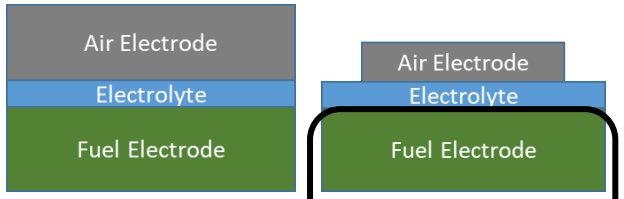
# CELL LEVEL DEVELOPMENTAL WORK

## DURABILITY INVESTIGATION, SOEC MODE

Cell Level

BOSTON UNIVERSITY SAINT-GOBAIN WPI

**Oxygen Electrode Investigations (Nickelates)**



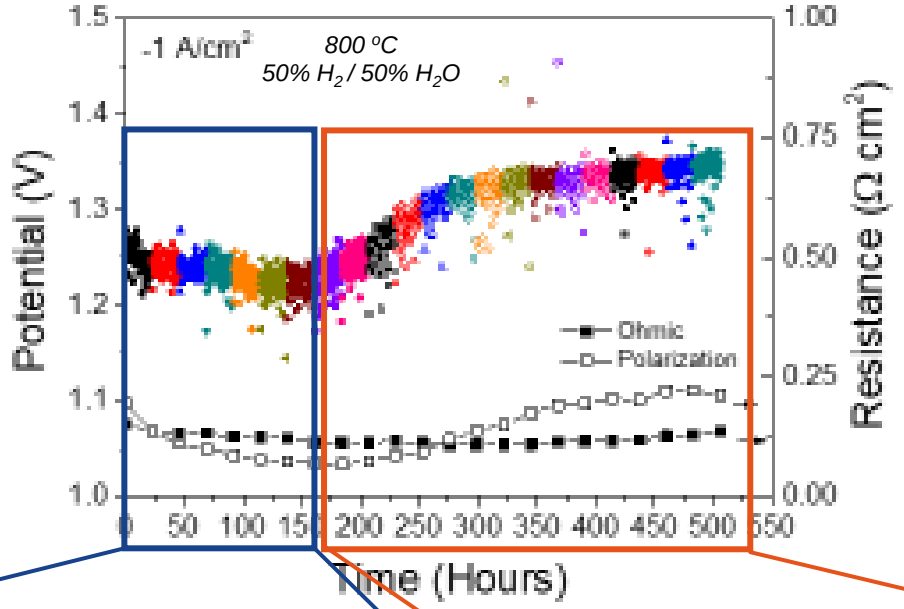

Stack Supported

Anode Supported

**Fuel Electrode Investigations (Nickel migration)**

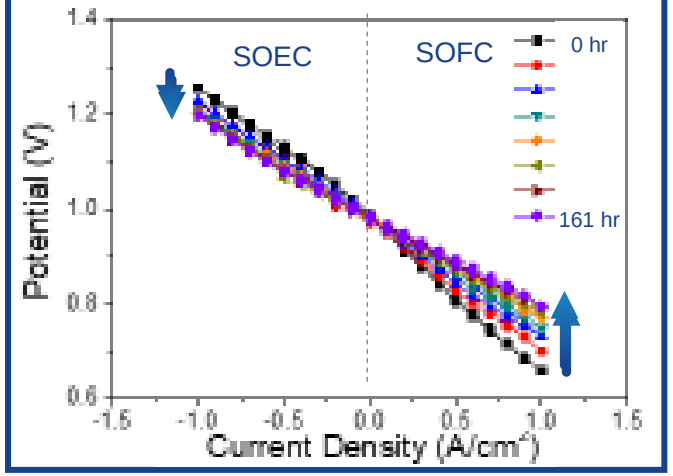
BOSTON UNIVERSITY SAINT-GOBAIN WPI

Performance shows improvement over first 150 hrs followed by a period of rapid degradation which slows over time



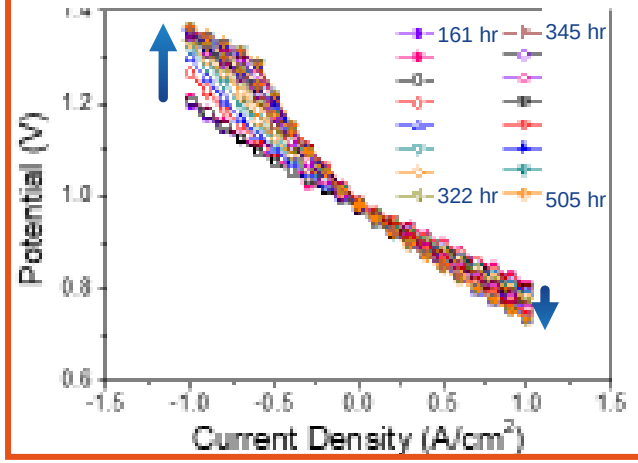

### Improvement

More significant in SOFC mode




### Degradation

more significant in SOEC mode






SAINT-GOBAIN

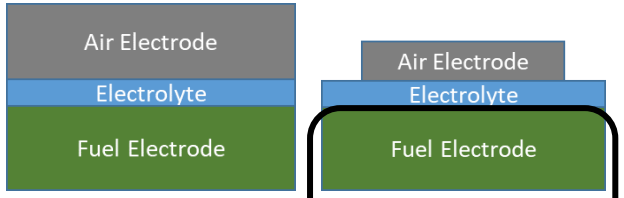
# QUANTIFICATION OF NICKEL MIGRATION

## CHANGE IN NICKEL PERCOLATION CORRELATES WELL WITH CHANGE IN CELL PERFORMANCE

Cell Level

BOSTON UNIVERSITY · SAINT-GOBAIN · WPI

**Oxygen Electrode Investigations (Nickelates)**




Stack Supported

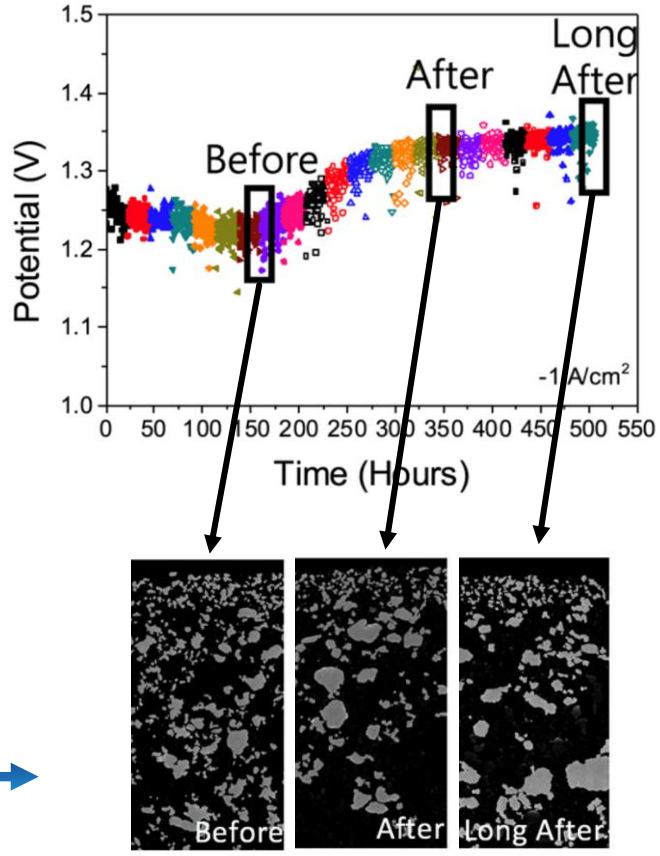
Anode Supported

**Fuel Electrode Investigations (Nickel migration)**

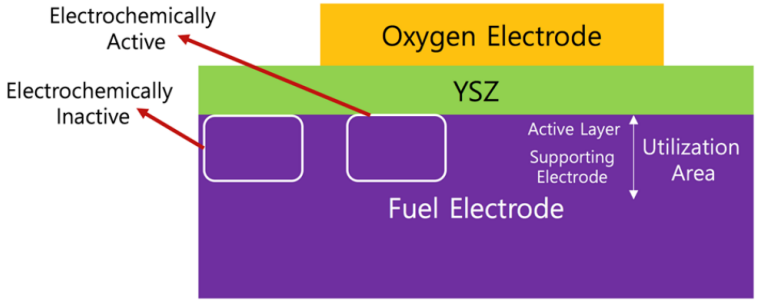
BOSTON UNIVERSITY · SAINT-GOBAIN · WPI

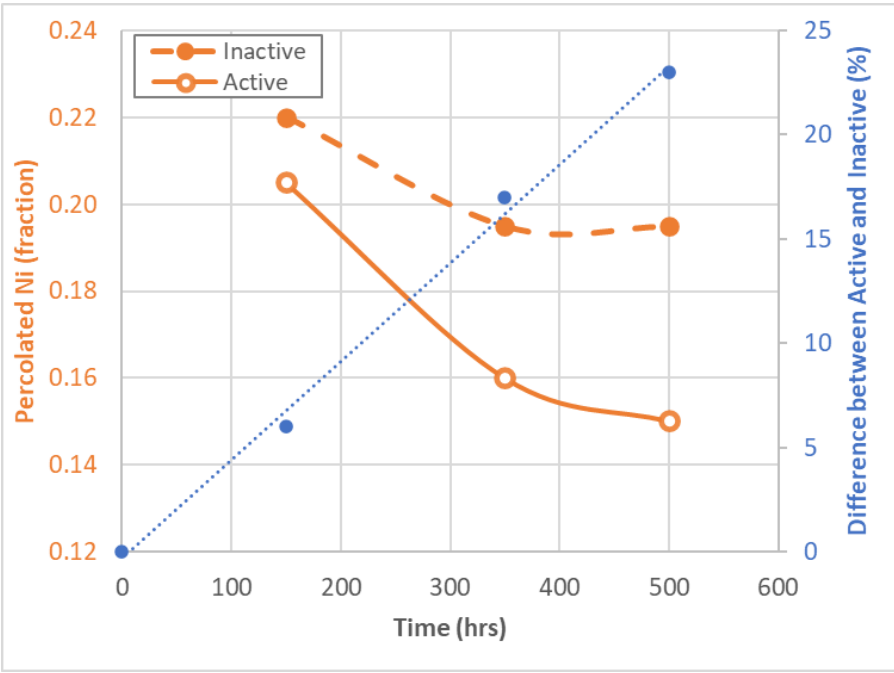
### Humidity and Electrochemical Activity Responsible for Change in Ni Connectivity

*Electrochemical Activity Continues to Drive Microstructure Change After 350 hrs of Operation*




Electrochemically Active Regions



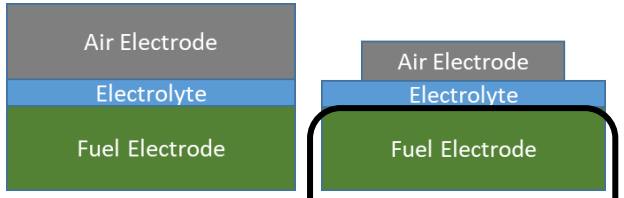
# GDC INFILTRATION EFFECTIVE AT PINNING NICKEL

## INFILTRATED STRUCTURE IS MORE STABLE AND SUPPRESSES CHANGE IN RESISTANCE

Cell Level

BOSTON UNIVERSITY SAINT-GOBAIN WPI

**Oxygen Electrode Investigations (Nickelates)**



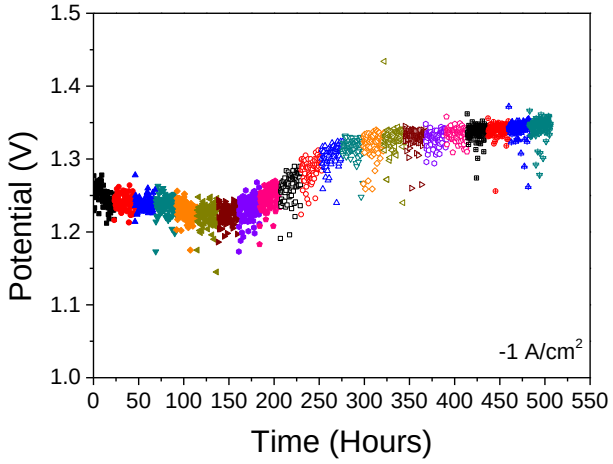

Stack Supported

Anode Supported

**Fuel Electrode Investigations (Nickel migration)**

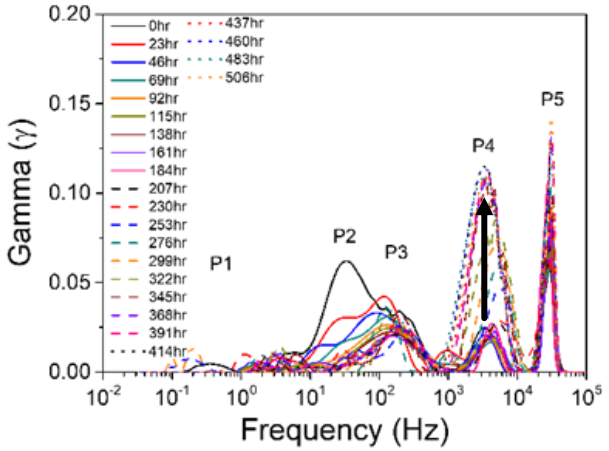
BOSTON UNIVERSITY SAINT-GOBAIN WPI

### NiO/YSZ electrode utilized without infiltration

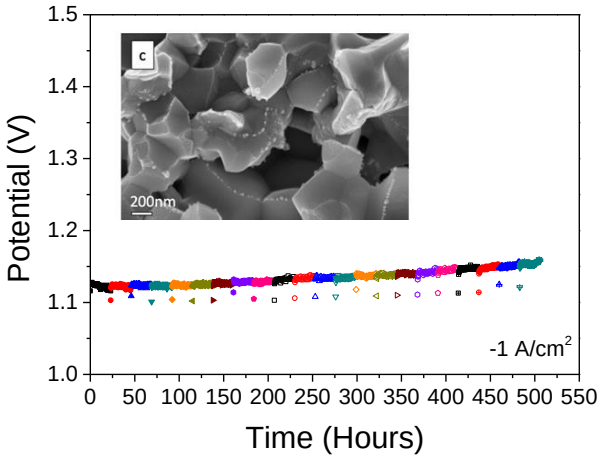


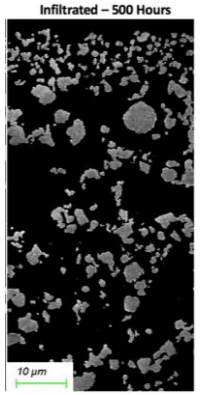


**Active region (500 hr)**

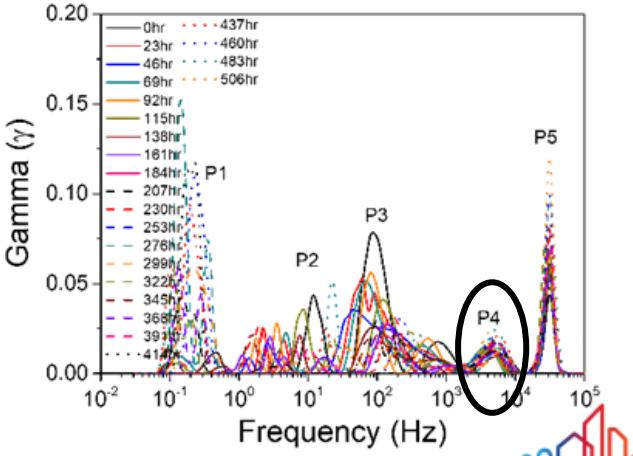



### NiO/YSZ electrode infiltrated with GDC prior to electrochemical testing




**Active region (500 hr)**




SAINT-GOBAIN

# CHALLENGES OF CELL-TO-CELL CONNECTIONS WITHIN A STACK

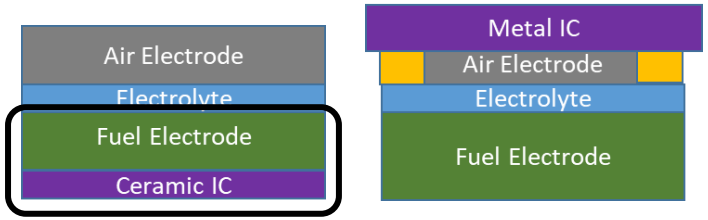
## EFFECT OF HIGH HUMIDITY ON THE CONDUCTIVITY OF THE CERAMIC INTERCONNECT

Stack Level

WPI

West Virginia University

**Nickelate – Cr interconnect interactions**



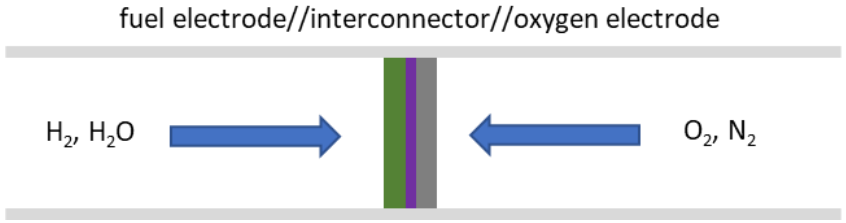

Stack Supported

Anode Supported

**Ceramic IC humidity interactions**

West Virginia University SAINT-GOBAIN

Ceramic interconnect being tested with relevant atmospheres and externally imposed current density to simulate fuel cell and electrolysis operational modes

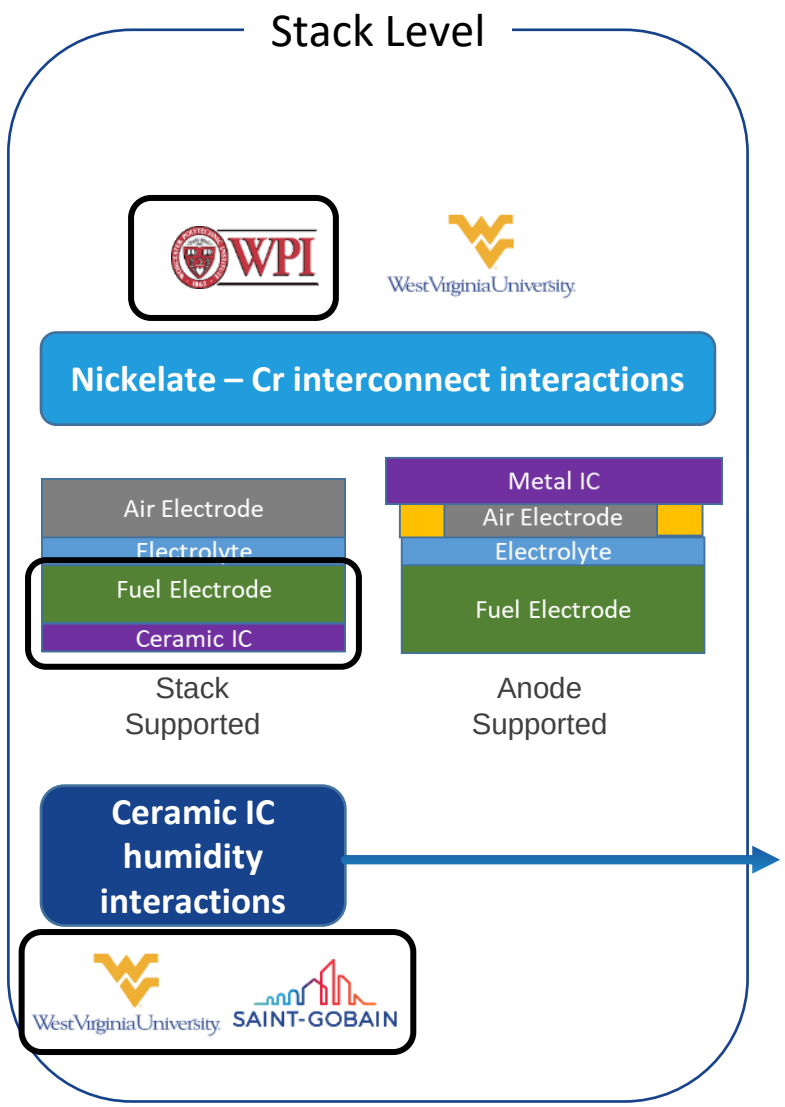



| Mode | Gas to air electrode | Gas to fuel electrode | Current density (A/cm2) | Status | Duration |
|---|---|---|---|---|---|
| SOFC | 18 vol.% $O_2$ balanced by $N_2$, 300sccm total | 10vol.% humid $H_2$, 90sccm $H_2$+10sccm steam | 0.5 | 1st run Finished | ~2200 h |
| SOFC | 18 vol.% $O_2$ balanced by $N_2$, 300sccm total | 10vol.% humid $H_2$, 90sccm $H_2$+10sccm steam | 1 | 1st, 2nd run Finished | ~200 h ~600 h |
| SOEC | 40 vol.% $O_2$ balanced by $N_2$, 300sccm total | 20vol.% humid $H_2$, 80sccm $H_2$+20sccm steam | -0.5 | Finished | 1000h |
| SOEC | 40 vol.% $O_2$ balanced by $N_2$, 300sccm total | 20vol.% humid $H_2$, 80sccm $H_2$+20sccm steam | -1 | ongoing | ~660h |
| Reversible SOFC-SOEC | Switching gas between SOFC/SOEC mode every 300h | Switching gas between SOFC/SOEC mode | 0.5 switch To -0.5 | ongoing | ~1750h |
| Reversible SOFC-SOEC | Switching gas between SOFC/SOEC mode every 300h | Switching gas between SOFC/SOEC mode | 1 switch to -1 | | |

SAINT-GOBAIN

# CHALLENGES OF CELL-TO-CELL CONNECTIONS WITHIN A STACK

## SIMULATION OF CERAMIC INTERCONNECT INITIATED TO AID IN MATERIAL EXPLORATION



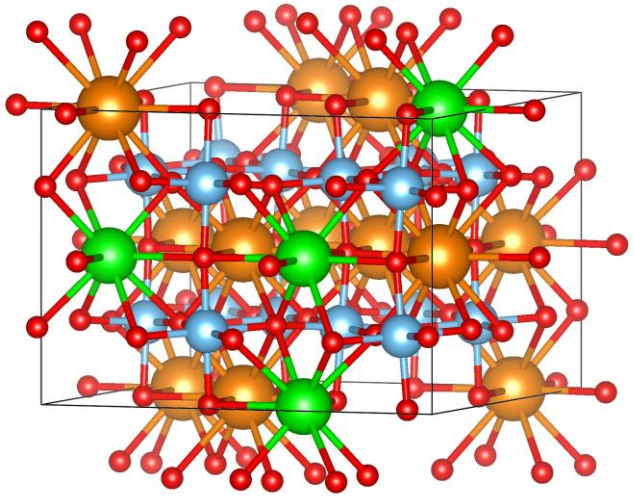

### Simulation of perovskite interconnect A

- Electronic conductivity vs. T
- Electronic conductivity vs. PO2
- Electronic conductivity vs. doping element concentration

### Simulate the role of dopants in perovskite interconnect B

- Electronic conductivity of 2-4 doping elements

Supercell Constructed




80-atom SQS structures for CI-A

SAINT-GOBAIN

# ENABLING STACK AGNOSTIC VOLUME PRODUCTION

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

### Powder Production

- Multiple commercial paths for powder production ✓
- Reproducible phase purity ✓
- Particle size control ✓

| Oxygen functional powders | Phase purity | Size effect |
|---|---|---|
| Lanthanum nickelate (LNO) | Complete | Awaiting data |
| 50 mol-% Lanthanum-doped ceria (LDC50) | Complete | Awaiting data |
| Neodymium nickelate (NNO) | Complete | In progress |
| 50 mol-% Neodymium-doped ceria (NDC50) | Complete | In progress |

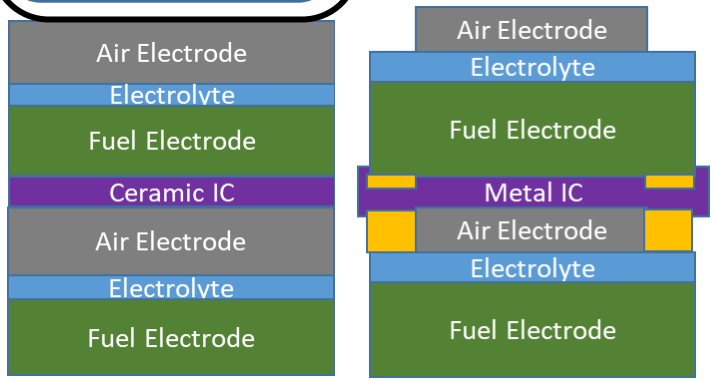
Next step: incorporation into cell/stack design


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

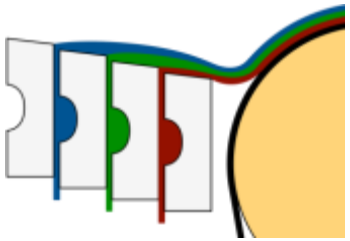
SAINT-GOBAIN

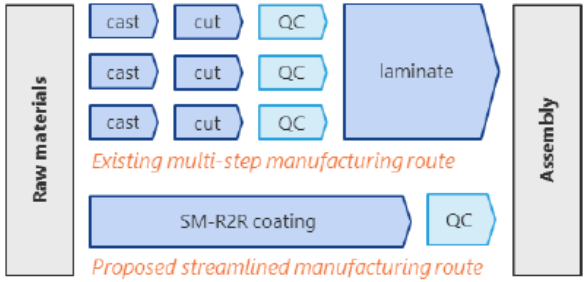
# SIMULTANEOUSLY CAST MULTILAYER STRUCTURES

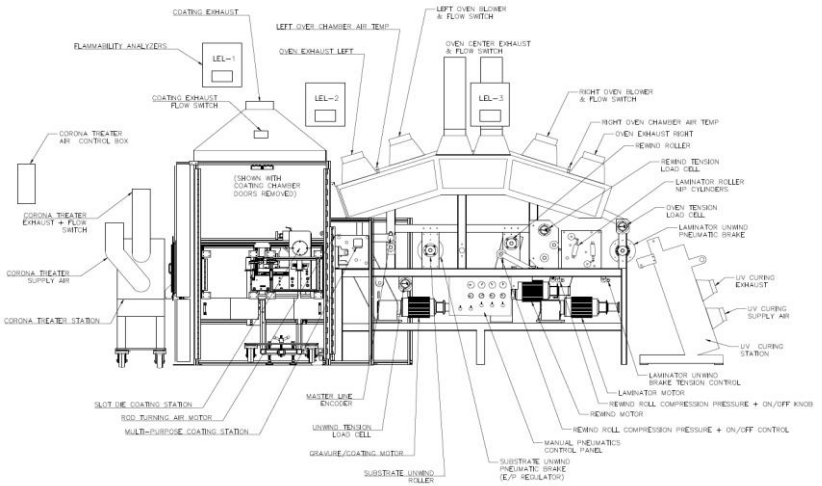
## ELIMINATES TAPE CASTING INDIVIDUAL LAYERS FOLLOWED BY PUNCHING, STACKING, AND LAMINATION

### Benefits

- Higher throughput, economical advantages
- Improved adhesion between layers
- Ultra-thin sub-layers < 30 um (wet) possible





Scale Up

SAINT-GOBAIN

WestVirginiaUniversity.

**Solution integration and stack testing**

| Stack Supported | Anode Supported |
|---|---|
| Air Electrode | Air Electrode |
| Electrolyte | Electrolyte |
| Fuel Electrode | Fuel Electrode |
| Ceramic IC | Metal IC |
| Air Electrode | Air Electrode |
| Electrolyte | Electrolyte |
| Fuel Electrode | Fuel Electrode |

**Techno-economic analysis**

Gaia Energy Research Institute LLC (Gaia)

Energy, Environmental, and Engineering Research

WPI

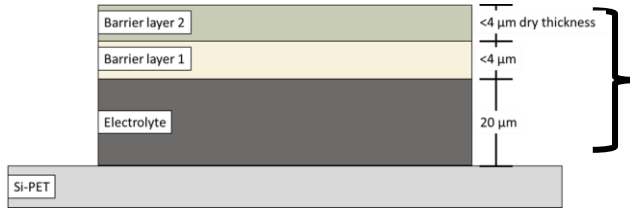
SAINT-GOBAIN

SAINT-GOBAIN

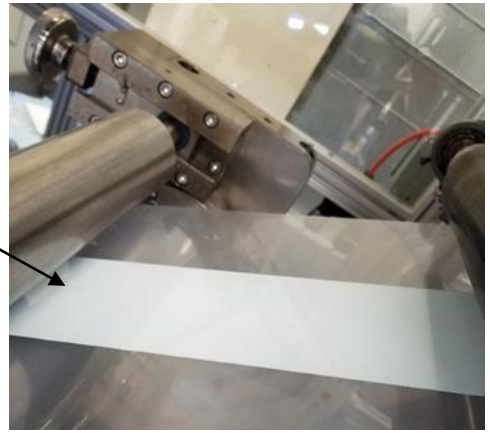
# PILOT PRODUCTION OF MULTILAYER STRUCTURES

## SIMULTANEOUS MULTI-LAYER WET COATING WITH HIGH QUALITY INTERFACES AND UNIFORM MICROSTRUCTURE

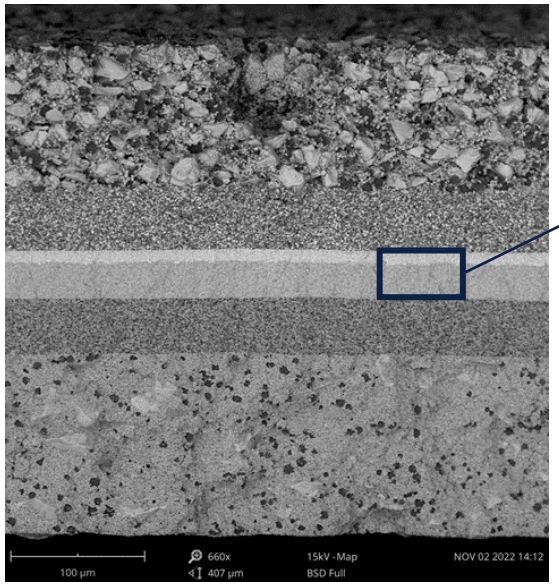
### Target Design



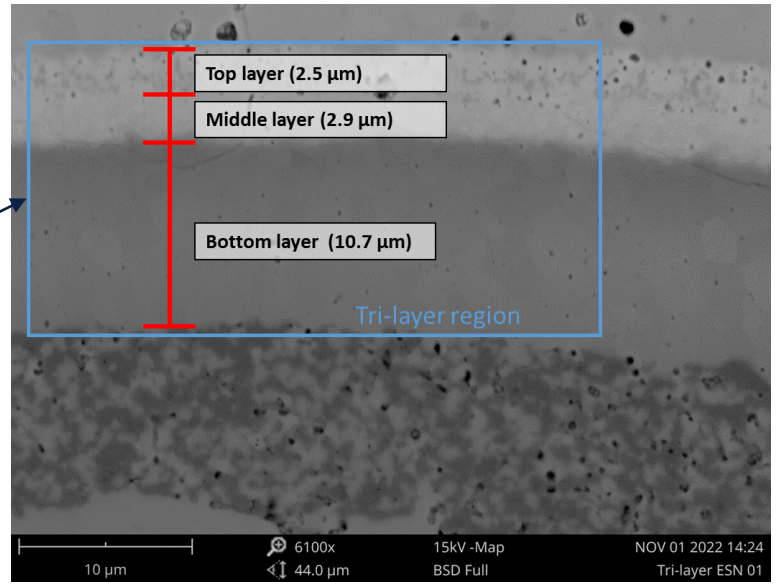

### Multilayer Wet Coating



### Laminated Green Structure



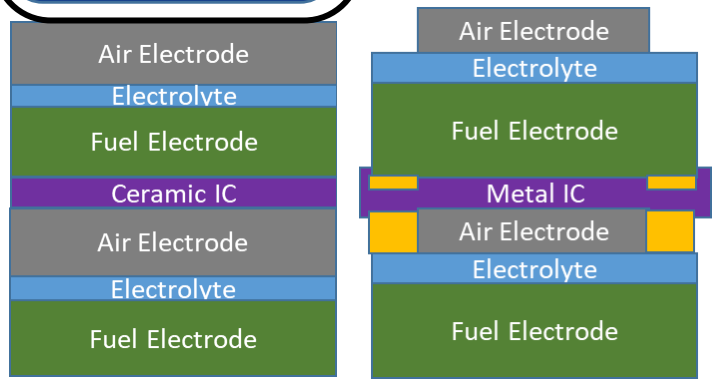

### Final Sintered Microstructure




### Scale Up

SAINT-GOBAIN  West Virginia University

**Solution integration and stack testing**




**Techno-economic analysis**

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

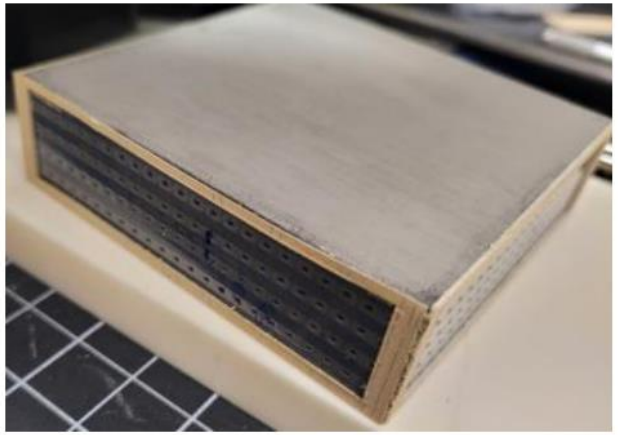
WPI  SAINT-GOBAIN

SAINT-GOBAIN

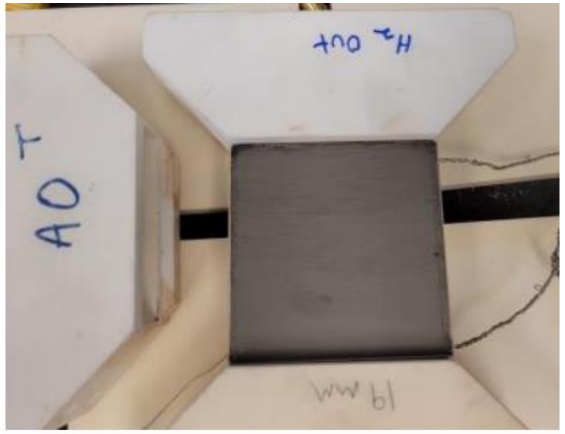
# SHORT STACK TESTING

## STACKS PRODUCED, MANIFOLDED AND SHIPPED TO WVU FOR TESTING

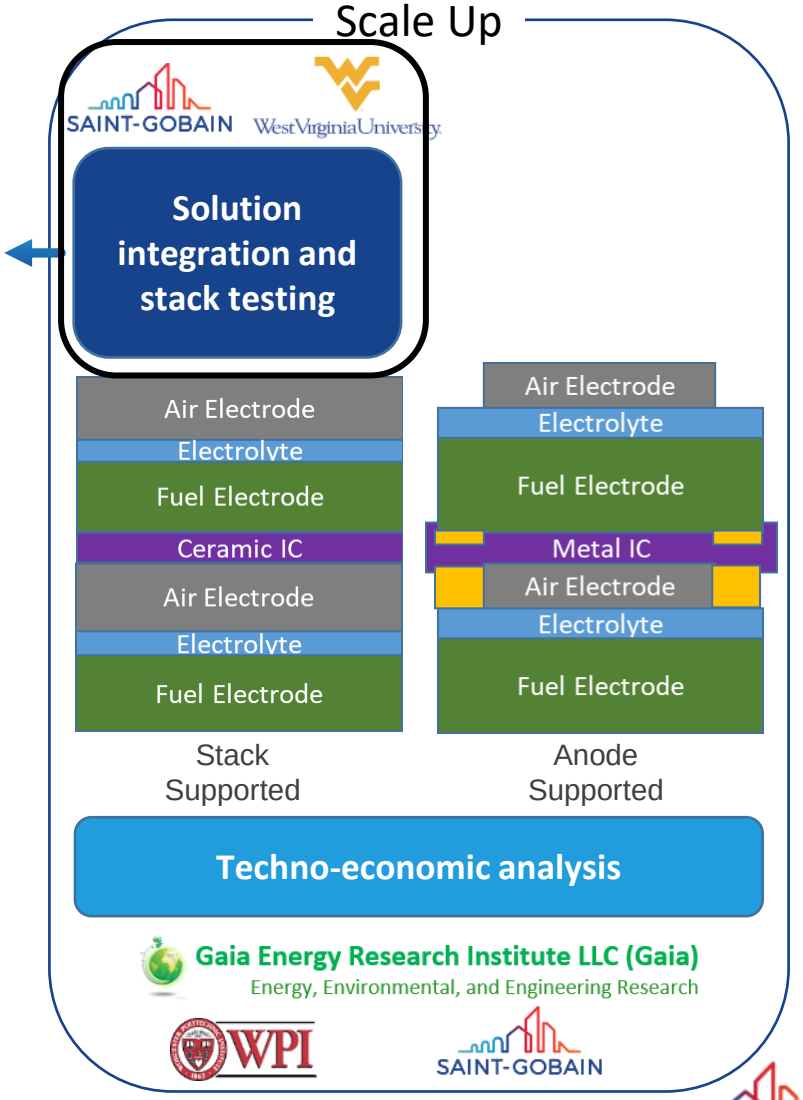
4-cell short stack fit with gaskets



Short stack assembly into gas manifolds



Assembled stack shipped for testing




SAINT-GOBAIN

# SUMMARY & NEXT STEPS

## Key Takeaways

- Completed ionic conductivity calculations from first principles as a function of temperature
- Linked cell degradation to reduction in percolated nickel
- Demonstrated technique to successfully suppress nickel migration
- Began investigation into effect of humidity on electronic conductivity of ceramic interconnect
- Demonstrated use of simultaneous multi-layer casting of ceramic slurries in cells and stacklets
- Begun short stack testing

## Next Steps

- Produce powders and test cells with advanced air electrode compositions and optimized PSD
- Fabricate short stack samples with different advanced air electrodes for testing
- Incorporate air electrode compositions into the multilayer casting configuration
- Use model to predict electrical conductivity of perovskites under varying T, pO2, doping elements
- Continue infiltration and operational condition techniques to mitigate Ni migration
- Utilize performance data for system level performance and cost modeling