Degradation modeling and electrode engineering of SOFCs SOECs and R-SOCs

NETL FWP 1022411

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Project manager: Jai-Woh Kim (FECM)

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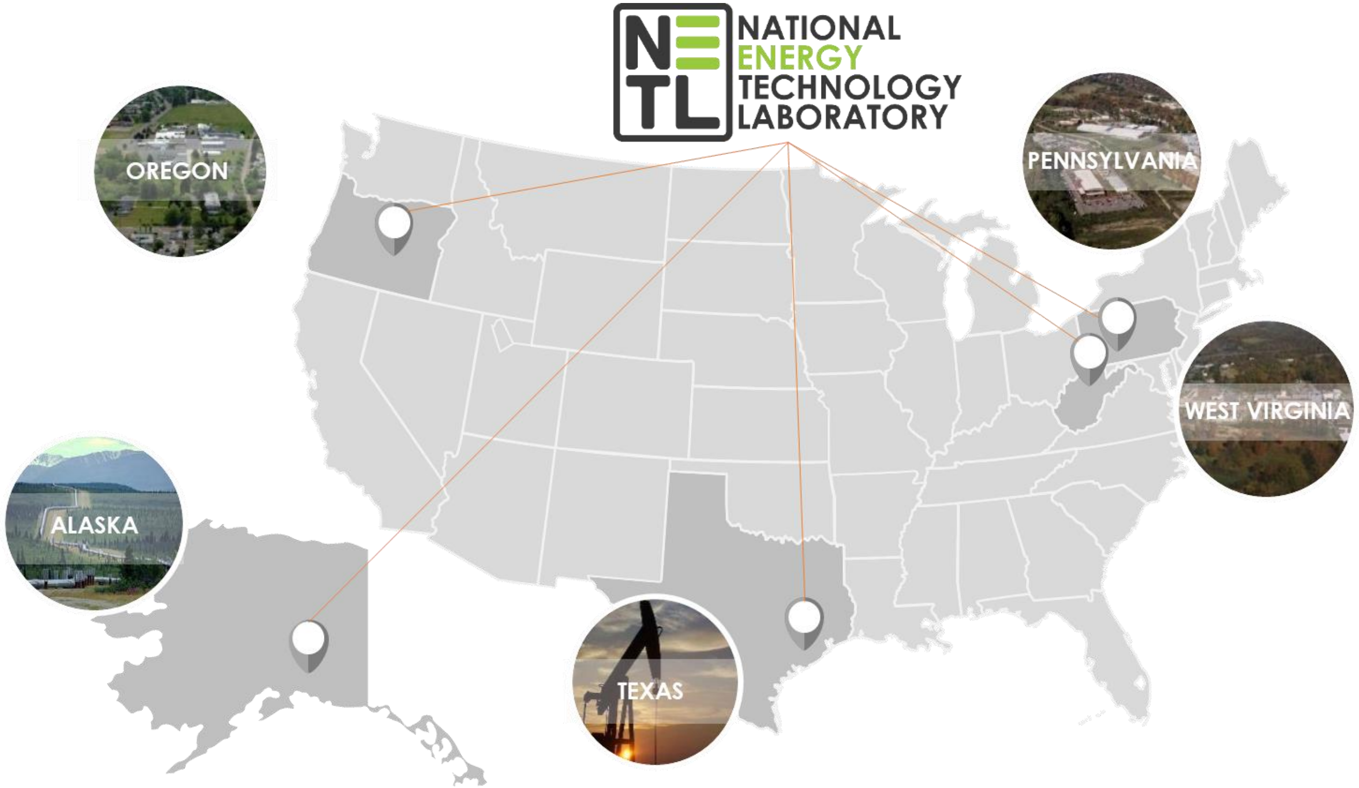
Disclaimer

Outline

- **Introduction**
- **Recent Progress**
  - Computational materials design
  - Electrode design and engineering
  - Cell degradation modeling
- **Wrap-Up**



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- **Harry Abernathy (TPL, MGN)**
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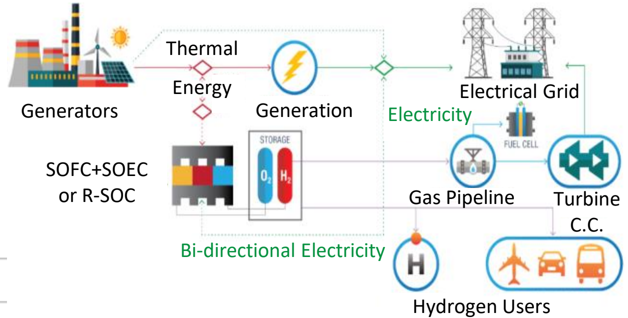
- **Yu Zhong (MME)**

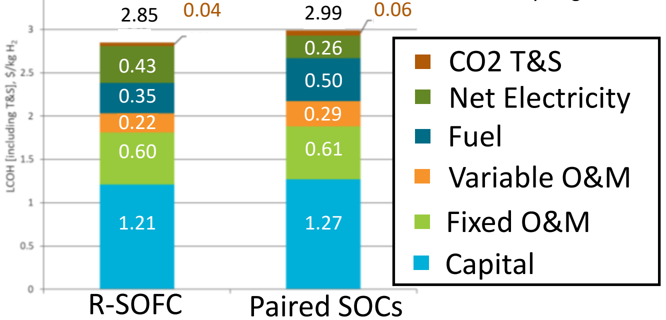
NETL SOC Capability Overview

CHALLENGE: SOC technology is cost prohibitive due to long-term performance degradation
APPROACH: Develop degradation modeling and mitigation tools to improve performance / longevity of SSEC

Systems Engineering and Analysis

- Techno-Economic Analysis
- Hybrid configuration assessment
- R&D Goals Evaluation

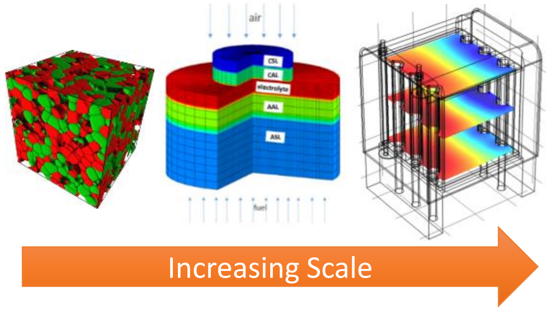


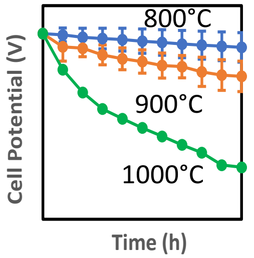


Levelized Cost of H_2 at $20/MWh Electricity

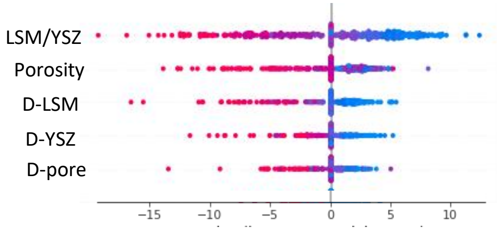
Performance Degradation Modeling

- Degradation prediction tools
- Atoms-to-System scale bridging
- Experimental validation
- Advanced Gas, Temperature Sensors





Machine learning-informed design tools

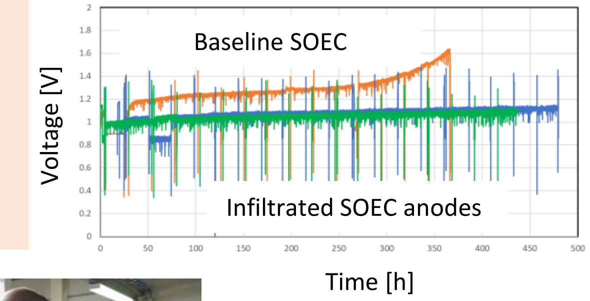


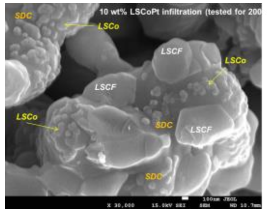
Impact of microstructural features on lifetime performance

Electrode Engineering

- Degradation mitigation
- Microstructure optimization
- Technology transfer to industry
- System demonstrations

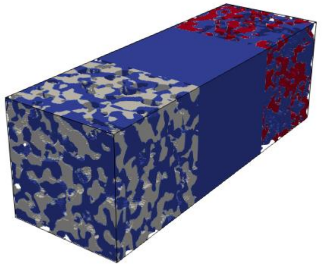
Infiltrated Cells from **6** Partners

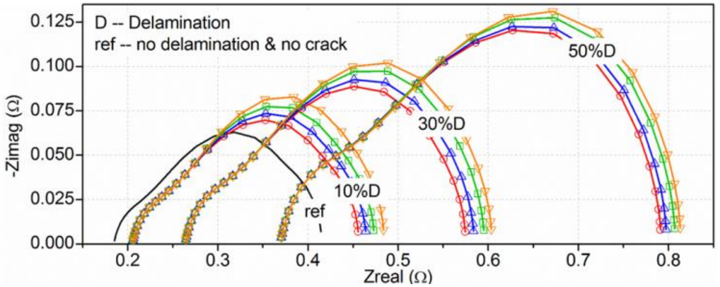


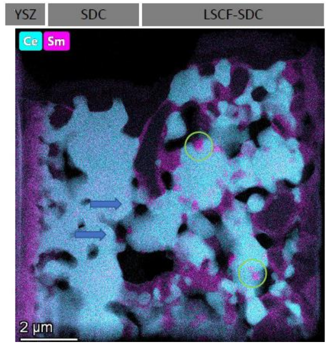


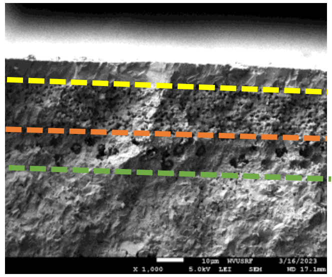
NETL SOFC Work Plan Tasks

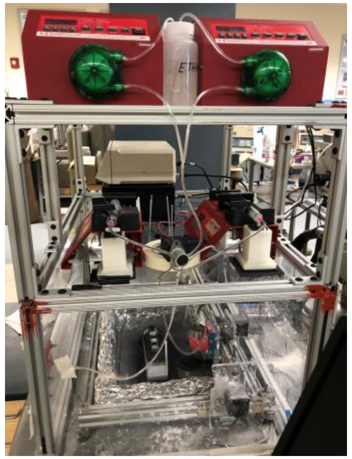
- **Task 2: Cell and Stack Degradation Evaluation and Modeling**
  - Performance and degradation model development
  - Microstructural analysis and analysis methods
  - Machine learning for materials studies, electrode design
- **Task 3: Electrode Engineering**
  - Infiltration for degradation mitigation
  - R-SOC characterization
  - Protonic SOC materials characterization and development
  - Advanced electrode design and manufacturing
  - S/TEM analysis of cell degradation
- **Task 4: Strategic Systems Analysis and Engineering**
  - R-SOC, SOEC system studies
  - SOFC scaling study, H_2-fueled SOFC market study
- **Task 5: Cyber Physical Modeling**
  - 1D real-time SOEC stack model development
  - Controls design for dynamic operation of SOC stacks











See *"Recent Progress in Solid Oxide Cell Technology Analysis at NETL"* by **Greg Hackett at 9:30 AM**

See *"Cyber-physical Simulation of Solid Oxide Cell Hybrid Systems"* by **Biao Zhang at 2:45 PM**

Computational materials design

Discovering higher performing, more stable materials

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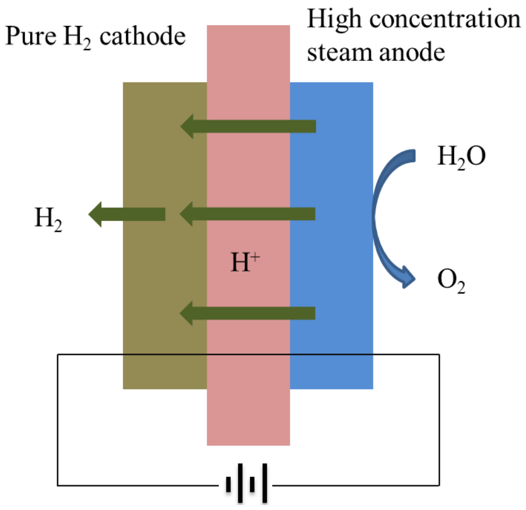
Proton-conducting SOCs

Expanding the degradation modeling framework's material sets

- Proton-conducting SOCs can help lower operating temperatures, increase operational stability, and don't have a diluted H_2 stream
- Materials needs:
  - More active electrodes
  - Electrolytes with higher σ, higher H^+ transference number
  - Less expensive thermal processing
- SOEC, SOFC performance model code options created for proton-conducting systems
- **NOTE:** NETL's available SOC stack manufacturing cost tool includes options for P-SOCs
  - **OSTI ID:1842511**



Proton-conducting SOEC

Triple-Conducting-Perovskite Defect Model Released

Electrode/Electrolyte materials

- A defect model solver was developed to allow incorporation of nonlinear δ-dependent defect reaction energies and entropies for calculating defect concentration of the triple-conducting perovskites **$(La,Ba)Fe_{1-x}M_xO_{3-\delta}$**
- **H_2 incorporation through $[OH_O^{\bullet}]$ and $[H_O^{\bullet}]$ (hydride) defects.**
- Octave-based Script publicly available on NETL' s EDX Server:

  doi.org/10.2172/2328139

Input variables:
C_{La}, C_M, Q_M
ΔH^{def} (δ, C_{La}, C_M),
ΔS^{def} (δ, C_{La}, C_M),
for 4 defect reactions:
Hydration, Reduction, Disproportionation, Hydride formation

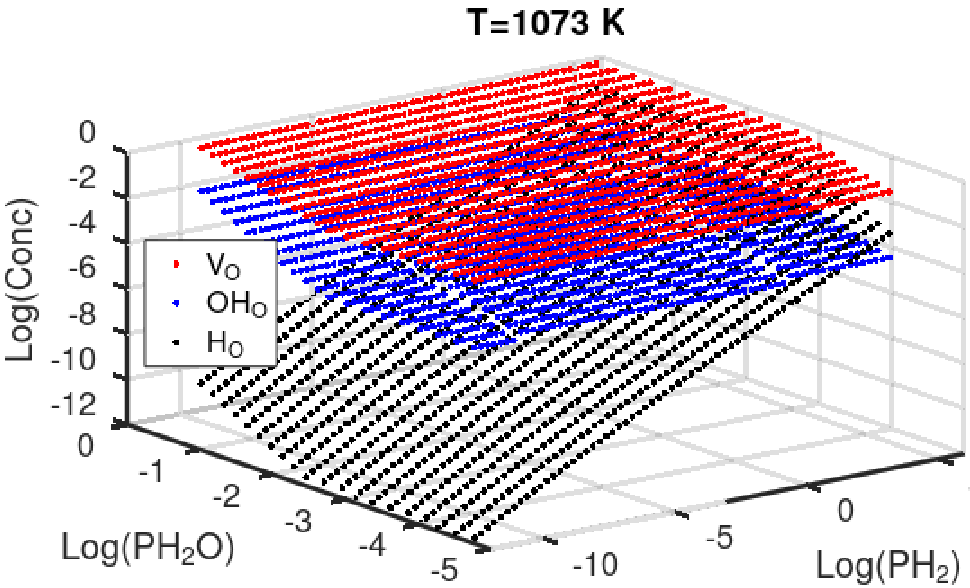
Program loops to output the defect model solution in a defined grid of T, $P(O_2)$, $P(H_2O)$ and their equivalent $P(H_2)/P(H_2O)$

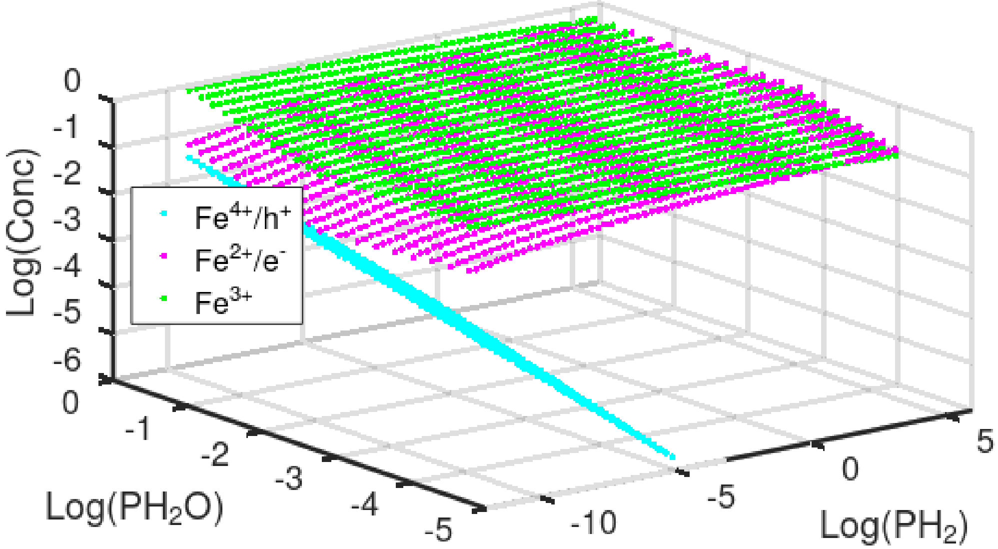
Subroutines to generate plots:

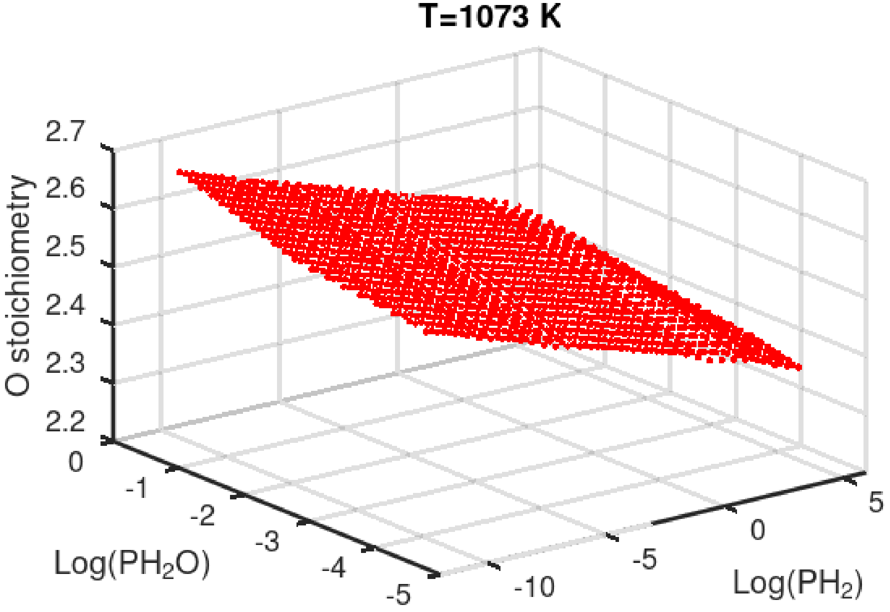
1. Brower diagrams (C^{def}, $P(O_2)$, $P(H_2O)$)
2. Brower diagrams (C^{def}, $P(H_2)$, $P(H_2O)$) at T=873~1273K
3. C^{def} temperature dependences at specified $P(O_2)/P(H_2O)$
4. C^{def} temperature dependences at specified $P(H_2)/P(H_2O)$

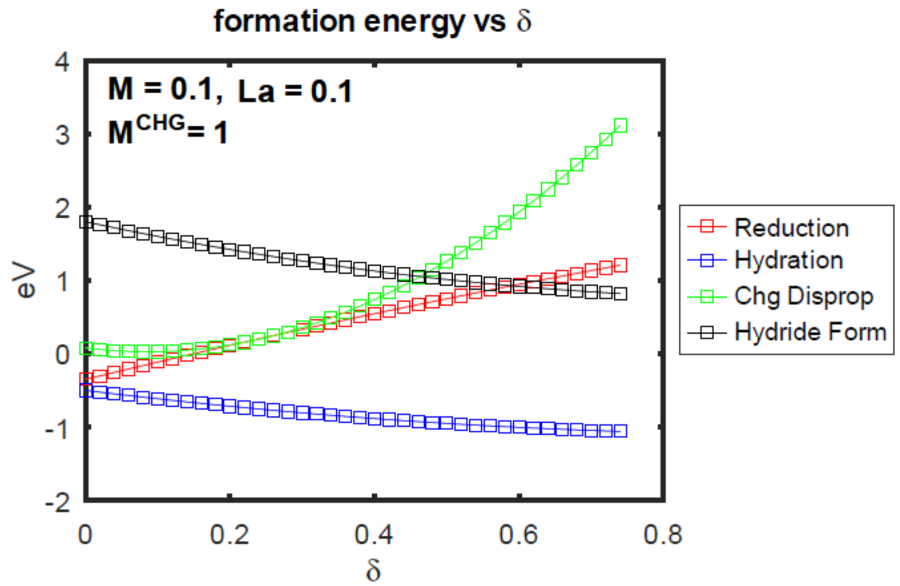
Y.L. Lee, et al., Submitted to **ECS Transactions** Vol. 111 for SOFC XVIII, 2023.

$P(H_2)/P(H_2O)$ Brouwer Diagrams of $(La_{0.1}Ba_{0.9})(Fe_{0.9}M_{0.1})O_{3-\delta}$ at T=1073K

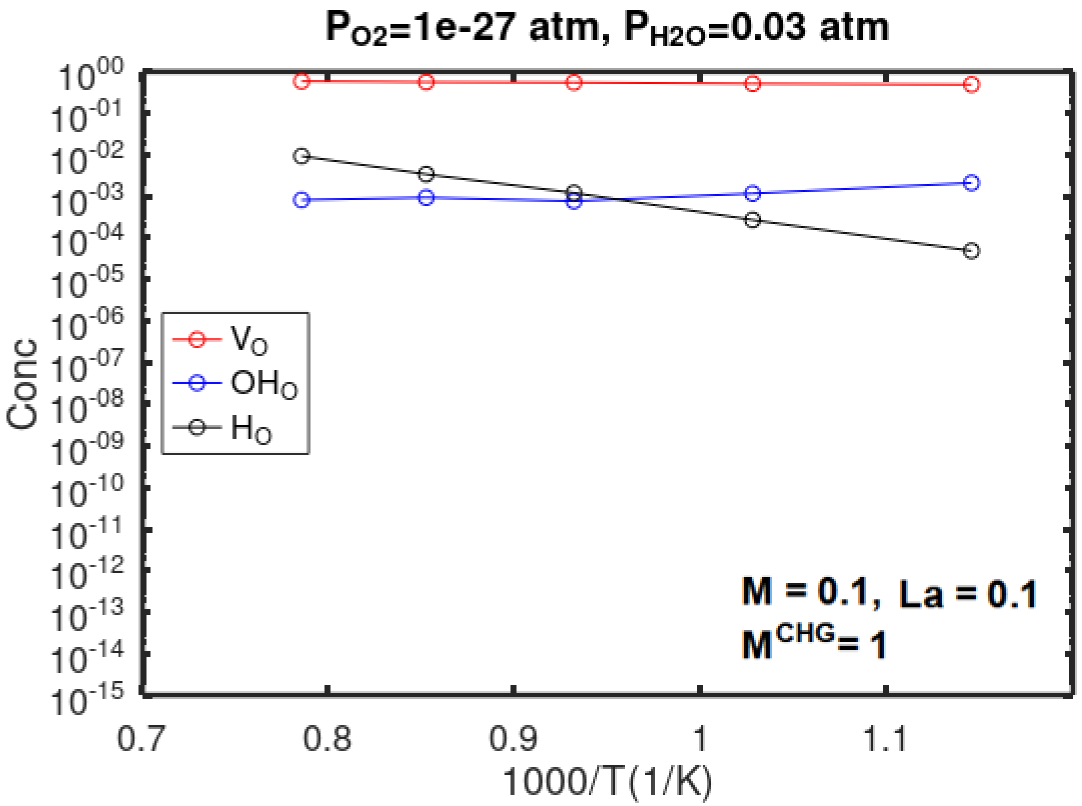


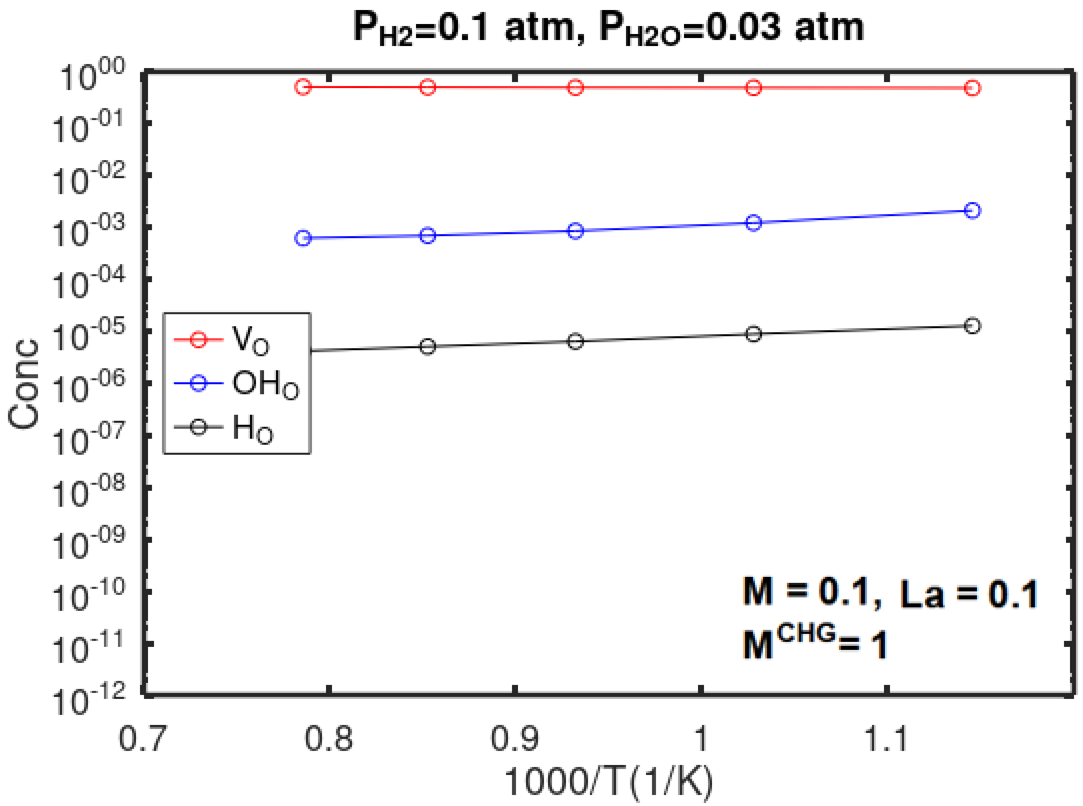






Temperature dependencies of $V_O^{''}$, $OH_O^{\bullet}$, and $H_O^{\bullet}$





Proton diffusion in Ba(Co, Fe, Zr, Y)$O_{3-\delta}$ (BCFZ/Y)

- **Hydride ($[H_O^\bullet]$) defect formation added for proton conduction at low P(O_2)**

- Defect reaction equations

1. Hydrogenation (proton) reaction

$$H_2^{(g)} + 2[O_O^{x}] + 2[M_B^\bullet] \rightleftharpoons 2[OH_O^\bullet] + 2[M_B^{x}]$$

2. Oxidation reaction

$$\frac{1}{2}O_2^{(g)} + [V_O^{\bullet\bullet}] + 2[M_B^{x}] \rightleftharpoons [O_O^{x}] + 2[M_B^\bullet]$$

3. Hydride formation

$$[O_O^{x}] + 2\,[M_B^\bullet] + [H_O^\bullet] \rightleftharpoons [V_O^{\bullet\bullet}] + 2[M_B^{x}] + [OH_O^\bullet]$$

- Calculating c_H and σ_H

1. Hydrogen concentration

$$[OH_O^{\cdot}] + [H_O^\bullet] \rightarrow c_H$$

2. Hydrogen conductivity

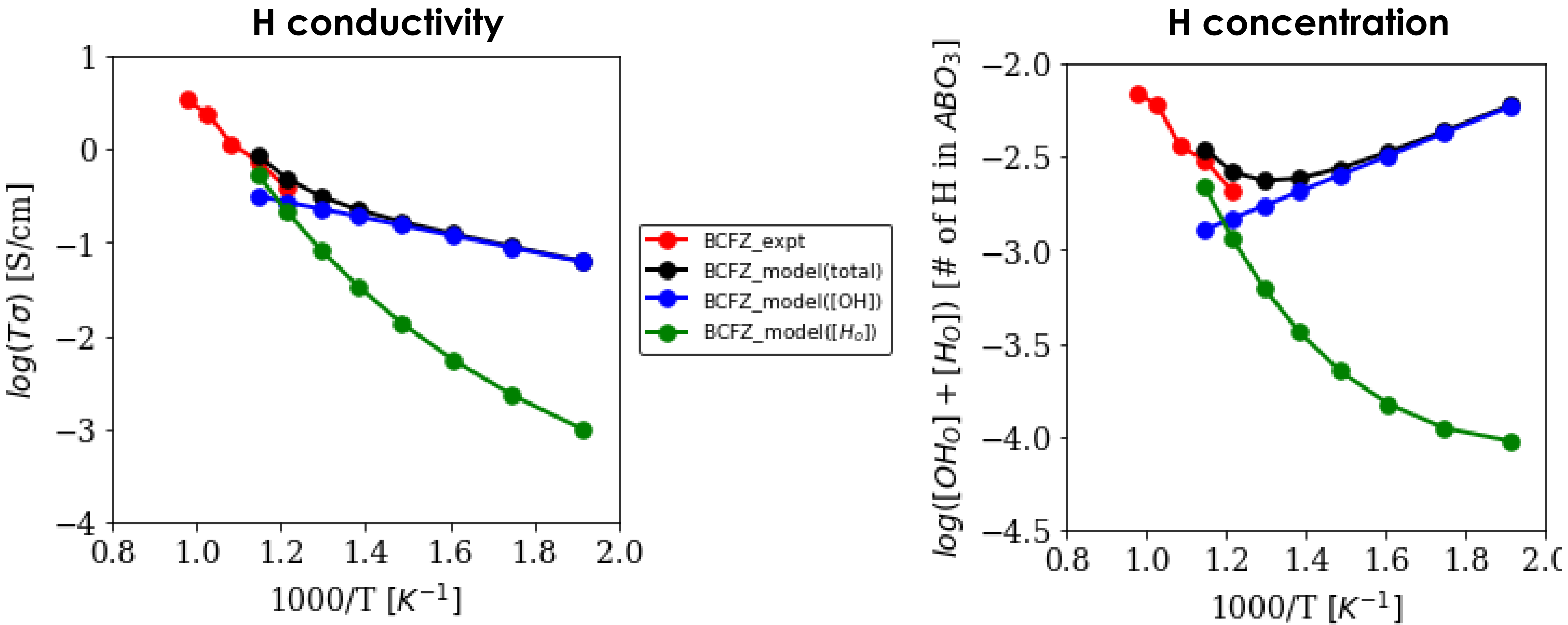
J_H: Hydrogen permeation flux
F: Faraday constant
R: gas constant
k: Boltzmann constant
L: membrane thickness
c_H: Hydrogen conc. [atoms/m^3]
D_H: self-diffusion coefficient [m^2/s]
z: charge number

$$\underbrace{\sigma_H}_{\text{Goal}} = \underbrace{\frac{4F^2L\cdot J_{H_2}}{RT}\ln\frac{P''_{H_2}}{P'_{H_2}}}_{\text{Experiments (Clemson)}} = \underbrace{\frac{z^2e^2c_HD_H}{kT}}_{\text{DFT (UW-Madison)}}$$

Zohourian, R., Merkle, R., & Maier, J. (2017). *Solid State Ionics*, 299, 64–69.
Lee, Yueh-Lin, et al. *ECS Transactions* 111.6 (2023): 1823.
Duffy, Jack H., et al., *Membranes* 11.10 (2021): 766.

Proton diffusion in Ba(Co, Fe, Zr)$O_{3-\delta}$ (BCFZ)

Initial partial pressures: PH_2: 0.05 atm , PO_2: 0.002 atm

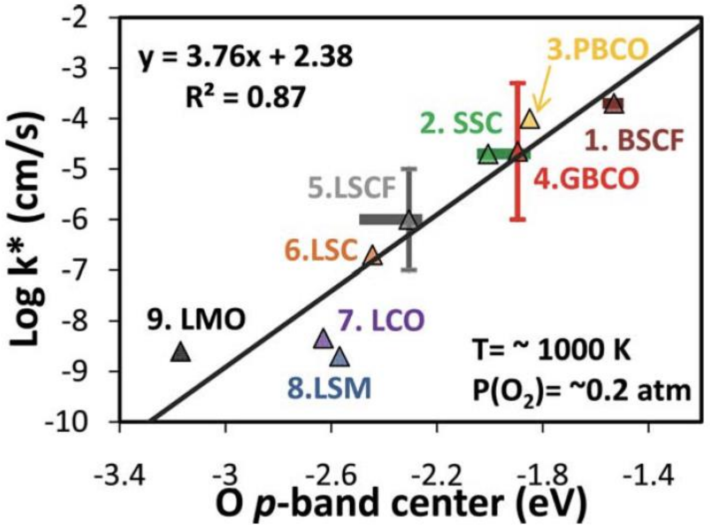


Hydride formation improves match with experimental data

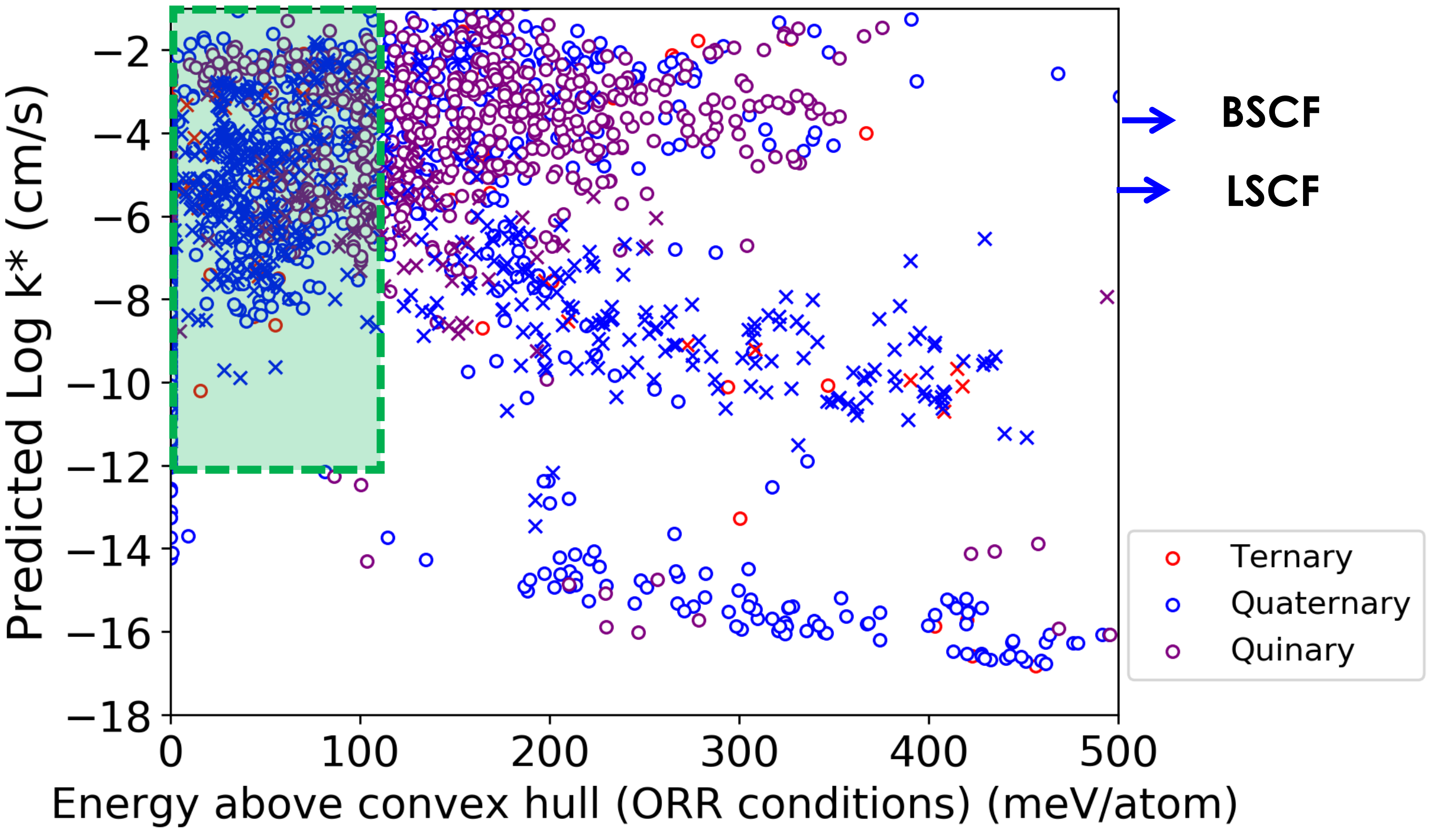
Developing materials through DFT

O p-band correlates well with air electrode material properties

ORR surface exchange [1]



From predicted k* using DFT-calculated O p-band center of >2100 perovskites, NETL examined Ba(Fe, Co, Zr)O_3 (BFCZ) materials



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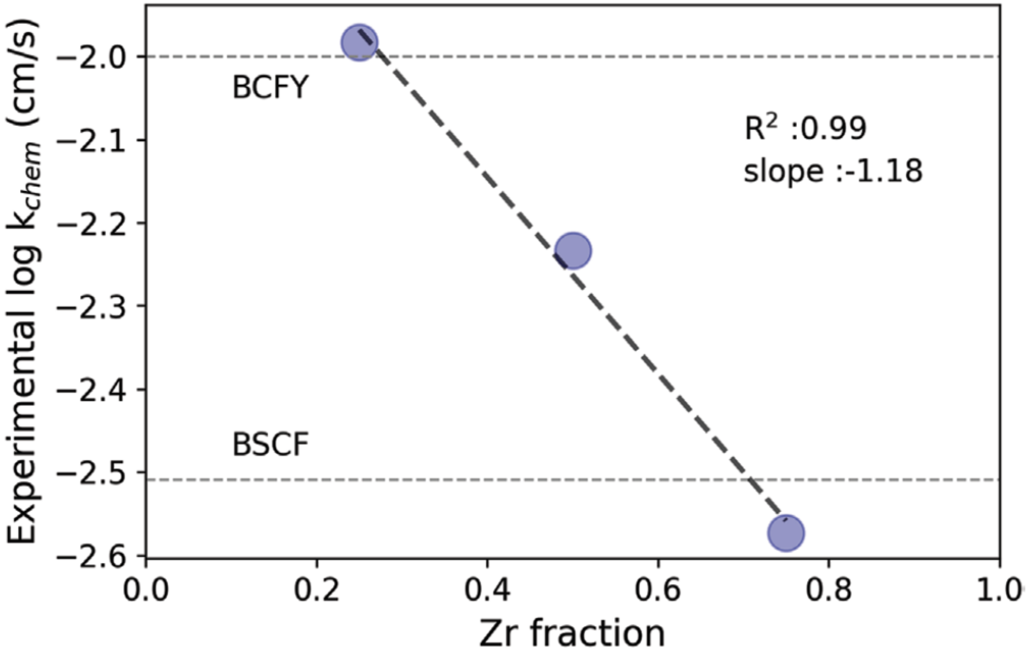
[1] Lee, et al., ***Eng Env Sci*** (2011)
[2] Jacobs, et al., ***Adv. Eng. Mat.*** (2018)
[3] Jacobs, et al., ***Chem. Mat.*** (2019)

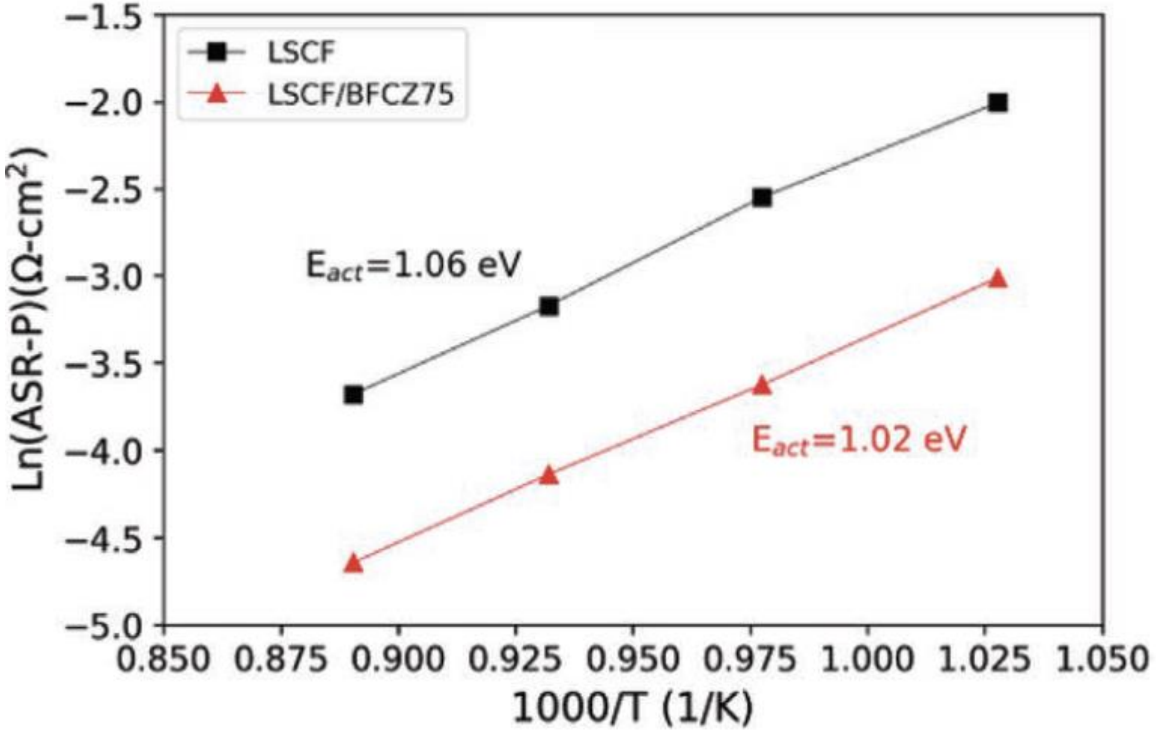
BFCZ (Zr = 25, 50, 75%) Performance

Higher k_{chem}, improved stability, not enough σ_{el}



All BFCZ compositions highly active, on par with BSCF, with only 0.5 log k_{chem} difference over entire Zr range

LSCF/BFCZ75 composite



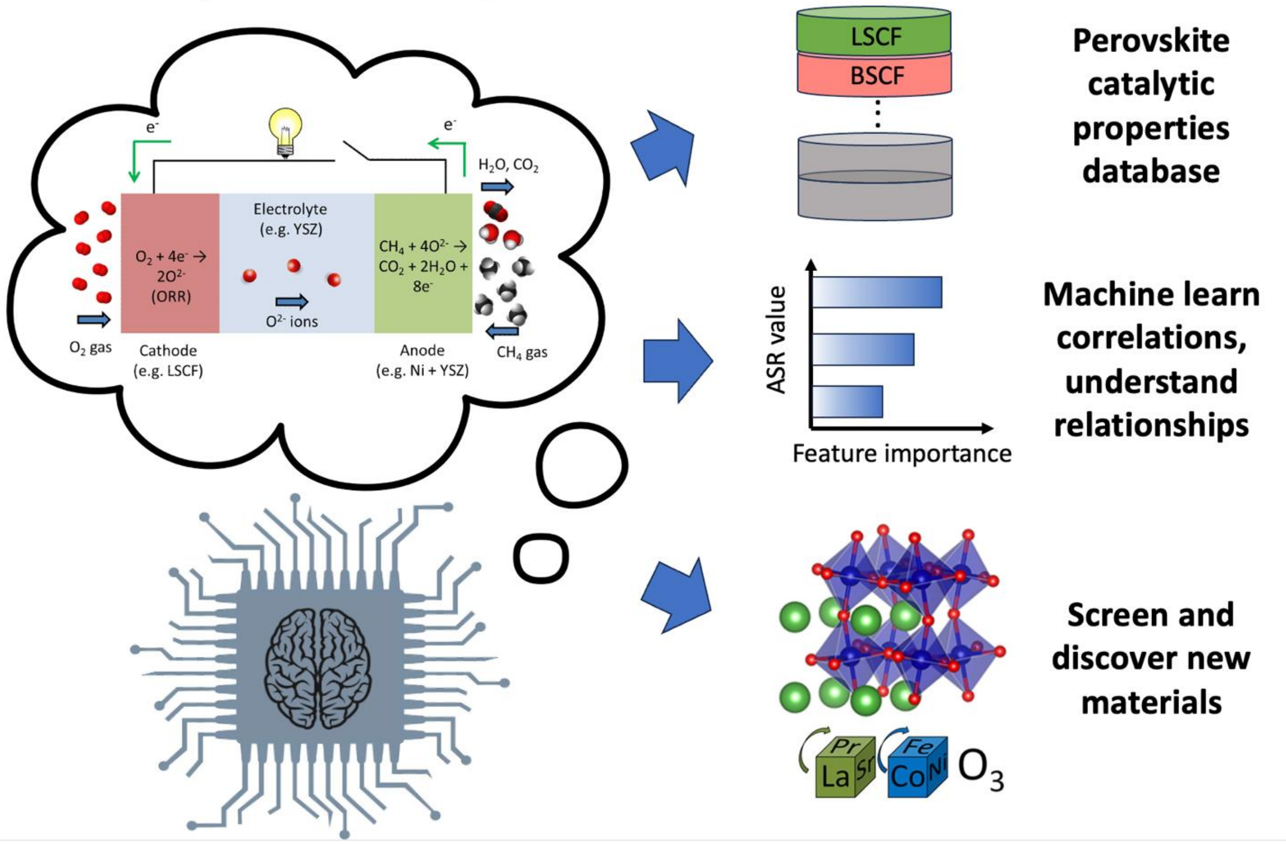
LSCF/BFCZ75 composite shows about 9x reduction in ASR at 800 °C, 65% less performance degradation vs. LSCF

[1] Jacobs, R., et al. Adv. Eng. Mat. (2022)

Machine learning prediction of properties

Using machine learning for faster calculations, larger sampling space

Data-centric ORR/OER perovskite catalytic materials design



Perovskite catalytic properties database

Machine learn correlations, understand relationships

Screen and discover new materials

- 749 data points from 313 studies for 299 unique perovskite compositions
- Elemental features calculated using MAST-ML (UW-M) instead of using DFT
- **19 million perovskite oxides** were examined using ML model

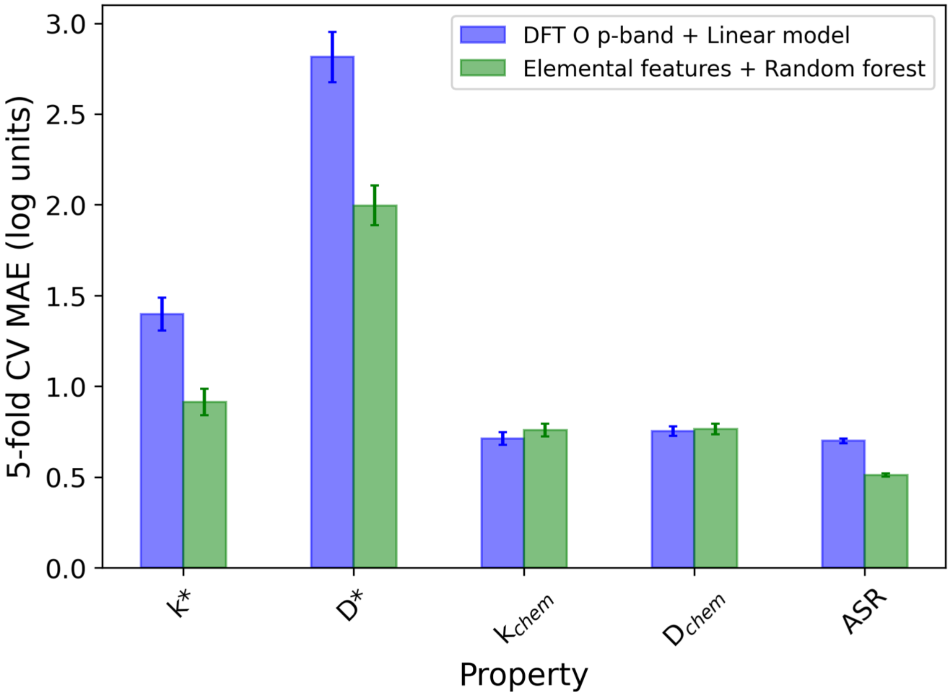
Property	Number of studies examined	Number of measurements extracted	Number of unique materials
k_{chem}	70	98	62
D_{chem}	56	83	58
k*	39	80	48
D*	37	66	42
ASR	235	422	257

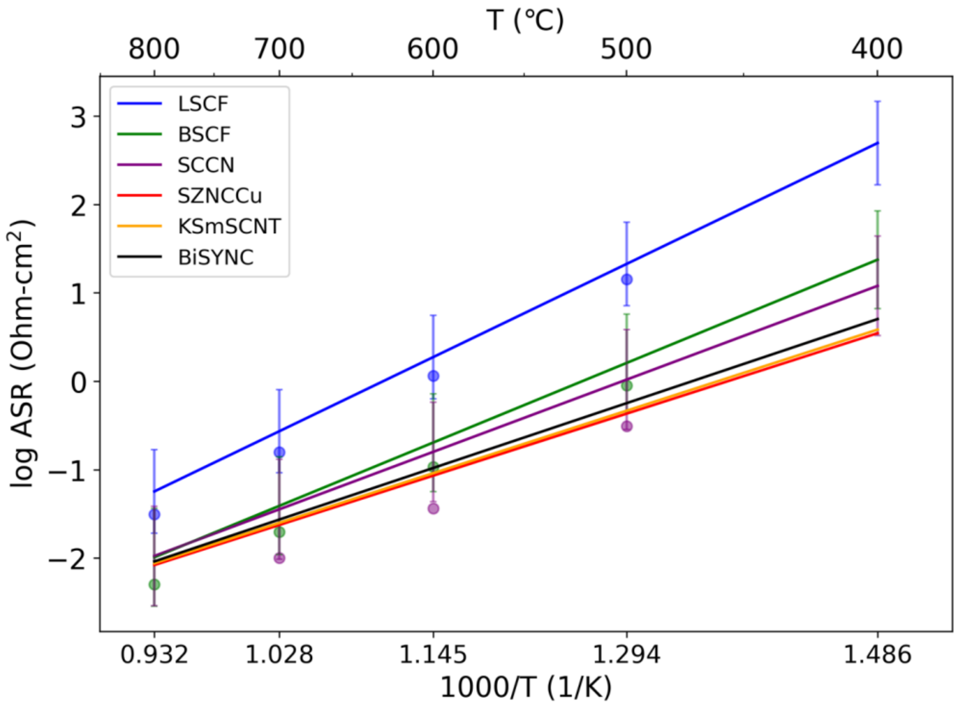
Jacobs, R., et al. Adv. Eng. Mat. ***(2024), just accepted***

Data points discussed: Jacobs, R., et al., **ACS Applied Energy Materials** 7(8), 3366-3377, 2024.

Machine learning predicted electrode materials

- Trained machine learning model could predict properties faster and at least as accurately than DFT-based study and could cover a larger space containing traditionally less-explored elements (e.g., K, Bi, Y, Ni, Cu).





$SrZr_{0.125}Nb_{0.125}Co_{0.625}Cu_{0.125}O_3$ (SZNCCu),
$K_{0.25}Sm_{0.125}Sr_{0.625}Nb_{0.125}Ta_{0.125}Co_{0.75}O_3$ (KSmSCNT)
$Bi_{0.125}Sr_{0.875}Y_{0.125}Ni_{0.125}Co_{0.75}O_3$ (BiSYNC)

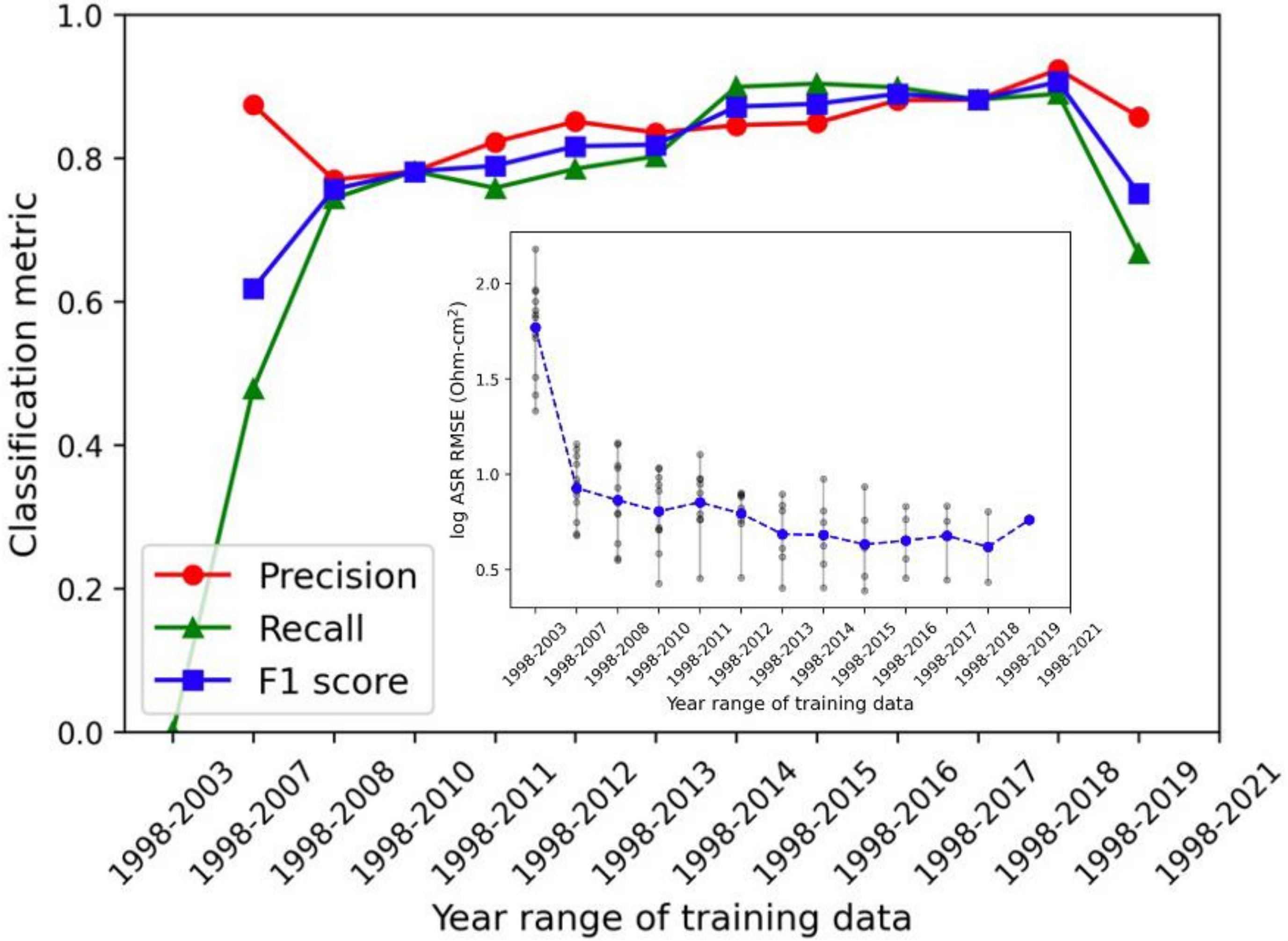
SCCN: Xin, H. Nature Energy (2022), Zhai, S., et al. Nature Energy (2022)7(8), 3366

Jacobs, R., et al. ***Adv. Energy. Mat***, 2303684 **2024.** *(doi.org/10.1002/aenm.202303684)*
Jacobs, R., et al. ***ACS Applied Energy. Mat***, 7(8), 3366, **2024.** *(doi.org/10.1021/acsaem.4c00125)*

Time dependent cross-validation

Examining how many data points would be necessary to make advances

Training on materials known *prior to 2003* suggests high performing materials in the Ba(Fe, Co, Zr)O_3 space, suggesting BSCF and BFCZ could have been predicted at the time using machine learning



Electrode Design and Engineering

Building better performing, longer lasting electrodes

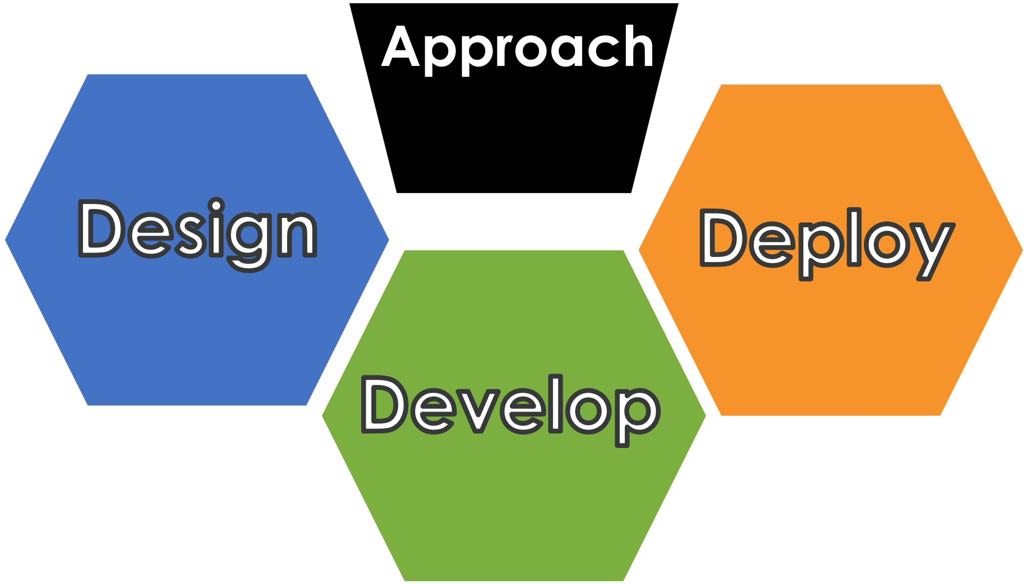
SOC Electrode Design and Engineering

Objectives

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

Benefits

- Cell/stack cost reduction
- Cell overpotential reduction
- Increased thermo-chemical/thermo-mechanical stability
- Reduced cost-of-electricity and/or cost of hydrogen



Approach

Design

Develop

Deploy

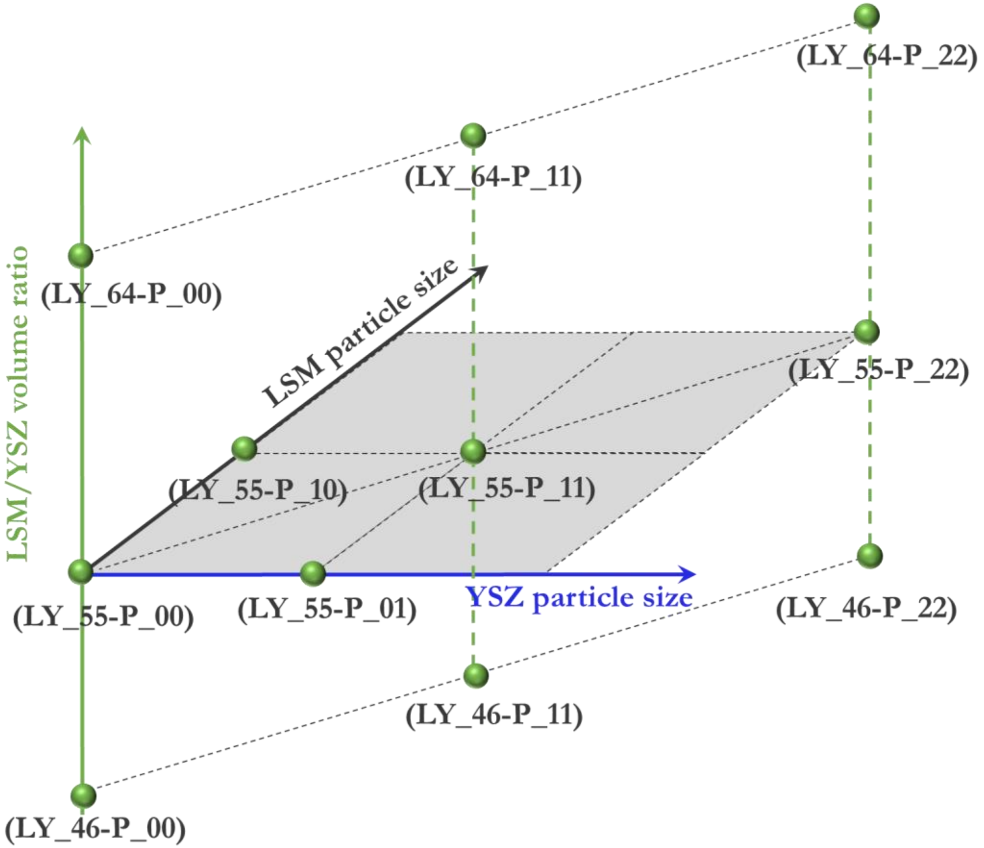
DESIGN new materials and structures

DEVELOP tailored electrode designs

DEPLOY in commercial SOC systems

Simulating infiltrated electrodes

- Using in-house multiphysics code to optimize cathode performance by controlling infiltration and backbone structure
  - Optimal catalytic properties for a given backbone composition/structure
  - Optimal backbone composition/structure for given catalytic properties
  - Optimal lifetime performance based on backbone/infiltrated particle degradation



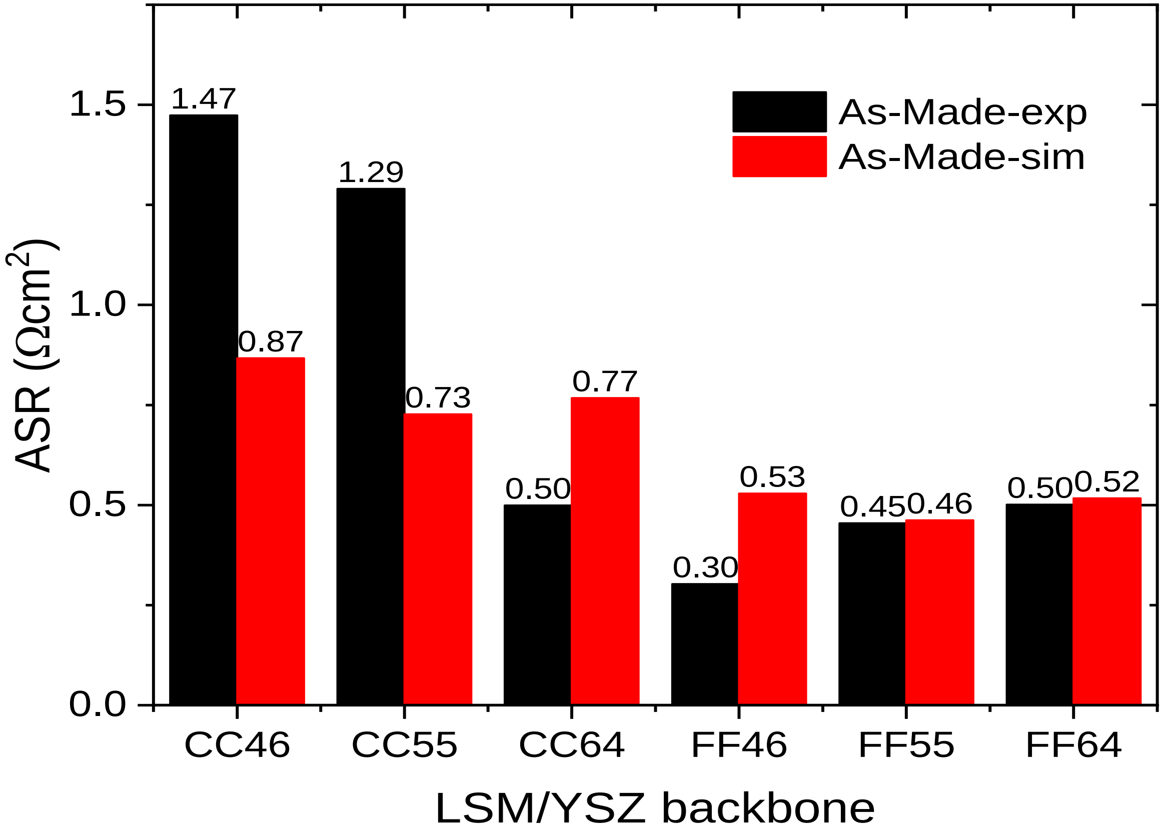
Volume fractions:	**Particle size:**
40:60	-1,0,1,2,3,4
50:50	0 = Baseline
60:40	-1 = Coarser
	1-4 = Finer

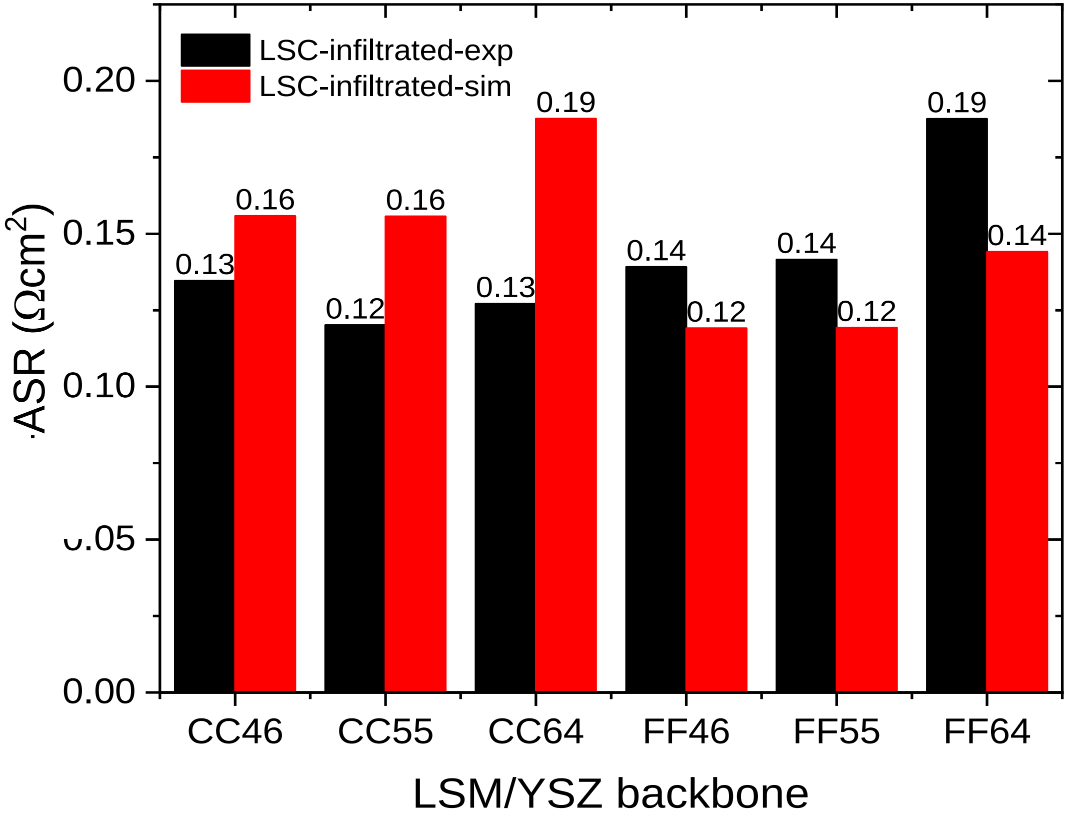
Notation: Backbone_LSM:YSZ-LSM/YSZ Particle size

Experiments vs. Simulations

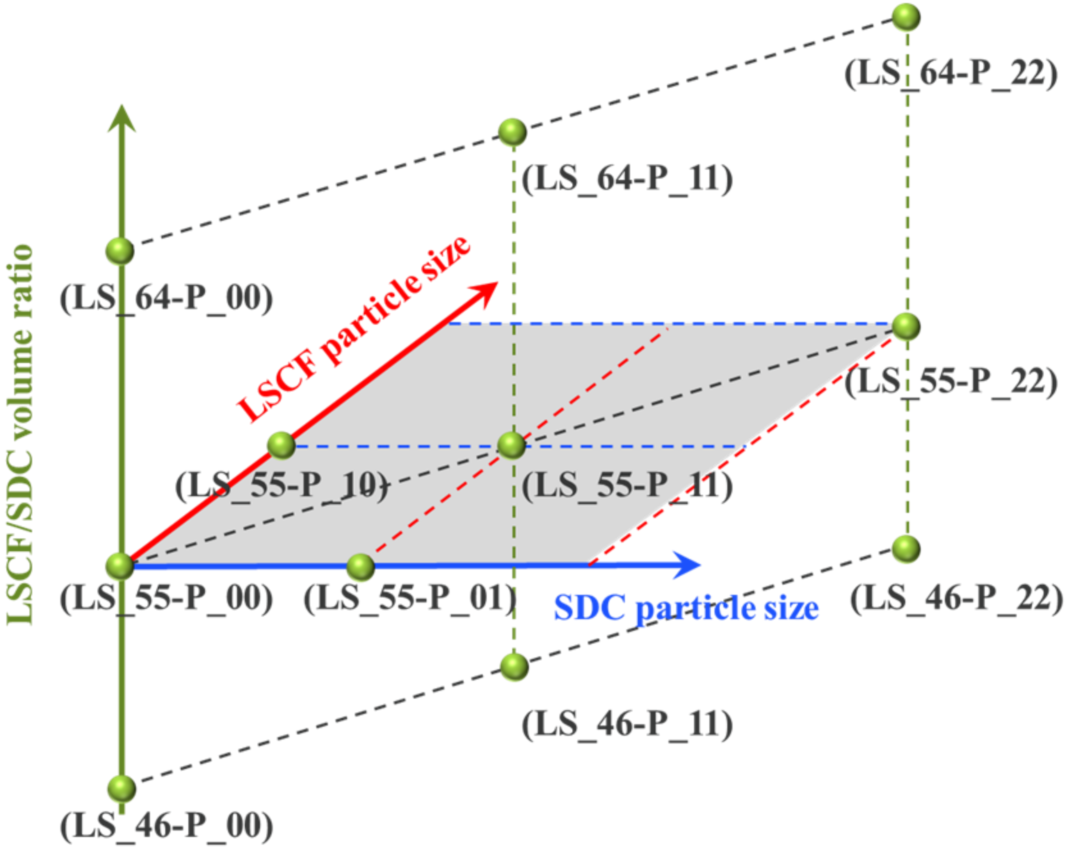
2023 Results for LSM/YSZ air electrodes

- LSC infiltration into LSM/YSZ overcame the spread in performance from the different backbones.





2024 Update: LSCF/SDC Electrodes



Parameters:

1) Volume Fraction

$$V_{LSCF}:V_{SDC} = 40\%:60\%,\ 50\%:50\%,\ \text{or}\ 60\%:40\%$$

2) Grain Size

$\beta = 0$: Coarse grain(P-0)

$\beta = 1$: Fine grain (P-1)

$$D_{LSCF} = 0.52^{\beta_{LSCF}} \cdot D_{LSCF,ref}$$

$$D_{SDC} = 0.38^{\beta_{SDC}} \cdot D_{SDC,ref}$$

$$D_{LSCF,ref} = 0.68\ \mu m$$

$$D_{SDC,ref} = 0.63\ \mu m$$

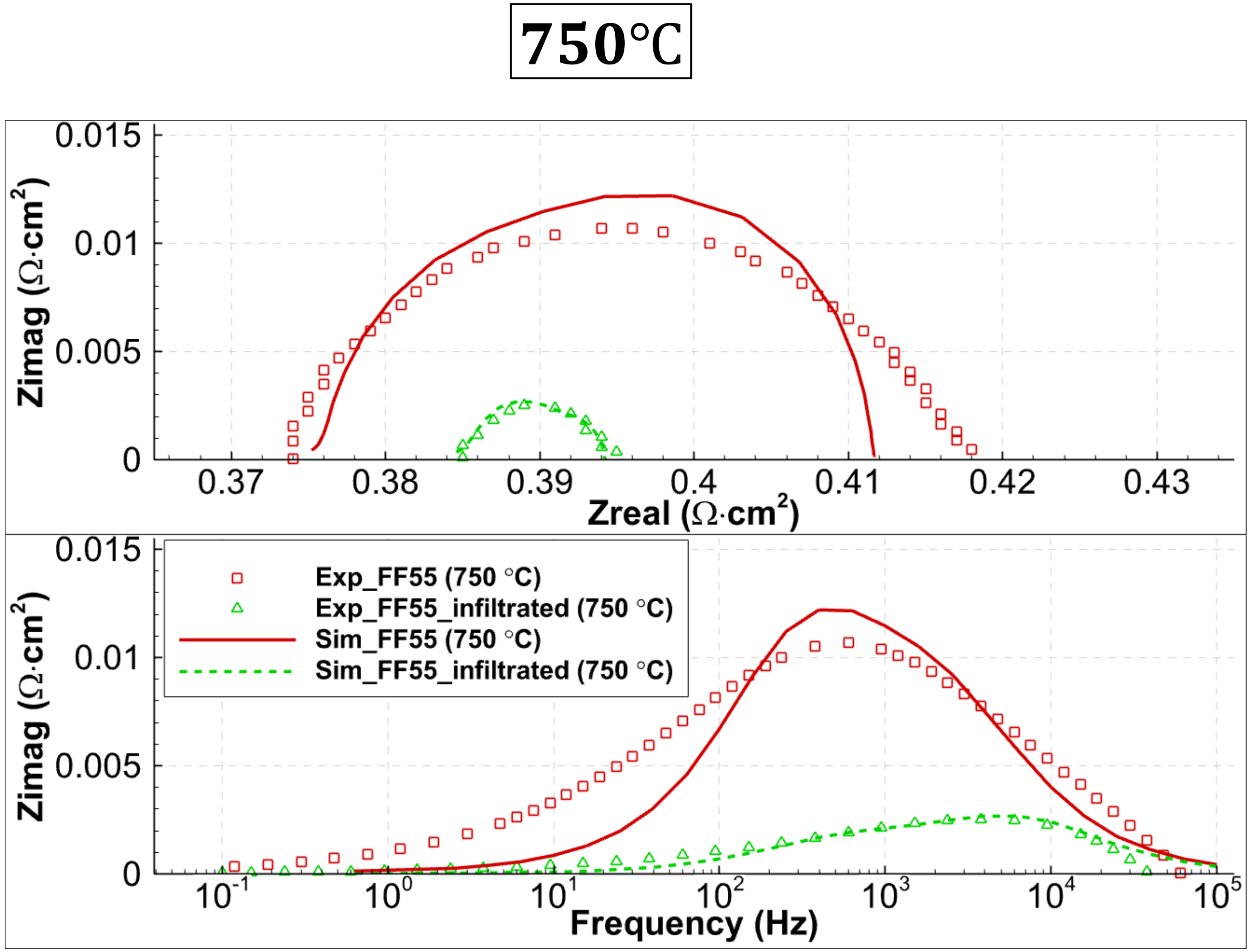
Sample Notation

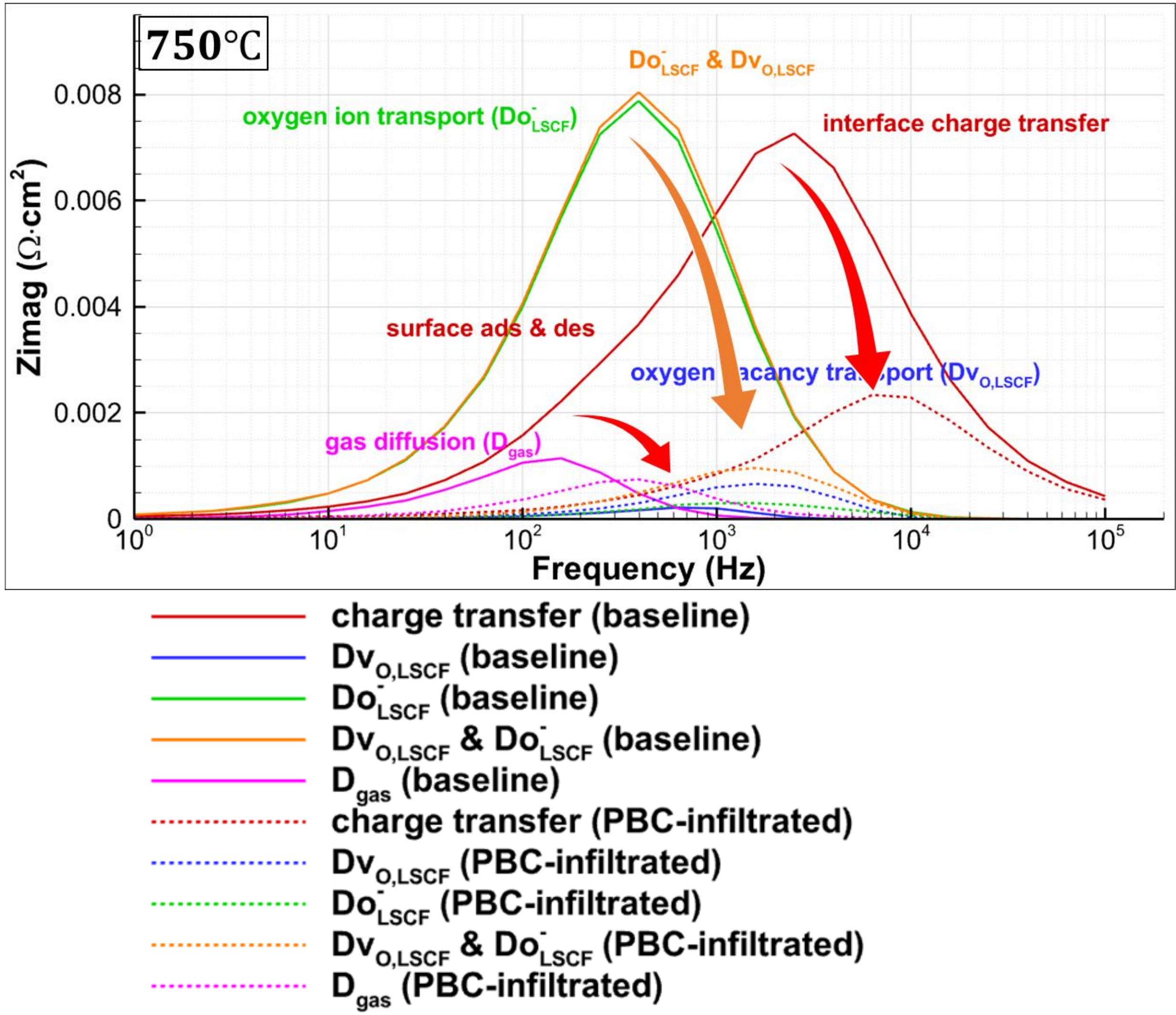
ex) LS_46-P_01 (CF46)

volume fraction: $V_{LSCF}:V_{SDC} = 40\%:60\%$

grain size: $\beta_{LSCF} = 0$, $\beta_{SDC} = 1$

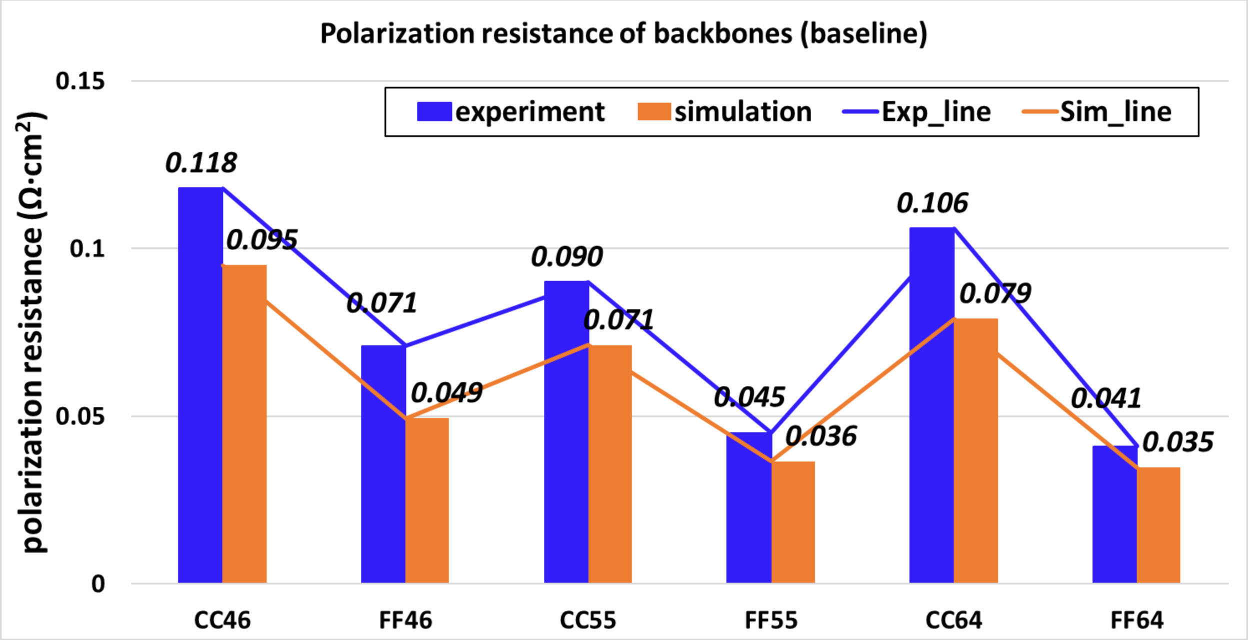
Calibration of Numerical Model



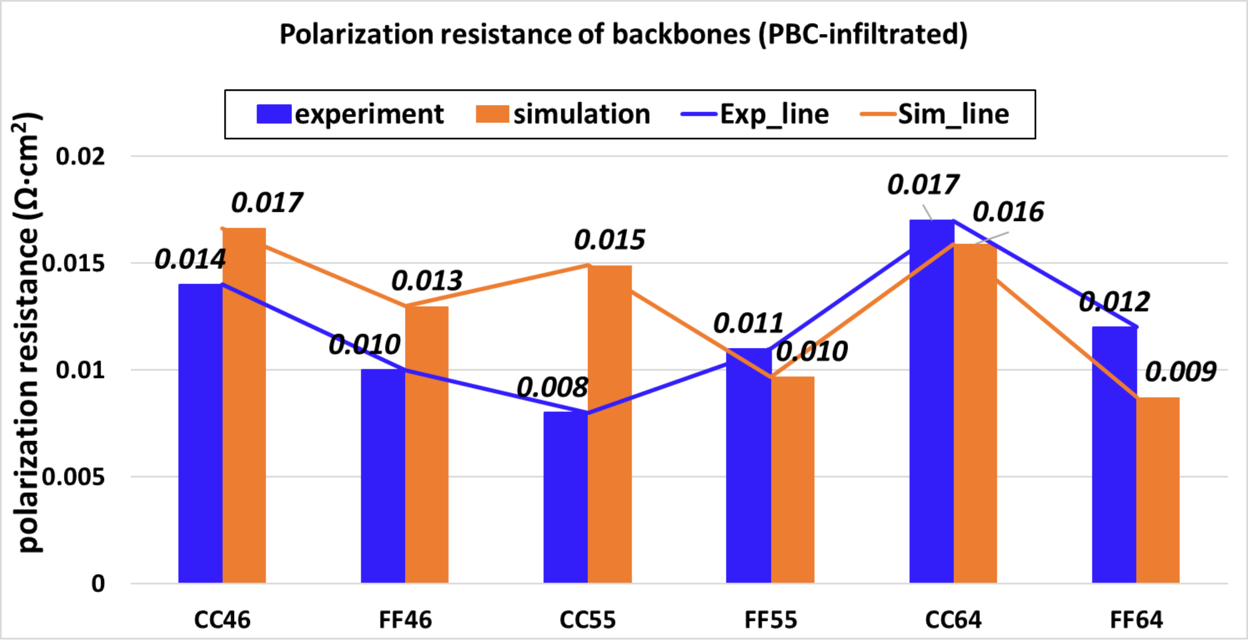


PBC Infiltration of LSCF/SDC Backbones

Baseline



PBC-infiltrated



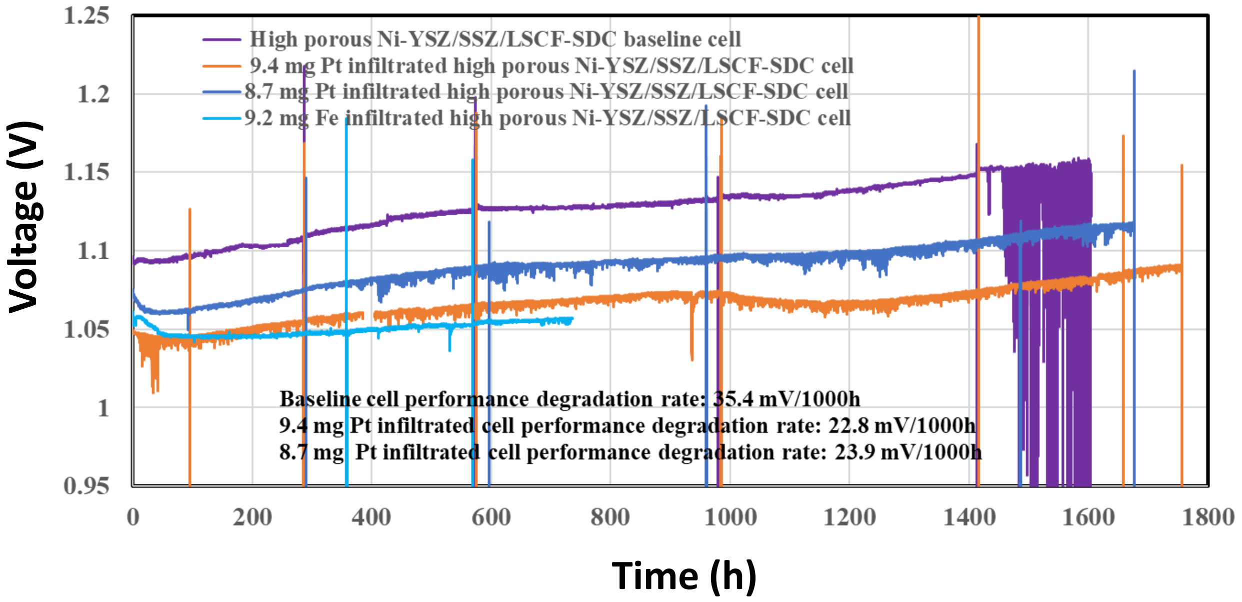
- As with LSM/YSZ, infiltration levelized the performance of all the backbones.
- Still suspect that microstructural discrepancies exist between real and simulated microstructures

Additional Infiltration Results

Recent focus on SOEC infiltration before transition to R-SOCs

- Ni-YSZ/SSZ/LSCF/SDC Samples operated at 1 A/cm^2 at 850°C
- Post-mortem analysis is ongoing
- Infiltrated R-SOC testing beginning this quarter

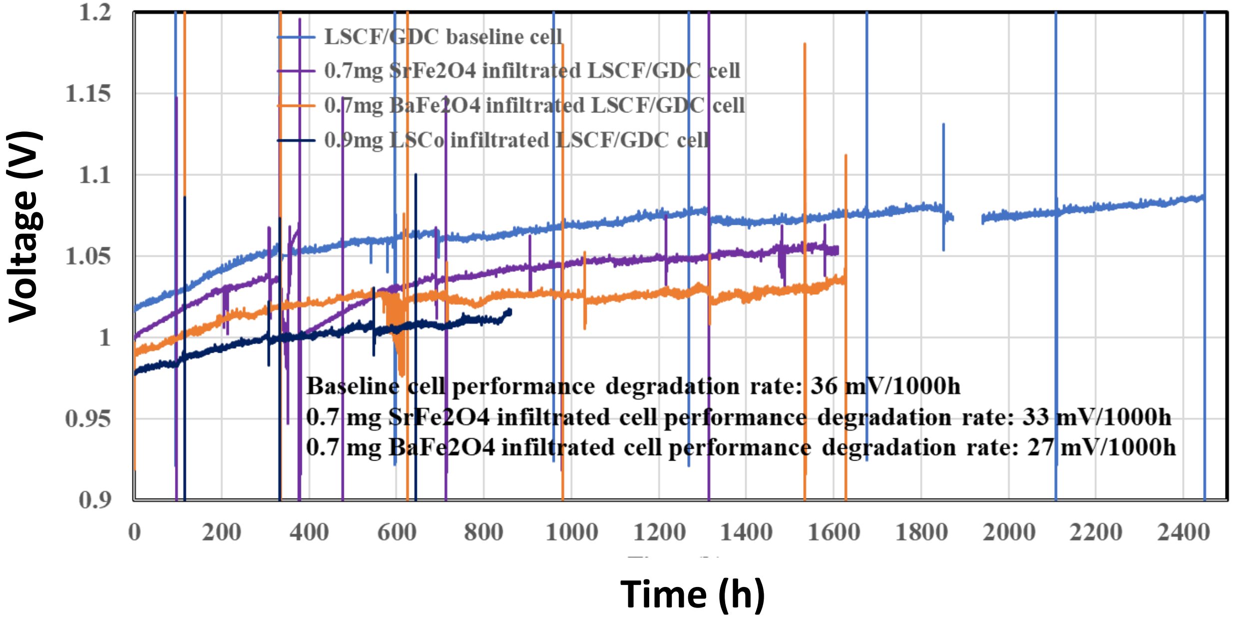
Infiltration into Steam Electrode



Baseline cell performance degradation rate: 35.4 mV/1000h
9.4 mg Pt infiltrated cell performance degradation rate: 22.8 mV/1000h
8.7 mg Pt infiltrated cell performance degradation rate: 23.9 mV/1000h

Fe infiltration more effective and cheaper than Pt

Infiltration into Air Electrode



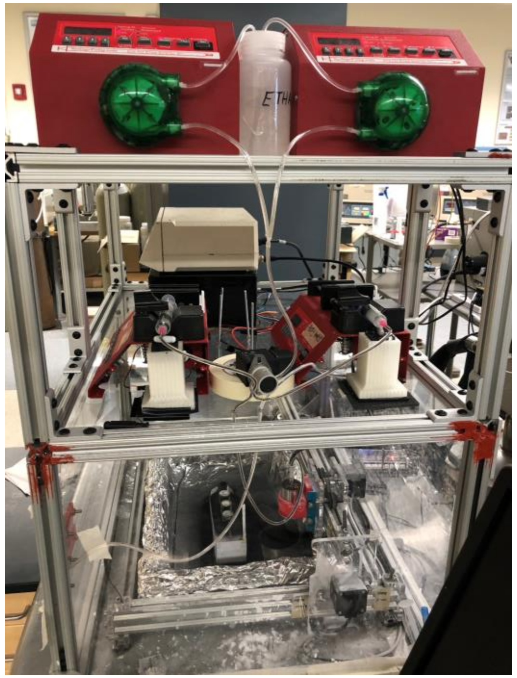
Baseline cell performance degradation rate: 36 mV/1000h
0.7 mg SrFe2O4 infiltrated cell performance degradation rate: 33 mV/1000h
0.7 mg BaFe2O4 infiltrated cell performance degradation rate: 27 mV/1000h

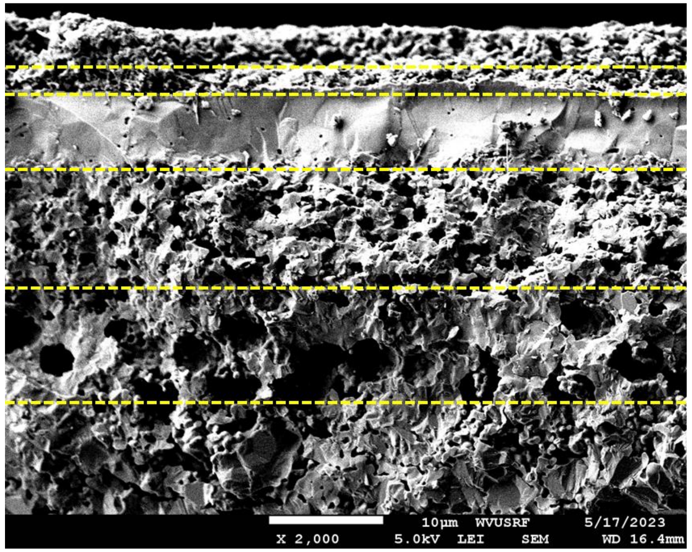
In terms of performance, LSC > $BaFe_2O_4$ > $SrFe_2O_4$

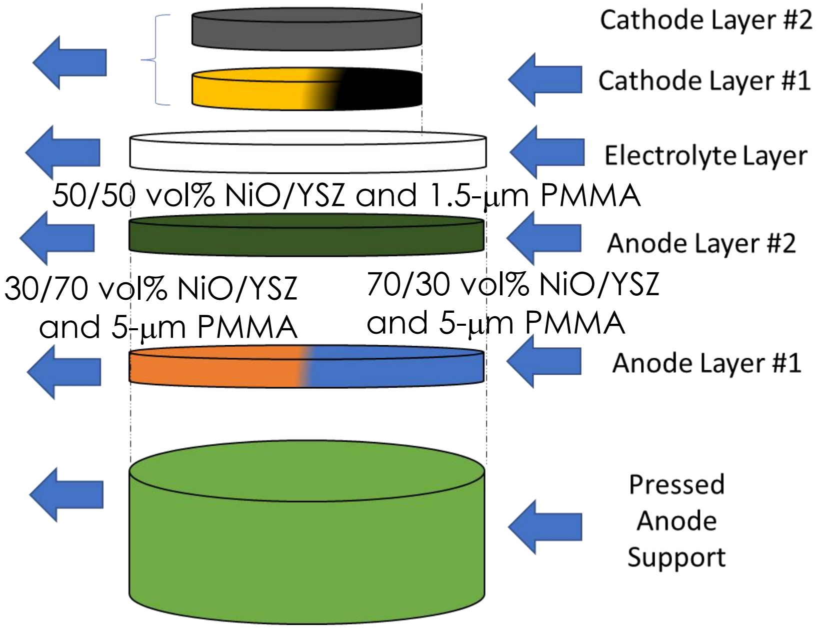
Additive Manufacturing of SOCs

Creating 3D microstructure gradients to control gradients in T, V

- Automated spray deposition system built at WVU used to apply active anode, electrolyte, and active cathode layers. Deposition parameters adjusted to improve quality/performance
  - Cathode polarization resistance at 800°C improved from 0.377 ohm-cm^2 down to 0.0381 ohm-cm^2
- Finer resolution nozzle installed, deposition width of 1.21 mm vs. 10.95 mm







Cathode Layer #2

Cathode Layer #1

Electrolyte Layer

50/50 vol% NiO/YSZ and 1.5-µm PMMA

Anode Layer #2

30/70 vol% NiO/YSZ and 5-µm PMMA

70/30 vol% NiO/YSZ and 5-µm PMMA

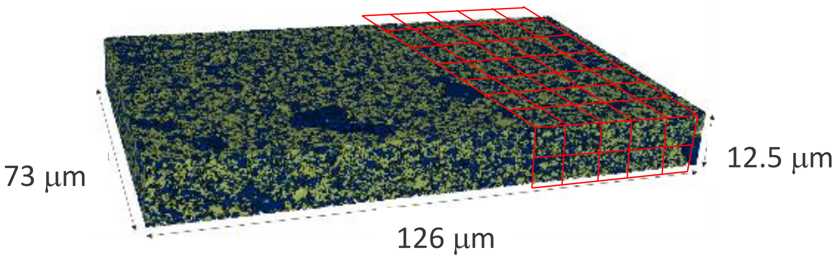
Anode Layer #1

Pressed Anode Support

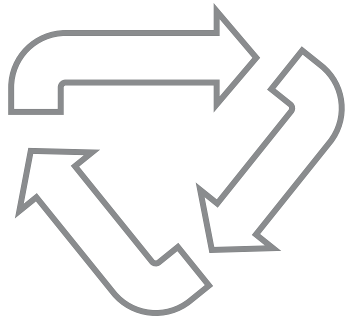
Cell and Stack Degradation Modeling

Simulation-driven design of advanced SOCs

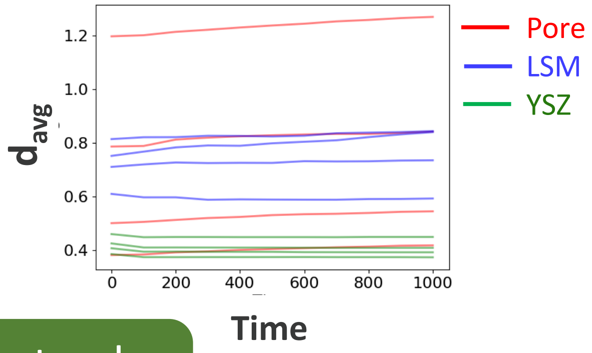
Integrated Cell Degradation Model



3D Electrode Microstructures



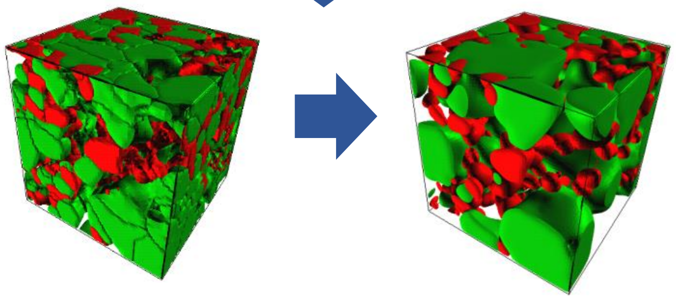
Microstructural Analysis



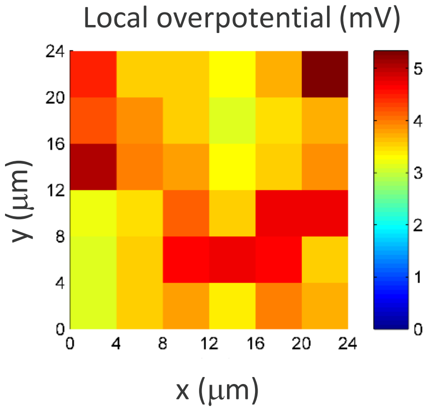
- Particle sizes
- Volume fraction
- Distributions
- Heterogeneity
- Tortuosity

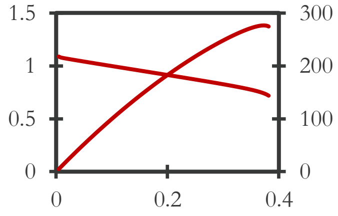
Degradation models

- Coarsening
- Secondary phases
- Poisoning
- Interdiffusion
- Cracking/delamination



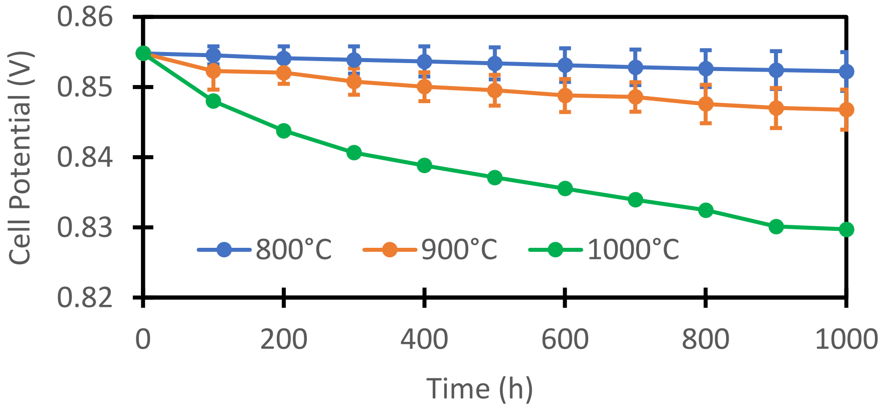
Multiphysics Performance Model





- Polarization curves
- Impedance spectra
- T, P Distributions
- Hotspots

Cell Lifetime Prediction



SOFC degradation from **coarsening** shown. Framework can be used in SOFC, SOEC, and r-SOC mode with multiple modes.

Analyzing performance degradation

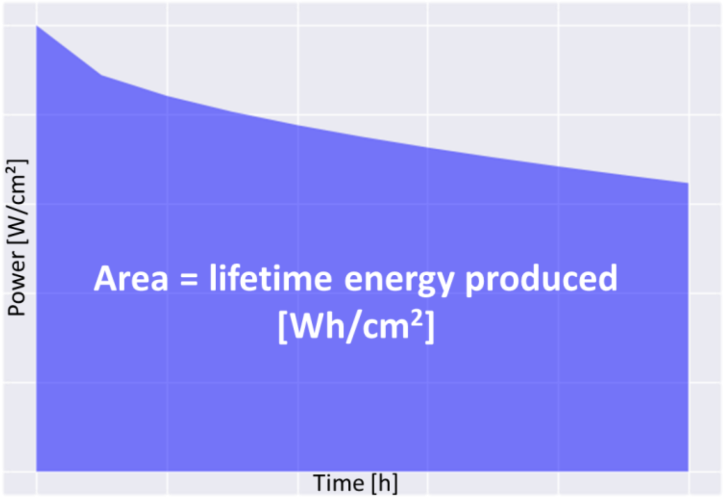
How to determine what's a good or bad electrode?

- SOC Simulations run on database of **1000s** of synthetic microstructure covering large matrix of microstructural parameter combinations (particle sizes, phase fractions, particle size distribution, phase fraction distribution, etc.)

Need a single figure-of-merit that captures **both** **initial performance** and **stability**

Lifetime energy production chosen.

Presently: operation at a given current density, up to a given time



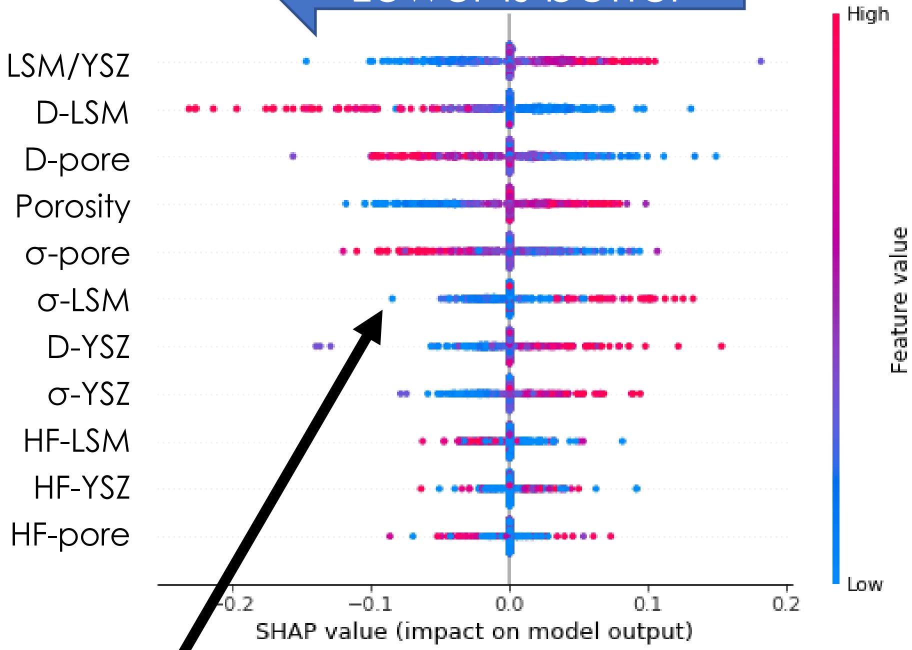
NETL Microstructure Resources

- **SOC Synthetic Electrode Microstructure Database**
  - **1,970 unique 3-phase electrode microstructure files**
  - **DOI: 10.18141/1988063**
- **PFIB-SEM 3D reconstructions of real SOFC electrodes:**
  DOI: 10.18141/1425617

SOFC Cathode Feature Importance Ranking

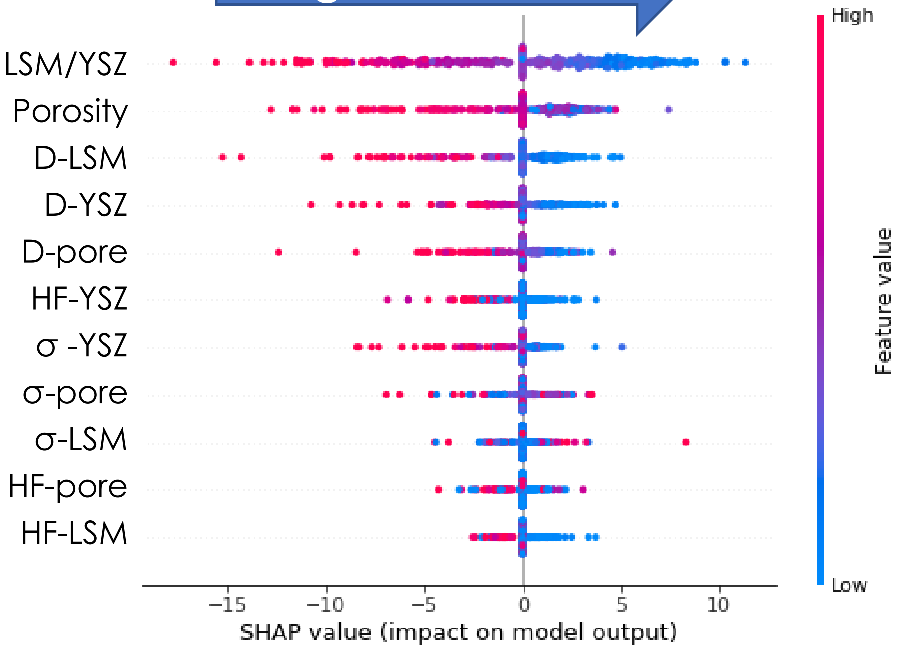
Impact on voltage decay [%/khr]

Lower is better



Impact on lifetime energy [Wh/cm^2]

Higher is better



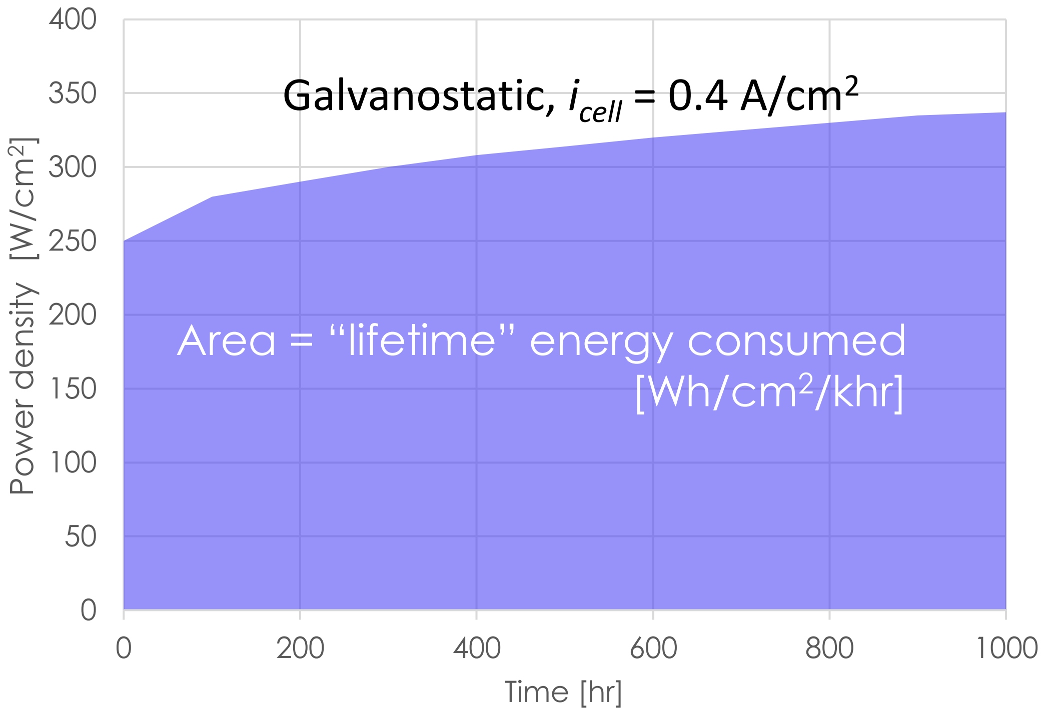
Each point represents a feature value from a specific simulated electrode microstructure

Small LSM particle sizes are bad for voltage decay, but net good for lifetime performance - **worthwhile tradeoff.** Lower LSM/YSZ ratio is good for both metrics

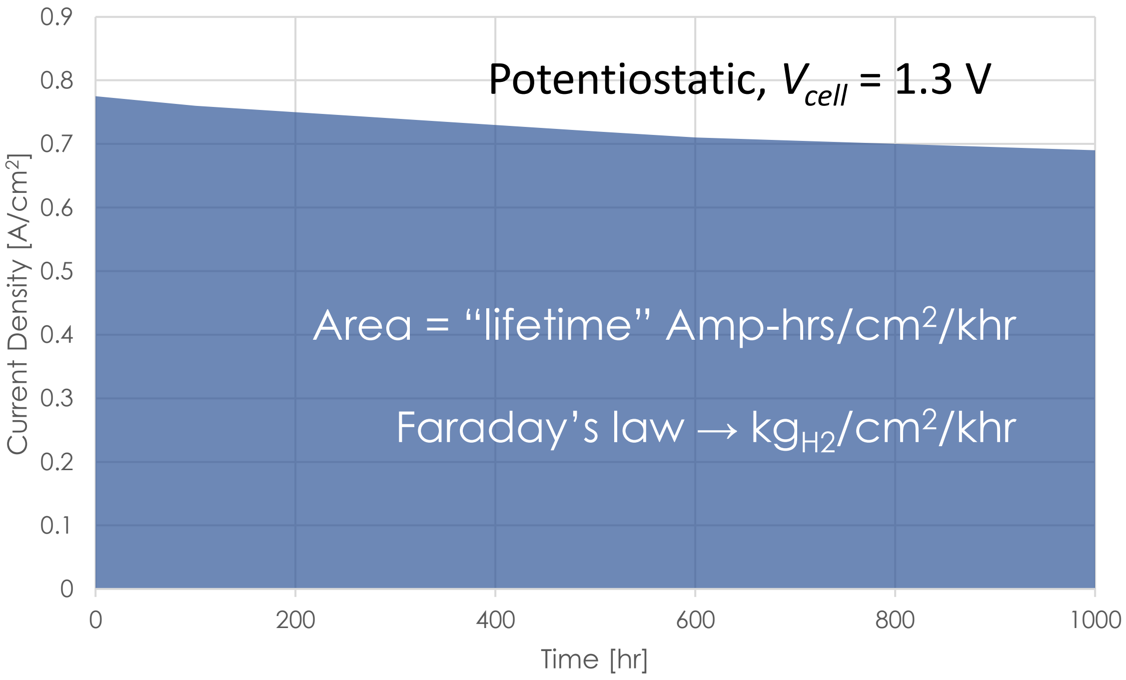
W. K. Epting et al., ***ECS Trans.***, 103, 909 **(2021)**.

SOEC Figures of Merit

Linking SOEC lifetime performance to economics



"Lifetime" energy consumed – at a given current density (and hence H_2 rate)

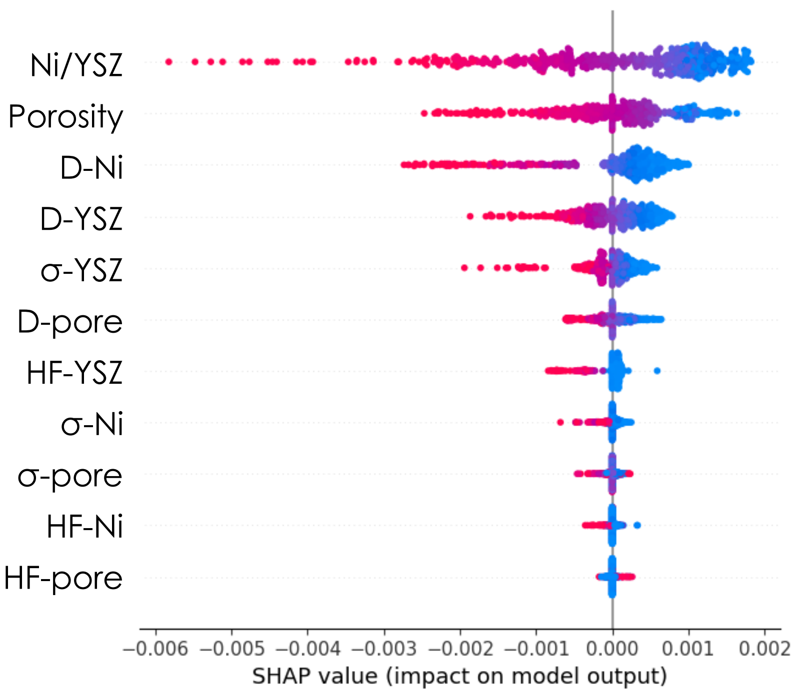


"Lifetime" H2 produced – at a given voltage (chosen roughly thermoneutral)

Feature Importance

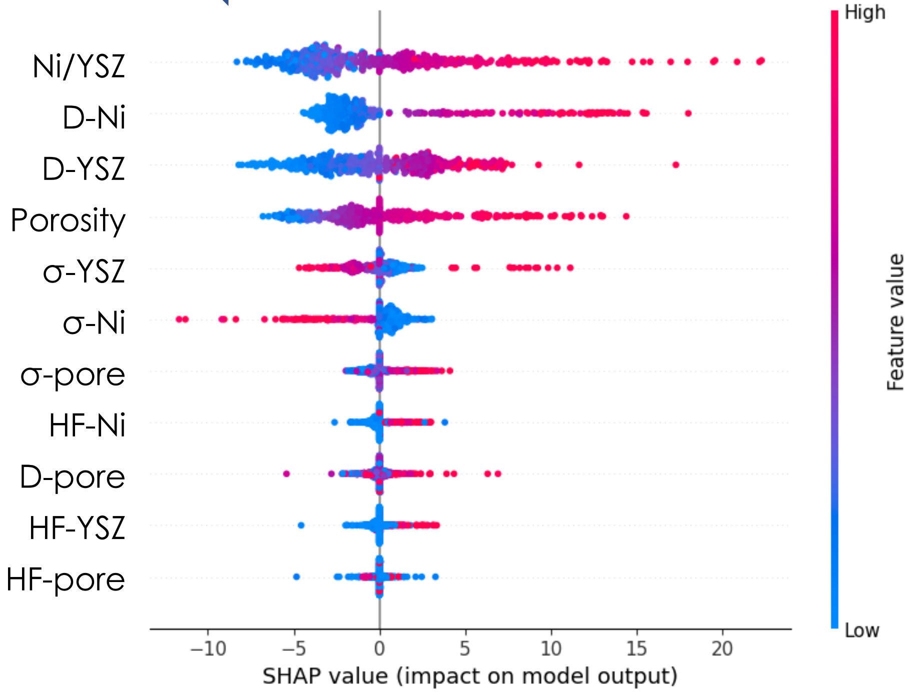
Impact on H_2 Produced [$kg/cm^2/khr$]

Higher is better



Impact on energy consumed [$Wh/cm^2/khr$]

Lower is better



Low Ni/YSZ ratio, low porosity, small solid particles beneficial for both, but rankings are different

Other figures of merit (e.g. degr. only) may show different dependence

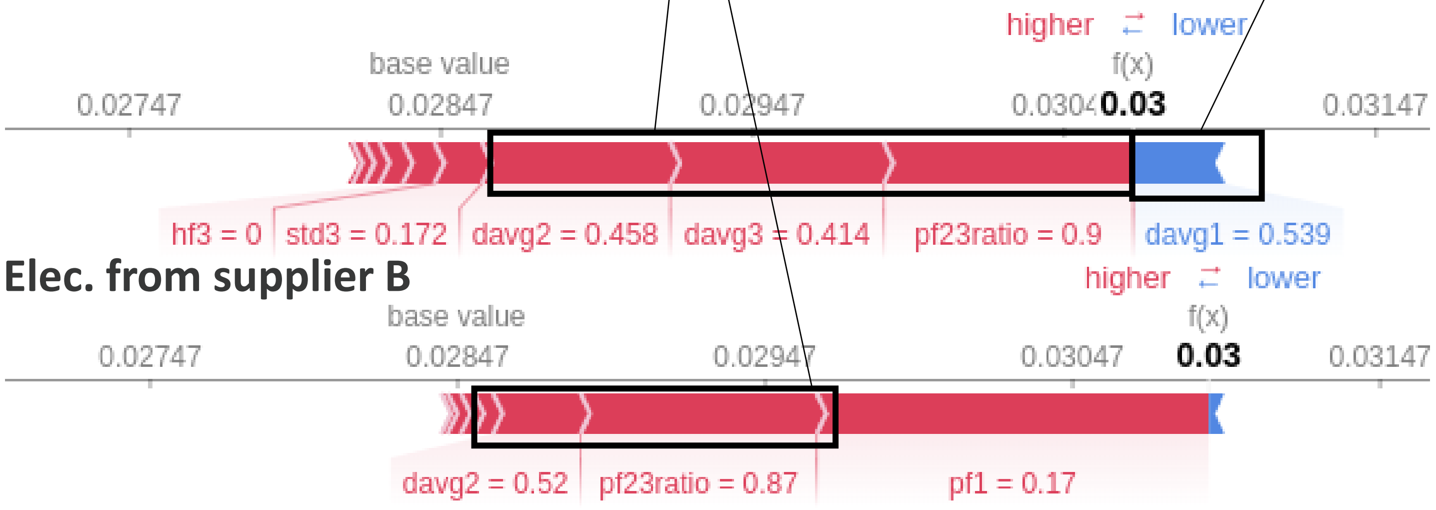
Making specific recommendations: SOEC

Commercial electrodes' initial microstructural parameters (measured) → ML model trained on bank of results from synthetic microstructures → SHAP analysis to estimate feature impact in commercial electrodes

Low Ni/YSZ ratio, small solid particles were good choices

Biggest drain was pore size

Fuel Elec. from supplier A



Fuel Elec. from supplier B

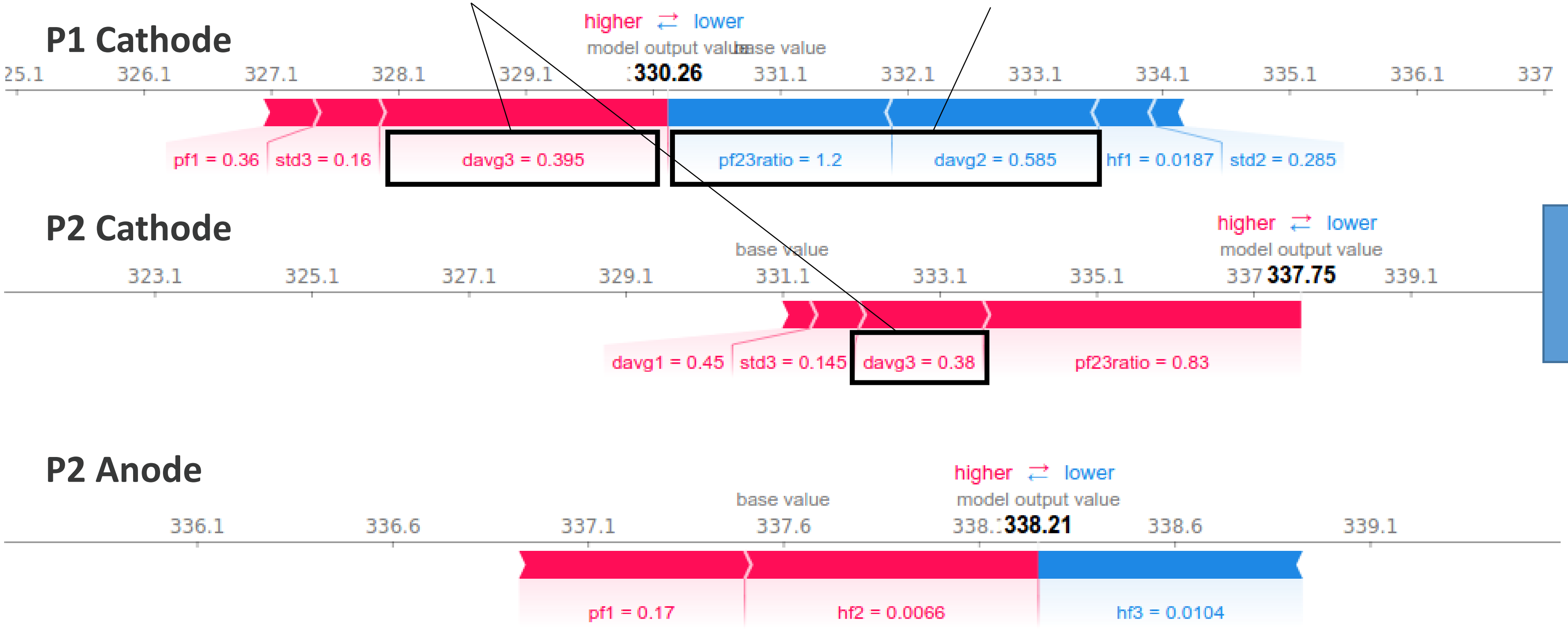
Chosen metric was H_2 produced

SOFC Recommendations

- Samples from Materials Systems Research Inc. (MSRI, Salt Lake City, UT).

Small YSZ powder was a good choice

Biggest drains are LSM/YSZ ratio and D_{LSM}


P1 Cathode

P2 Cathode

P2 Anode

Chosen metric was lifetime energy

Wrap Up

Conclusions

- Materials discovery using machine learning can screen an even larger parametric space than previous high throughput methods
- Modeling is useful tool for deeper interpretation of performance data, designing more durable electrodes, and providing context to literature results
- NETL continues to develop advanced electrode design and fabrication tools for more optimized lifetime performance

How can NETL help you?

- NETL’s synthetic microstructure database, real 3D microstructures, microstructural analysis tools, and defect modeling tools are available to the public
- NETL can collaborate with partners, using partner data and conditions to run performance degradation and optimization simulations

2024 FECM Project Review Posters

- “Cation Migration and LSCF Decomposition Related to Long Term Operation Mode as Revealed by Electron Microscopy” – **Yoosuf Picard**
- “In-House Developed Multiphysics Simulation for the Performance of Solid Oxide Cells (SOCs)” – **Jian Liu**
- “Defect Thermodynamics and Transport Properties of Perovskite and Fluorite Materials for Solid-Oxide and Proton Conducting Oxide Cells Evaluated Based on Density Functional Theory Modeling” – **Yueh-Lin Lee**
- “Pathway Study for Large-Scale Hydrogen Production from Solid Oxide Electrolysis Cell Technology” - **Kyle Bucheit and Alex Noring**
- “Modeling Ni Coarsening under Humid Atmosphere in Electrode of Solid Oxide Cells” - **Yves Mantz**
- “Inter- & Intra-Granular Nanostructure Degradation of YSZ in Electrolyte Under SOEC Operation“ **– Yun Chen and Xueyan Song**

Recent Publications

1. R. Jacobs, et al., "A Critical Assessment of Electronic Structure Descriptors for Predicting Perovskite Catalytic Properties" **ACS Applied Energy Materials** 7(8), 3366-3377, 2024.
2. H. Kim, et al., “Systematic and Predictive Trends to Chromium Poisoning in Solid Oxide Fuel Cell Cathodes,” **Journal of Power Sources** 603, 234390, 2024.
3. B. Guan, et al., “Unraveling the conundrum of electronic leakage in protonic ceramic cells: Operation-specific insights and rational design strategies,” **Journal of Power Sources** 601, 234454, 2024.
4. R. Jacobs, et al., “Machine Learning Design of Perovskite Catalytic Properties,” **Advanced Energy Materials** 2303684, 2024.
5. K. L. Buchheit, et al., “Techno-Economic Analysis of a Thermally Integrated Solid Oxide Fuel Cell and Compressed Air Energy Storage Hybrid System,” **Energies**, 17(1), 42, 2023.
6. Y. Fan, et al., “Enabling durable hydrogen production and preventing the catastrophic delamination in the solid oxide electrolysis cells by infiltrating SrFe$_2$O$_{4-\delta}$ solutions into LSM/YSZ -based air electrode**,” J Power Sources** 580, 233389, 2023.
7. J.H. Mason, et al., “Fundamental study of gas species transport in the oxygen electrode of solid oxide fuel and electrolysis cells,” **Int. J. Hydrogen Energy** 50(b), 1142-1158, 2024.
8. J.H. Duffy, H. Abernathy, K. Brinkman, “Tuning Proton Kinetics in BaCo$_{0.4}$Fe$_{0.4}$Zr$_{0.2-X}$Y$_X$O$_{3-\delta}$ Triple Ionic-Electronic Conductors via Aliovalent Substitution” Accepted by **Journal of Materials Chemistry A** 2023.
9. B. Zhang, et al., “A real-time multiphysics model of a pressurized solid oxide electrolysis cell (SOEC) for cyber-physical simulation,” **Energy Conversion and Management** 298, 117778, 2023.
10. Y. Mantz, Y. L. Lee, “Surface Energies of LaMnO$_3$ High-Index Surfaces Obtained from Density-Functional Theory,” **J. Phys. Chem C** 127, 20, 9814–9822, 2023.
11. Y. L. Lee, et al., “Defect Thermodynamic Modeling of Triple Conducting Perovskites (La,Ba)Fe1-xMxO3-δ for Proton-Conducting Solid-Oxide Cells,” **ECS Transactions** 111(6), 1823, 2023.
12. Y. Lei, et al. “Modeling the Distribution of Oxygen Partial Pressure in the Electrolyte of Solid Oxide Cells and Its Implication on Microstructure Evolution in the Hydrogen Electrode,” **ECS Transactions** 111(6), 965, 2023.
13. A. Noring, et al., “Techno-Economic Analysis of Reversible and Paired Solid Oxide Cell Systems for Hydrogen Production,” **ECS Transactions** 111(6), 2445, 2023.
14. T.L. Cheng, et al., “Oxidation of nickel in solid oxide cells during electrochemical operation: Experimental evidence, theoretical analysis, and an alternative hypothesis on the nickel migration,” **Journal of Power Sources** 569, 232991, 2023.
15. X. Fei et al., “Phase-field modeling of crack growth and mitigation in solid oxide cells”, **International Journal of Hydrogen Energy** 48, 9845, 2023.

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NETL Resources

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