Improving Durability and Performance of Solid Oxide Electrolyzers by Controlling Surface Composition on Oxygen Electrodes

Award DE FE0032102

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In collaboration with Elango Elangovan, Tyler Hafen and Jenna Pike, OxEon Energy

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Project Meeting with DOE-NETL, April 23rd, 2024

Sr segregation and Cr & S poisoning of $La_{0.6}Sr_{0.4}Co_{0.2}Fe_{0.8}O_3$

Cr Poisoning Nucleation Theory:

$CrO_{3(g)} + \mathbf{SrO_{(s)}} \rightarrow Cr{-}Sr{-}O_{(nuclei)(s)}$

$Cr{-}Sr{-}O_{(nuclei)(s)} + CrO_{3(g)} \rightarrow Cr_2O_{3(s)}$

$Cr{-}Sr{-}O_{(nuclei)(s)} + CrO_{3(g)} + SrO_{(s)} \rightarrow SrCrO_{4(s)}$

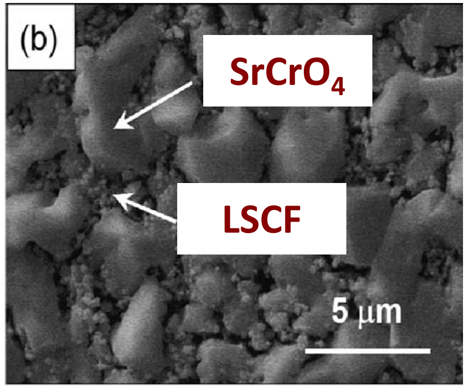
S.P. Jiang, X. Chen, *Int. J. of Hydrogen Energy,* 2014, 39.
K. Chen, S.P. Jiang, *Electrochemical Energy Reviews* 2020, 3.

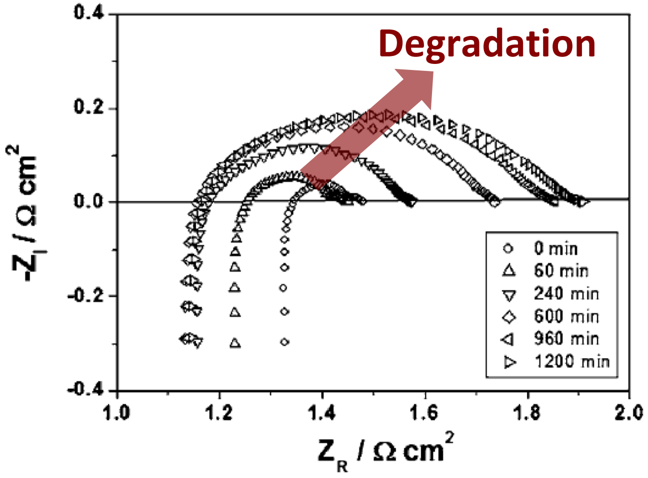
S Poisoning Nucleation Theory:

$SO_{2(g)} + \mathbf{SrO_{(s)}} \rightarrow SrSO_{4(s)}$

S.P. Jiang, X. Chen, *Int. J. of Hydrogen Energy,* 2014, 39.
K. Chen, S.P. Jiang, *Electrochemical Energy Reviews* 2020, 3.

LSCF-GDC electrodes after operation at 200 mA/cm^2, 900˚C:

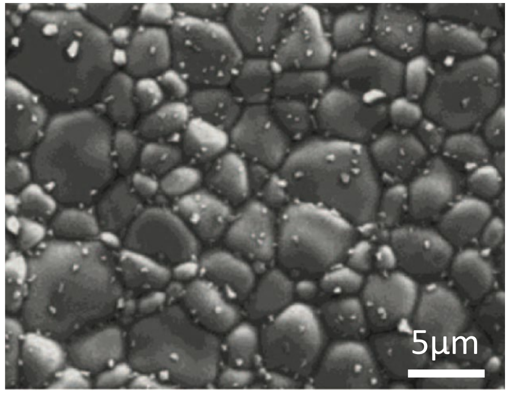


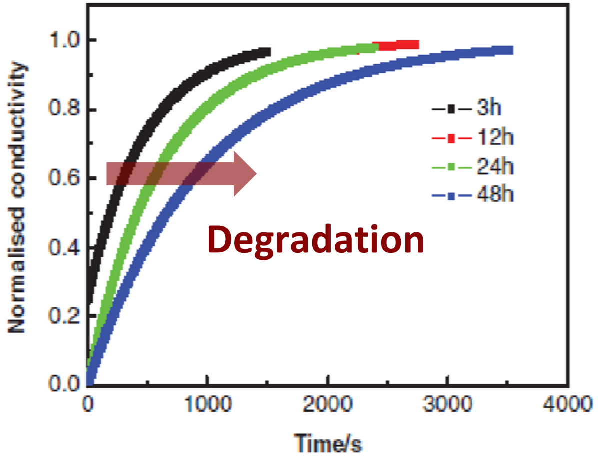


XRD, EDS, SEM → **formation of $SrCrO_4$ and Cr_2O_3 on LSCF surface.**

S.P. Jiang et al. *J. Electrochem. Soc.*, 2006,153, A127

LSCF bar samples in the presence of **20 ppm SO_2** at 900˚C:

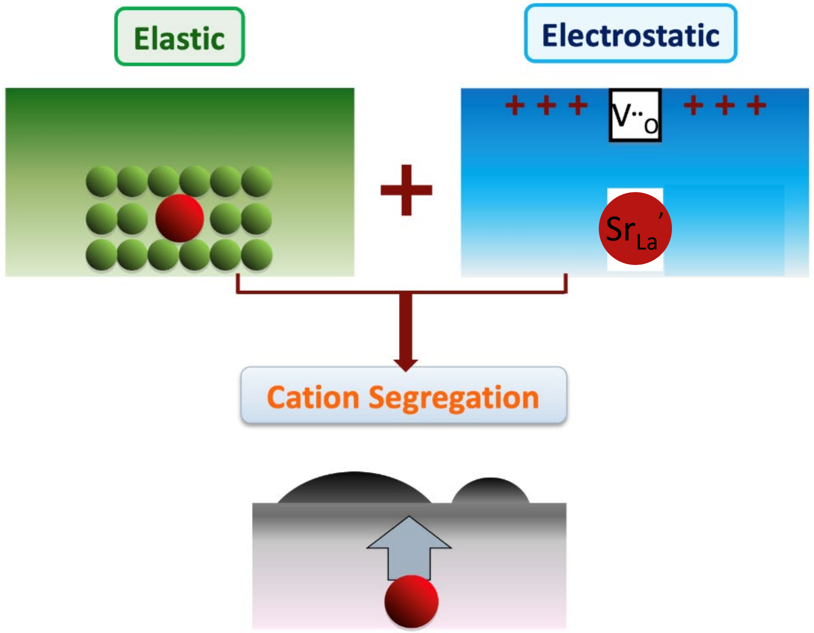


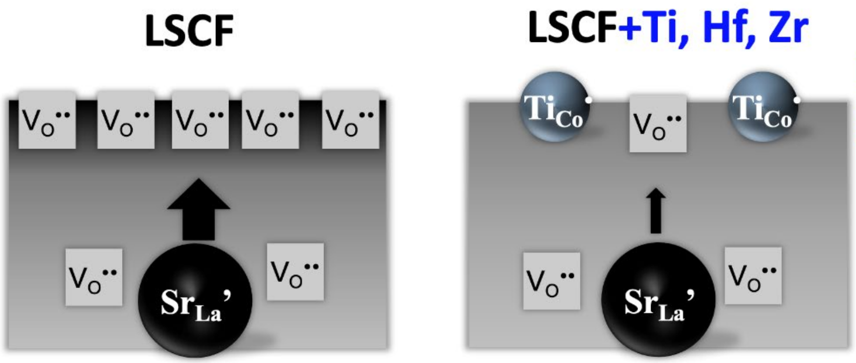


XRD, EDS, Raman → **formation of $SrSO_4$ on LSCF surface.**

C. C. Wang et al. *J. Electrochem. Soc.*, 2014, 161

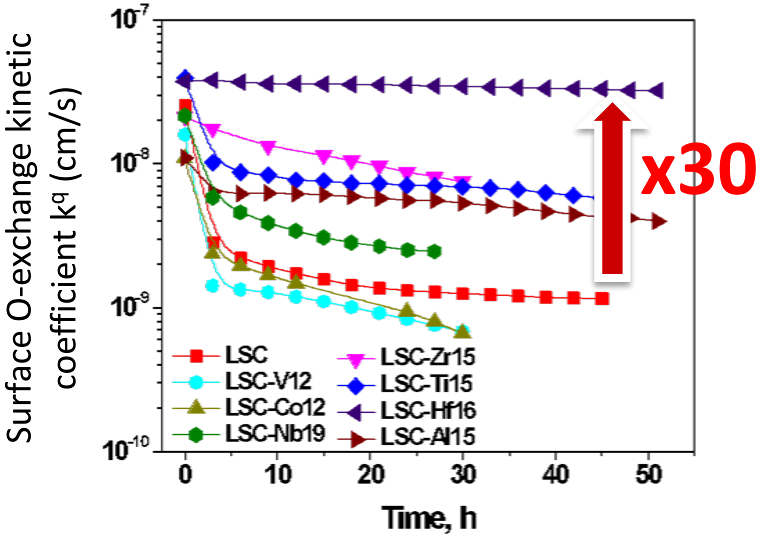
Perovskite oxide surface more stable and has faster oxygen exchange kinetics with oxidizable surface-cations.

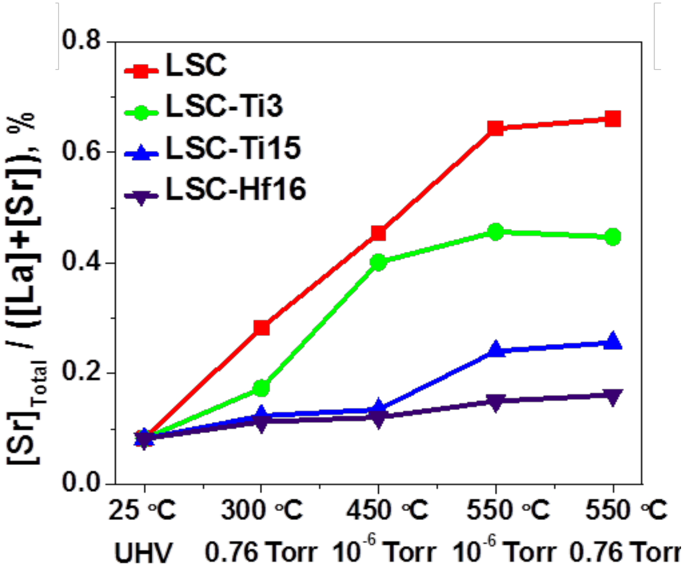




More oxidized surface

Same bulk

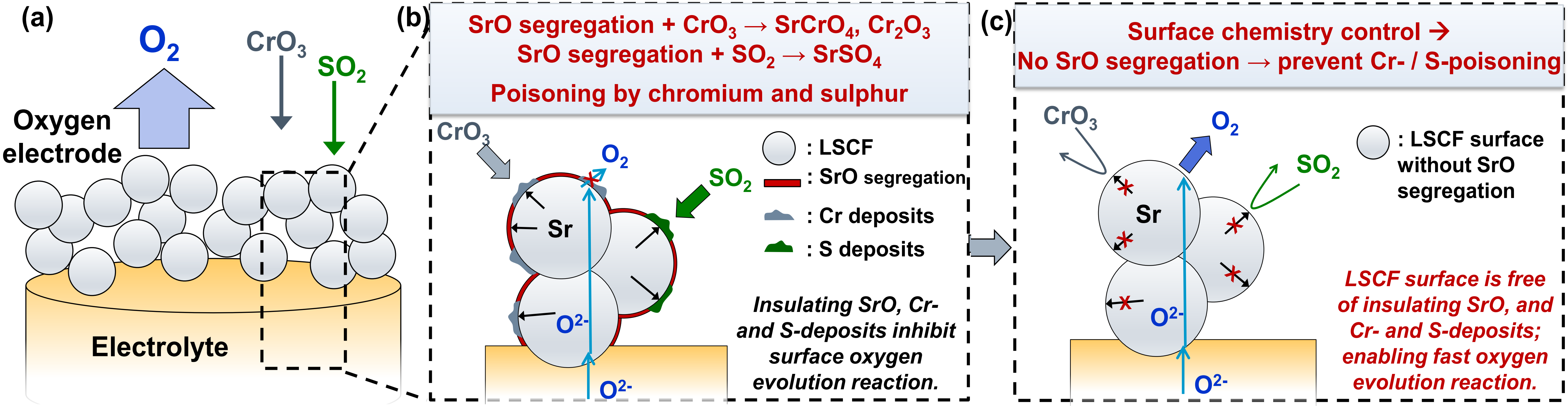




Lee et al. Yildiz, *J. Am. Chem. Soc.*, 2013, *135*.
Kim, Bliem, Hess et al. Yildiz, *J. Am. Chem. Soc.*, 2020

LSC: Tsvetkov, Lu, Sun, Crumlin, Yildiz, *Nature Materials*, 2016, 15 (9).
LSM: Bliem, Kim, Yildiz, J. Mat. Chem. A. 2021

This project aims to attack the degradation pathway coupling surface chemistry to impurity poisoning on perovskite oxygen electrodes, taking LSCF as a state-of-the-art electrode.



SrO segregation + $CrO_3 \rightarrow SrCrO_4, Cr_2O_3$
SrO segregation + $SO_2 \rightarrow SrSO_4$
Poisoning by chromium and sulphur

Insulating SrO, Cr- and S-deposits inhibit surface oxygen evolution reaction.

Surface chemistry control →
No SrO segregation → prevent Cr- / S-poisoning

LSCF surface is free of insulating SrO, and Cr- and S-deposits; enabling fast oxygen evolution reaction.

1. **Improve the chemical and electrochemical stability** of the surface of LSCF, both the initial oxygen exchange kinetics and durability.
2. Develop **infiltration chemistries** to enable the surface modifications, to **suppress the Sr-segregation and the Cr- and S-poisoning processes**.
3. Advance our understanding of **the role of operational parameters on oxygen-electrode surface chemistry and performance,** combining experiments and computations.

Methods to achieve project objectives

Collaboration with PNNL using well-established button cell fabrication process.

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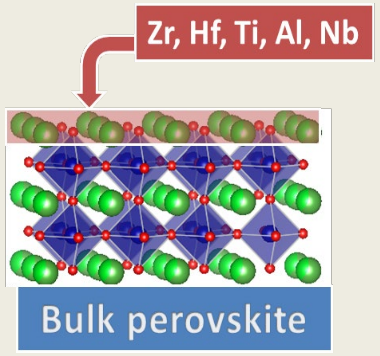
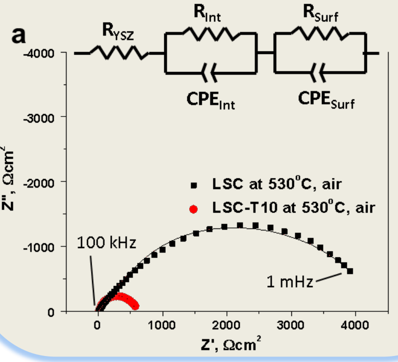
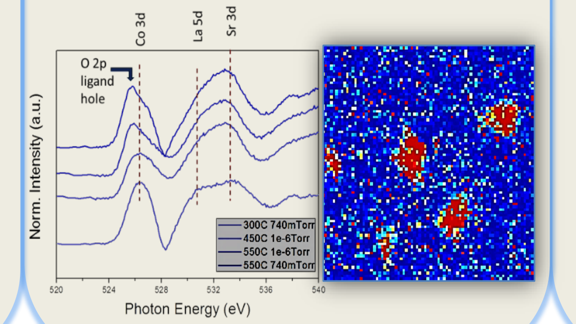
LSCF oxygen electrode surface chemical modification by solution infiltration.



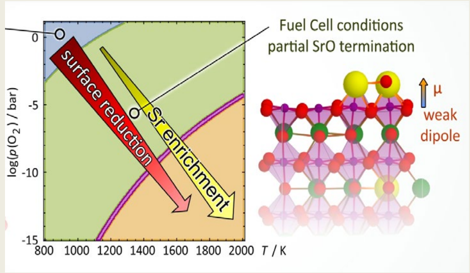
Electrochemical characterization of the electrode performance and stability.



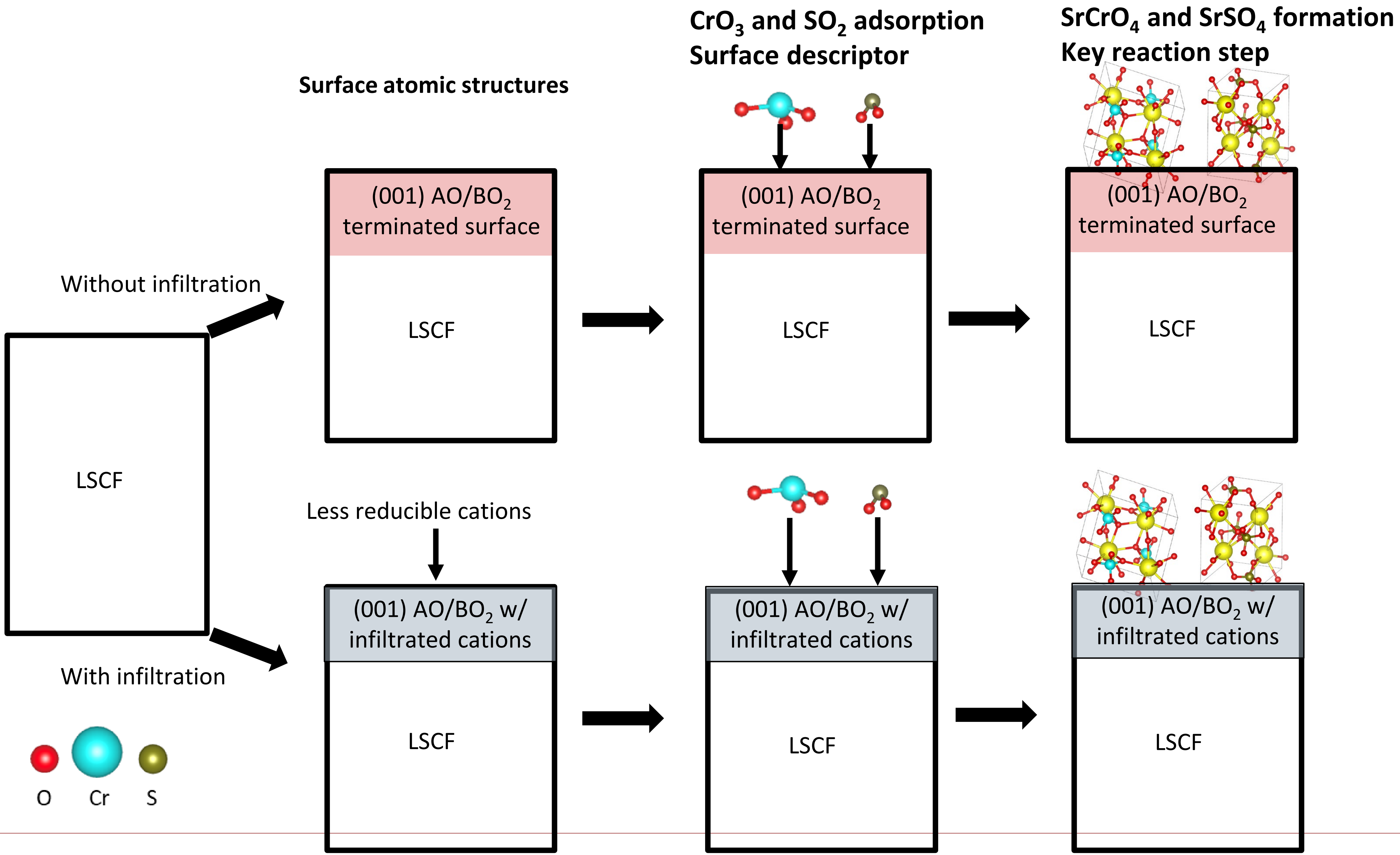
Characterization of surface chemistry (Sr-segregation, Cr- / S-poisoning) by XPS and AES, oxidation and bonding (XANES, EXAFS).



First-principles-based modeling of surface phase and reaction mechanisms. Guide design of best surfaces.



Computational investigation of Cr & S poisoning on model LSCF surfaces



Surface atomic structures

CrO_3 and SO_2 adsorption
Surface descriptor

$SrCrO_4$ and $SrSO_4$ formation
Key reaction step

LSCF

Without infiltration

(001) AO/BO_2 terminated surface

LSCF

(001) AO/BO_2 terminated surface

LSCF

(001) AO/BO_2 terminated surface

LSCF

With infiltration

Less reducible cations

(001) AO/BO_2 w/ infiltrated cations

LSCF

(001) AO/BO_2 w/ infiltrated cations

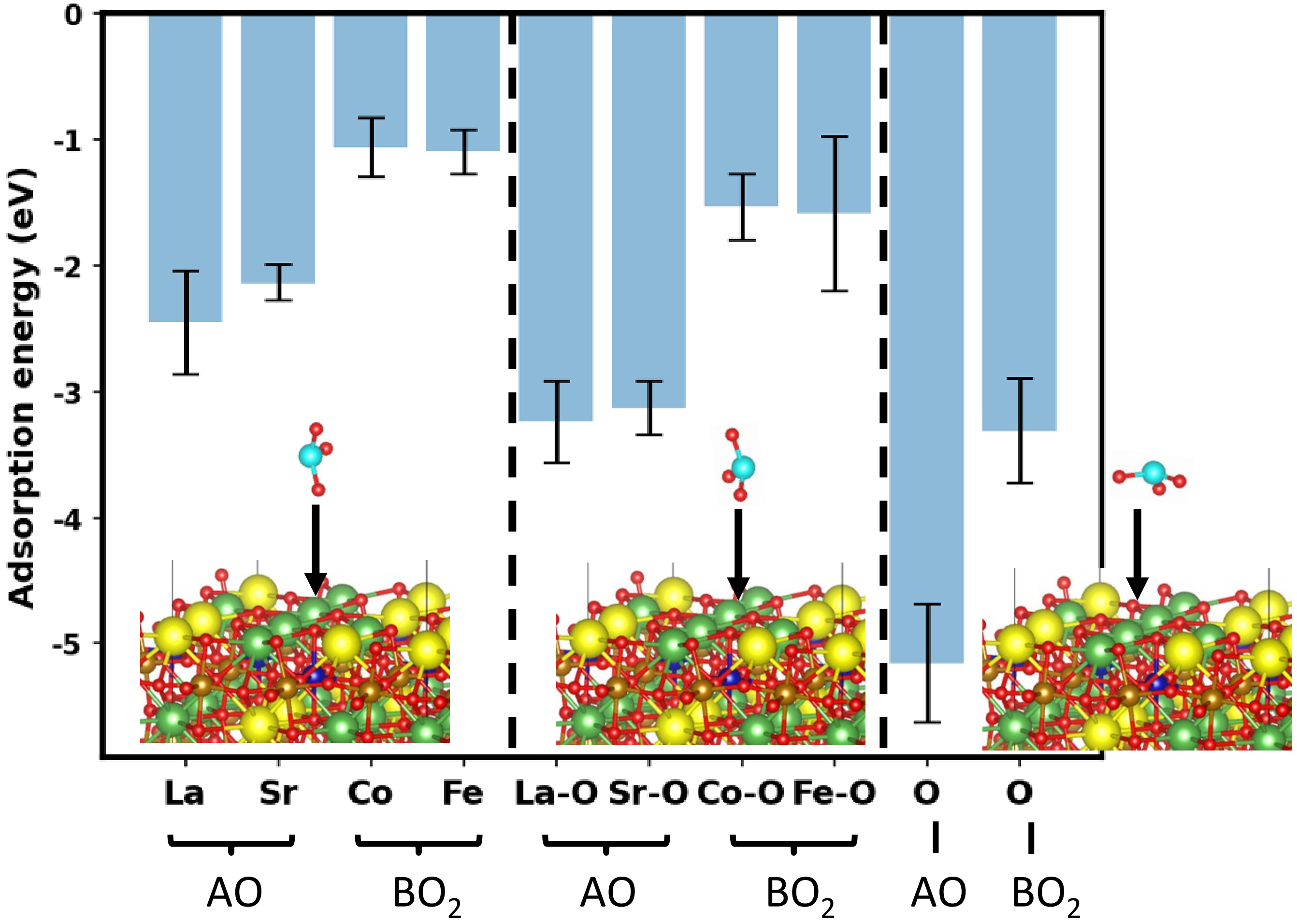
LSCF

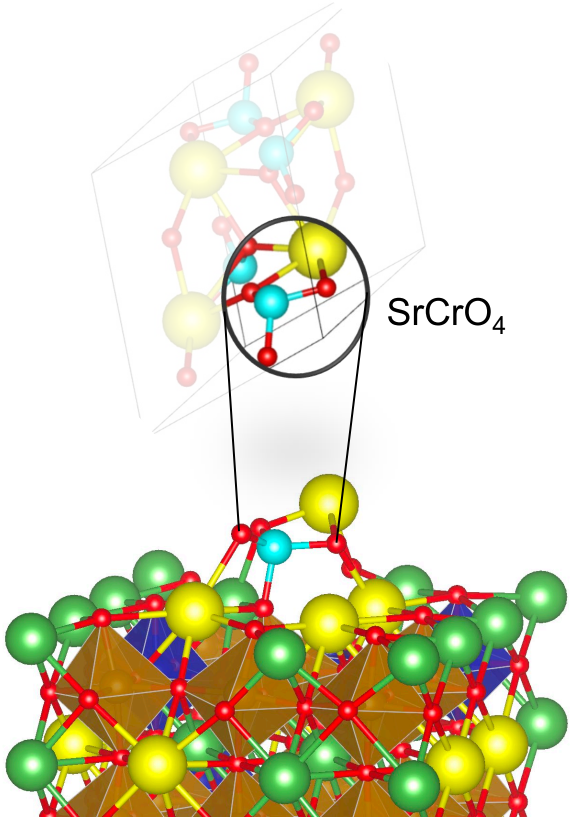
(001) AO/BO_2 w/ infiltrated cations

LSCF

● O
● Cr
● S

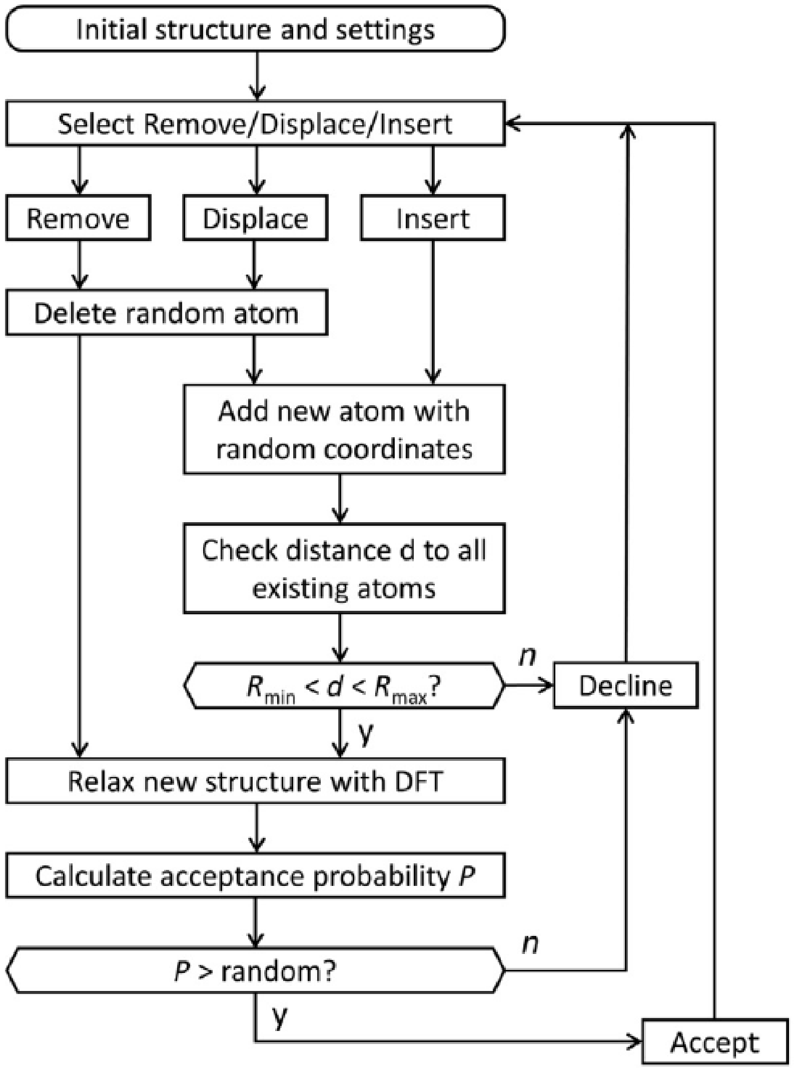
CrO_3/SO_2 favorable adsorption on LSCF lead to nucleation structures





- ○ CrO_3 adsorption energies are all negative for pristine LSCF slabs, whereas SO_2 is more positive
- ○ AO-terminated slabs have larger adsorption energies – more susceptible to poisoning
- ○ **Key descriptors – Bader charge of surface O atoms (electrostatic), oxygen p-band center in sub-surface (charge transfer)**
- ○ The protruded Sr surface exhibits notably lower adsorption energies and leads to nuclei structure

Grand Canonical Monte Carlo with IAP for resolving perovskite surface reconstructions (LSF example)

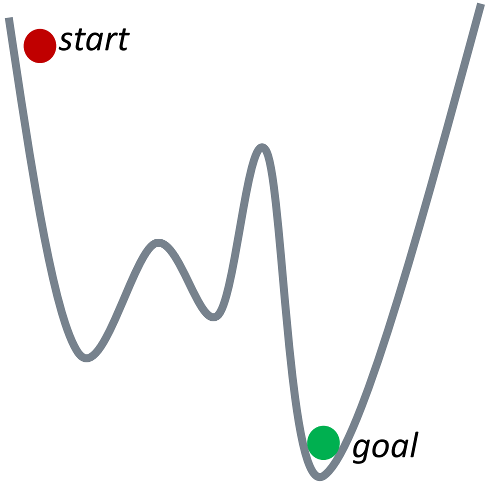

Initial structure and settings
Select Remove/Displace/Insert
Remove
Displace
Insert
Delete random atom
Add new atom with random coordinates
Check distance d to all existing atoms
$R_{min} < d < R_{max}$?
n
y
Decline
Relax new structure with DFT
Calculate acceptance probability P
$P >$ random?
n
y
Accept

- ☐ **Objective: To investigate $La_{0.6}Sr_{0.4}FeO_3$ (LSF) bulk and surface phase diagrams using realistic surface/interface atomic structures**
- ☐ Given the vast LSCF surface configuration space involved, we need an automated search for the minimum energy structure
- ☐ Grand canonical Monte Carlo (GCMC) is a physically motivated scheme to **identify the most stable surface oxide configuration at a given chemical potential and temperature.**

$$P_{\mathrm{move}} = \min\{1,\ e^{-\Delta U/k_B T}\}$$

$$P_{\mathrm{add}} = \min\left\{1,\ \frac{V}{(N+1)\Lambda^3} e^{-(\Delta U - \mu)/k_B T}\right\}$$

$$P_{\mathrm{remove}} = \min\left\{1,\ \frac{N\Lambda^3}{V} e^{-(\Delta U + \mu)/k_B T}\right\}$$

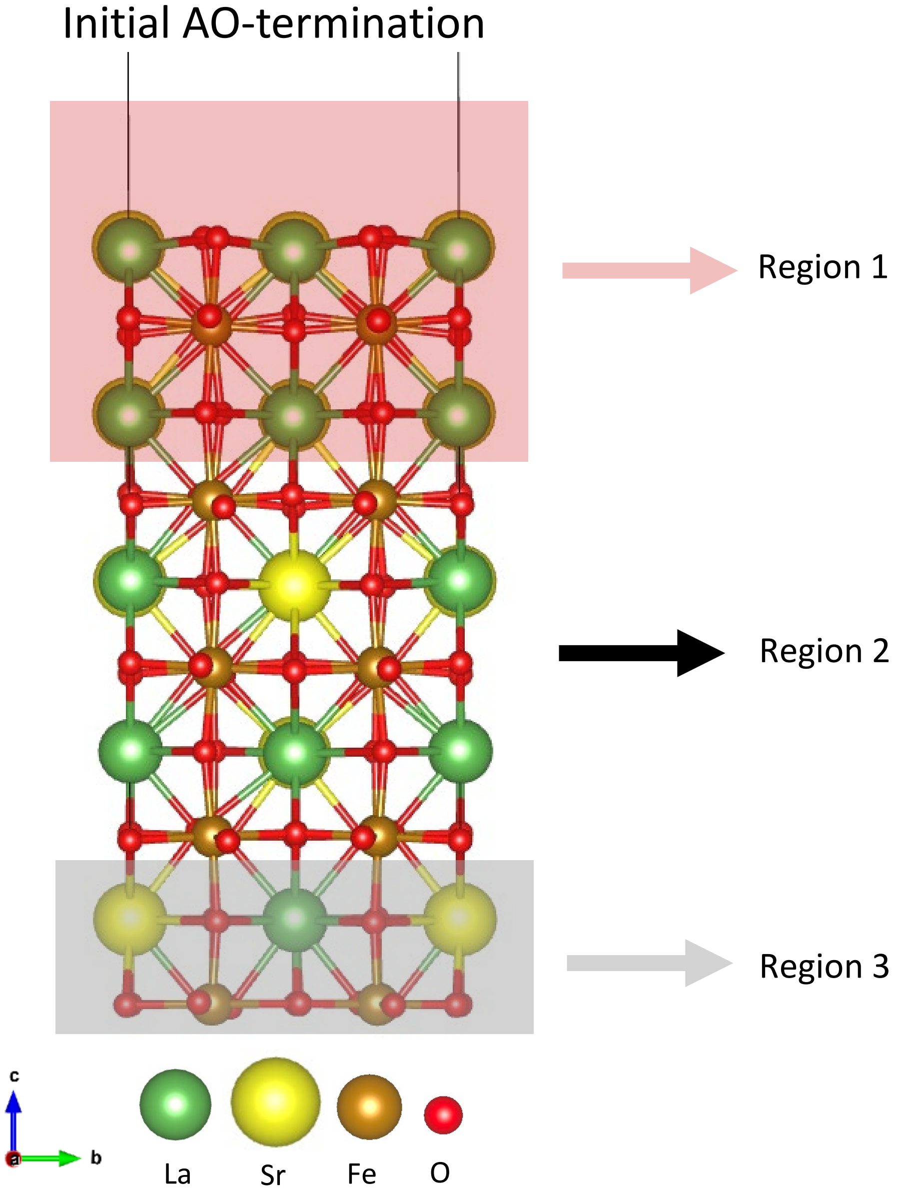


P. Wang and T. P. Senftle, AIChE Journal, 2021
V. Somjit and B. Yildiz, ACS Appl. Mater. Interfaces, 2022

LSF: GCMC setup to investigate surface atomic structures configurational space

☐ Objective: To investigate **$La_{0.6}Sr_{0.4}FeO_3$ (LSF)** bulk and surface phase diagrams using realistic surface/interface atomic structures

☐ Grand canonical Monte Carlo (GCMC) is a physically motivated scheme to **identify the most stable surface oxide configuration at a given chemical potential and temperature.**



- Region 1:
  - Full GCMC on oxygen and A/B sites
  - MC exchange on A-site
- Region 2:
  - Full GCMC on oxygen
- Region 3:
  - Fix atoms in positions

What phases might be on the surface: competing compound phases when LSF is stable at 800 C

LSF is stable: $\sum_i c_i E^i_{product} > E_{LSF}$

$$\mu_O(T, P_{O_2}) = \frac{1}{2}\left(E^{IAP}_{O_2} + \mu^0_{O_2}(T, P^0) + k_B T ln\left(\frac{P_{O_2}}{P^0}\right)\right)$$

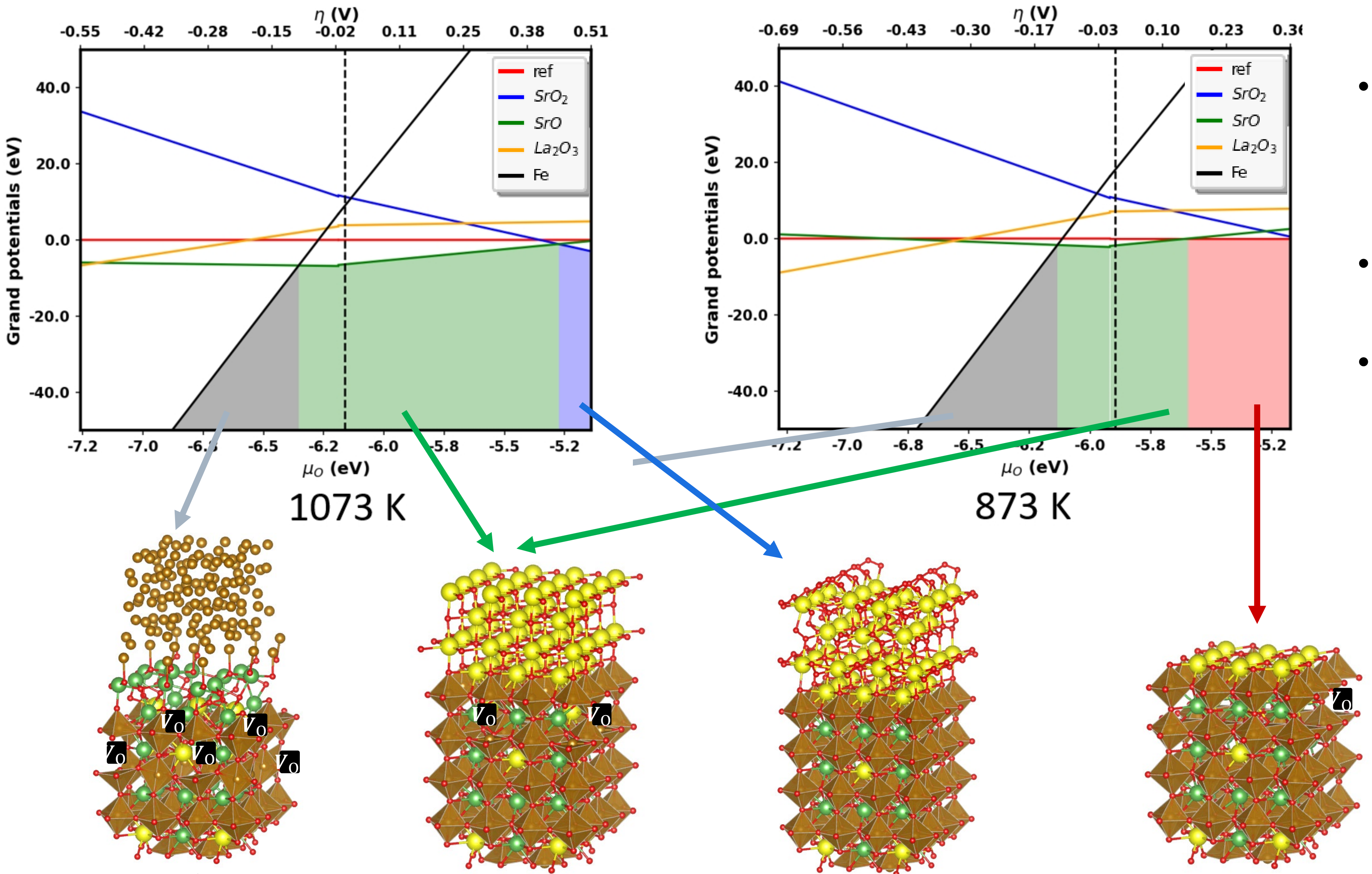
Competing phases: $\sum_j s^{i,j}_{product}\mu_i > E^i_{product}$

$$P_{O_2} = P_{O_{2,gas}} exp\left(\eta\frac{4F}{RT}\right)$$

μ_O range (eV)	P_{O2} range (atm)	η range (V)	Competing phases
$-5.23 < \mu_O < -5.14$	$10^{8.07} < P_{O2} < 10^{8.83}$	$0.47 < \eta < 0.51$	SrO_2, LFO
$-5.43 < \mu_O < -5.23$	$10^{6.18} < P_{O2} < 10^{8.07}$	$0.37 < \eta < 0.47$	SrO, LFO
$-7.26 < \mu_O < -5.43$	$10^{-11.02} < P_{O2} < 10^{6.18}$	$-0.55 < \eta < 0.37$	SrO, La_2O_3, Fe

- Stable competing phases can inform interfacial structures on LSF perovskite surface surfaces

Surface phase diagram shows different segregation regimes



Fe exsolution

SrO segregation

SrO_2 segregation

Reference perovskite SrO-termination

- The dominant regime for SrO segregation is around ambient pressure, more dominant in SOEC region
- Sr segregation more dominant in anodic condition
- The LSF phase becomes more stable at lower temperature

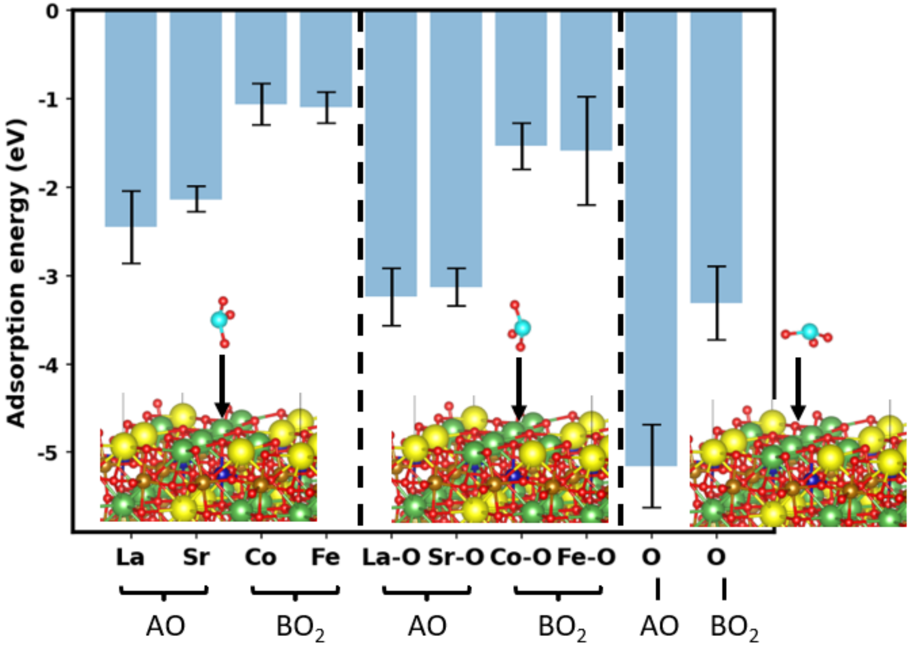
Conclusion and Future work

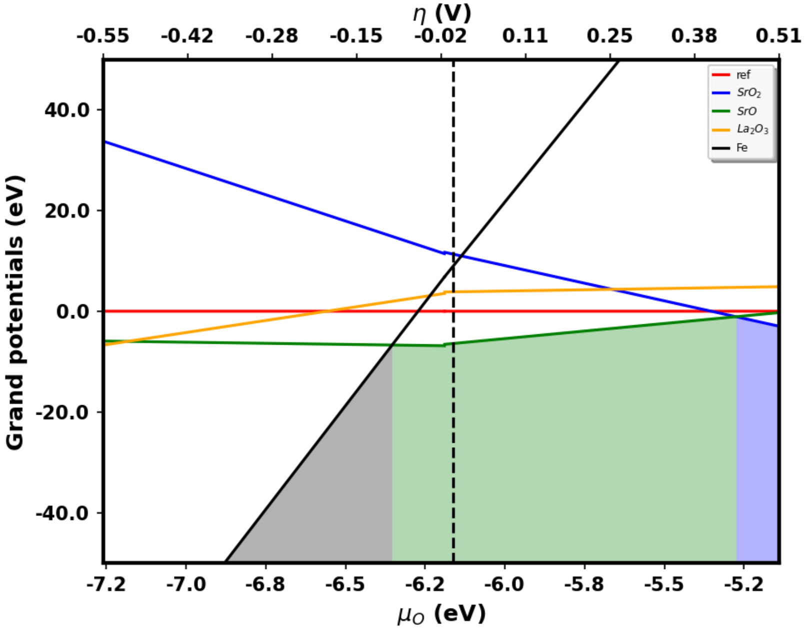
Conclusion

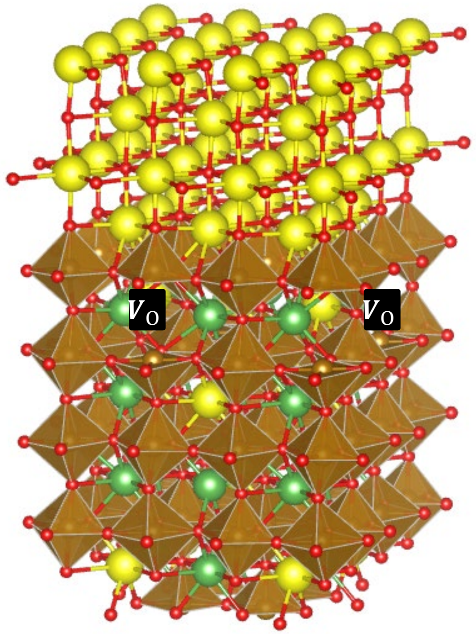
- AO-terminated surfaces more susceptible to CrO_3 and SO_2 adsorption
- Derived thermodynamic surface phase diagram refined by GCMC atomic structures
- The dominant regime for SrO segregation is around ambient pressure, more dominant in SOEC region
- The LSF phase becomes more stable at lower temperature

Next step

- Introduce infiltrates
  - Investigate structure and electronic property change when introduce infiltrates

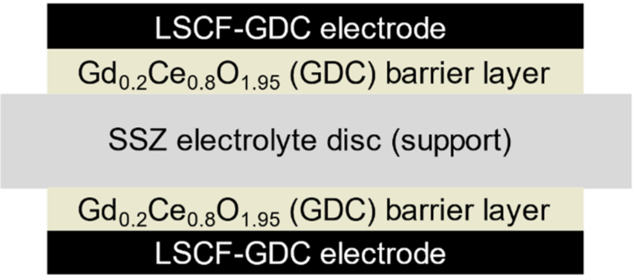






Surface modification of LSCF to suppress Sr

Our current system:



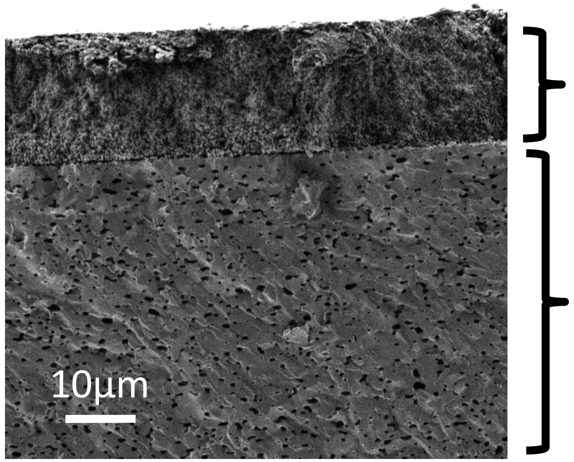
LSCF: $(La_{0.6}Sr_{0.4})_{0.95}Co_{0.2}Fe_{0.8}O_3$

Cells fabricated at PNNL with OxEon electrolytes
Cells fabricated at OxEon

Well-established button cell production process:

- Screen printing
- Sintering

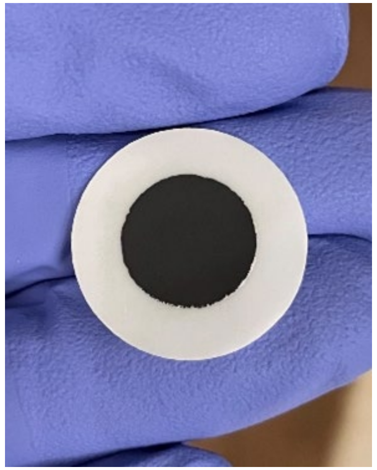
Microstructure



LSCF/GDC composite on GDC barrier layer

ScSZ electrolyte

Our approach controls surface chemistry through a physically-based and practical single-step infiltration:



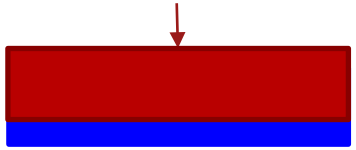
As-prepared LSCF

Infiltration

Aqueous solutions of metal chlorides.

Metals: Zr, Hf, Ti, Nb, Al
Infiltration parameters:
Reagent concentration, metal loading, surfactants.

Metal solution



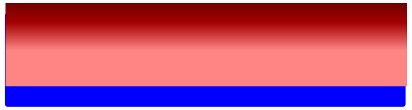
Infiltrated LSCF

Annealing

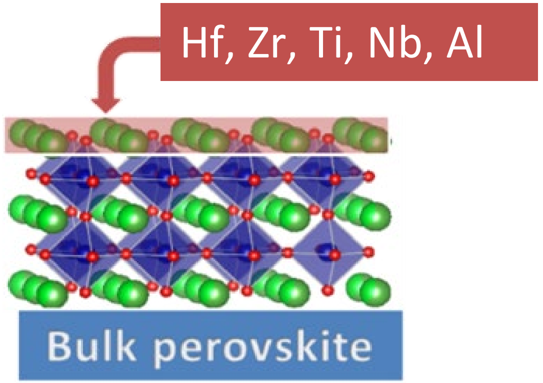
Anchoring of metal oxides on/into the LSCF surface

Annealing conditions:
Temperature, potential, gas.

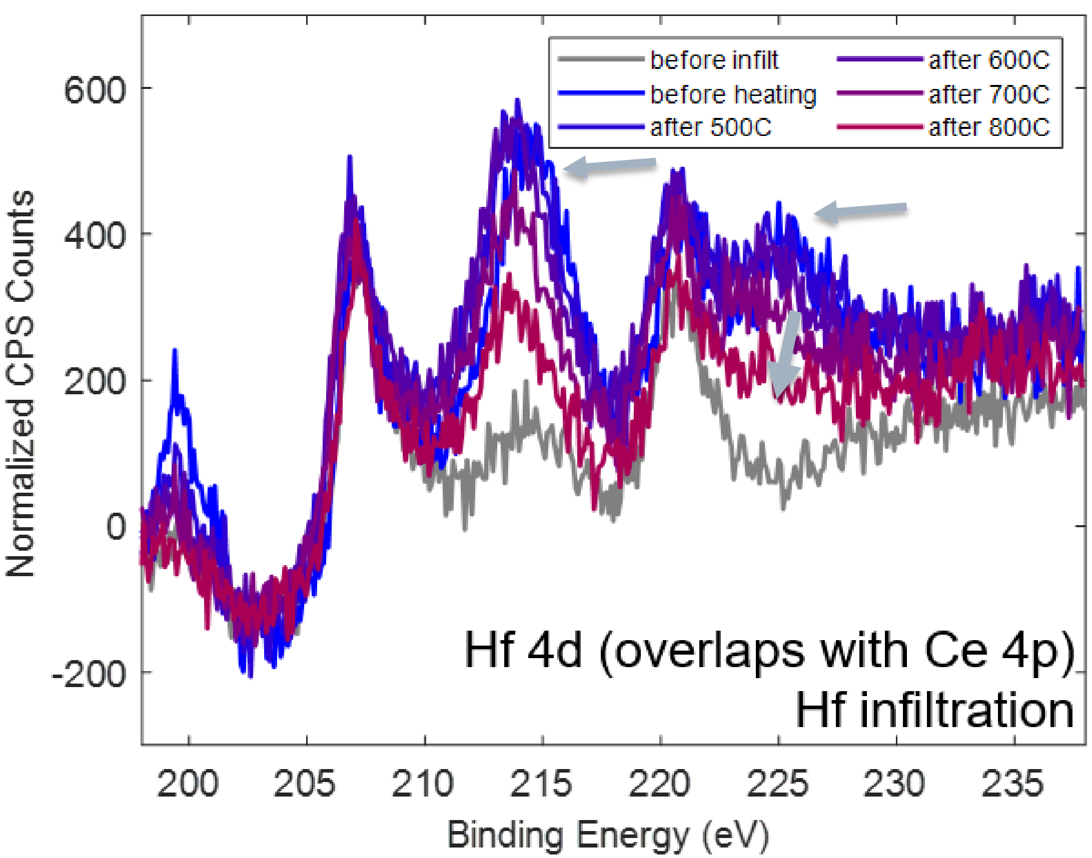
Near-surface doping

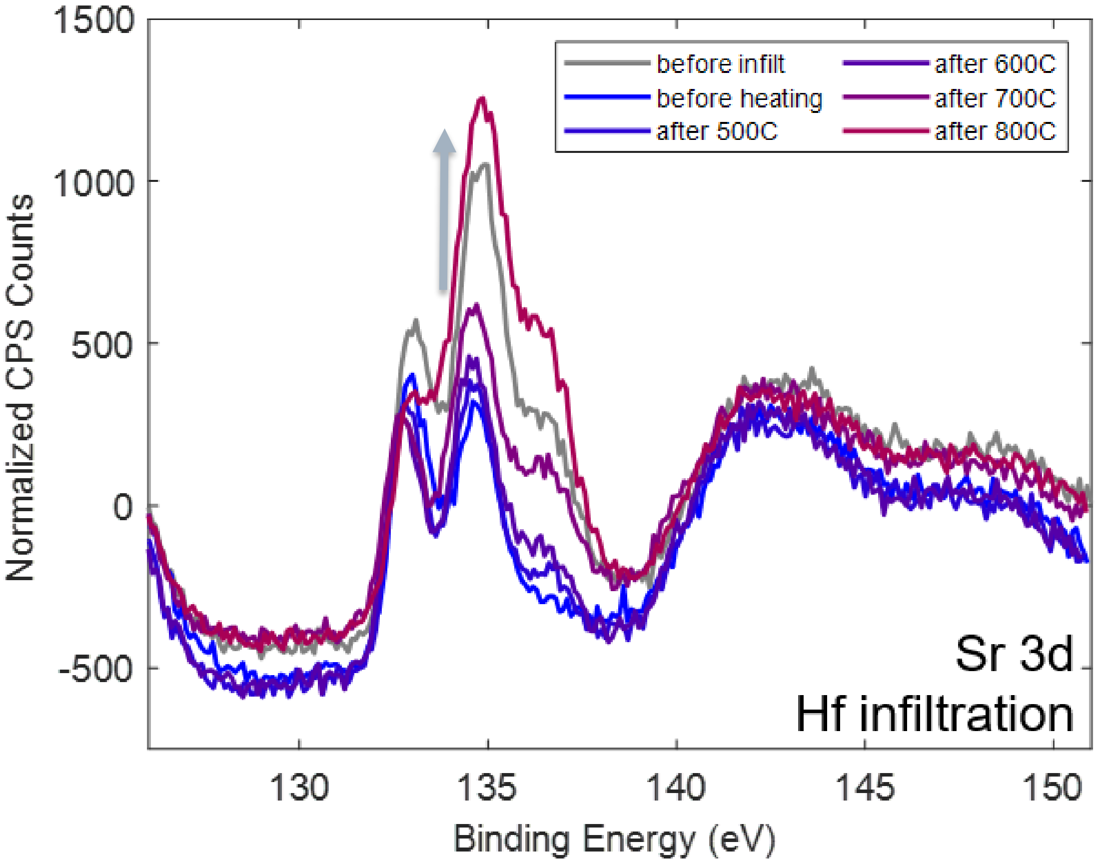


Surface-modified LSCF



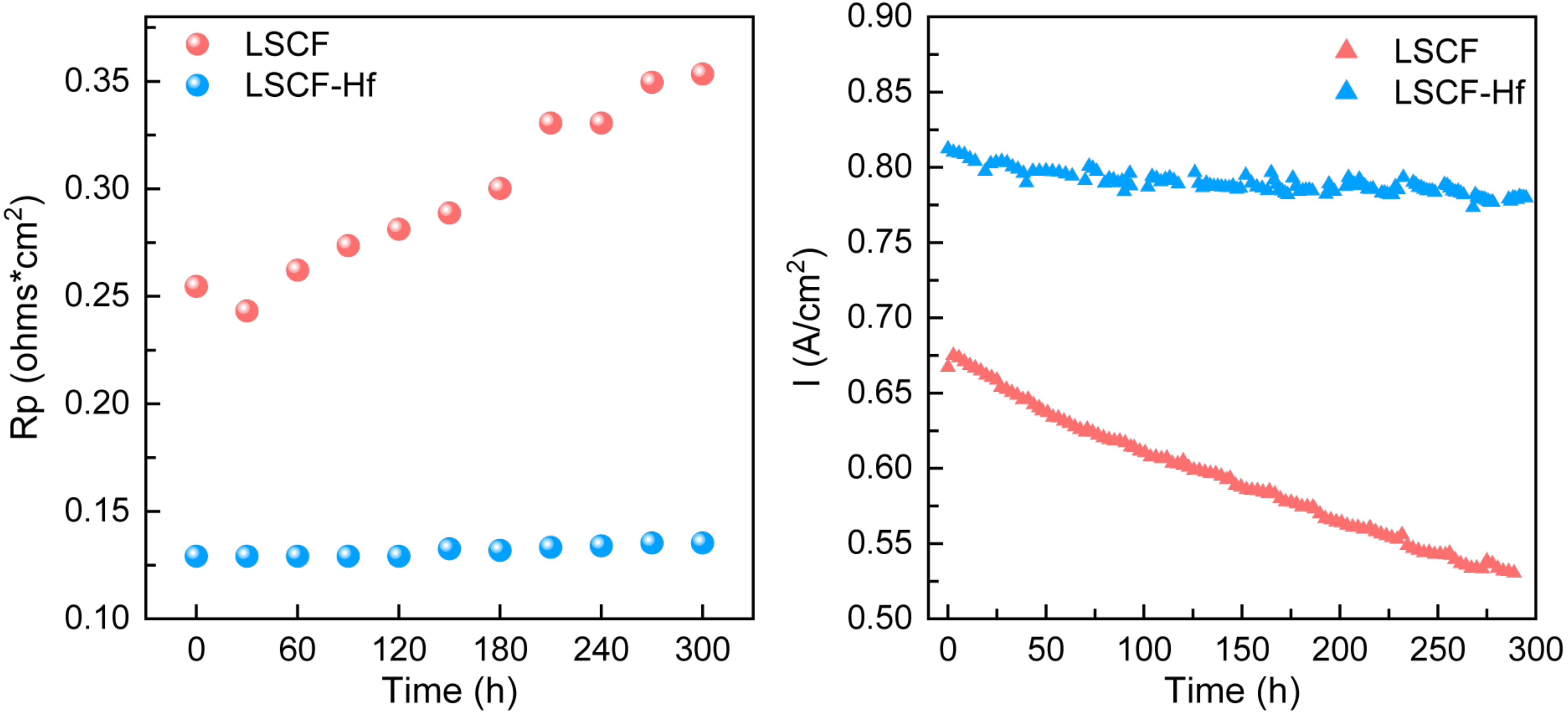
Previously: Hf and Zr dissolution at T > 700°C





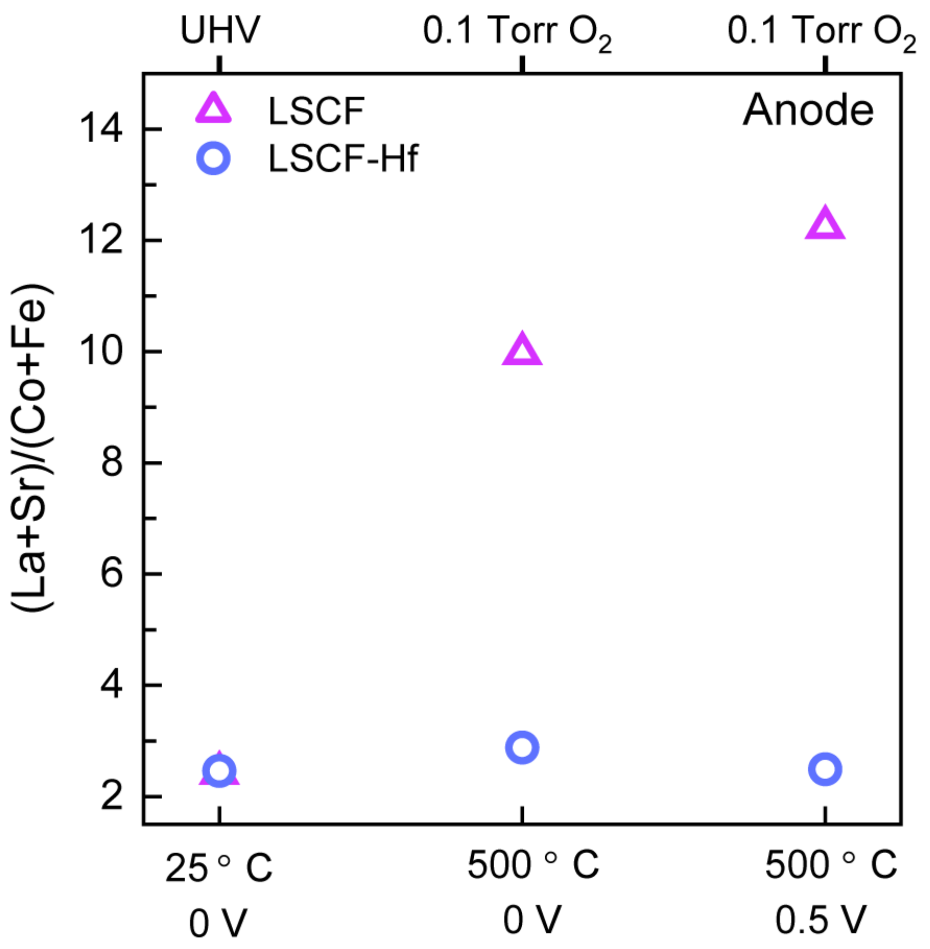
- ☐ Considerable Hf cation diffusion away from the surface is noticeable > 700℃.
- ☐ As Hf diffuses away from the electrode surface, Sr segregates more strongly to the surface

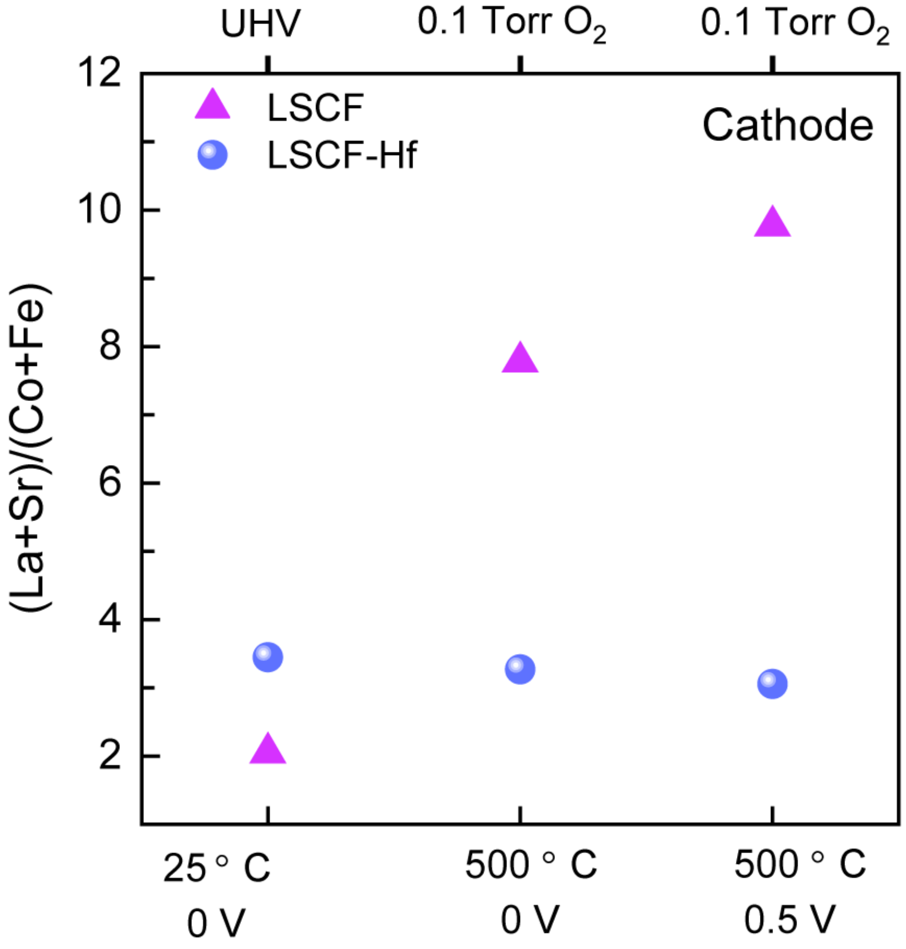
Improvement in electrochemical stability and activity with Hf infiltration in LSCF porous electrode (of OxEon)



Samples made by OxEon (Tyler Hafen and Elango Elangovan)

Improvement in chemical stability with Hf infiltration in LSCF porous electrodes (OxEon)





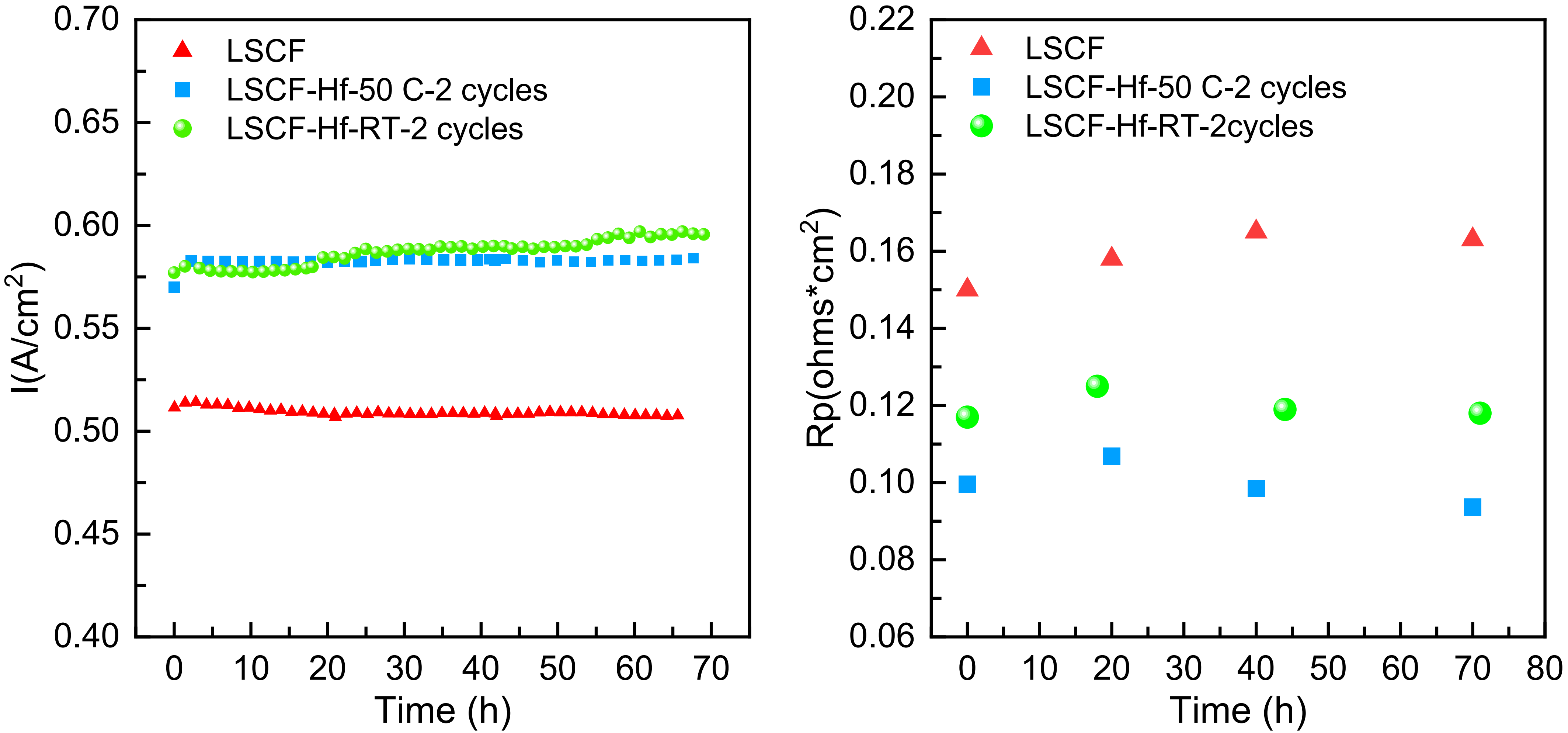
- Near ambient pressure XPS measured while polarizing the cells
- Performed at NSLS-II synchrotron of Brookhaven National Laboratory (beamline 23-ID-2).

Hf penetration depth with different parameters, symmetric cells of LSCF-GDC (PNNL)

Parameters	Temperature	Hf/(A+B)
LSCF-GDC	RT	0.046
LSCF-GDC-sub surface	RT	0.053
LSCF-GDC	50 C	0.082
LSCF-GDC-sub surface	50 C	0.14
LSCF-GDC-sub surface (close to electrolyte)	50 C	0.075

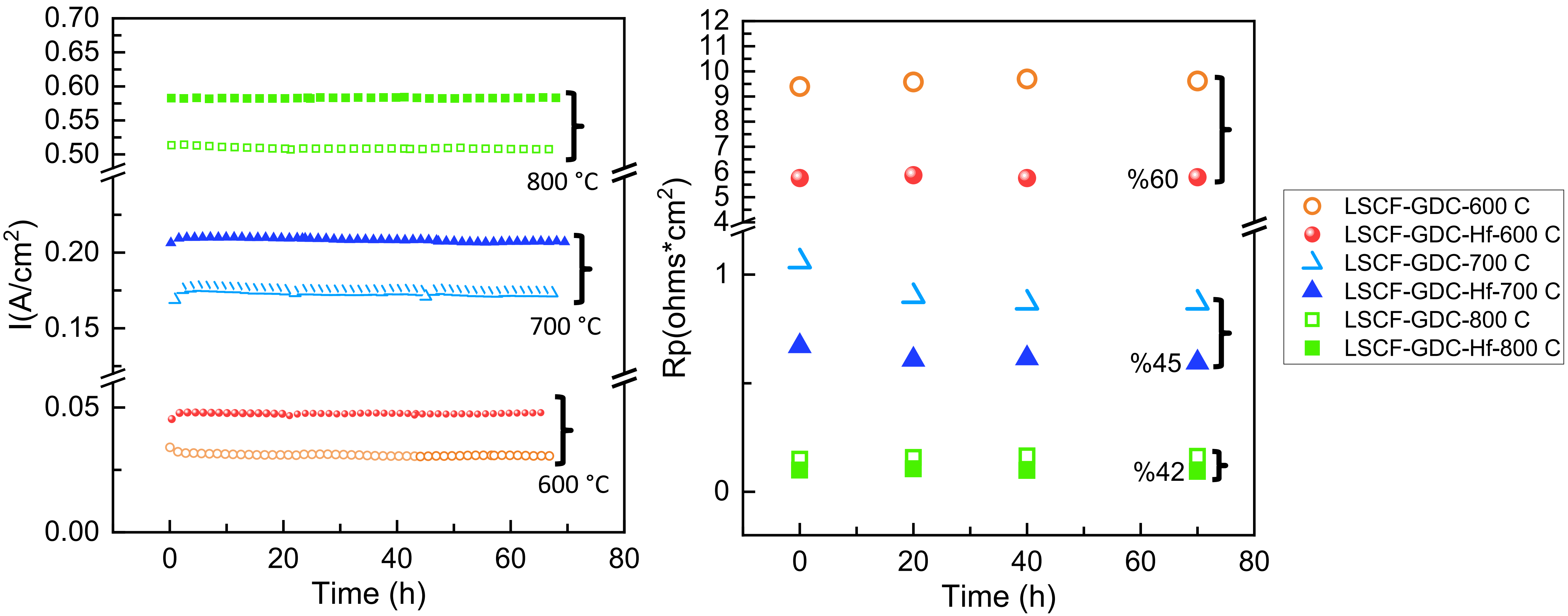
- XPS analysis shows Hf on the surface and sub-surface of LSCF-GDC
- XPS analysis on LSCF-GDC-subsurface was done after scratching the surface at different depths
- Increasing the temperature of Hf solution results in more penetration of Hf
- Sub-surface is achieved by scratching the surface and is not quantitative

Improvement of electrochemical performance with Hf infiltration



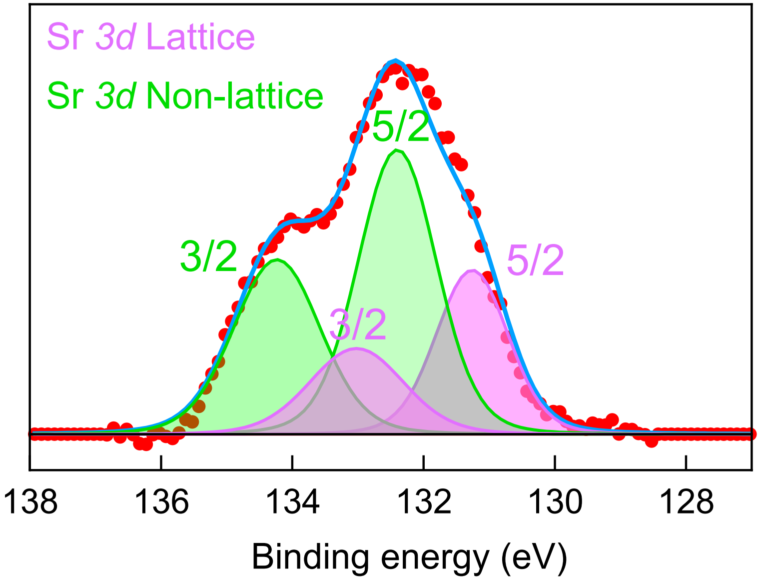
- EIS measurement carried out at 800 °C for symmetric cells of LSCF-GDC

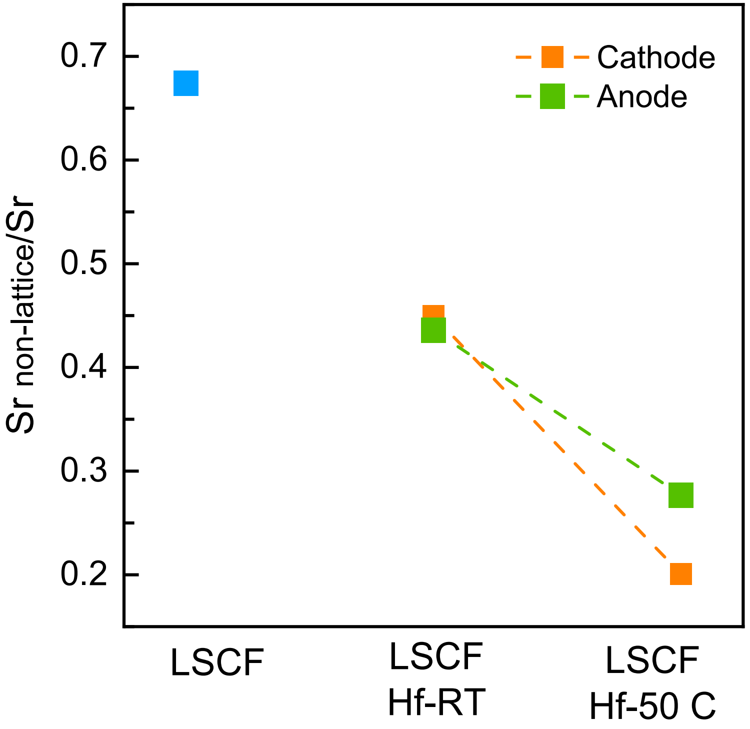
Improvement of electrochemical performance with Hf infiltration

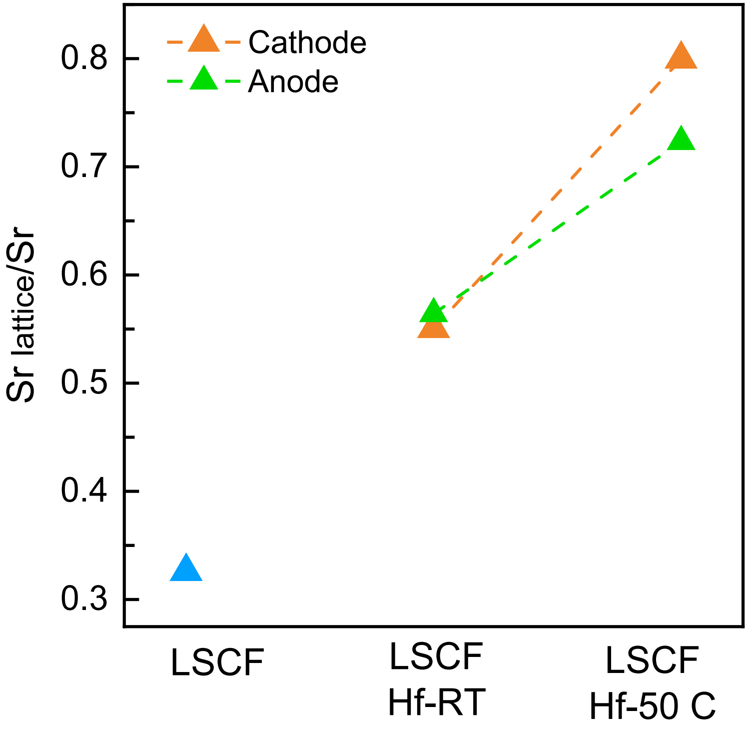


- EIS measurement carried out at different temperatures for symmetric cells of LSCF-GDC

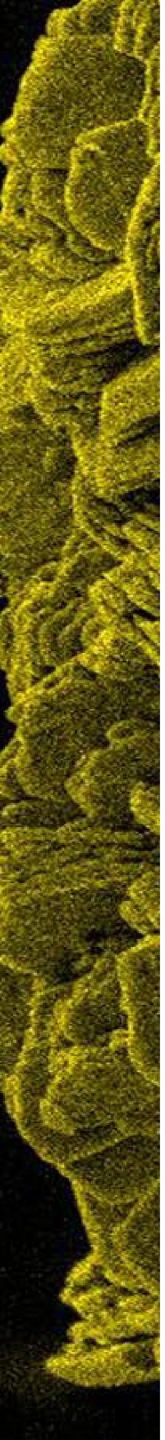
XPS analysis shows Hf infiltration reduces Sr segregation on LSCF-GDC







- XPS measurement carried out after quenching each cell from 800 C to room temperature
- XPS analysis shows that Hf infiltration reduces Sr segregation



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Update on Hafnium Infiltration

R. Springer, S. Karki, J. Escobar,
L. Seymour, O.A. Marina

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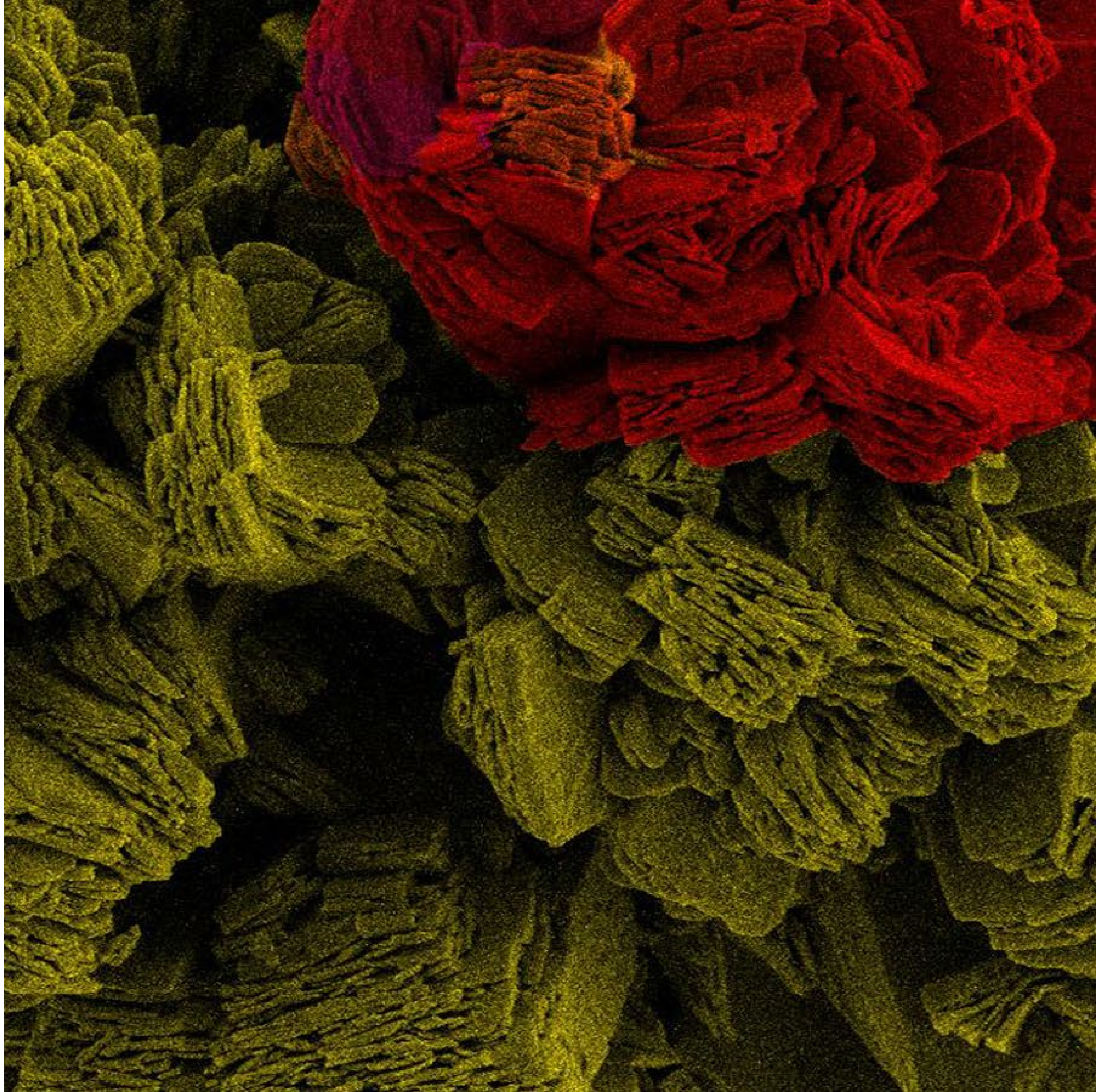
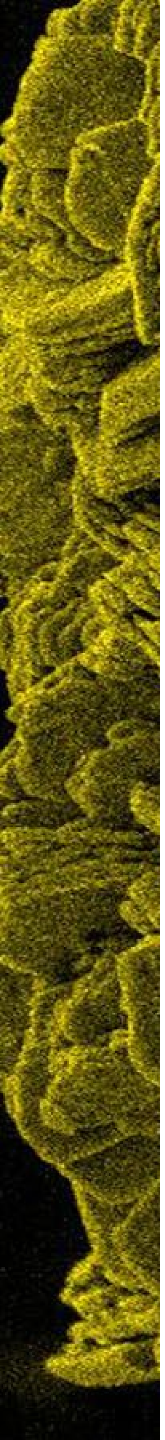
ENERGY
BATTELLE

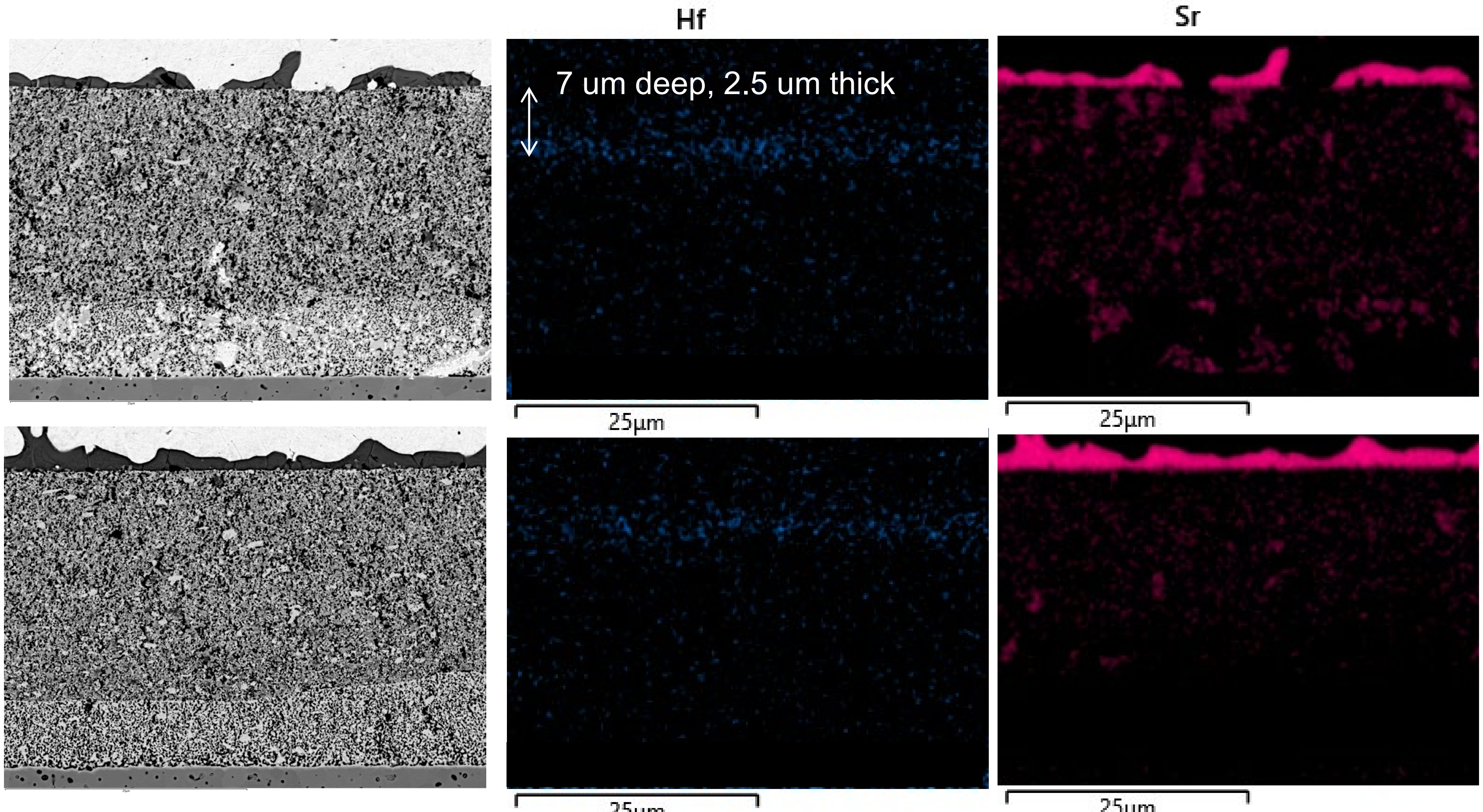


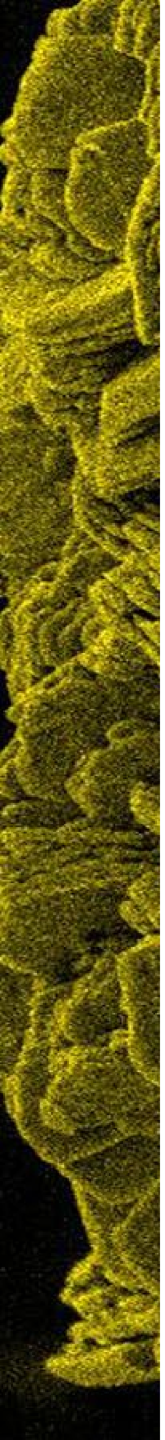
Hf Infiltration per Original MIT Recipe (0.08M): Hf was observed as a 2.5 μm band inside LSCF-GDC, 7 μm from the surface



SOEC: Hf infiltrated

SOFC: Hf infiltrated



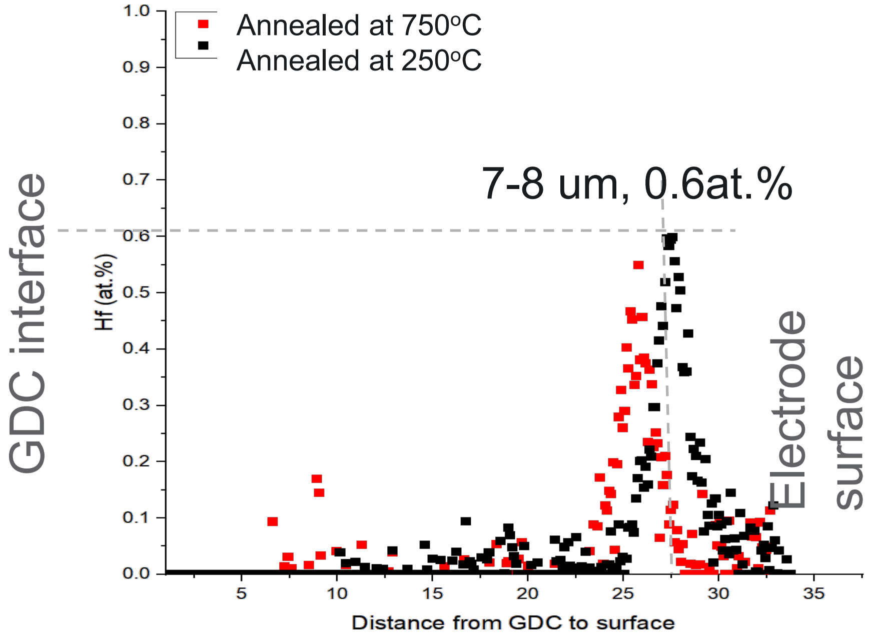


A slight increase in both concentration and depth of Hf for a fully submerged cell vs the dropper approach

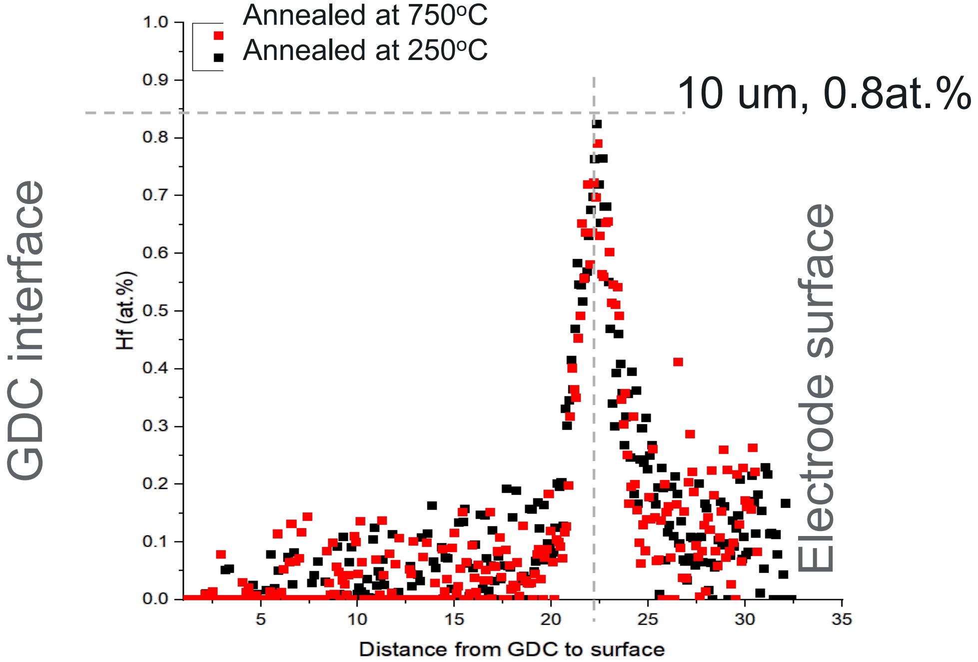
Modified Infiltration Process

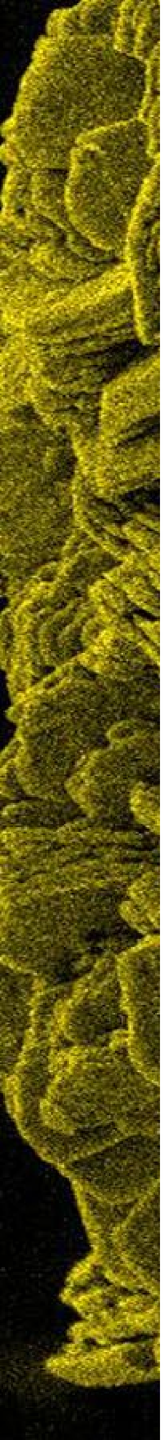
- Doubled $HfCl_4$ solution concentration and warmed it to 50°C
- Increased infiltration duration to 30 mins
- No rinsing after infiltration
- Infiltrated LSCF-GDC electrodes as follows:
  - 1 cell was fully submerged in solution
  - 1 cell had O_2 electrode exposed to solution via dropper method
  - Both cells were annealed at 250°C for 25 min
  - Cells were split in half, and each half was annealed at 750°C for 3 days (72 hr)

Dropper Approach

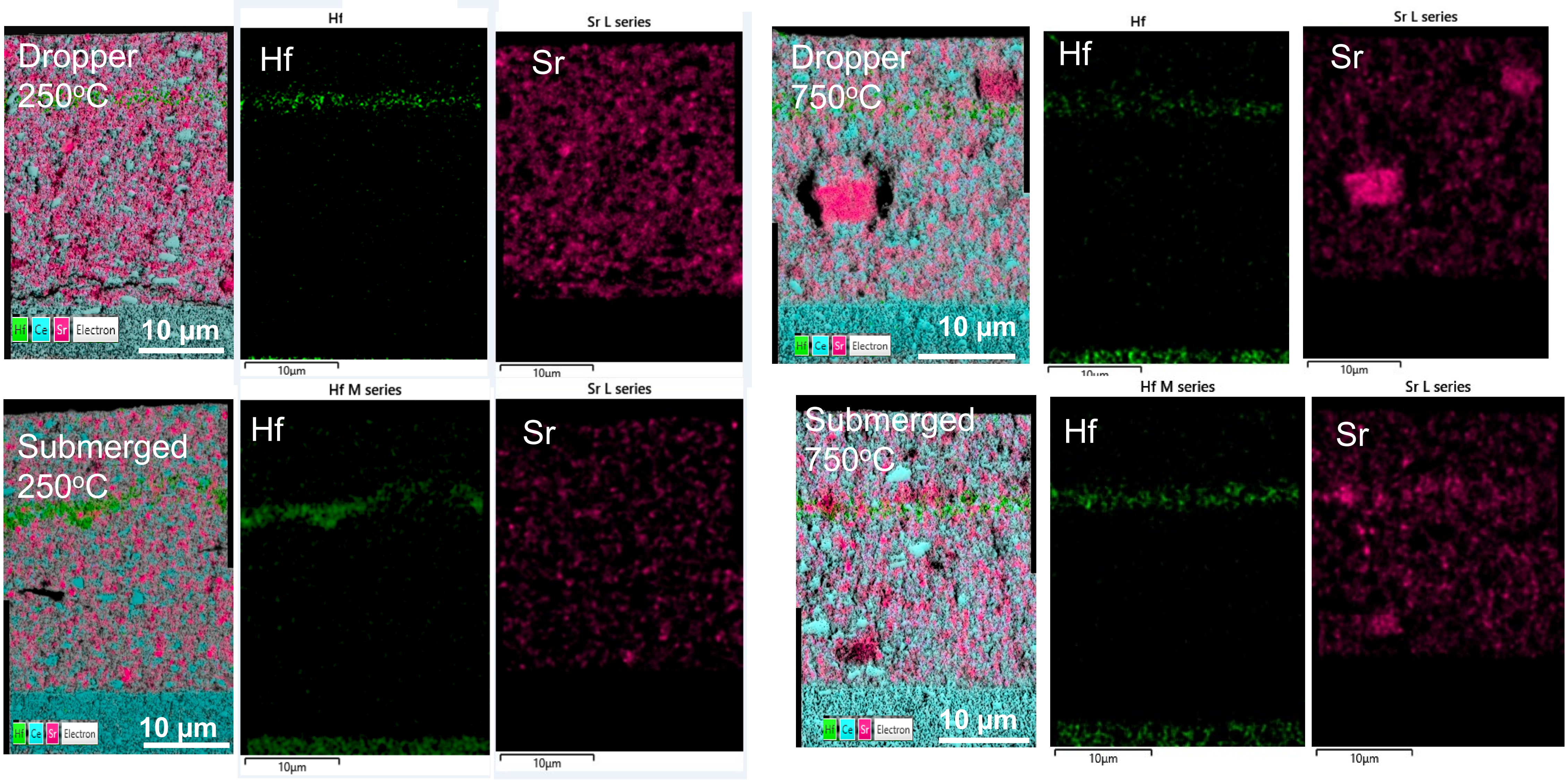


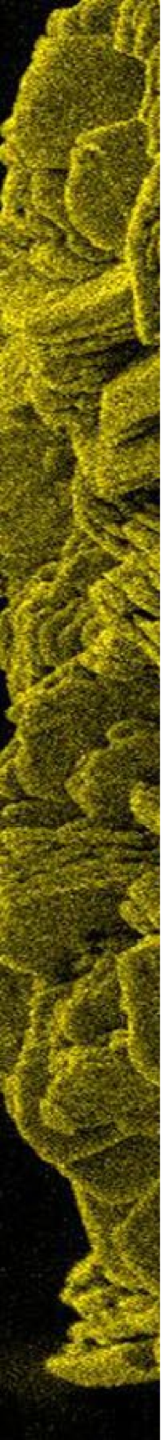
Fully Submerged





Hafnium is present as a thin band beneath the electrode surface; No correlation between Sr and Hf





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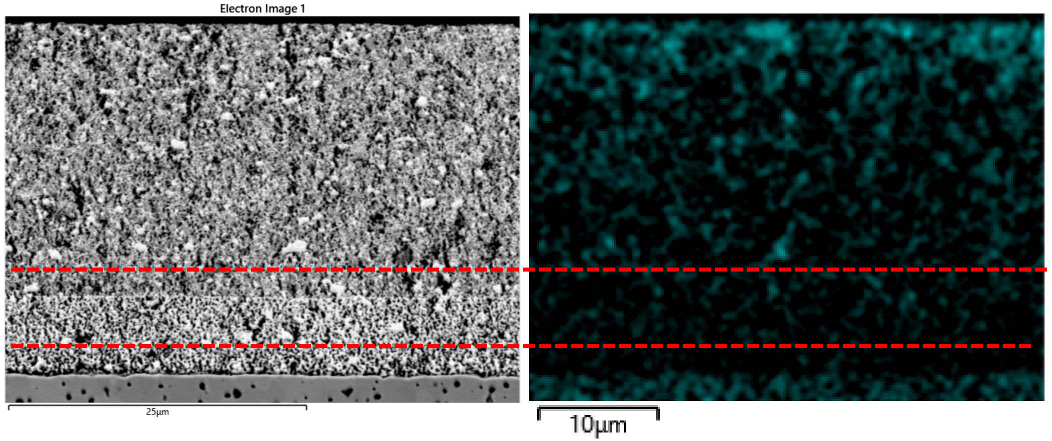
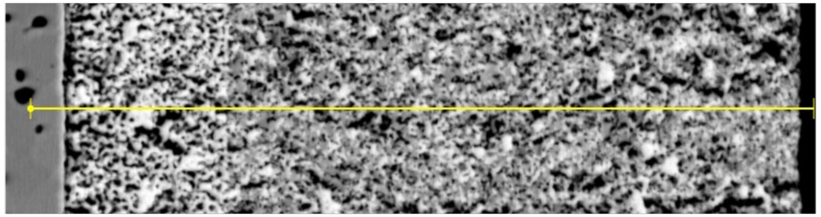
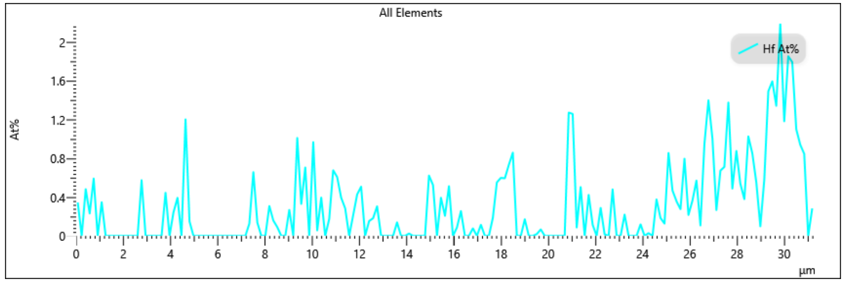
SEM/EDS Analysis of Hf-Infiltrated LSCF-GDC

Method 2: $HfCl_4$

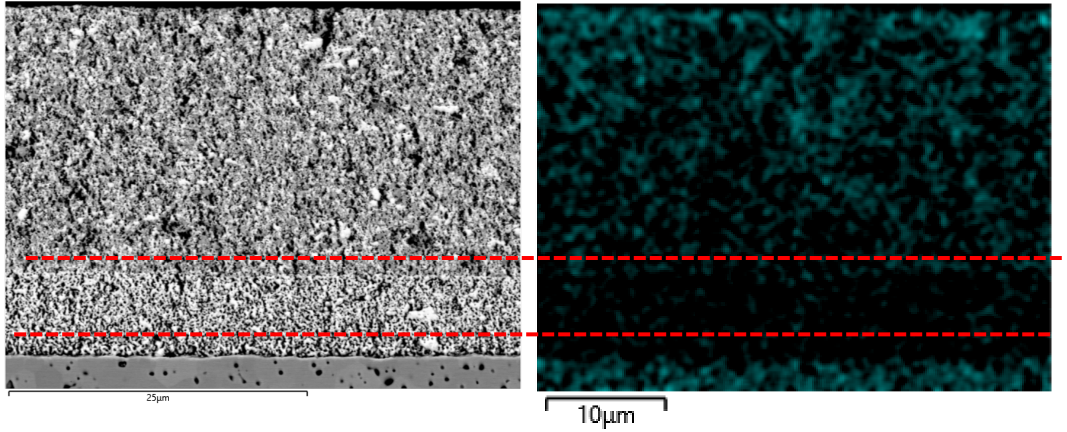


Line profile

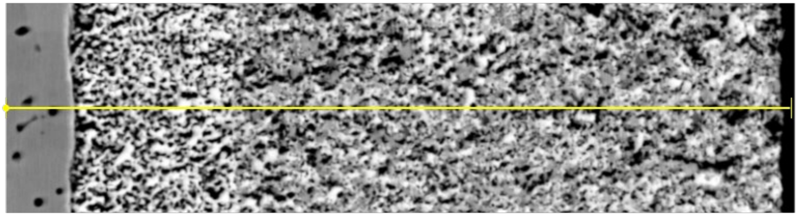


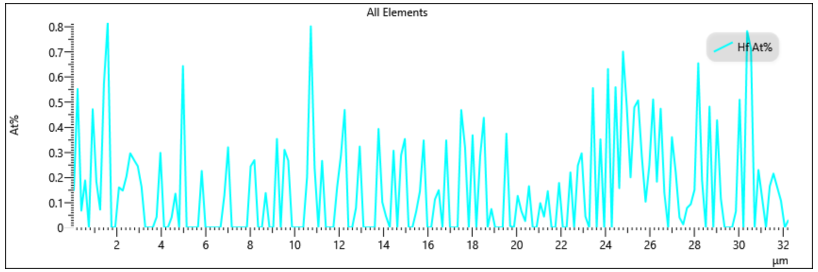


Method 3: $HfCl_4$+CA

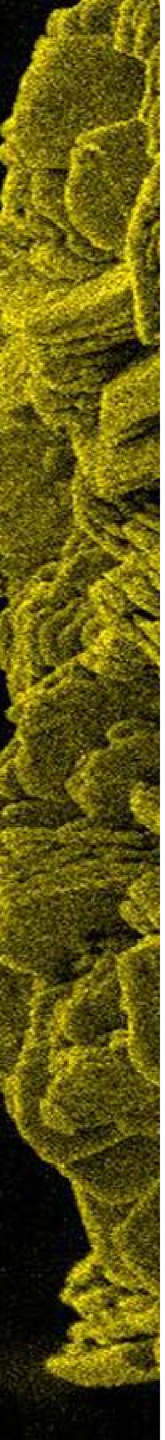


Line profile



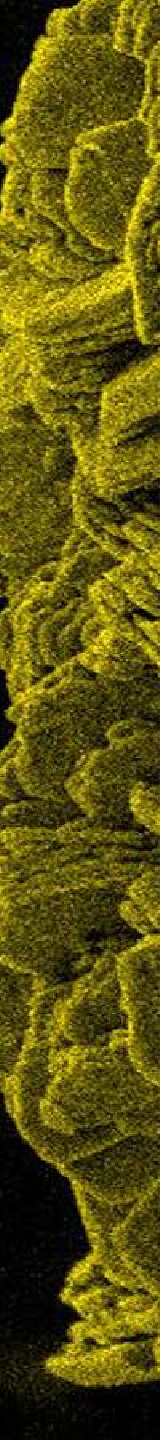


- Method 2: Hf is more concentrated at surface (~2 at.%) then decreasing to ~0.5 at.%
- Method 3: Hf is distributed more uniformly, ~0.5 to 0.8 at. %

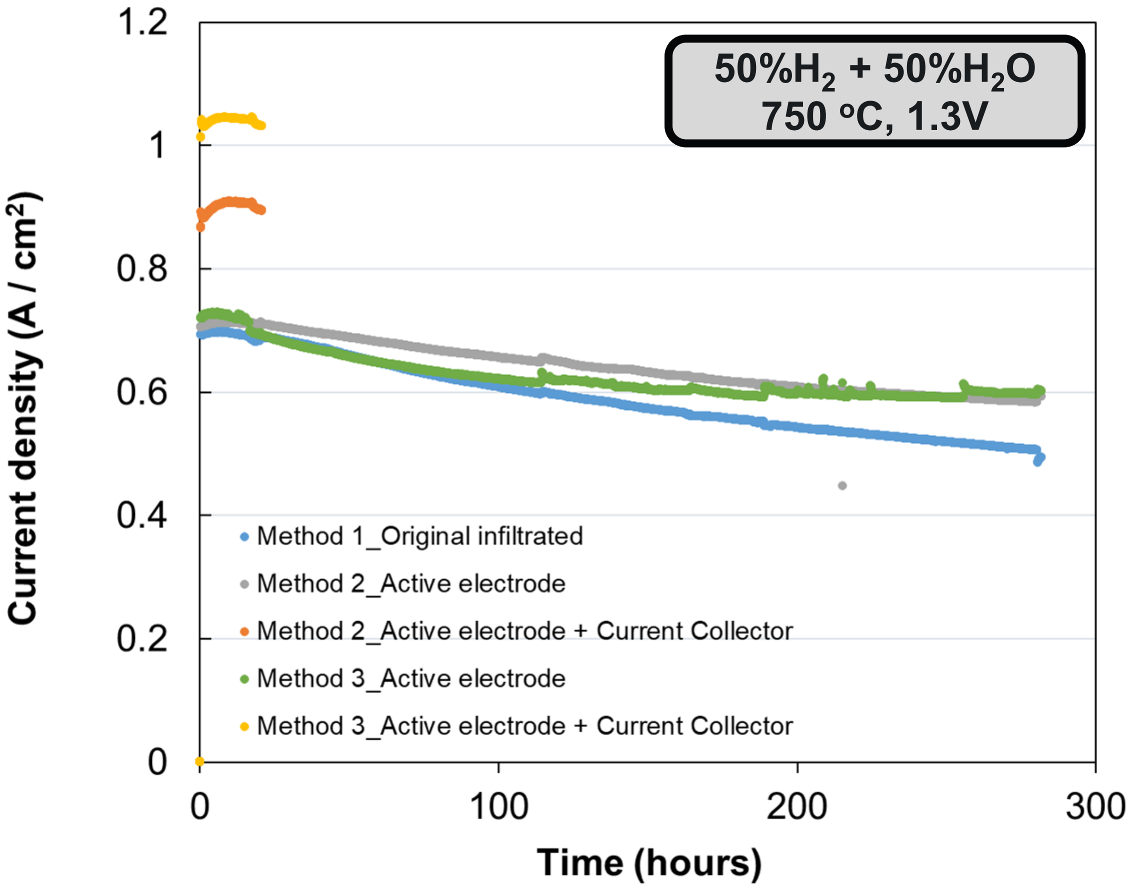


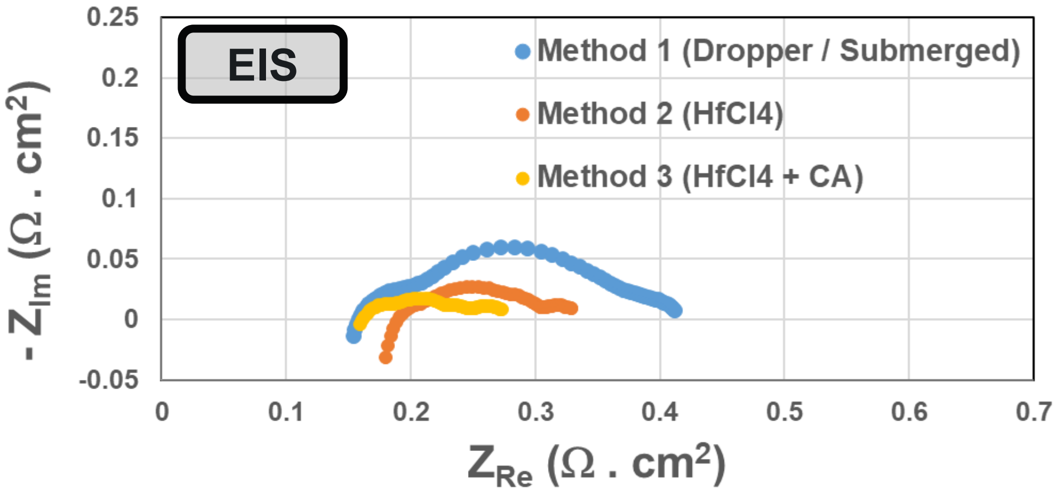
Additionally Modified Infiltration Process

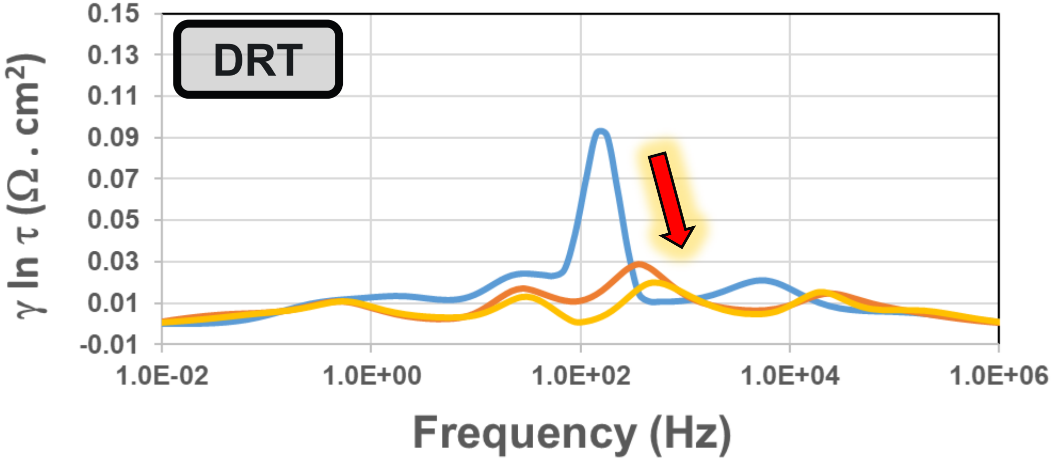
- 9 full Ni-YSZ electrode-supported cell made:
  - Method 2: 4 cells with 0.5M $HfCl_4$
  - Method 3: 4 cells with 0.5M $HfCl_4$ + CA
  - In 4 cell, only LSCF-GDC was infiltrated; LSC contact was applied on the top
  - In 4 cell, both LSCF-GDC electrode and LSC contact was co-infiltrated
  - 1 cell is a reference cell, without Hf

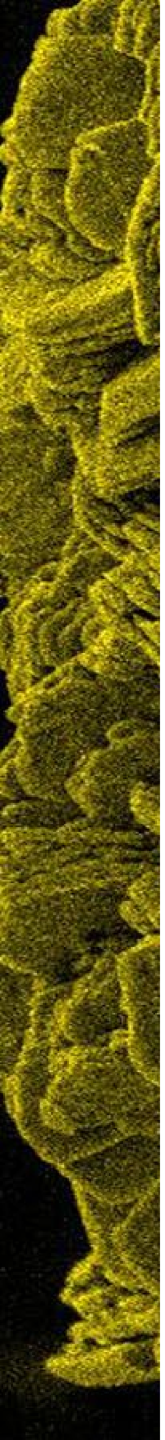


Modified Infiltration Strategy Brings Positive Impacts on Cell Performance



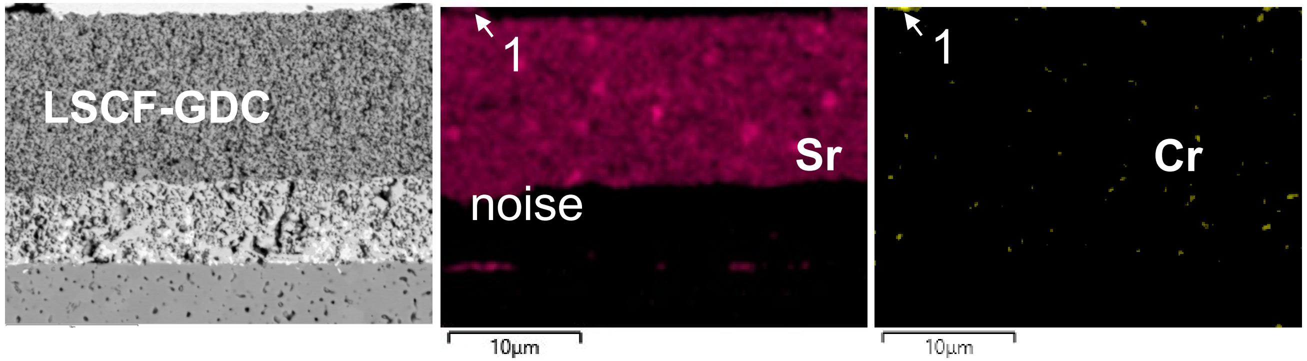






Pressurized SOEC Operation: 200 hrs at 90 psi 750°C at 1.3 V in 50%H_2O-50%H_2

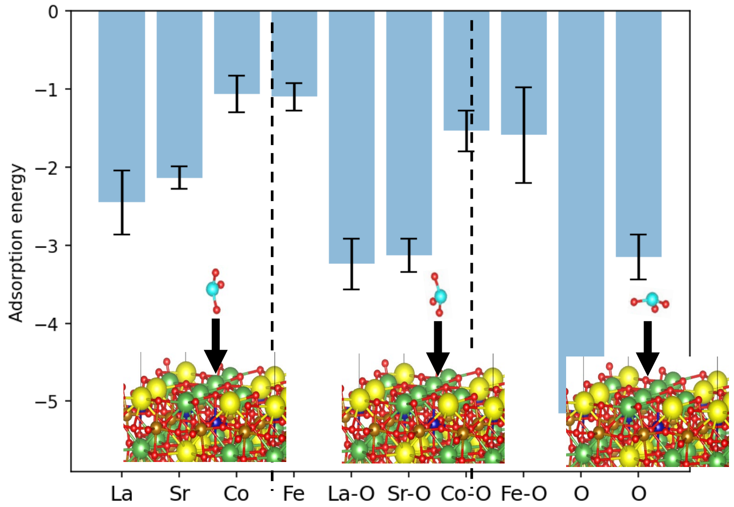
- Cr was not added intentionally, but could come from metal tubing
- LSCF-GDC was not infiltrated with Hf (baseline)
- Cr was noted on the LSCF surface, but not in the LSCF electrode

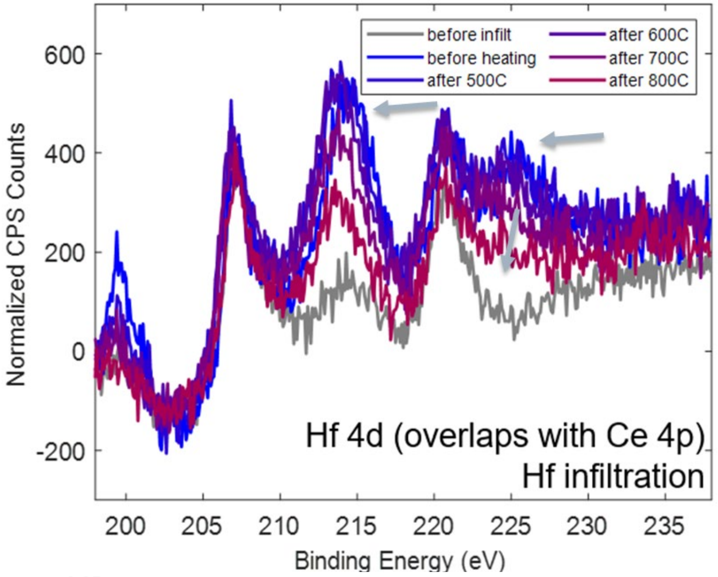


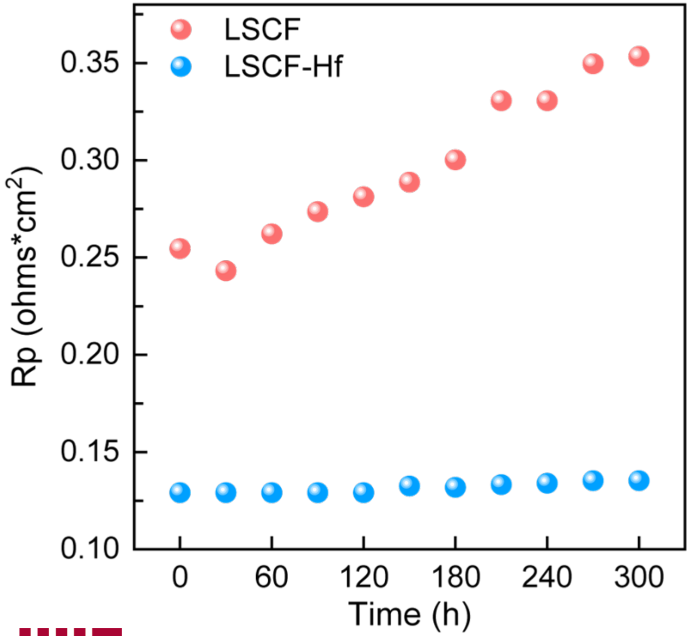
At. %	O	Cr	Sr
1	59.03	23.59	17.39

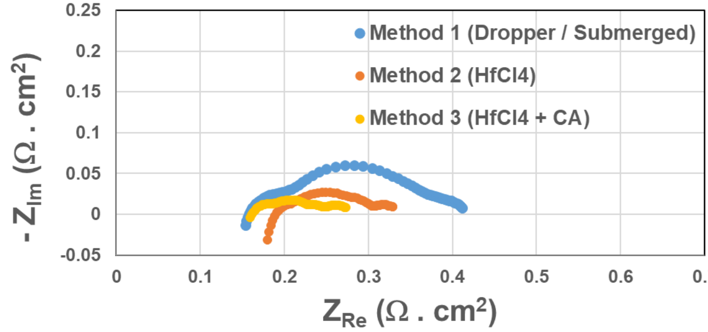
- $SrCrO_4$ noted at the LSCF surface
- No Cr in LSCF electrode

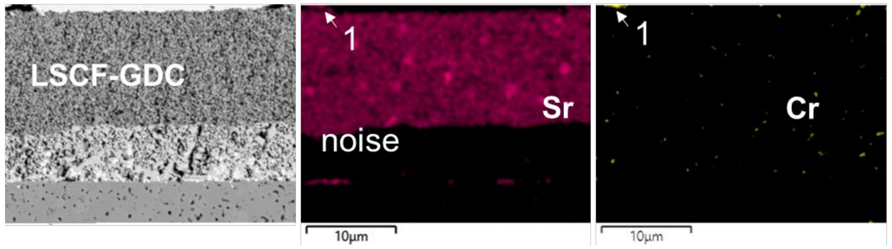
Summary and future work











Till now:

☐ AO-terminated surfaces more susceptible to CrO_3 and SO_2 adsorption.

☐ SrO layer/multilayer formation at surfaces possible, depending on oxygen chemical potential.

☐ LSCF surface chemistry gets more stable with Hf infiltration.

☐ Optimized Hf infiltration approach leads to enhanced electrochemical performance.

Next Steps:

☐ Testing of performance degradation in S environment after Hf treatment.

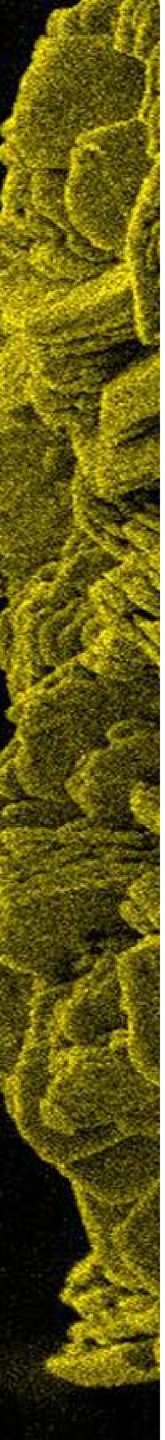
Acknowledgements

- This material is based upon work performed by the Massachusetts Institute of Technology and the Pacific Northwest National Laboratory, award DE FE0032102 supported by the Department of Energy’s Solid Oxide Fuel Cells program.

- We thank Sarah Michalik and Debalina Dasgupta for supporting this project.

- Disclaimer

Backup slides



Task Description

Task 2.1	• Fabrication of SOCs with LSCF electrodes: full cell and cells with symmetric electrodes	11/30/2021	Completed
Task 3.1	• LSCF electrochemical characterization for performance and durability without impurities. Post-test characterization by SEM/EDS	9/30/2022	Completed
Task 3.2	• LSCF electrochemical characterization for performance and durability in the presence of Cr impurities (durability test up to 1000 hours). Post-test characterization by SEM/EDS	3/30/2023	Completed
	• Effect of Hf infiltration	9/30/2023	Completed
Task 3.3	• LSCF electrochemical characterization for performance and durability in the presence of S impurities (durability test up to 1000 hours). Post-test characterization by SEM/EDS	9/30/2023	Completed
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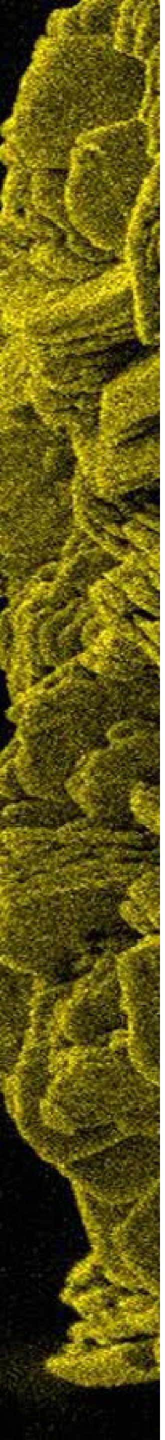
Task 3.4	• High-pressure button cell testing	9/30/2024	Completed



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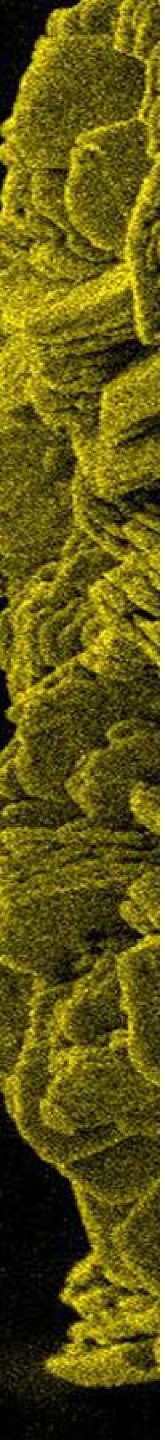
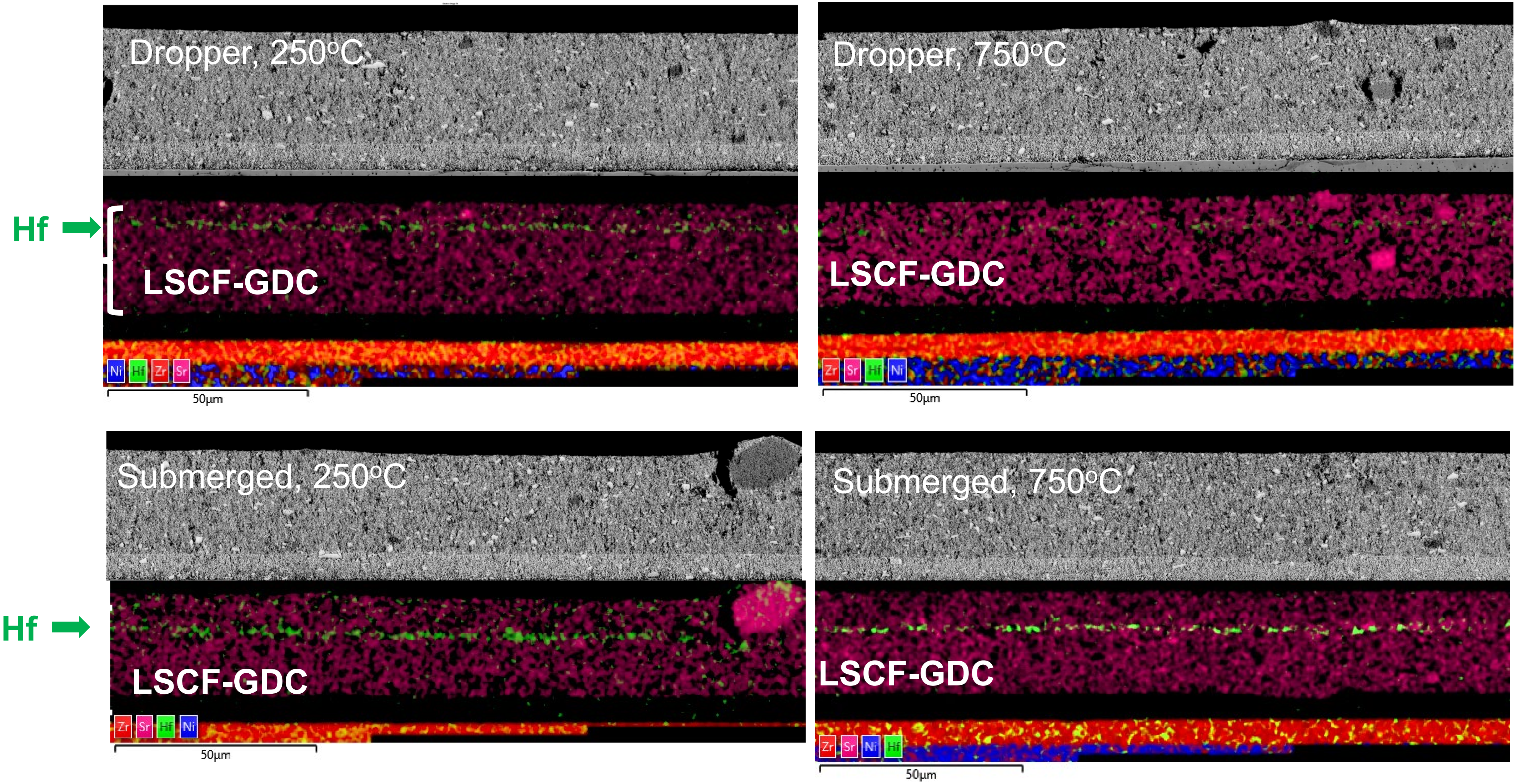


SEM/EDS Analysis of Large Electrode Areas



Acknowledgements

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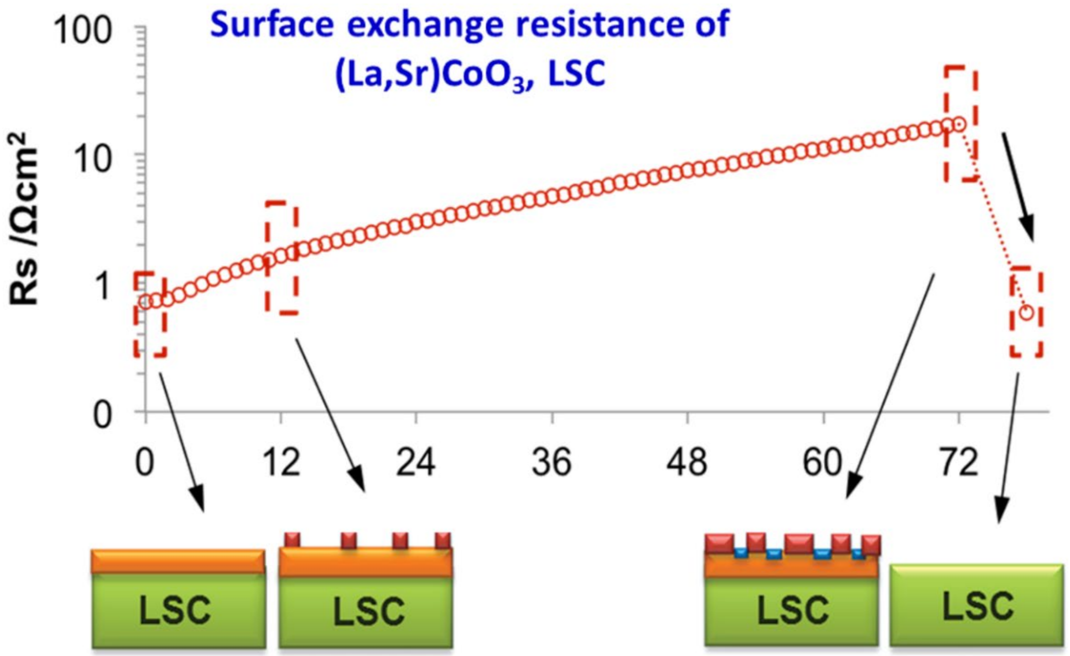
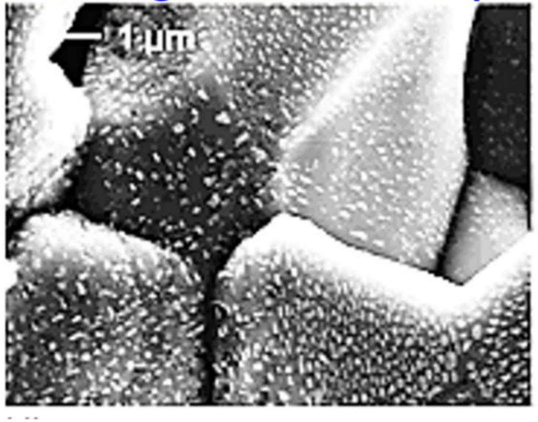
DEPARTMENT OF ENERGY · UNITED STATES OF AMERICA

Detrimental Sr-segregation and precipitation at perovskite oxide surfaces

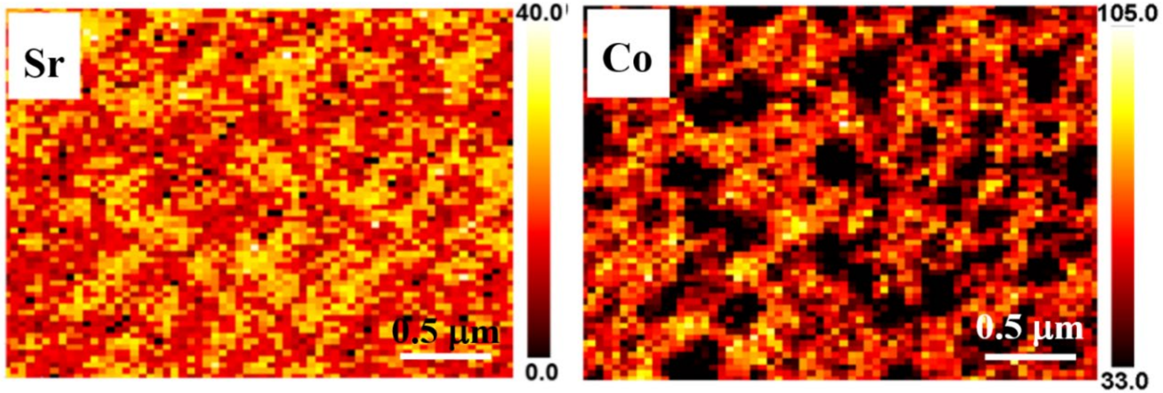
(a) Surface exchange resistance of $(La,Sr)CoO_3$, LSC



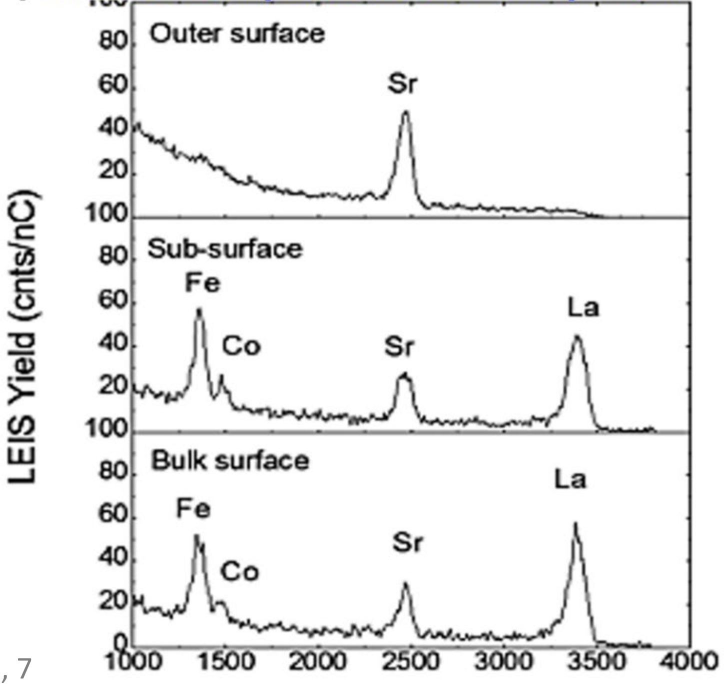
(c) Surface heterogeneities on LSCF pellet



(b) Surface heterogeneities on LSC



(d) Surface composition on LSCF pellet

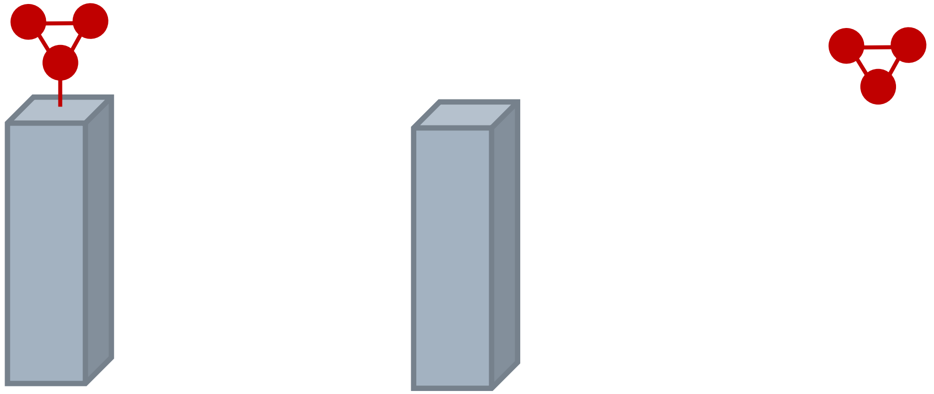


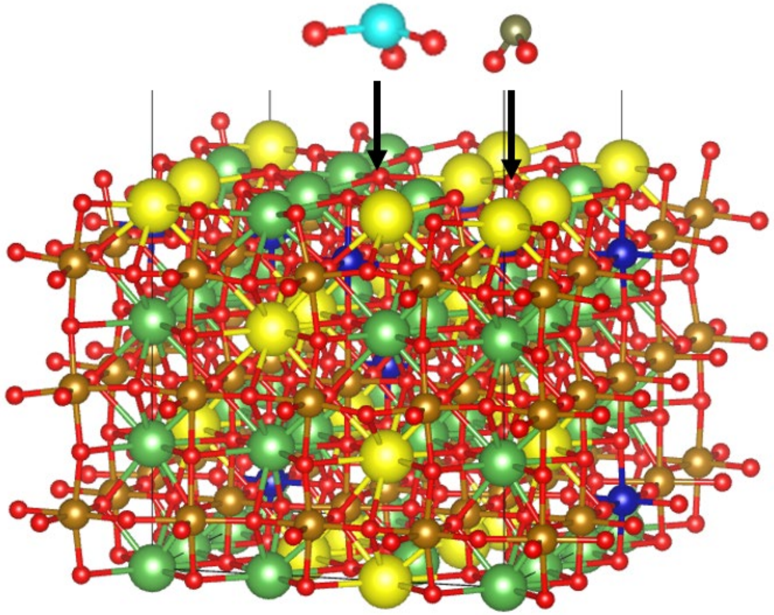
Z. Cai et al., *Chem. Mater.* 2011, 24 D. Oh et al., *J Mater Res.* 2012, 27 J. Druce et al. *Energ. Environ. Sci.*, 2014, 7

CrO_3 and SO_2 behavior on plane cut LSCF (001) surfaces

Adsorption energy:

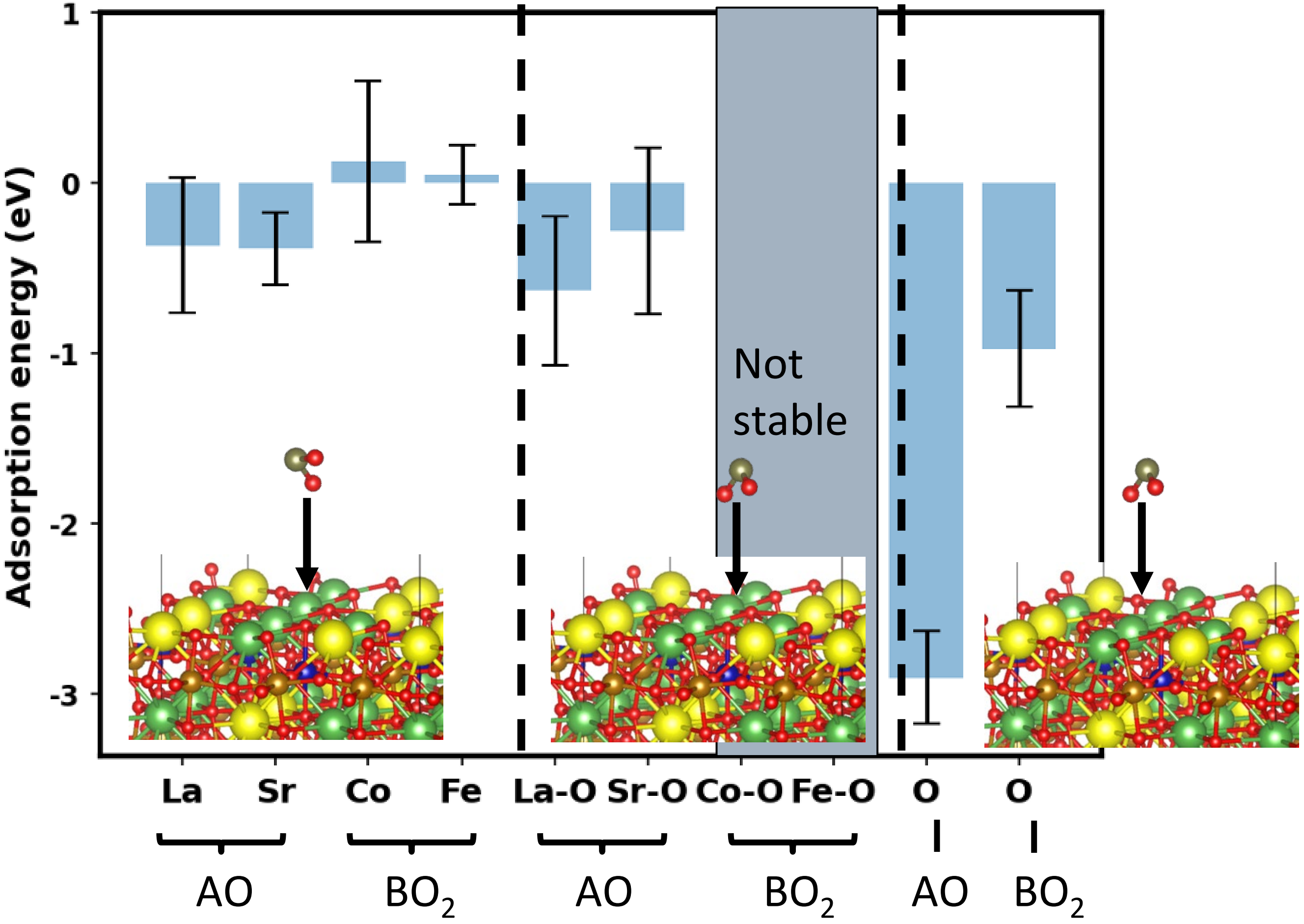
$$E_{ads} = E^{DFT}_{adsorbed} - E^{DFT}_{unadsorbed} - E^{DFT}_{molecule}$$

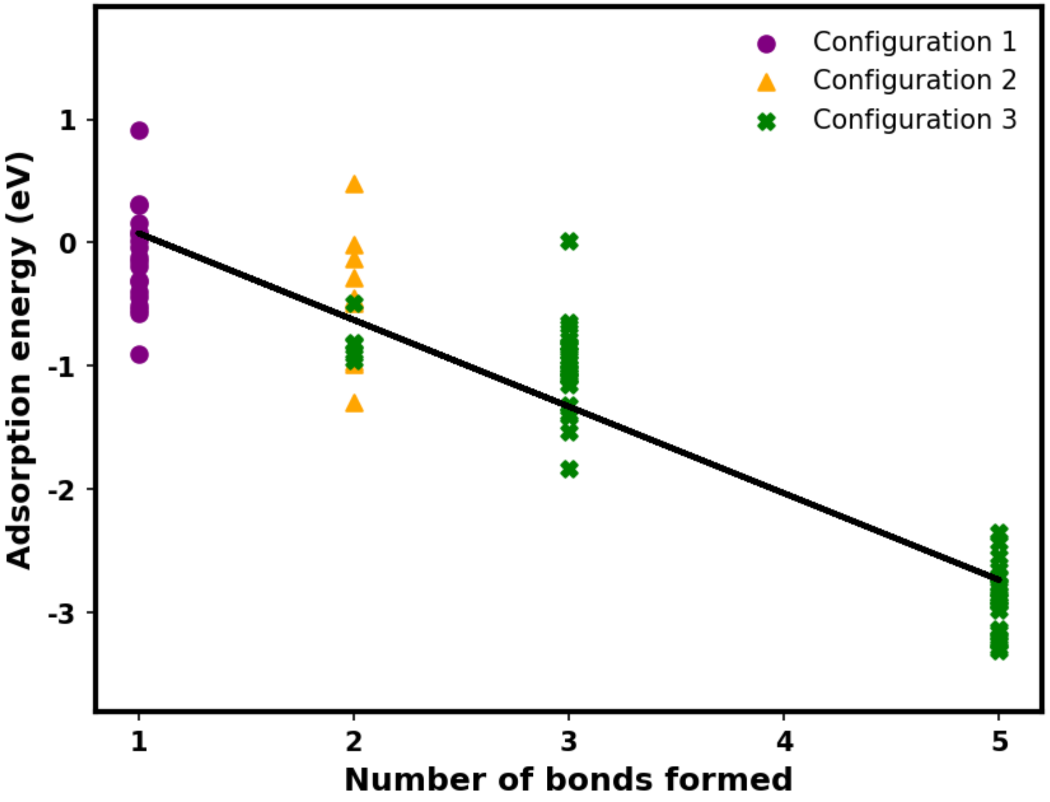




- First study the adsorption on pristine, non-polarized LSCF slabs
- Multiple adsorption configurations need to be studied

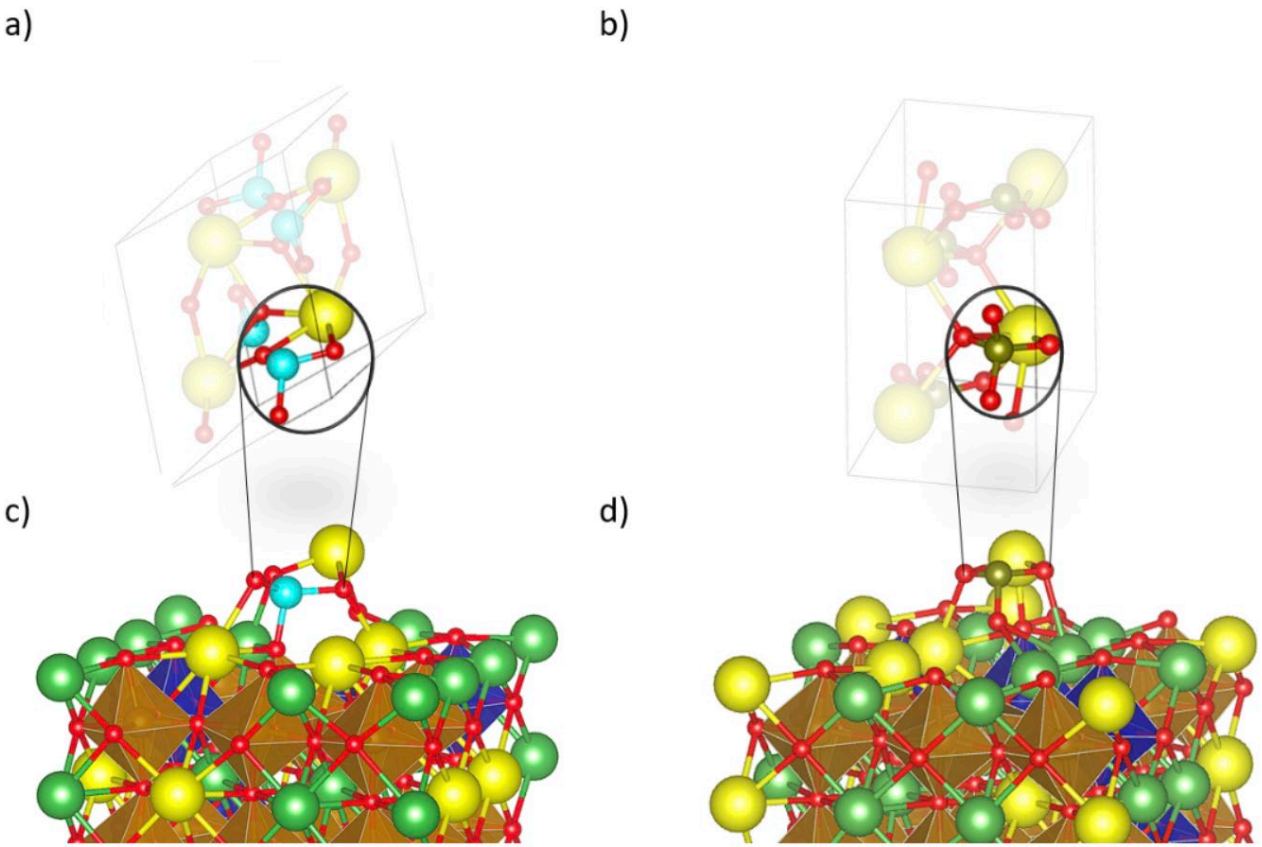
SO_2 adsorption energies vs configurations





- ○ CrO_3 adsorption energies are all negative for pristine LSCF slabs, whereas SO_2 is slightly more positive
- ○ Adsorption energies decrease as the number of bonds formed from the adsorption increase
- ○ BO_2-terminated slabs have higher (yet still negative) adsorption energies

Surface nuclei structure indicates $SrCrO_4$ and $SrSO_4$ compound formation

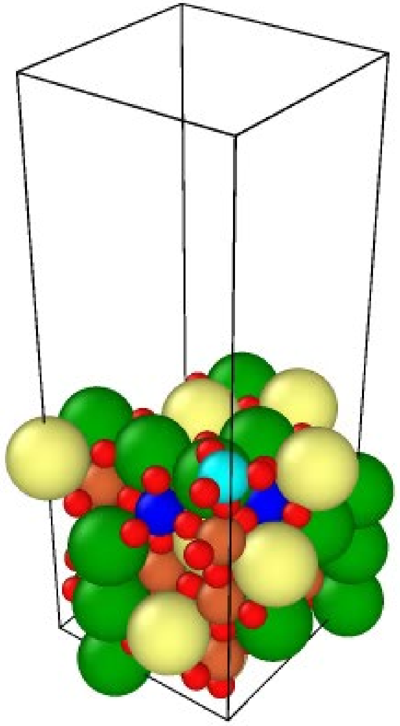


	Surface Sr-CrO_3 nuclei	$SrCrO_4$
Bond angle of O-Sr-O	68.68°	56.57°
Bond angle of O-Cr-O	106.45°	106.96°
Bond length of O-Sr	2.42 Å	2.95 Å
Bond length of O-Cr	1.73 Å	1.66 Å

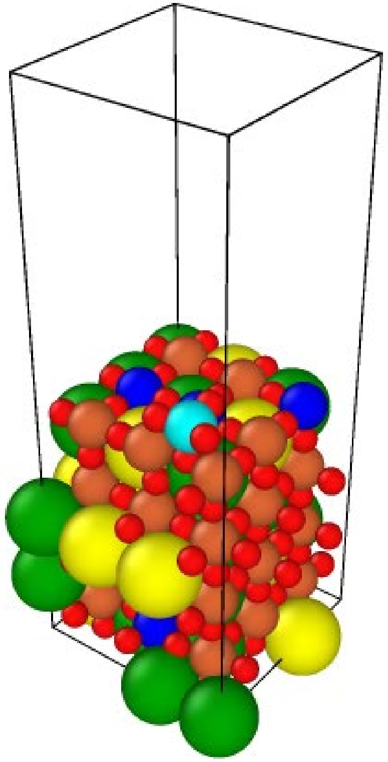
	Surface Sr-SO_2 nuclei	$SrSO_4$
Bond angle of O-Sr-O	58.41°	50.25°
Bond angle of O-S-O	102.18°	107.82°
Bond length of O-Sr	2.50 Å	2.86 Å
Bond length of O-S	1.57 Å	1.50 Å

CrO_3 more strongly bound in dynamic simulations, too.

T = 1300 K AO-termination



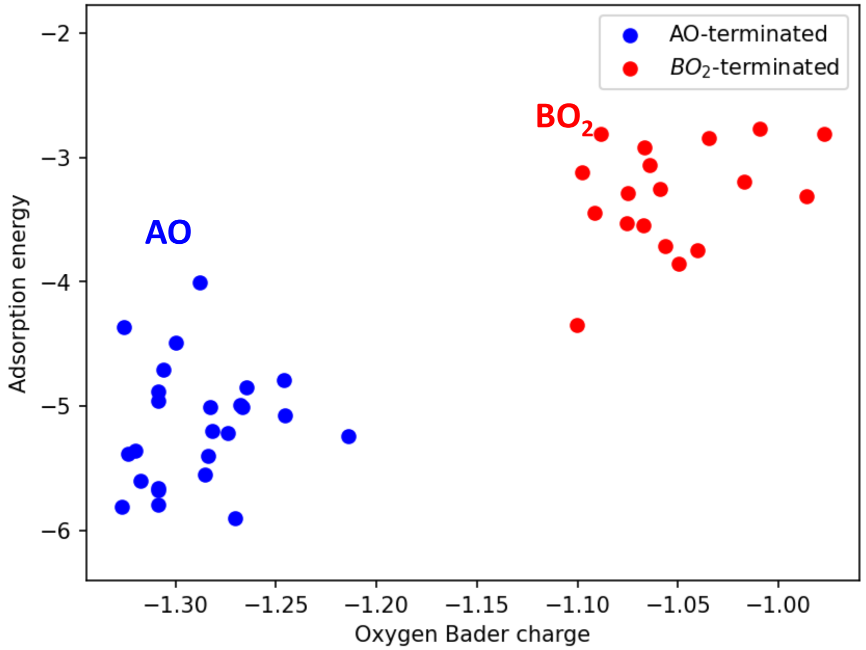
T = 1300 K BO_2-termination

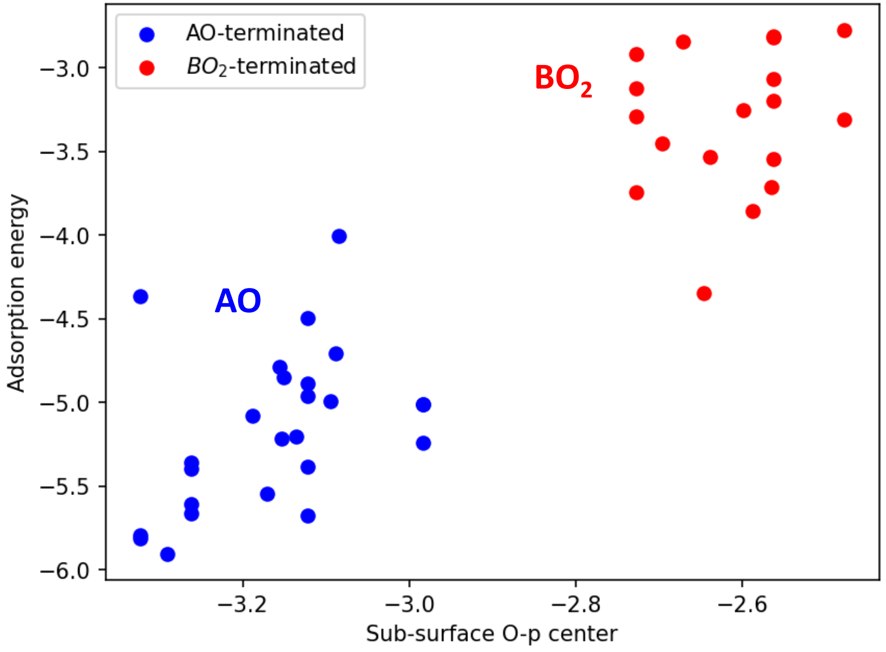


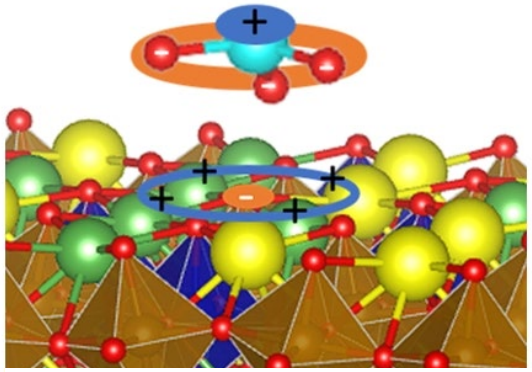
- AO-terminated surface: molecules are anchored on the adsorption site with a rotational degree of freedom
- BO_2-terminated surface: molecules are more likely to diffuse on the surface

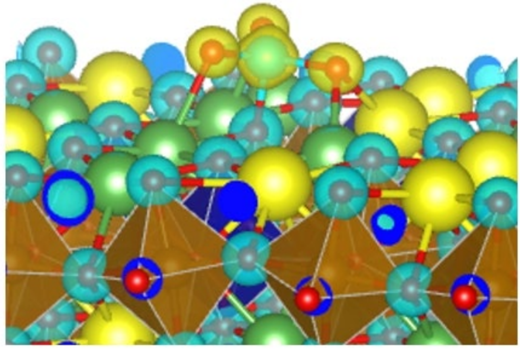
Oxygen site charge, and charge transfer as physical descriptors

1. Use as a **design principle** for future surface modifications
2. Gain **physical insights** behind adsorption on different surfaces





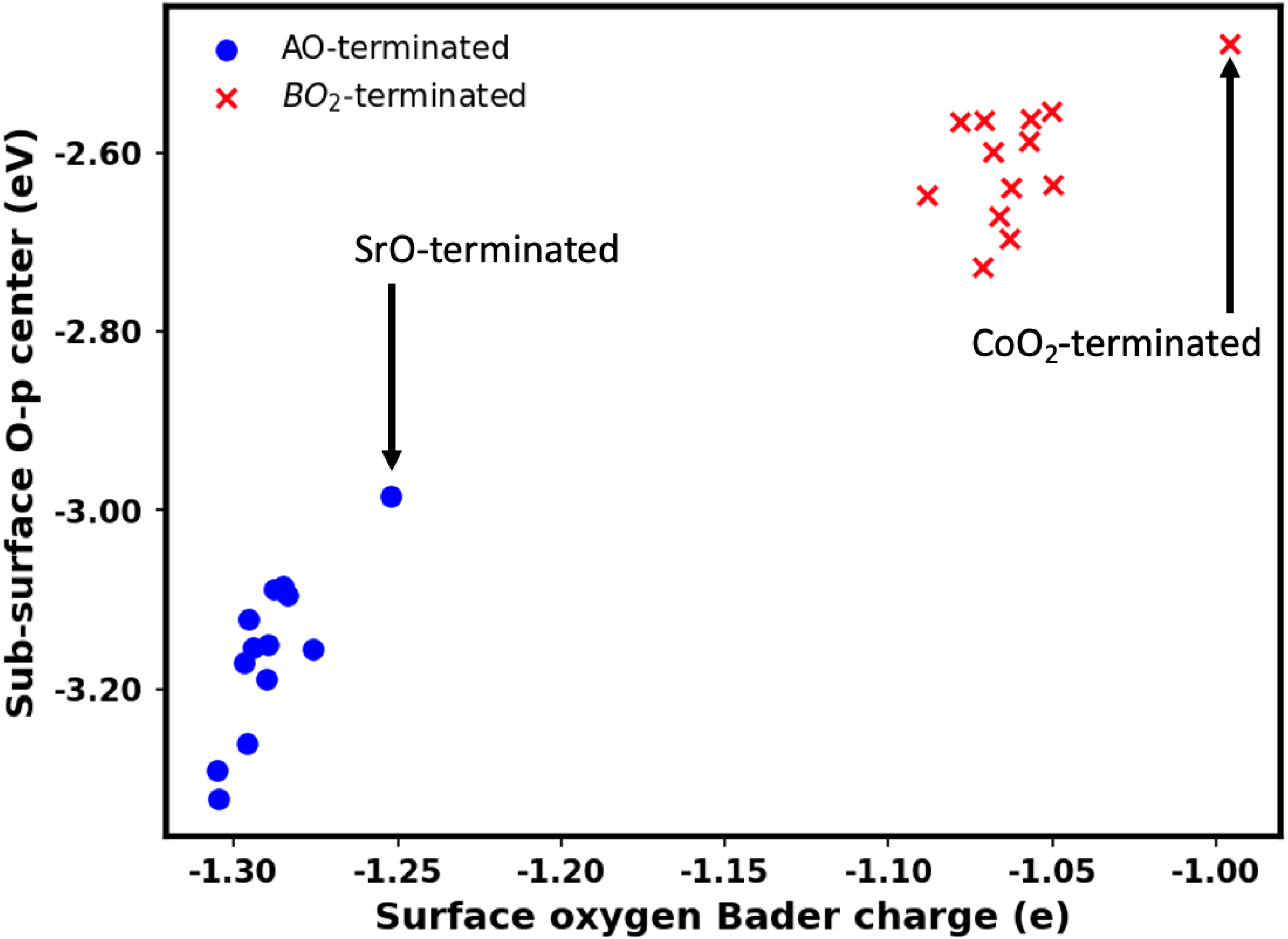




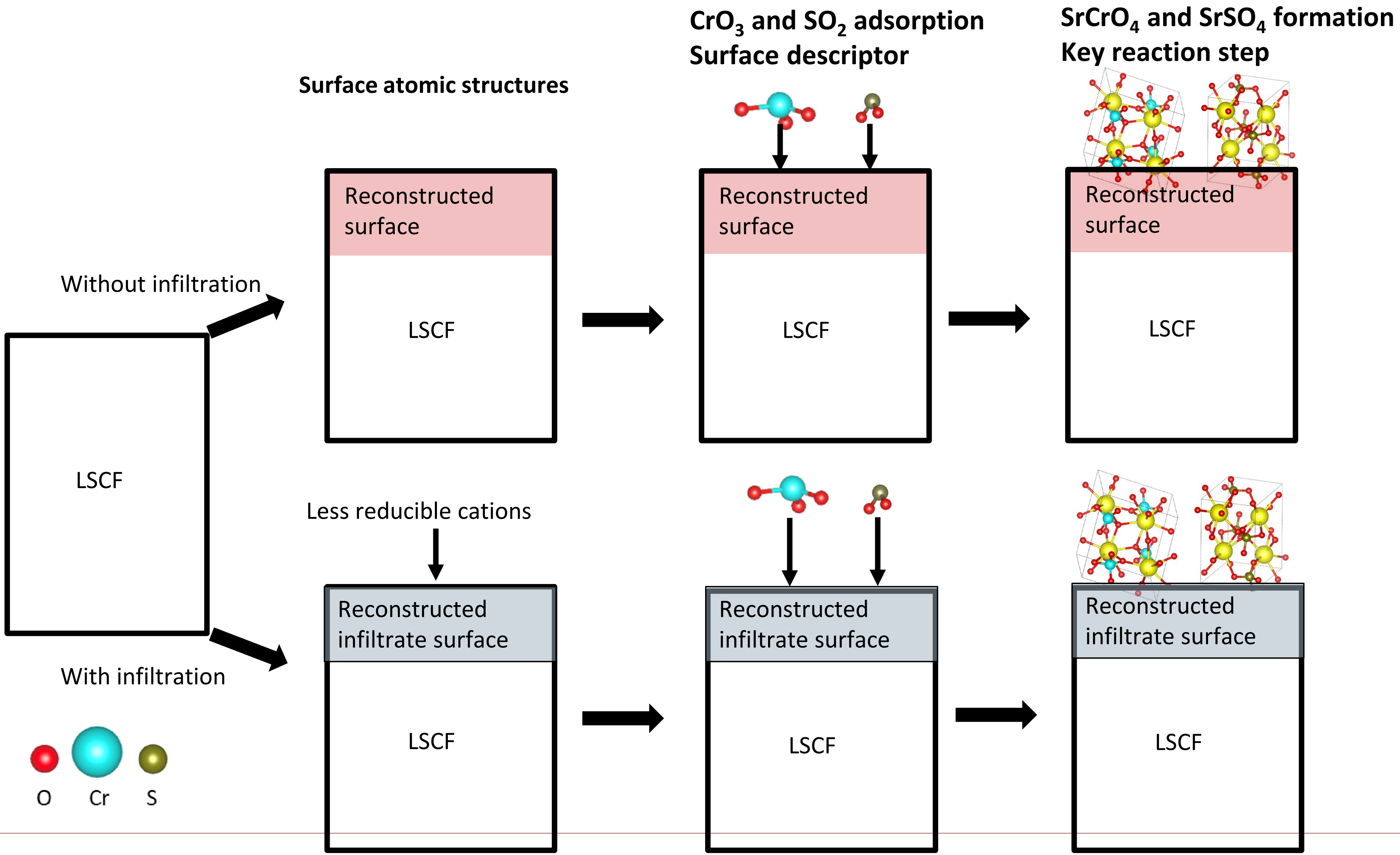
Oxygen charge (electrostatic) effect

Charge transfer effect $$\frac{\int_{-\infty}^{\infty} E D_{O2p}(E)dE}{\int_{-\infty}^{\infty} D_{O2p}(E)dE}$$

Surface O bader charge (electrostatic) and sub-surface O p center (charge transfer)



Computational investigation realistic LSCF surfaces



Surface atomic structures

CrO_3 and SO_2 adsorption
Surface descriptor

$SrCrO_4$ and $SrSO_4$ formation
Key reaction step

LSCF

Without infiltration

Reconstructed surface

LSCF

Reconstructed surface

LSCF

Reconstructed surface

LSCF

With infiltration

Less reducible cations

Reconstructed infiltrate surface

LSCF

Reconstructed infiltrate surface

LSCF

Reconstructed infiltrate surface

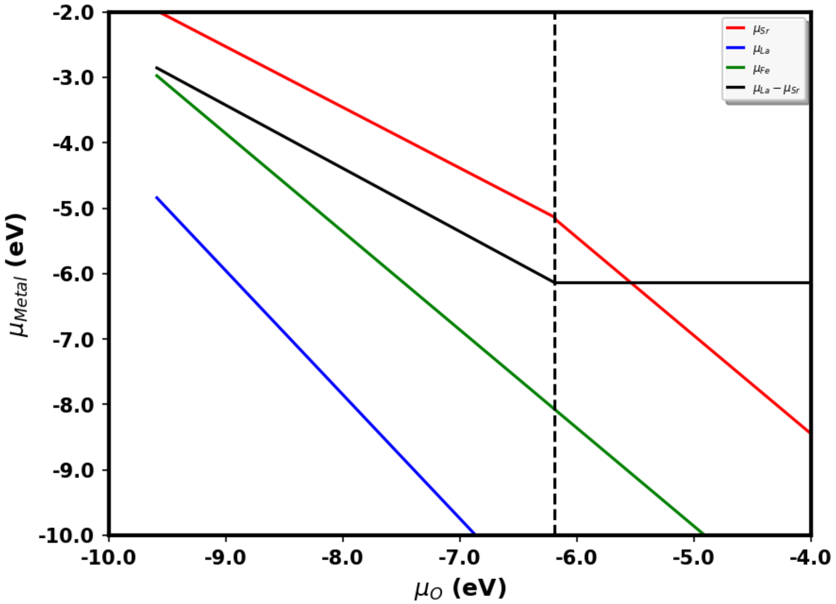
LSCF

Identify a physically meaningful path in chemical potentials space for GCMC simulation

• $E_{LSF}^{bulk} = \sum x_i \boldsymbol{\mu_i}$	Bulk equilibrium
• $\boldsymbol{\mu_O}(T, P_{O_2}) = \frac{1}{2}\left(E_{O_2}^{IAP} + \mu_{O_2}^0(T, P^0) + k_B T ln\left(\frac{P_{O_2}}{P^0}\right)\right)$	Slab is in equilibrium with environmental O_2
• $\boldsymbol{\mu_{Fe}}(T, \mu_O) = ½\ (E_{Fe_2O_3}^{bulk} - 3\mu_O(T, P_{O_2}))$	Slab is in equilibrium with Fe_2O_3
• $\boldsymbol{\mu_{Sr}} = E_{LSF}^{bulk}\left(VO\%, \frac{La}{Sr}\right) - E_{LSF-VSr}^{bulk}\left(VO\%, \frac{La}{Sr}\right) + \Delta\boldsymbol{\mu_{Sr}}$	$\Delta\mu$ as correction for E_{vac}(VO%)
• $\boldsymbol{\mu_{La}} = E_{LSF}^{bulk}\left(VO\%, \frac{La}{Sr}\right) - E_{LSF-VLa}^{bulk}\left(VO\%, \frac{La}{Sr}\right) + \Delta\boldsymbol{\mu_{La}}$	$\Delta\mu$ as correction for E_{vac} (VO%)
• Rearrange: $\Delta\boldsymbol{\mu_{Sr}} = E_{LSF-VSr}^{bulk} - E_{LSF}^{bulk} + \mu_{Sr}$	Physical meaning: equation for vacancy formation energy
• $\Delta\boldsymbol{\mu_{Sr}} - \Delta\boldsymbol{\mu_{La}} = k_B T ln\left(\frac{x_{Sr}}{x_{La}}\right)$	Based on Boltzmann distribution about 0.03eV
• $\boldsymbol{\mu_{Sr}} = \mu_{Sr}^{ref} + k_B ln(a_i)$	

We define the chemical potentials referenced in this way as Δμj, with Δμj = μj– μj (0), and the set comprising the values of this quantity for all species j involved in the **formation of the defect** is denoted by Δμj.

For $La_{0.6}Sr_{0.4}FeO_{3\text{-delta}}$:



Define simulation boundaries: linear programming to derive bulk phase diagram

$$\mathrm{La_{0.6}Sr_{0.4}FeO_3} \rightarrow$$
$$\mathrm{c_1La_2O_3 + c_2SrO + c_3SrO_2 + c_4Fe_2O_3 + c_5FeO}$$
$$\mathrm{+ c_6Fe + c_7LaFeO_3 + c_8SrFeO_3 + c_OO_2}$$

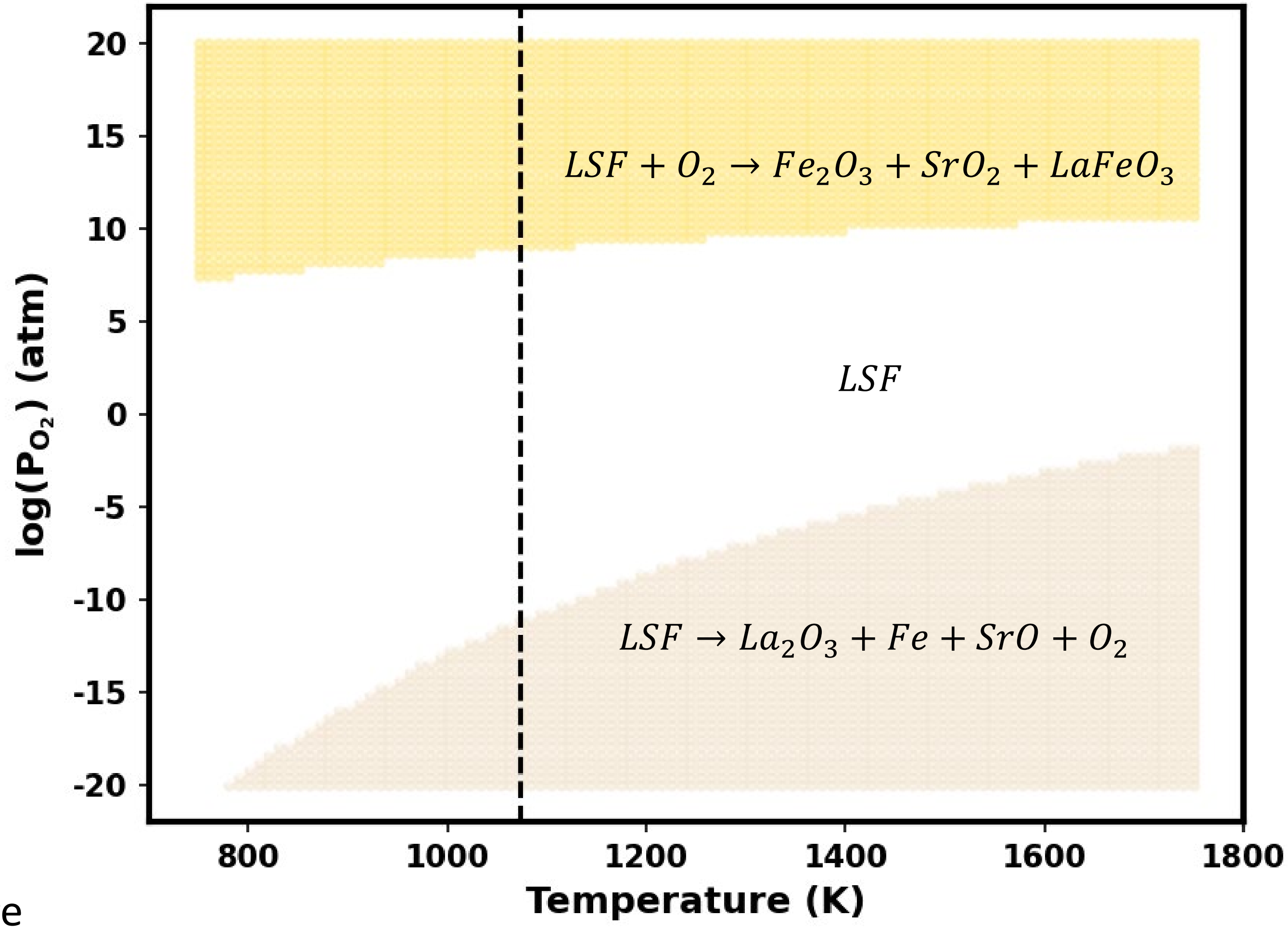
At a given μ_O:

$$min\left\{\sum_i c_i\, E^i_{product} - E_{LSF}\right\}$$

Subject to:

$\sum_i c_i s^{i,j}_{product} = s^j_{LSF}$ for all *j* in LSF

$c_i \geq 0$ for all *i* except for c_O

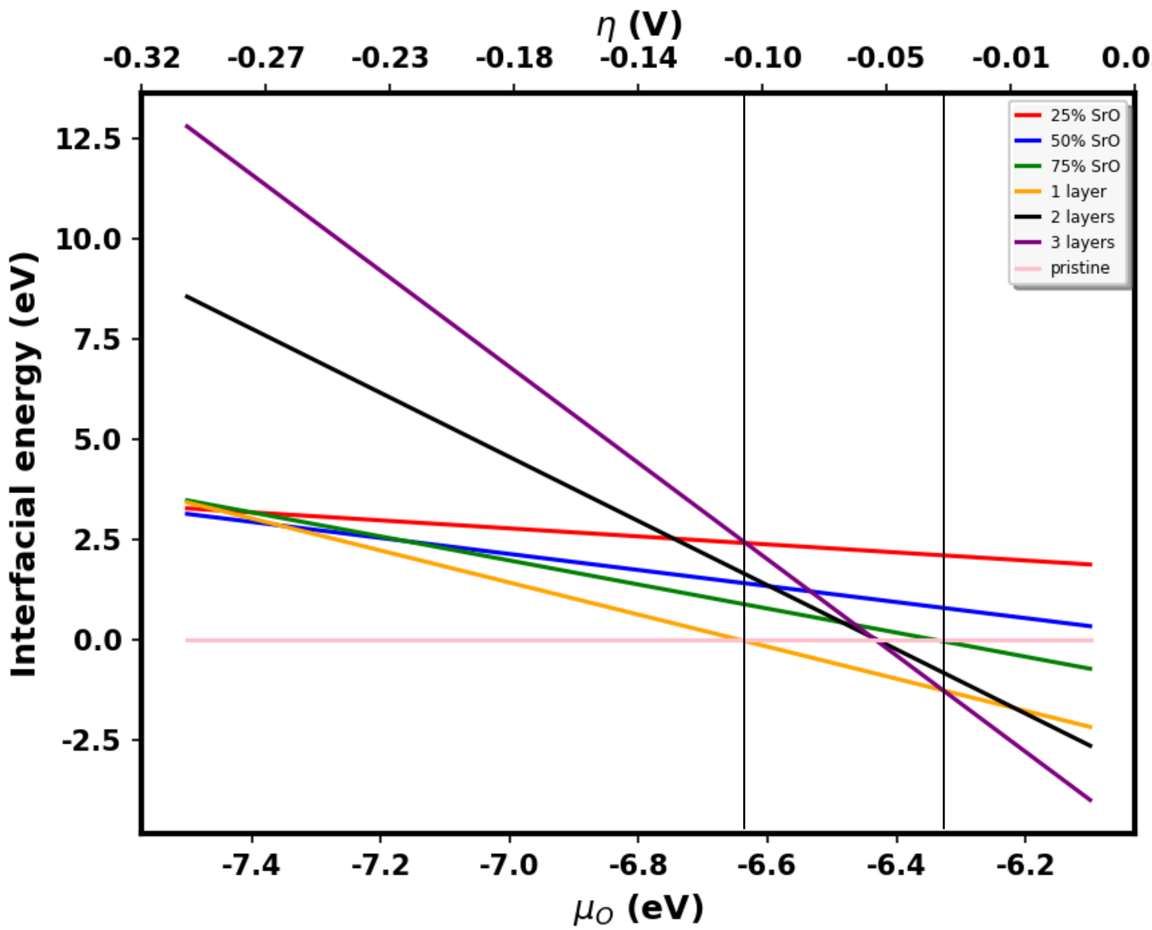


- The LSF phase becomes more stable at lower temperature

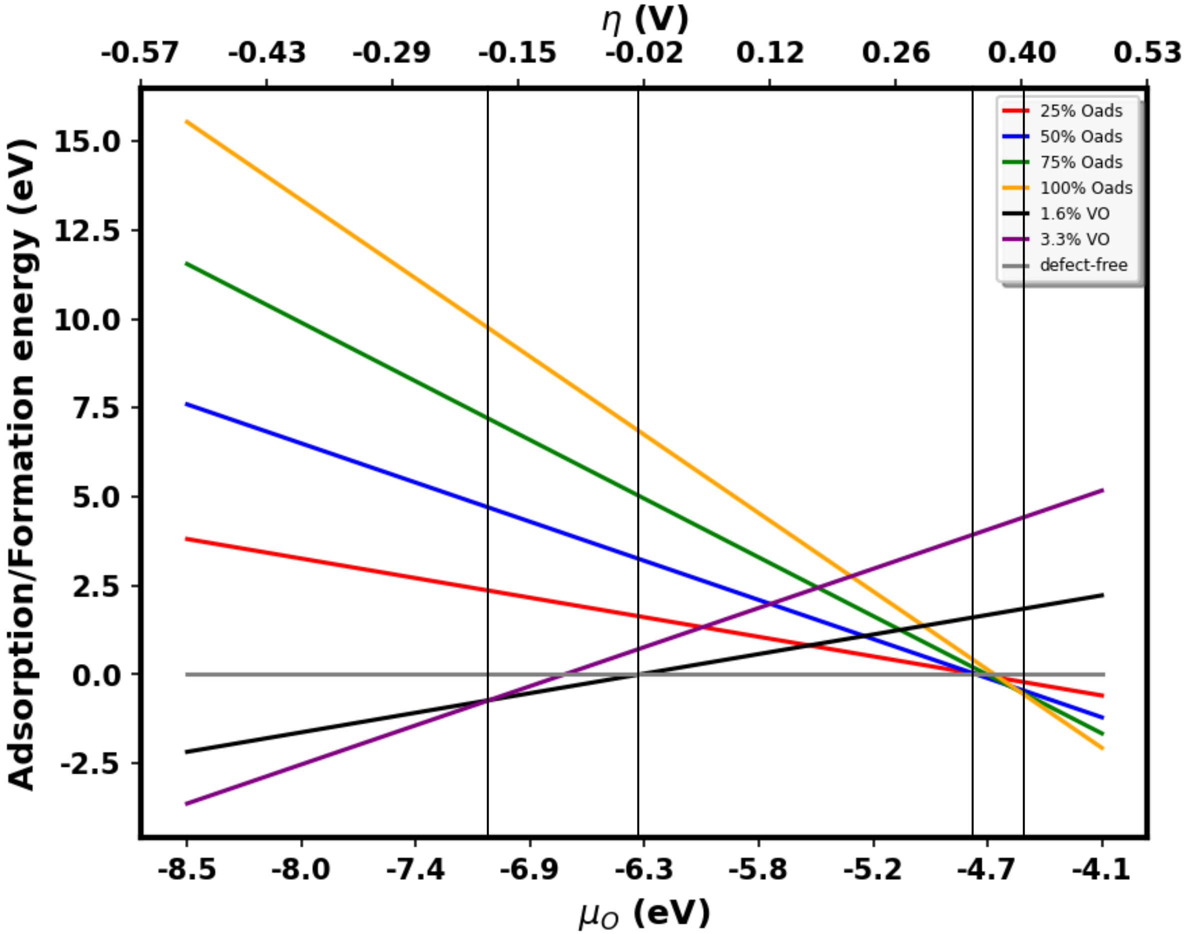
Modify chemical potential calculation to introduce Hf infiltrates

$E_{LSF}^{bulk} = \sum x_i \boldsymbol{\mu_i}$	Bulk equilibrium
$\boldsymbol{\mu_O}(T, P_{O_2}) = \frac{1}{2}\left(E_{O_2}^{IAP} + \mu_{O_2}^{0}(T, P^0) + k_B T ln\left(\frac{P_{O_2}}{P^0}\right)\right)$	Slab is in equilibrium with environmental O_2
$\boldsymbol{\mu_{Fe}(T, \mu_O)} = \frac{1}{2} \left(\boldsymbol{E_{Fe_2O_3}^{bulk} - 3\mu_O(T, P_{O_2})}\right)$	
$\boldsymbol{\mu_{Fe}} = E_{LSF}^{bulk}\left(VO\%, \frac{La}{Sr}\right) - E_{LSF-VFe}^{bulk}\left(VO\%, \frac{La}{Sr}\right) + \Delta\mu_{Fe}$	Slab is in equilibrium with Fe_2O_3
$\boldsymbol{\mu_{Hf}} = E_{LSF}^{bulk}\left(VO\%, \frac{La}{Sr}\right) - E_{LSF-VHf}^{bulk}\left(VO\%, \frac{La}{Sr}\right) + \boldsymbol{\Delta\mu_{Hf}}$	
$\boldsymbol{\mu_{Sr}} = E_{LSF}^{bulk}\left(VO\%, \frac{La}{Sr}\right) - E_{LSF-VSr}^{bulk}\left(VO\%, \frac{La}{Sr}\right) + \boldsymbol{\Delta\mu_{Sr}}$	$\Delta\mu$ as correction for E_{vac}(VO%)
$\boldsymbol{\mu_{La}} = E_{LSF}^{bulk}\left(VO\%, \frac{La}{Sr}\right) - E_{LSF-VLa}^{bulk}\left(VO\%, \frac{La}{Sr}\right) + \boldsymbol{\Delta\mu_{La}}$	$\Delta\mu$ as correction for E_{vac} (VO%)
Rearrange: $\boldsymbol{\Delta\mu_{Sr}} = E_{LSF-VSr}^{bulk} - E_{LSF}^{bulk} + \mu_{Sr}$	Physical meaning: equation for vacancy formation energy
$\boldsymbol{\Delta\mu_{Sr}} - \boldsymbol{\Delta\mu_{La}} = k_B T ln\left(\frac{x_{Sr}}{x_{La}}\right)$	Based on Boltzmann distribution about 0.03eV
$\boldsymbol{\mu_{Sr}} = \mu_{Sr}^{ref} + k_B ln(a_i)$	
$\Delta\mu_{Fe} - \boldsymbol{\Delta\mu_{Hf}} = k_B T ln\left(\frac{x_{Fe}}{x_{Hf}}\right)$	Based on Boltzmann distribution about 0.03eV

Oxidizing environment favors SrO phase formation



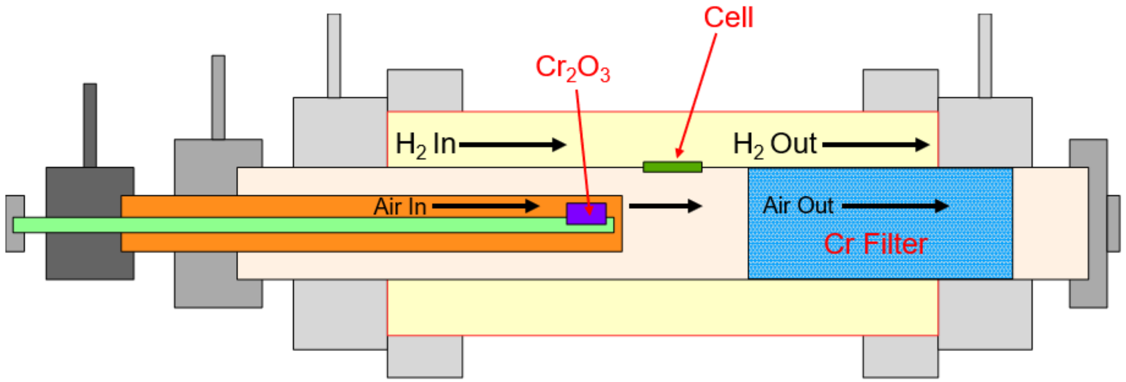
defect-free LSF with SrO

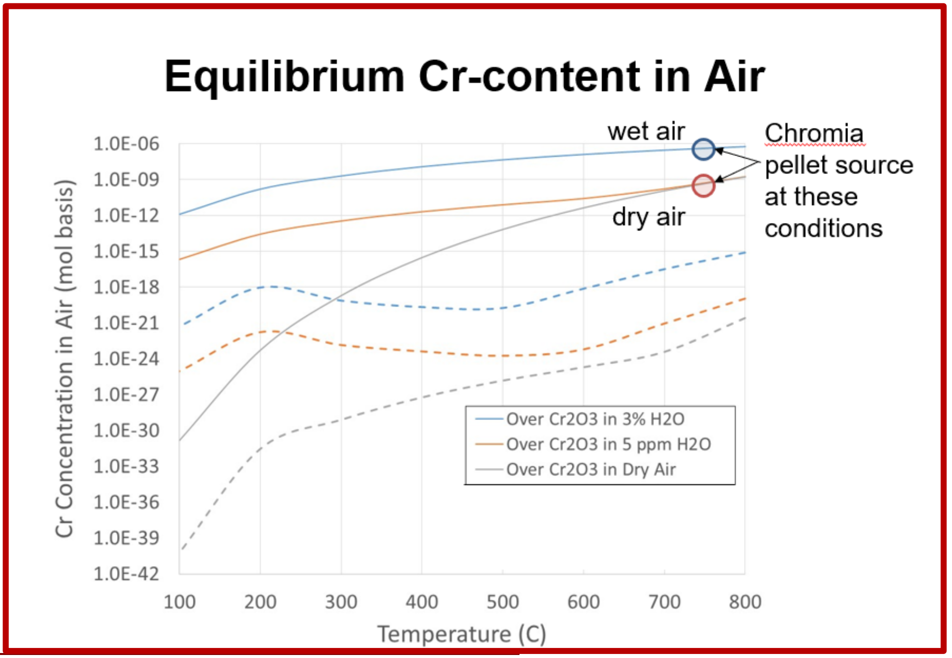


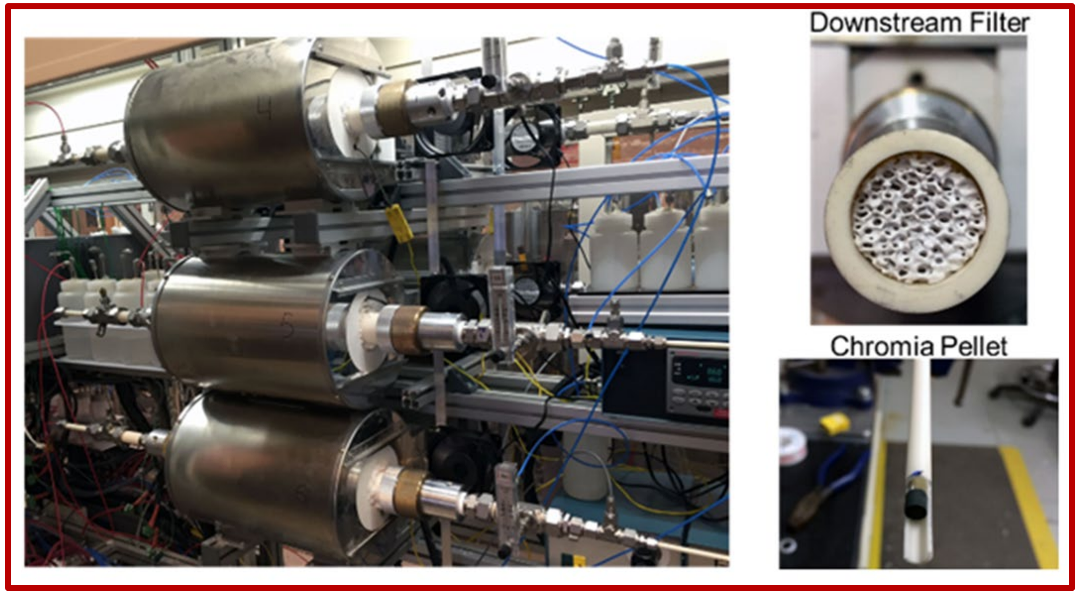
defect-free LSF with O

- Before oxygen being added as adatoms, SrO secondary phase would be favorable to form

PNNL team established controlled Cr poisoning testing capabilities

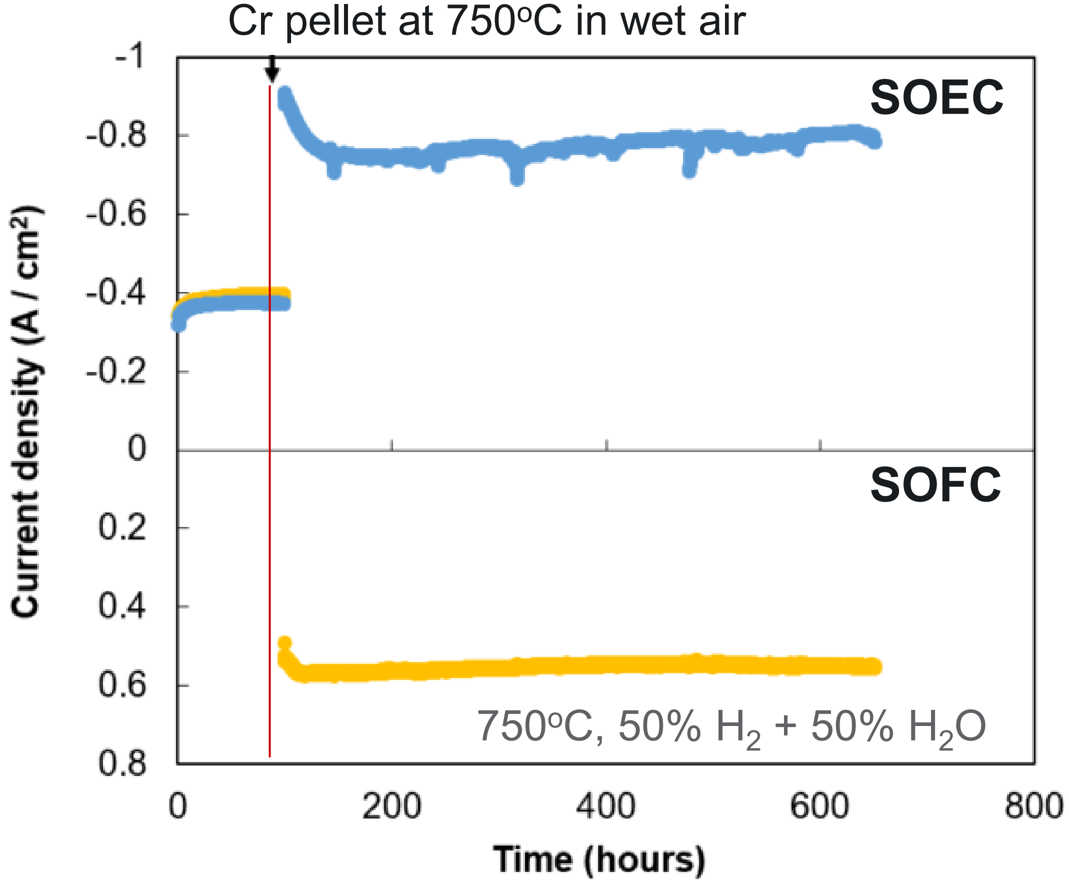


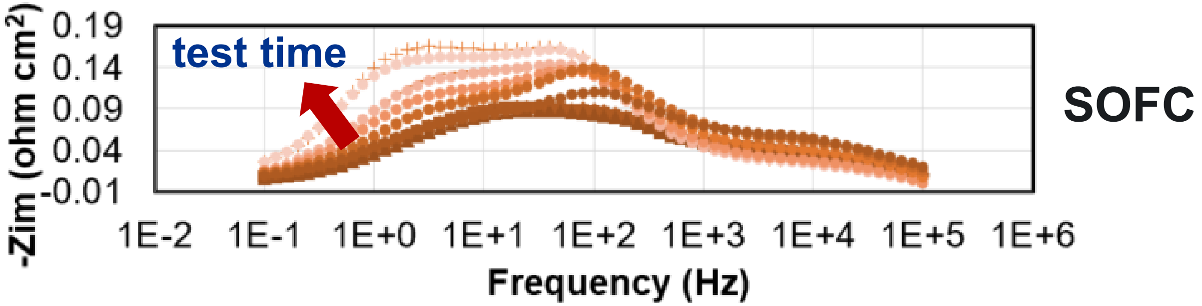


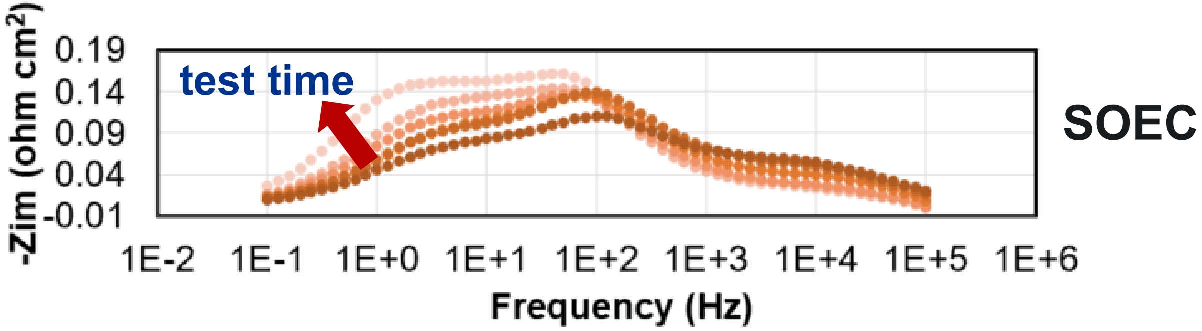


Cell: Ni/YSZ | YSZ | GDC – LSCF/GDC
T=750°C
H_2/H_2O=50/50

Cr Poisoning of SOFC and SOEC

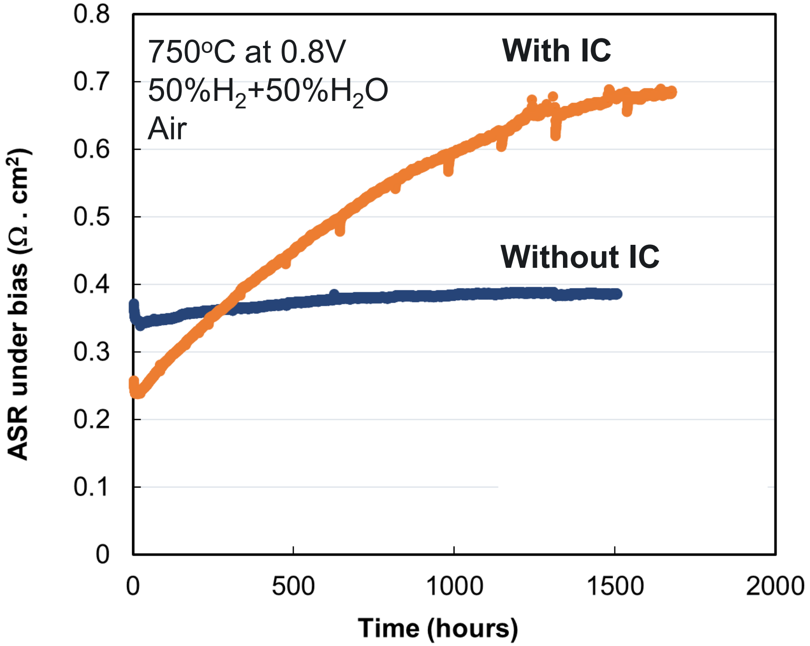


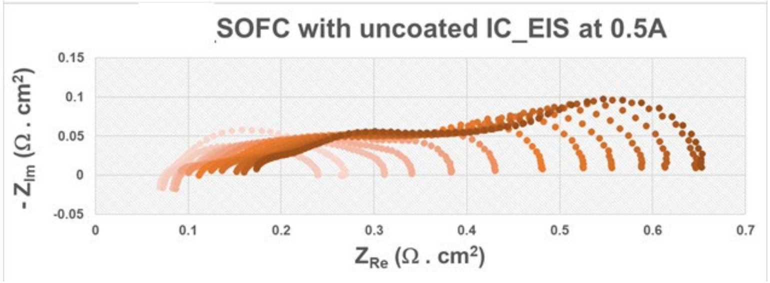


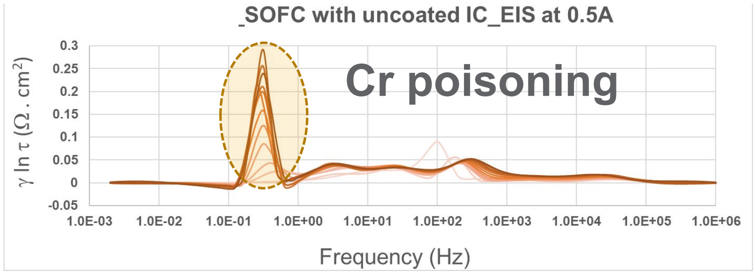


Low frequency process ASR is increasing during exposure to Cr in SOFC and SOEC

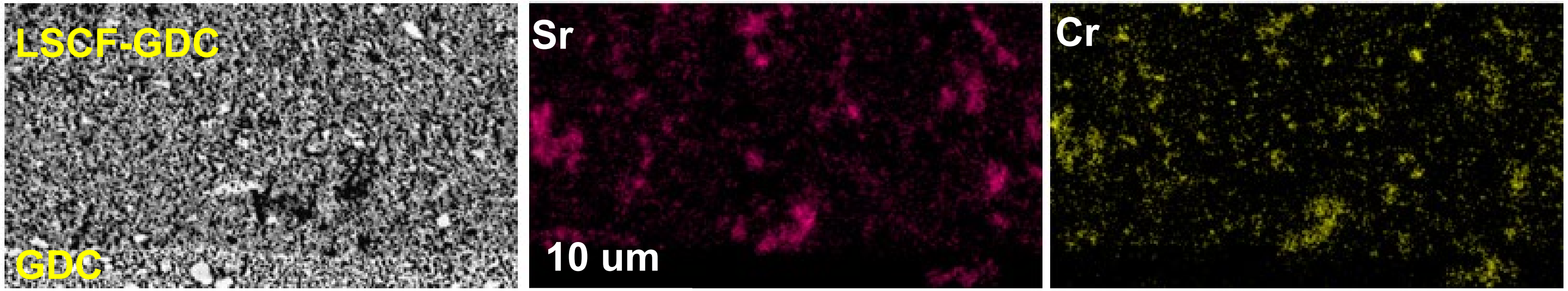
SOFC Testing With and Without Uncoated Interconnect





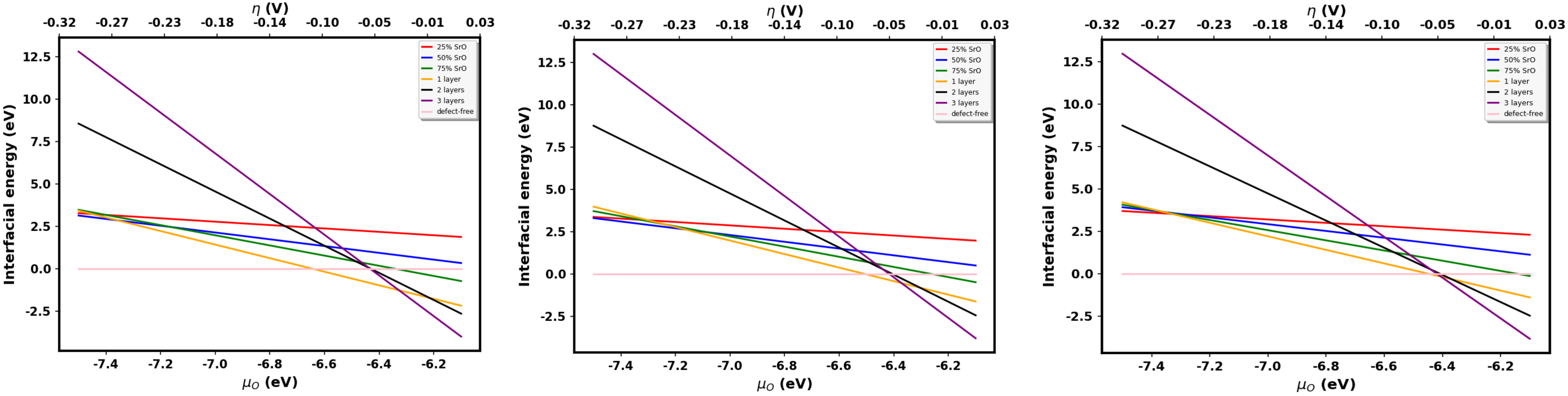


- **Low frequency process is related to oxygen adsorption or oxygen exchange**
- **Ohmic resistance significantly increased compared to test without interconnect**



- **Co-presence of Sr and Cr indicates $SrCrO_4$ precipitates.**

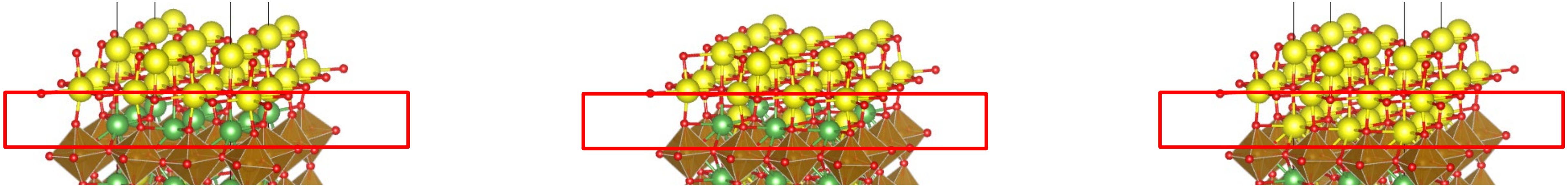
SrO termination favors multi-layer SrO secondary phase formation



LaO-termination

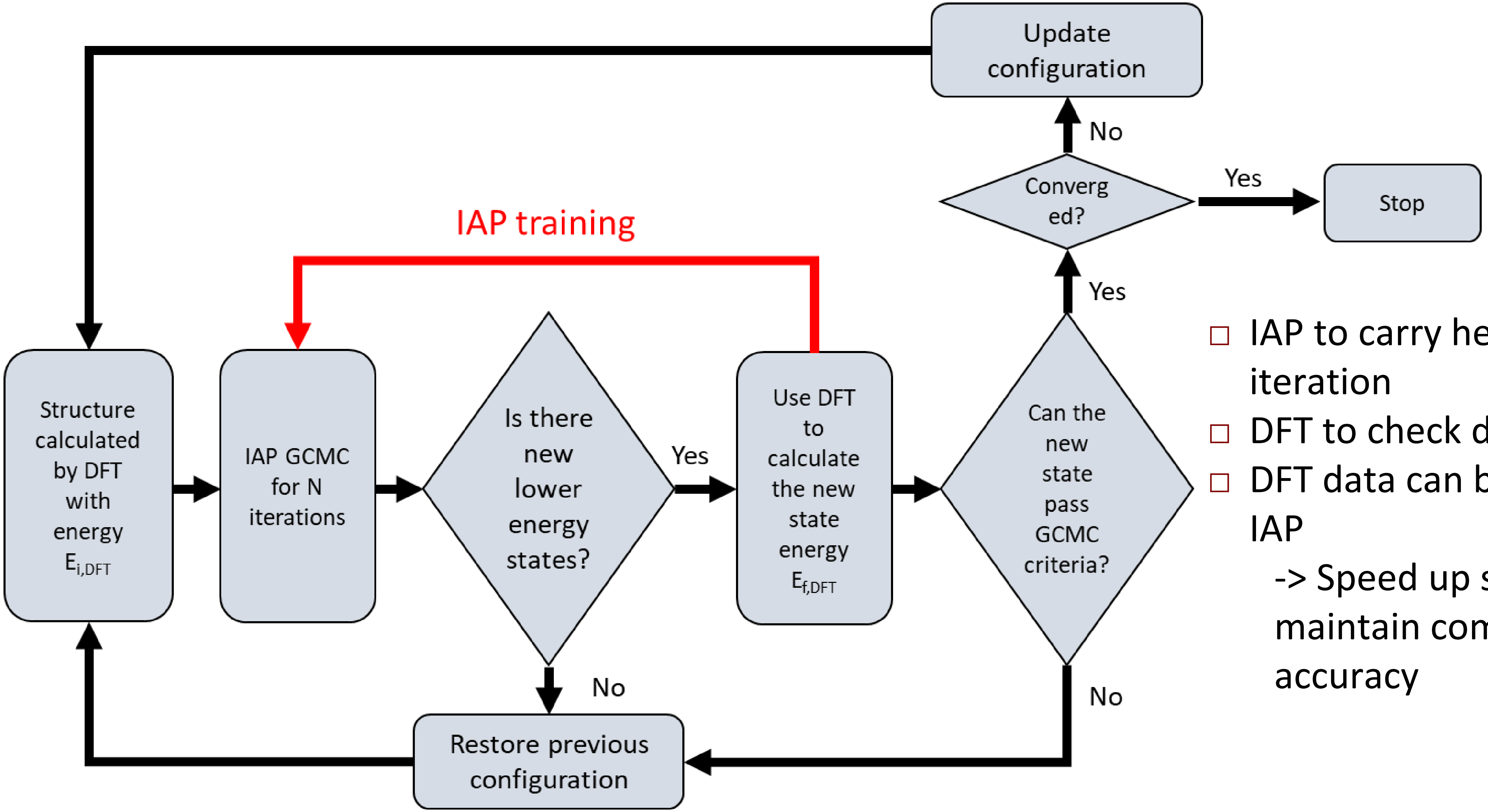
$(La_{0.5}Sr_{0.5})O$-termination

SrO-termination



- When surface Sr content increases -> multi-layer SrO phase formation favors
- Hypothesis is for elastic/chemical similarity of the interphase (SrO-termination/SrO vs LaO-termination/SrO)

Grand Canonical Monte Carlo with DFT + IAP for resolving LSCF surface reconstructions



- ☐ IAP to carry heavy GCMC iteration
- ☐ DFT to check data validity
- ☐ DFT data can be fed to train IAP
  - -> Speed up simulation while maintain computational accuracy

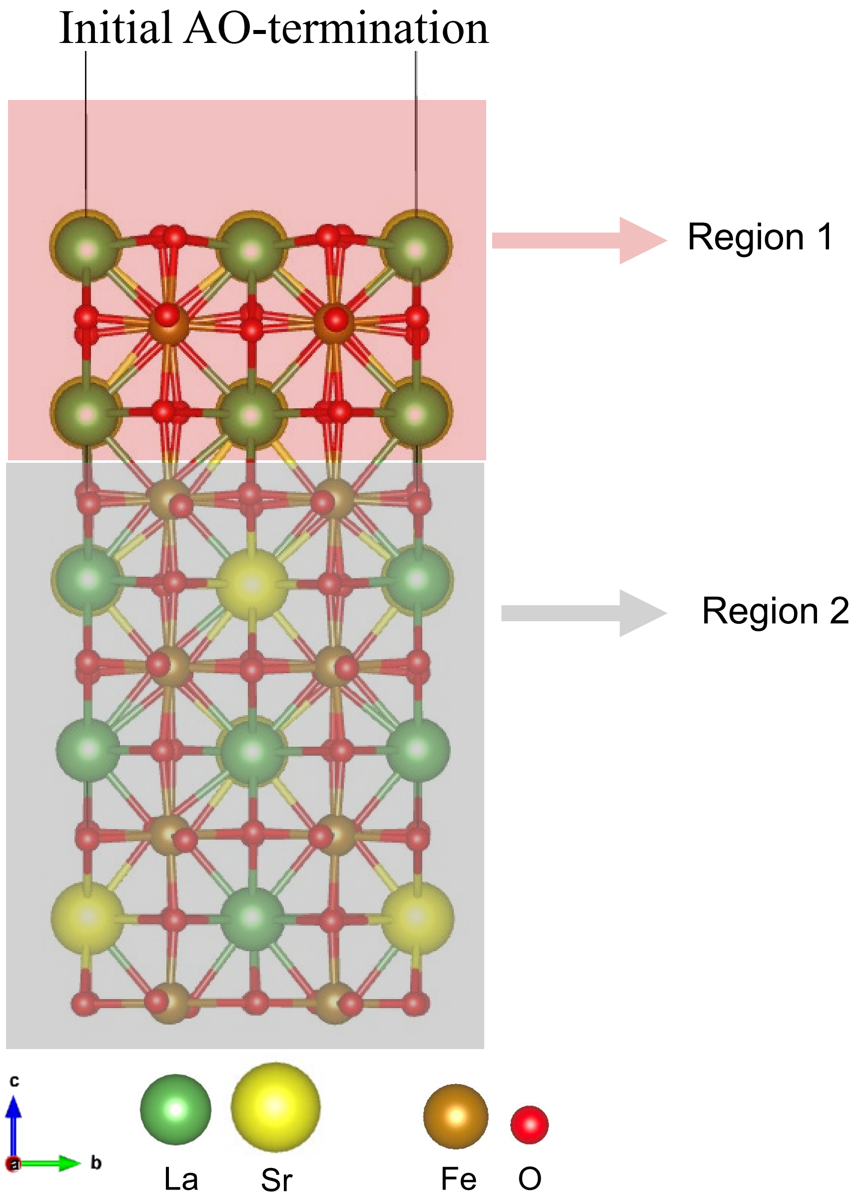
Identify thermodynamic condition for GCMC simulation

$$P_{O_2} = P_{O_{2,gas}} exp\left(\eta \frac{4F}{RT}\right)$$

TABLE I. Thermodynamic conditions for GCMC simulations.

Overpotential	Temperature	Effective partial pressure
0.42V	1073 K	10^7 atm
0.25 V	1073 K	10^4 atm
0 V	1073 K	0.2 atm
-0.18V	1073 K	10^{-4} atm
-0.34 V	1073 K	10^{-7} atm

Resolving surface structures on LSCF using Grand Canonical Monte Carlo with DFT and ML-IAP

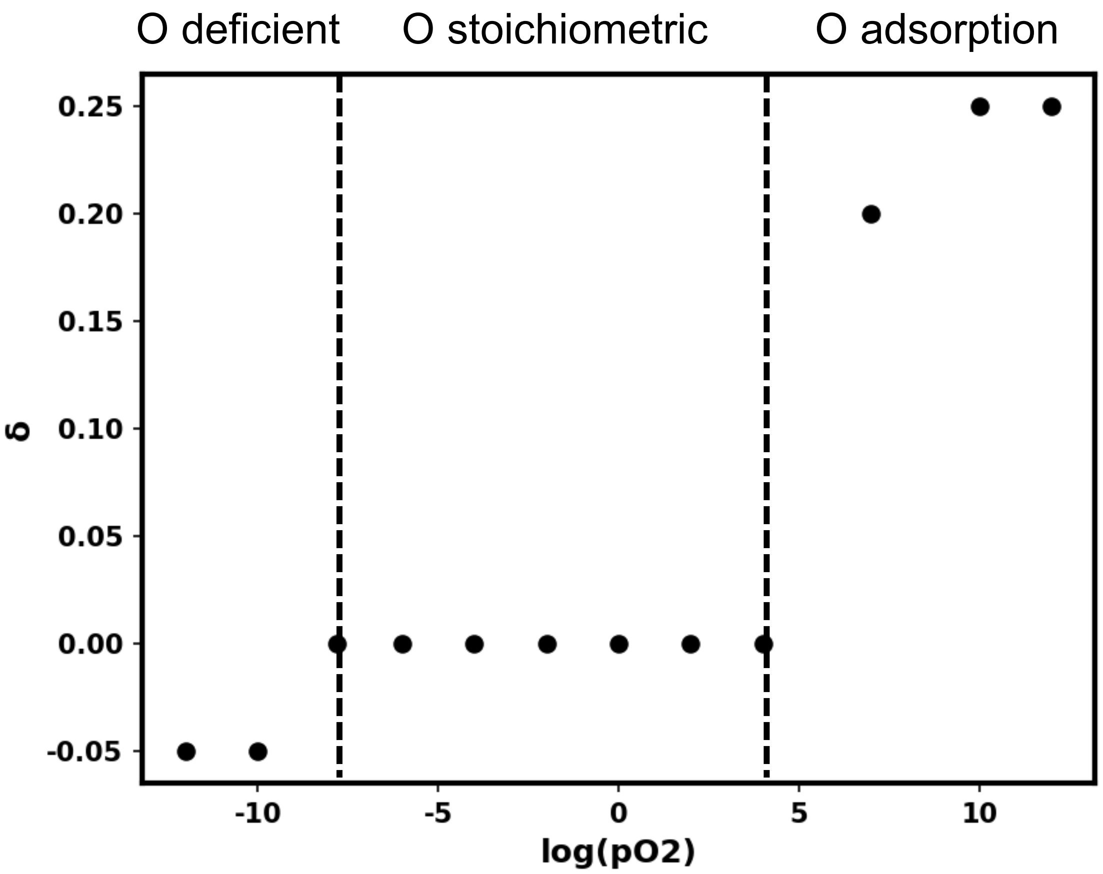


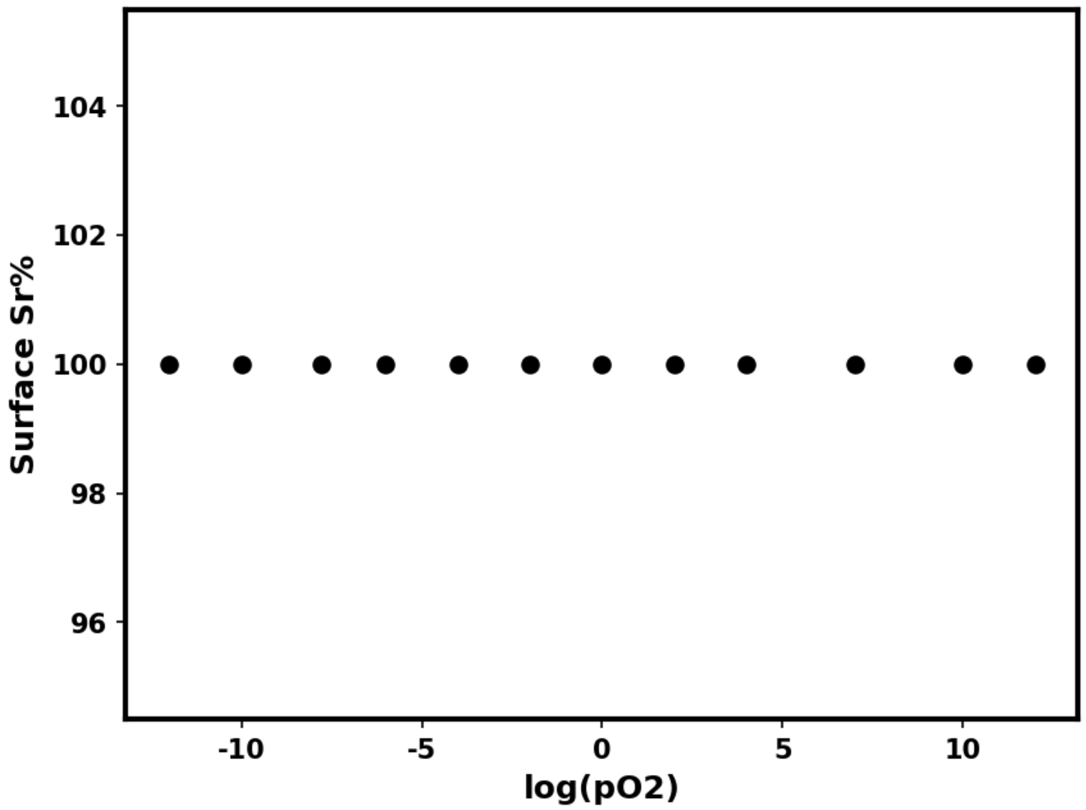
- Region 1:
  - Full GCMC on oxygen
  - MC exchange/displacement on metal
- Region 2:
  - Fix atoms in positions

-> The overall cation ratio is fixed

Oxygen nonstoichiometry and surface Sr coverage

ABO_{3+}

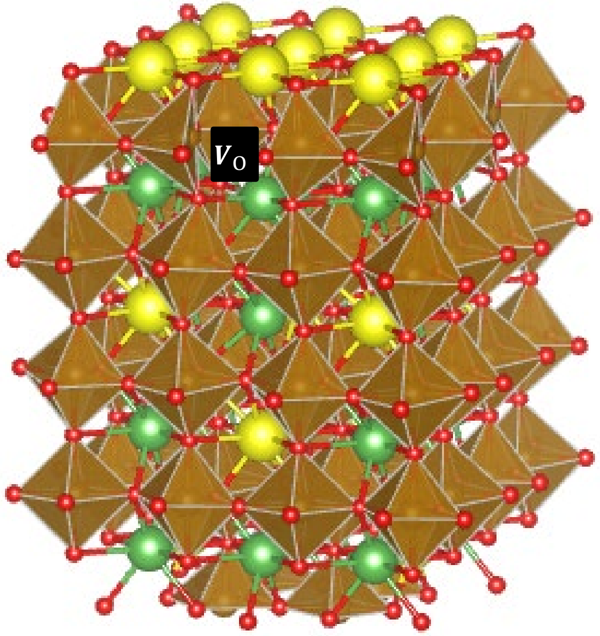




Representative LSCF structures

$pO2 < 10^{-11}$ atm

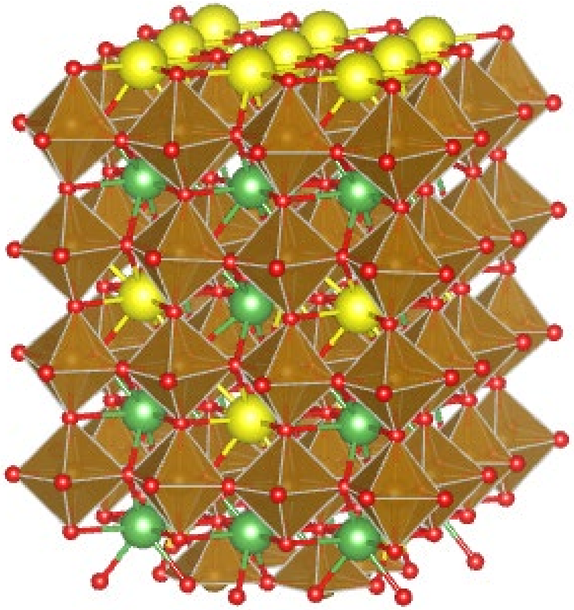
$\eta < -0.57$ V



- VO formed in sub surface
- 0.5% overall O deficiency

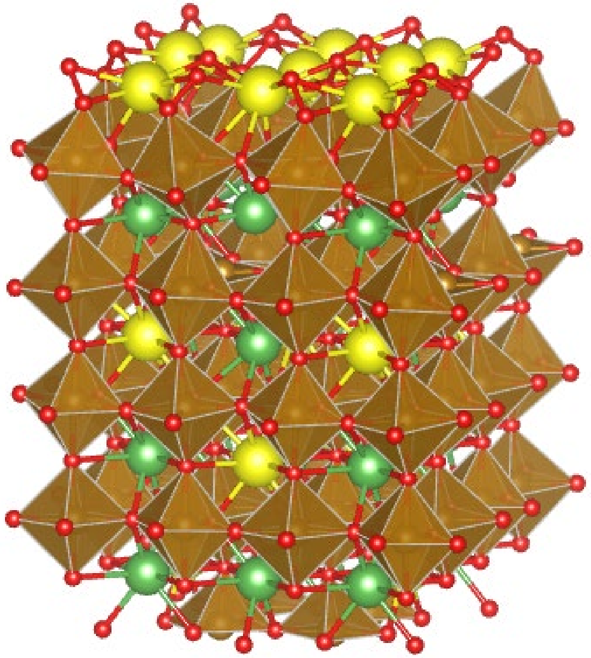
$pO2 = 10^{-11} \sim 10^{4}$ atm

$\eta = -0.57 \text{ V} \sim 0.25$ V



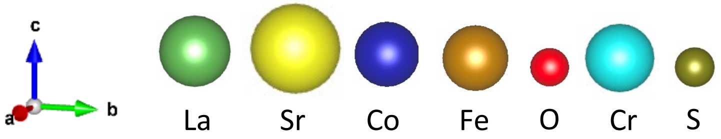
$pO2 > 10^{4}$ atm

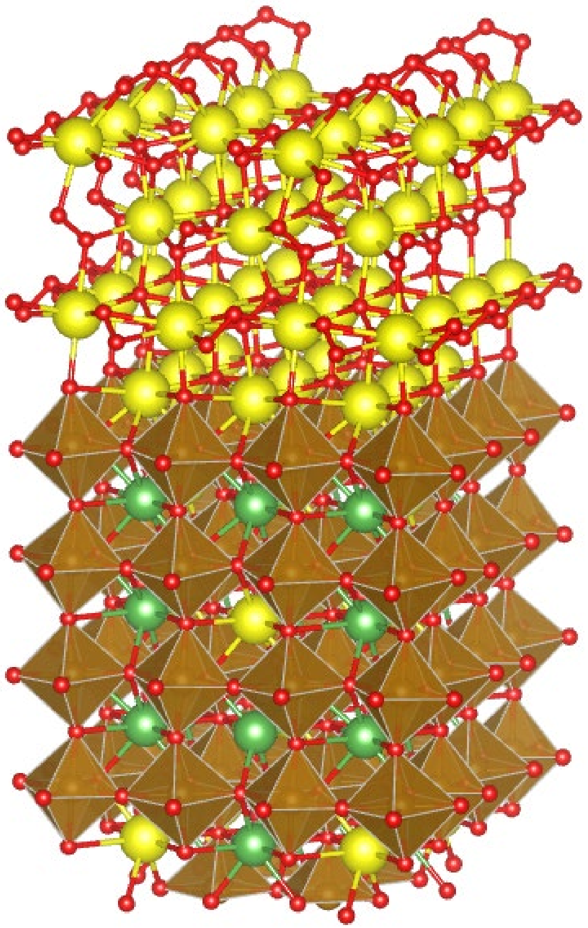
$\eta > 0.25$ V



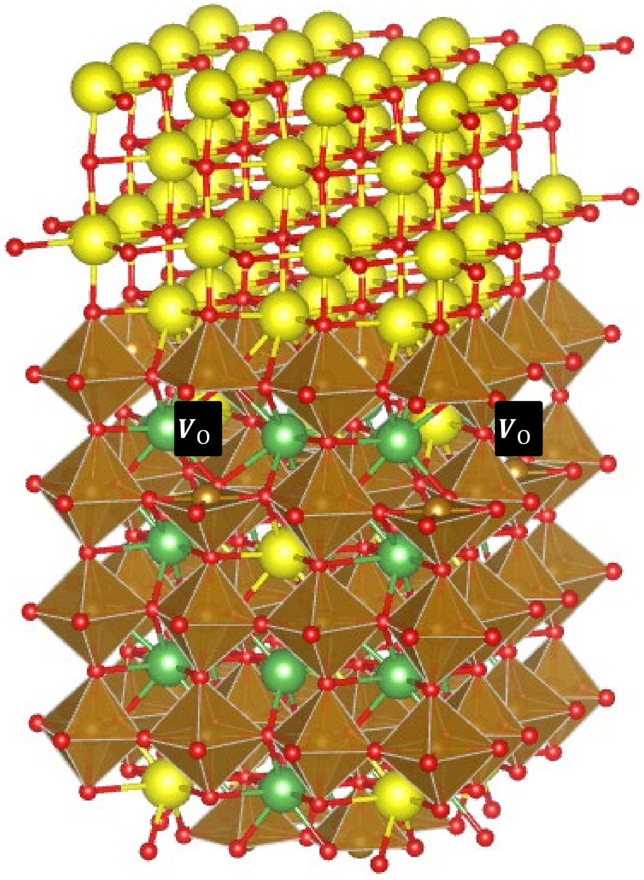
- Add O atoms on surface
- 2% O adsorption

Converged structures from GCMC setup

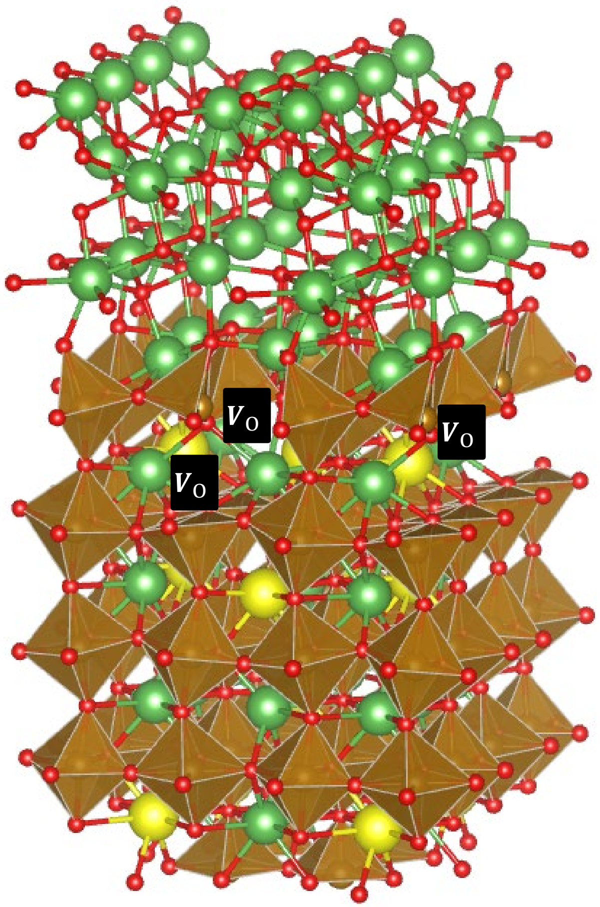




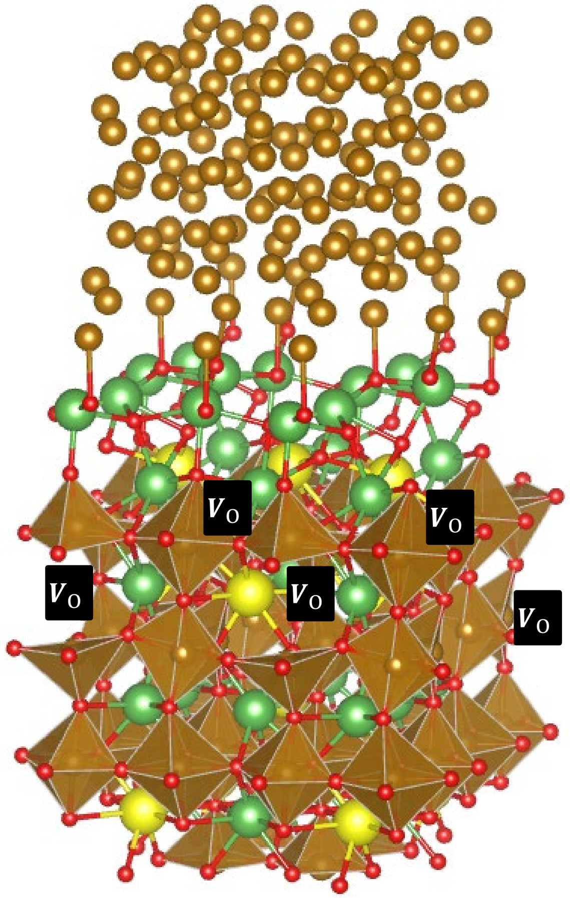
- muO = -5
- SrO2 surface phase
- Surface O ads



-6
SrO surface phase



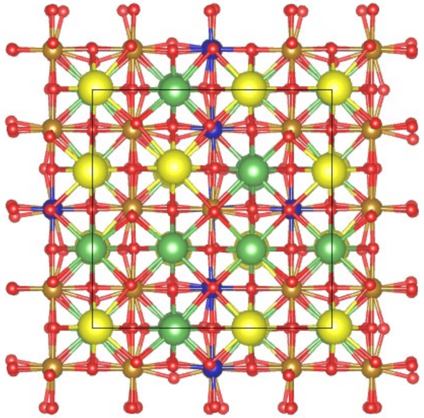
-7
La2O3 surface phase
VO formation at interface



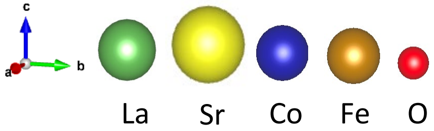
-8
Fe surface phase
Double perovskite layer

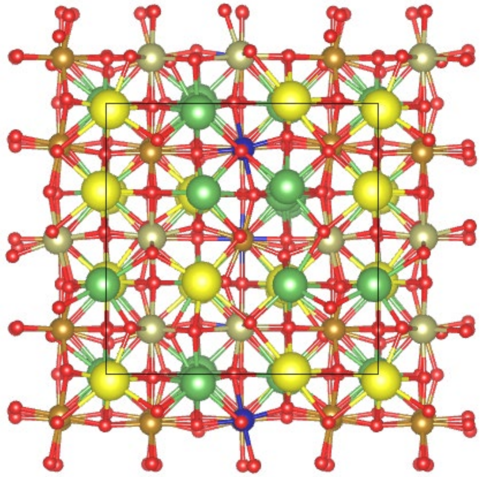
Current most stable structures

Initial structure



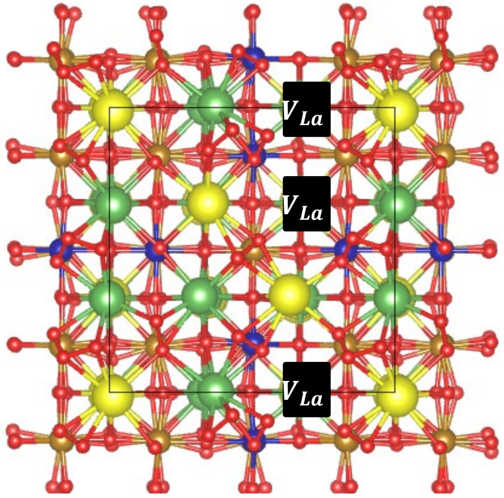
$(La_{0.56}Sr_{0.44})(Co_{0.33}Fe_{0.67})O_{3}$





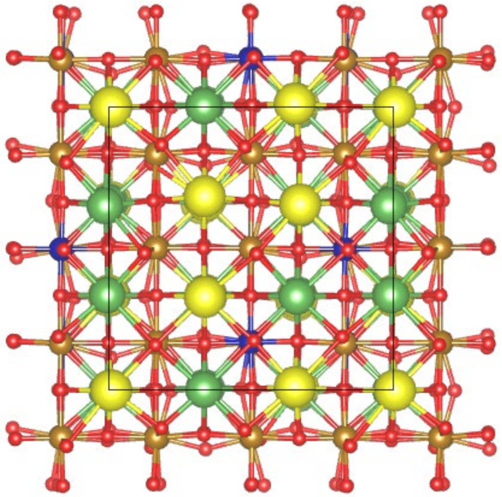
$(La_{0.56}Sr_{0.44})(Co_{0.11}Fe_{0.44}Hf_{0.44})O_{3}$

T = 1073 K, P_{O2} = 10^{10} atm (η = 0.5 V)



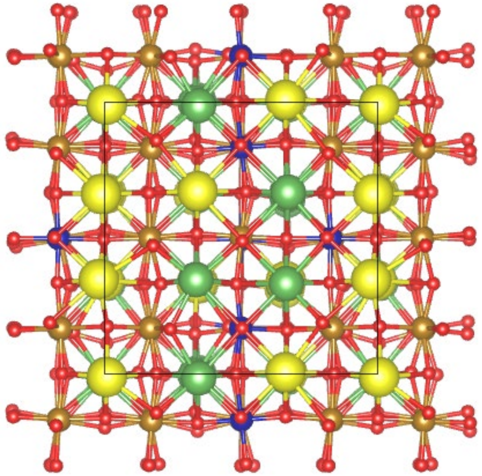
$(La_{0.58}Sr_{0.42})_{0.77}(Co_{0.44}Fe_{0.56})O_{4.2}$

T = 1073 K, P_{O2} = 0.2 atm (η = 0 V)



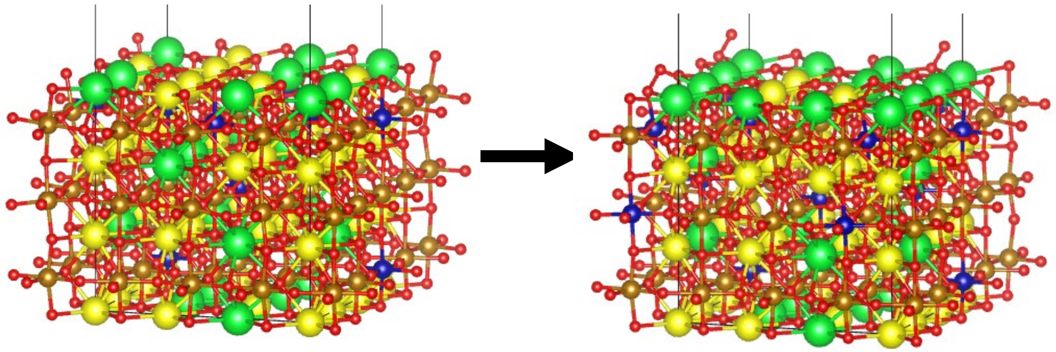
$(La_{0.44}Sr_{0.56})(Co_{0.22}Fe_{0.78})O_{3}$

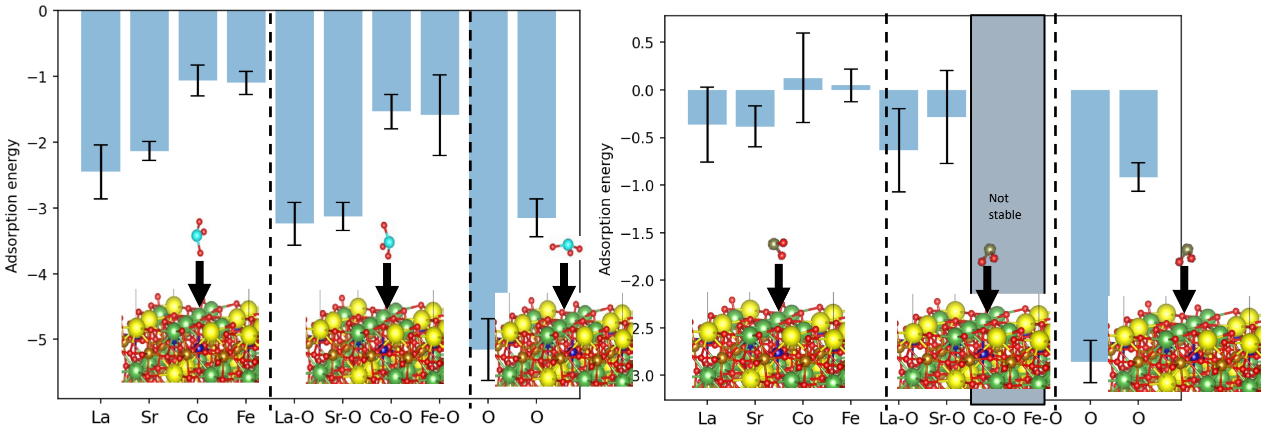
T = 1073 K, P_{O2} = 10^{-10} atm (η = -0.5 V)



$(La_{0.44}Sr_{0.56})(Co_{0.33}Fe_{0.67})O_{2.89}$

Computational summary – modeling for realistic LSCF slabs to investigate Cr & S poisoning pathways





☐ Key takeaways:

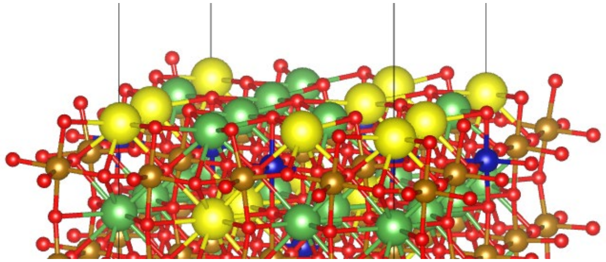
1. Established modified GCMC to identify the most stable surface oxide configuration
2. CrO_3 and SO_2 adsorption energies are mostly **negative** on pristine LSCF.
3. **Surface oxygen Bader charge** and **sub-surface oxygen p-band center** can describe the **electrostatic interaction** between molecules and surface.

☐ Future work:

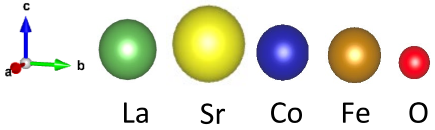
1. Investigate the surface infiltrants stability and how they modify the surface structure
2. Probe the Cr and S poisoning mechanisms on realistic LSCF surfaces with and without infiltrants

Current most stable structures

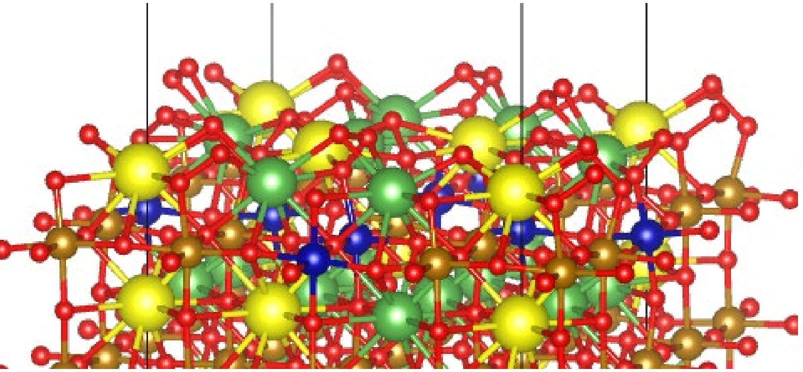
Initial structure



$(La_{0.56}Sr_{0.44})(Co_{0.33}Fe_{0.67})O_3$

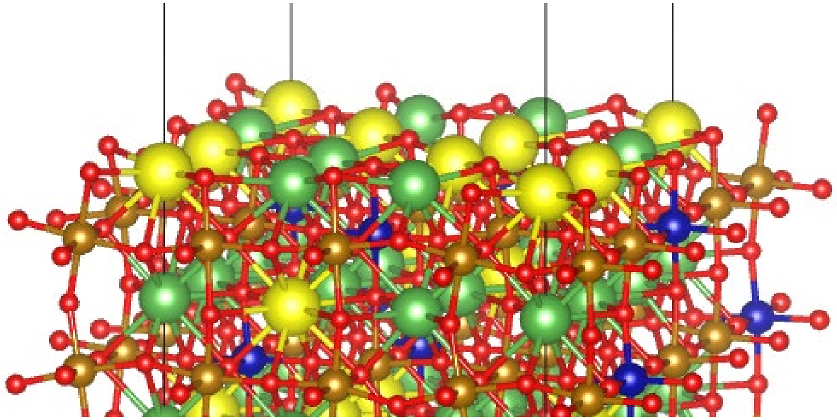


T = 1073 K, P_{O2} = 10^{10} atm (η = 0.5 V)



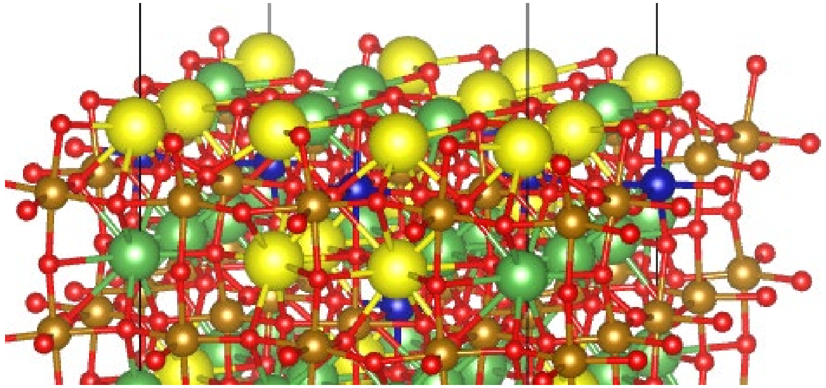
$(La_{0.58}Sr_{0.42})_{0.77}(Co_{0.44}Fe_{0.56})O_{4.2}$

T = 1073 K, P_{O2} = 0.2 atm (η = 0 V)



$(La_{0.44}Sr_{0.56})(Co_{0.22}Fe_{0.78})O_3$

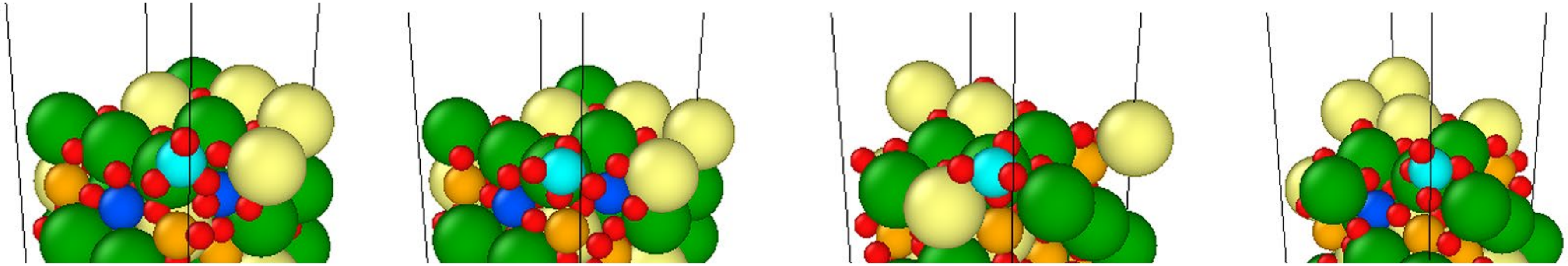
T = 1073 K, P_{O2} = 10^{-10} atm (η = -0.5 V)



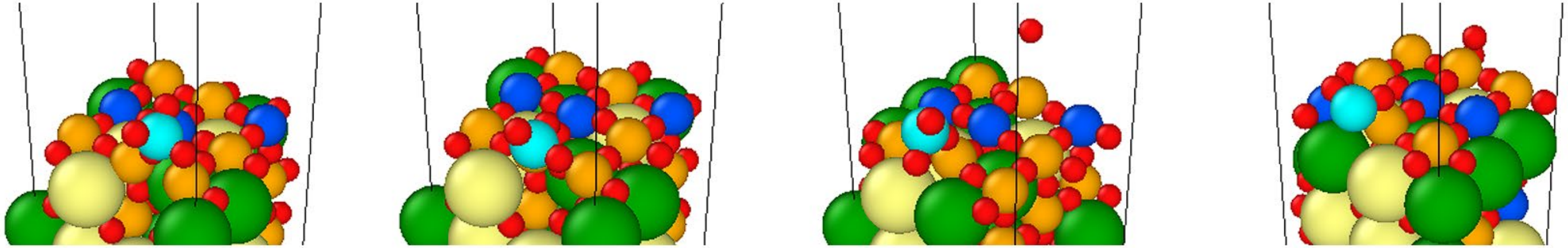
$(La_{0.44}Sr_{0.56})(Co_{0.33}Fe_{0.67})O_{2.89}$

AIMD to study the reaction steps beyond the initial adsorption

a) AO-termination



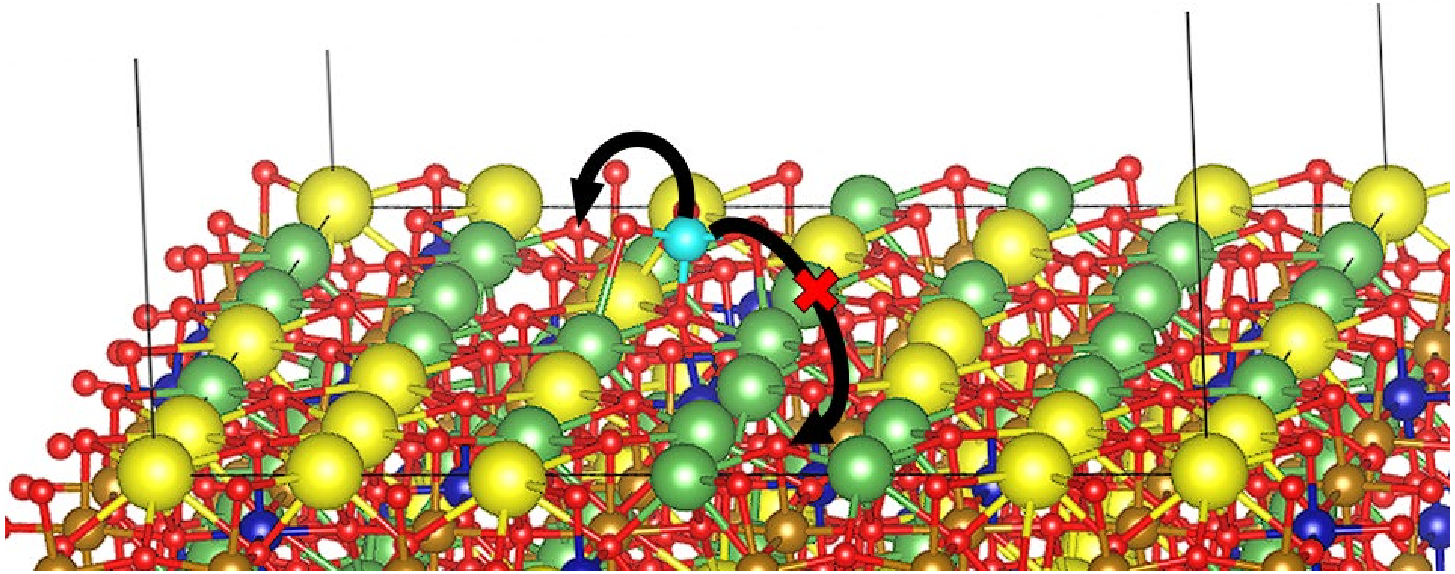
b) BO_2-termination

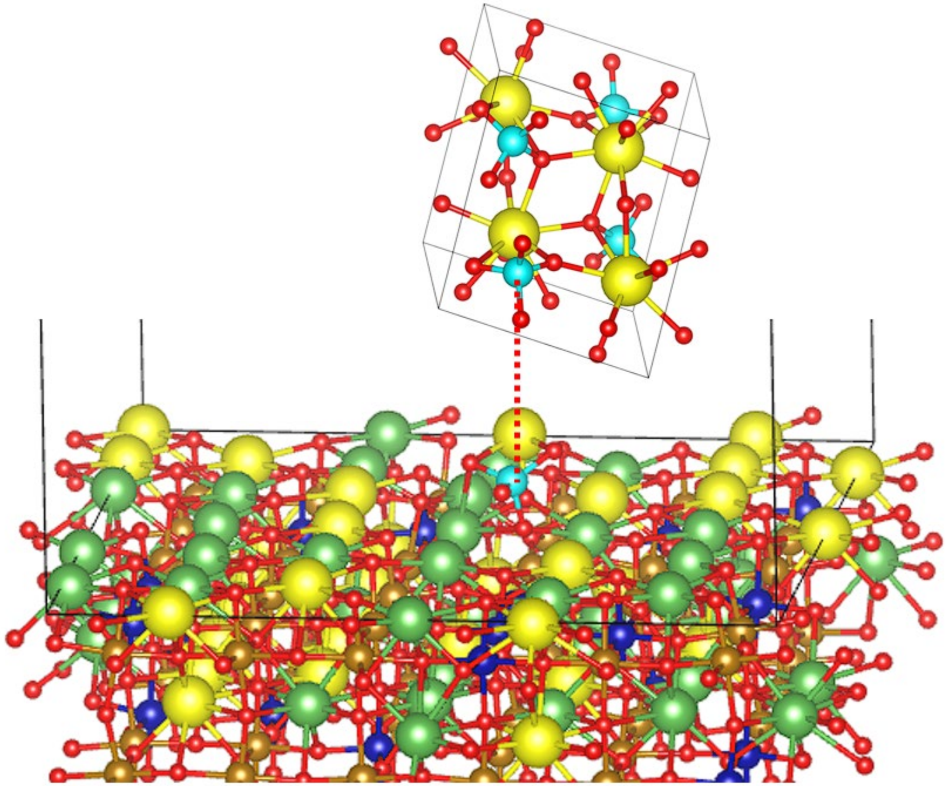


1.5 ps 2.7 ps 3.75 ps 4.95 ps

- AO-terminated surface: molecules are anchored on the adsorption site with a rotational degree of freedom
- BO_2-terminated surface: molecules are more likely to diffuse on the surface

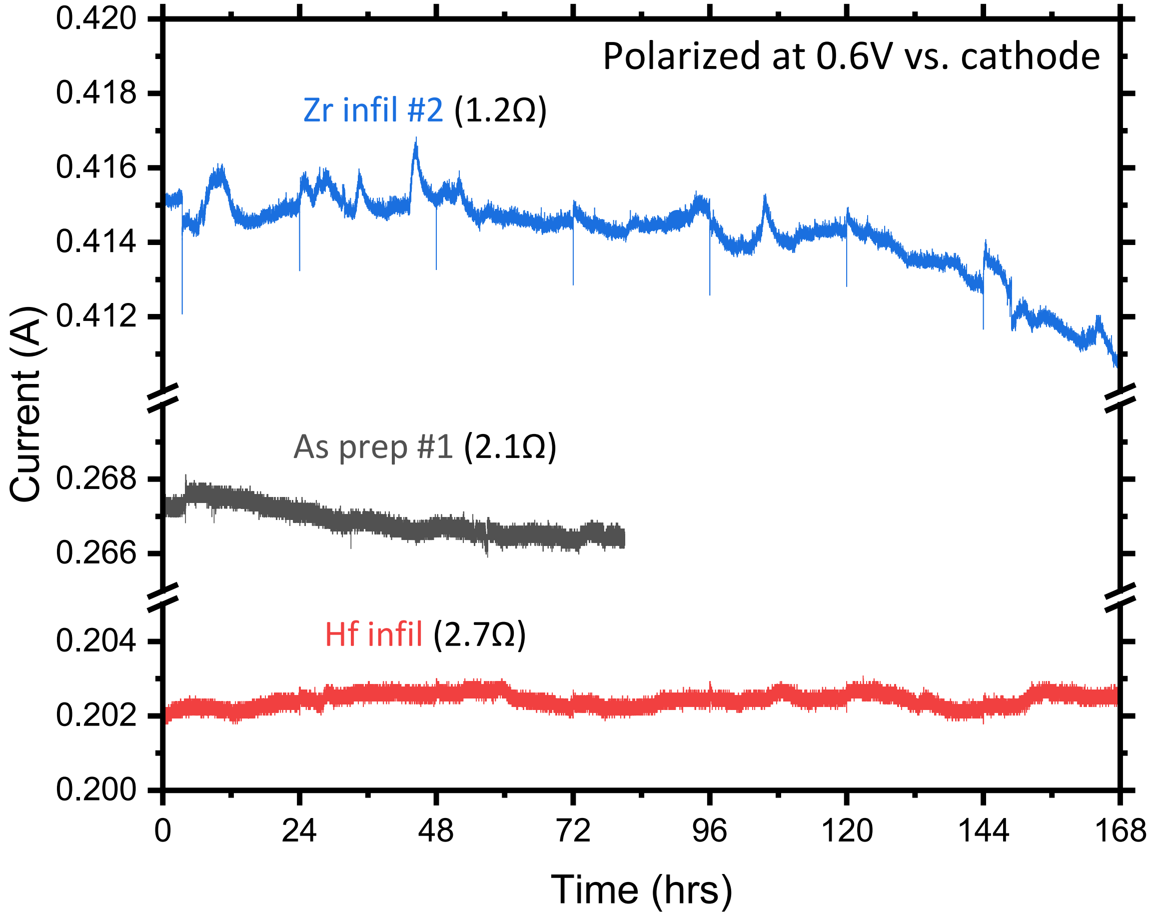
IAP MD to study the reaction steps beyond the initial adsorption

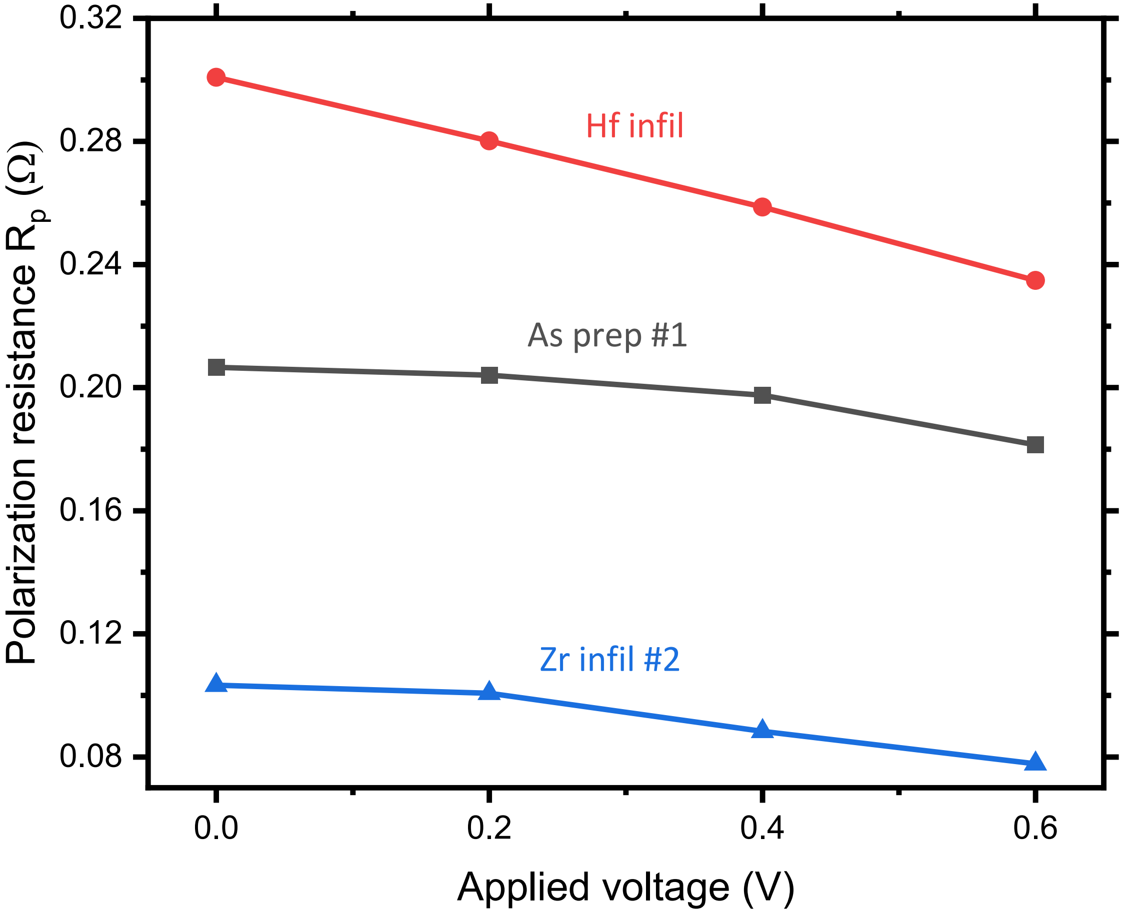




- □ Given a larger distance between molecules, molecule surface diffusion is also possible for AO-terminated surfaces. However, the molecule diffusion always require a Sr atom to be present next to the site where diffusion is taking place.
- □ Given a distorted LSCF slab with one surface Sr atom coming out of the surface, this distorted surface Sr atom can interact with the adsorbed molecules. The structure can be mapped to a $SrCrO_4$ unit.

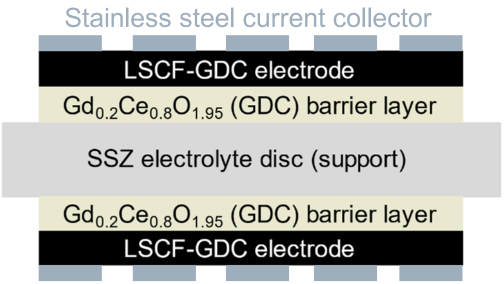
Chronoamperometry analysis – Hf infiltration stabilizes current profile



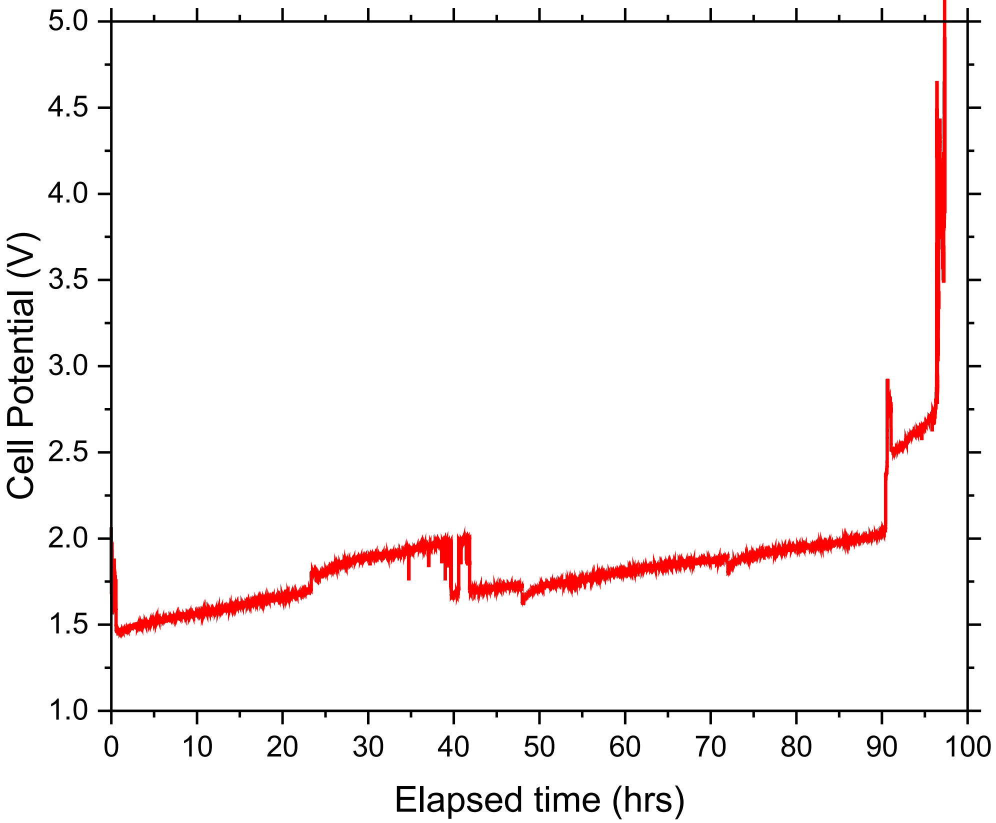


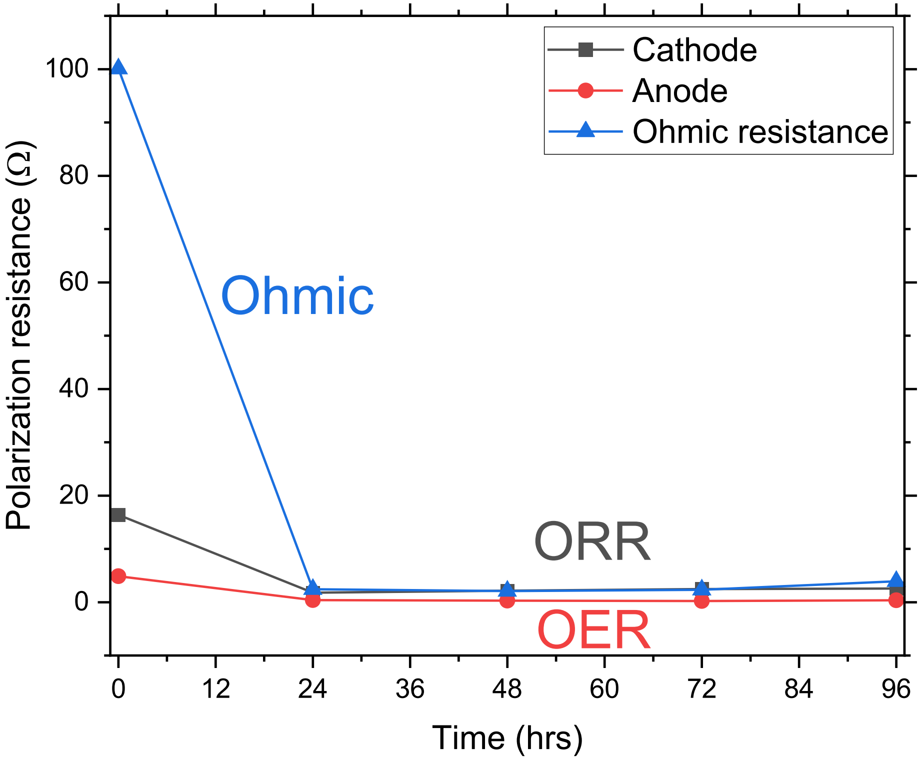
- ☐ As prepared cells show a steady decrease in current vs. time
- ☐ Hf infiltrated cell shows no current change over 7 days – fluctuations due to furnace
- ☐ Zr infiltrated cell shows initial stabilization but then sharp degradation
- ☐ All cells show a linear dependence of R_p on applied voltage

Cr poisoning initial tests – degradation source unknown



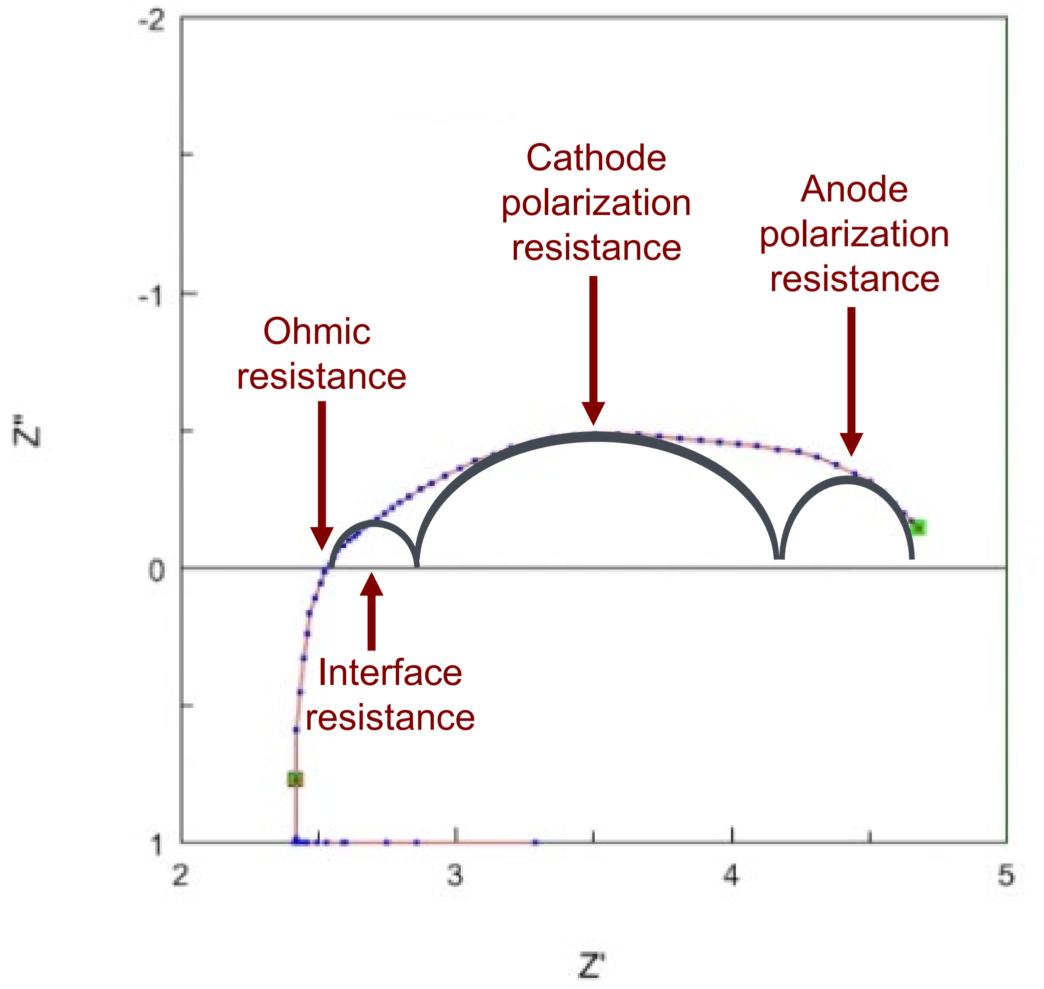
LSCF: $(La_{0.6}Sr_{0.4})_{0.95}Co_{0.2}Fe_{0.8}O_3$

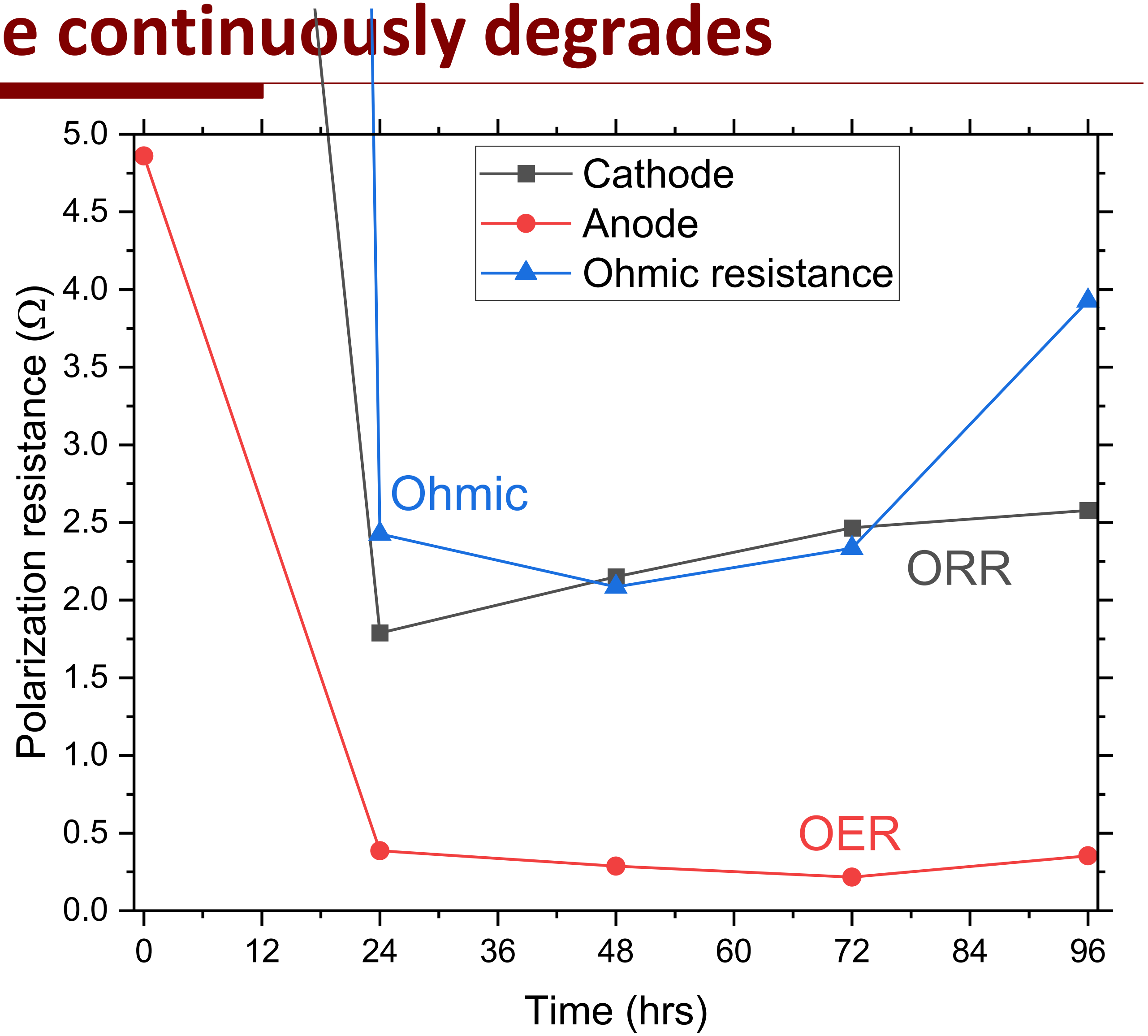




- Constant current measurements at 600mA
- Cell potential shows steady increase with time – degradation of cell?
- Analysis (see also next slide) indicates that initially cell improves a lot
  - Then see degradation of anode, cathode and stainless steel mesh

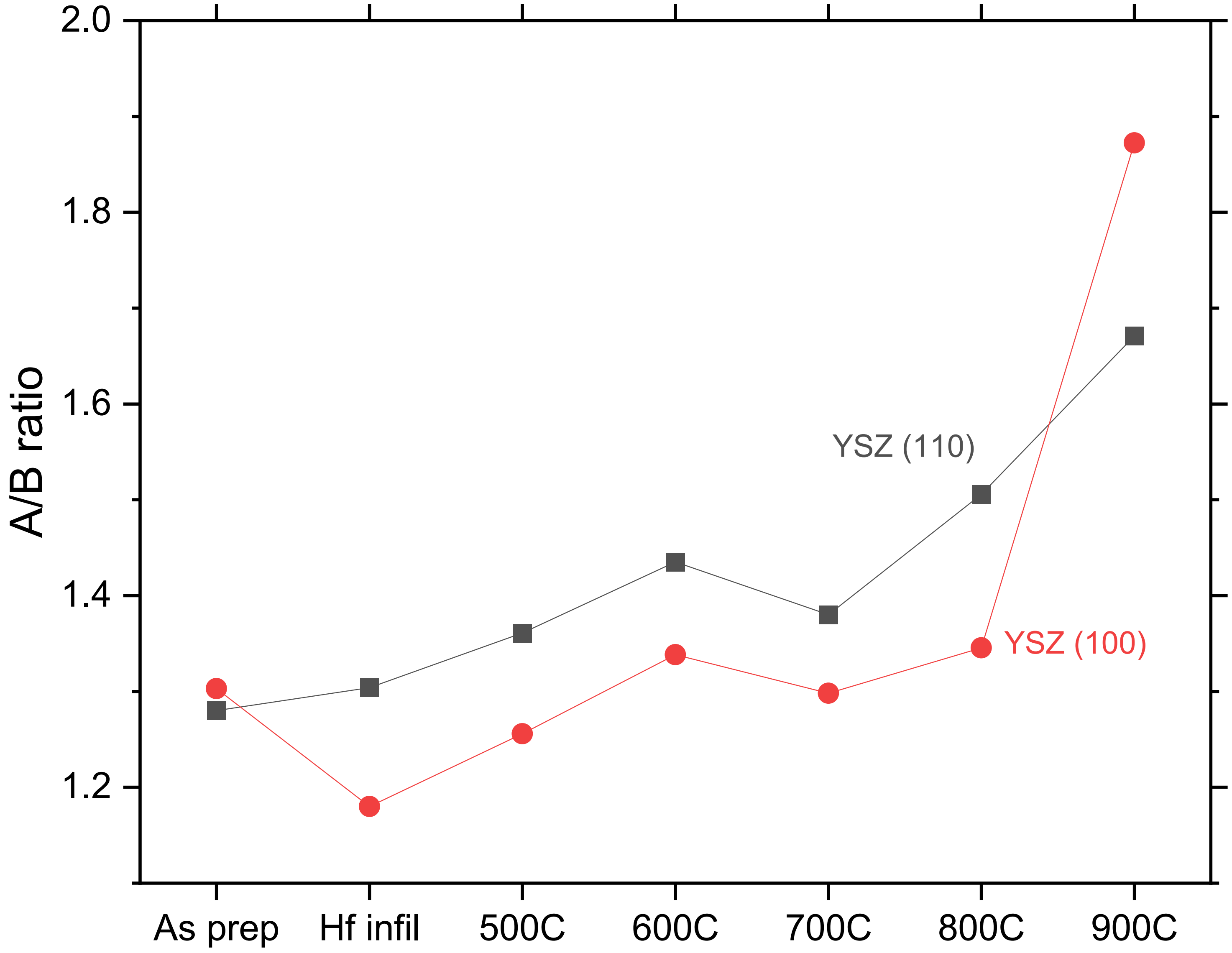
EIS analysis – ORR performance continuously degrades



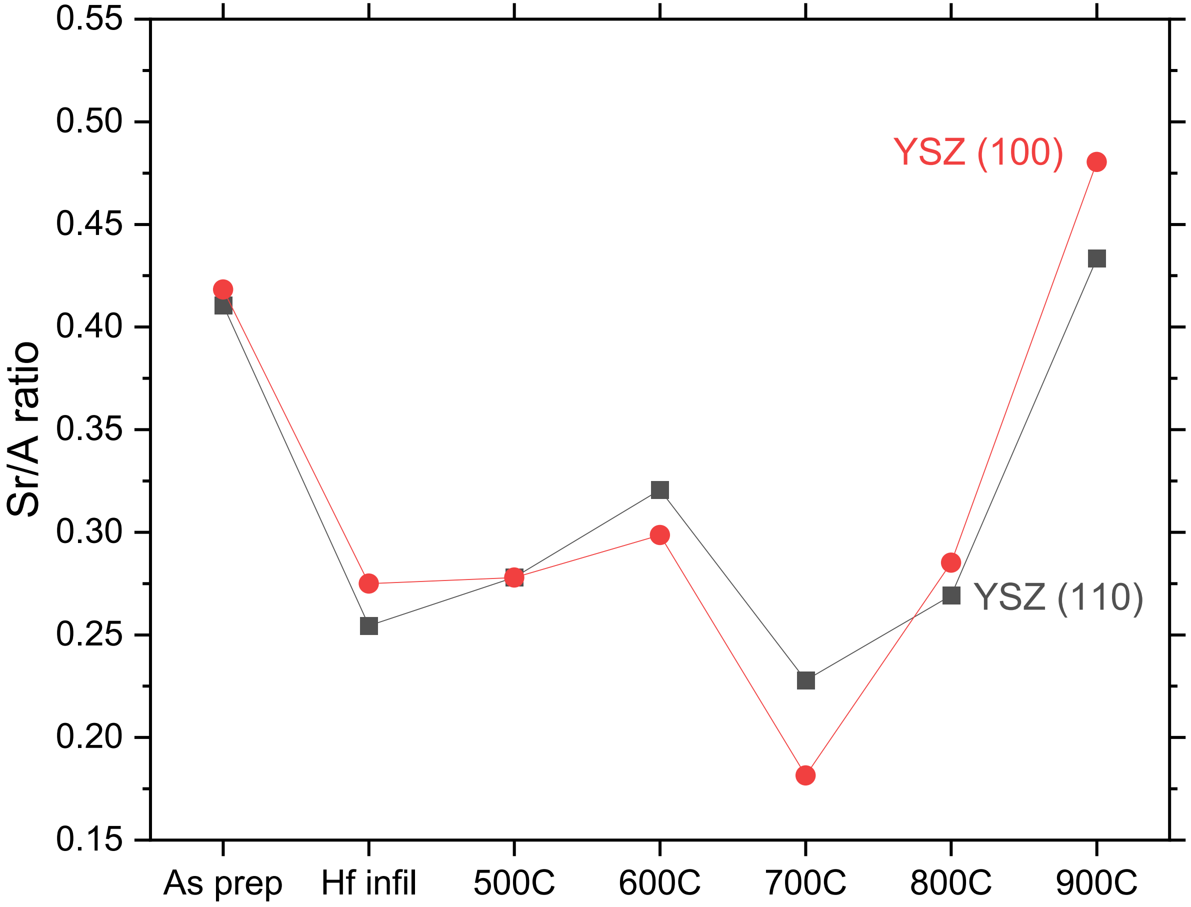


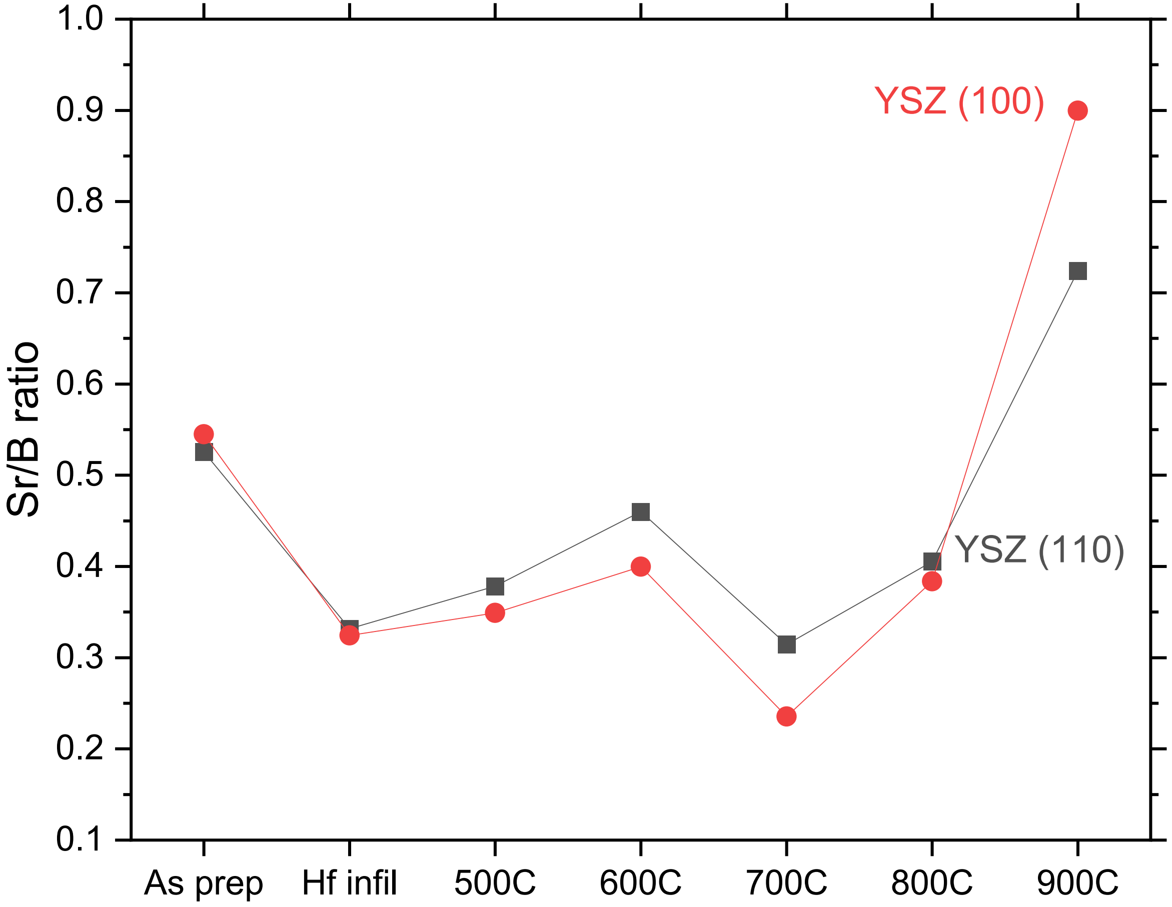
- □ ORR shows steady increase in polarization resistance – possibly due to Cr deposition

Comparison – YSZ100 (Hf) vs. YSZ110 (no Hf)

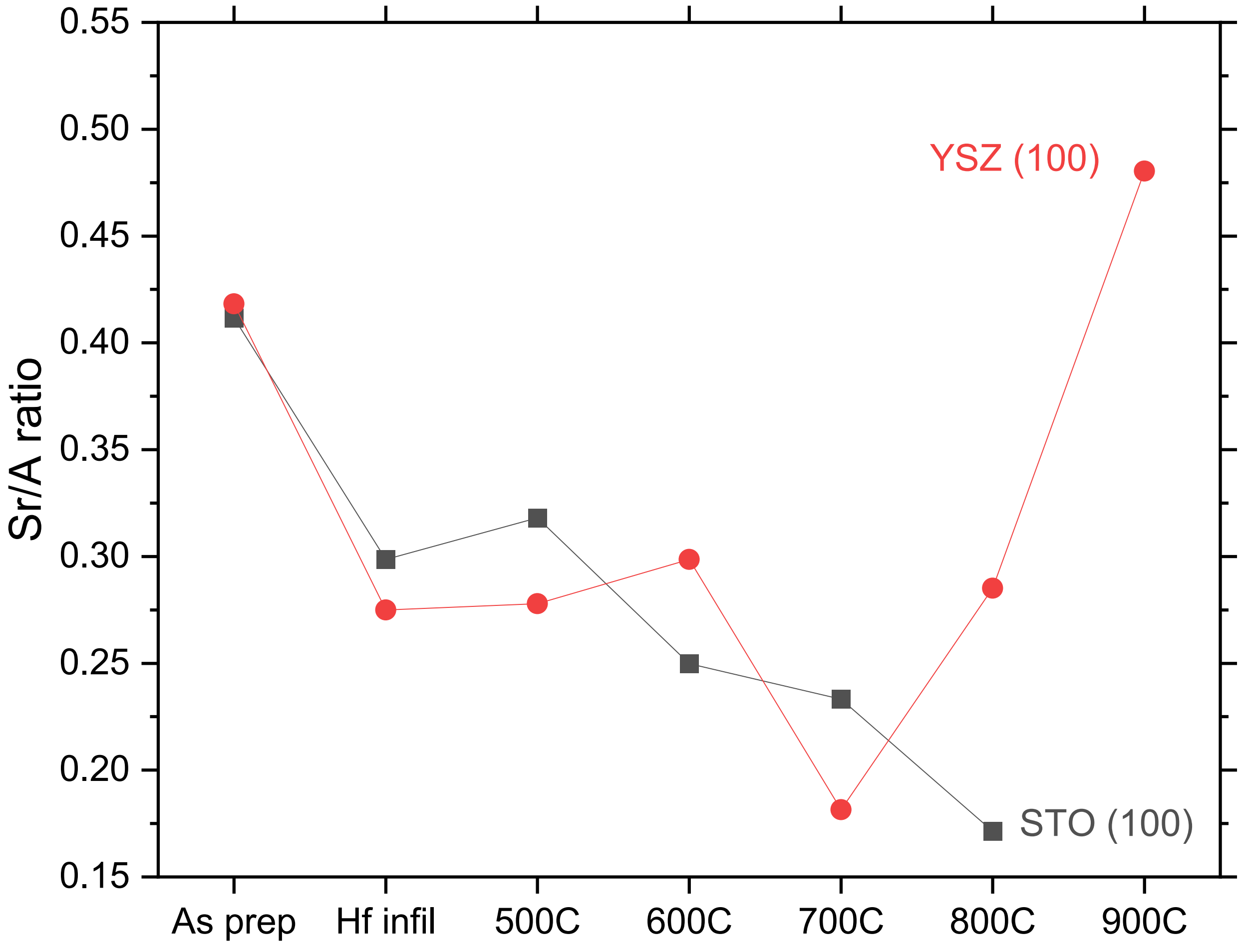


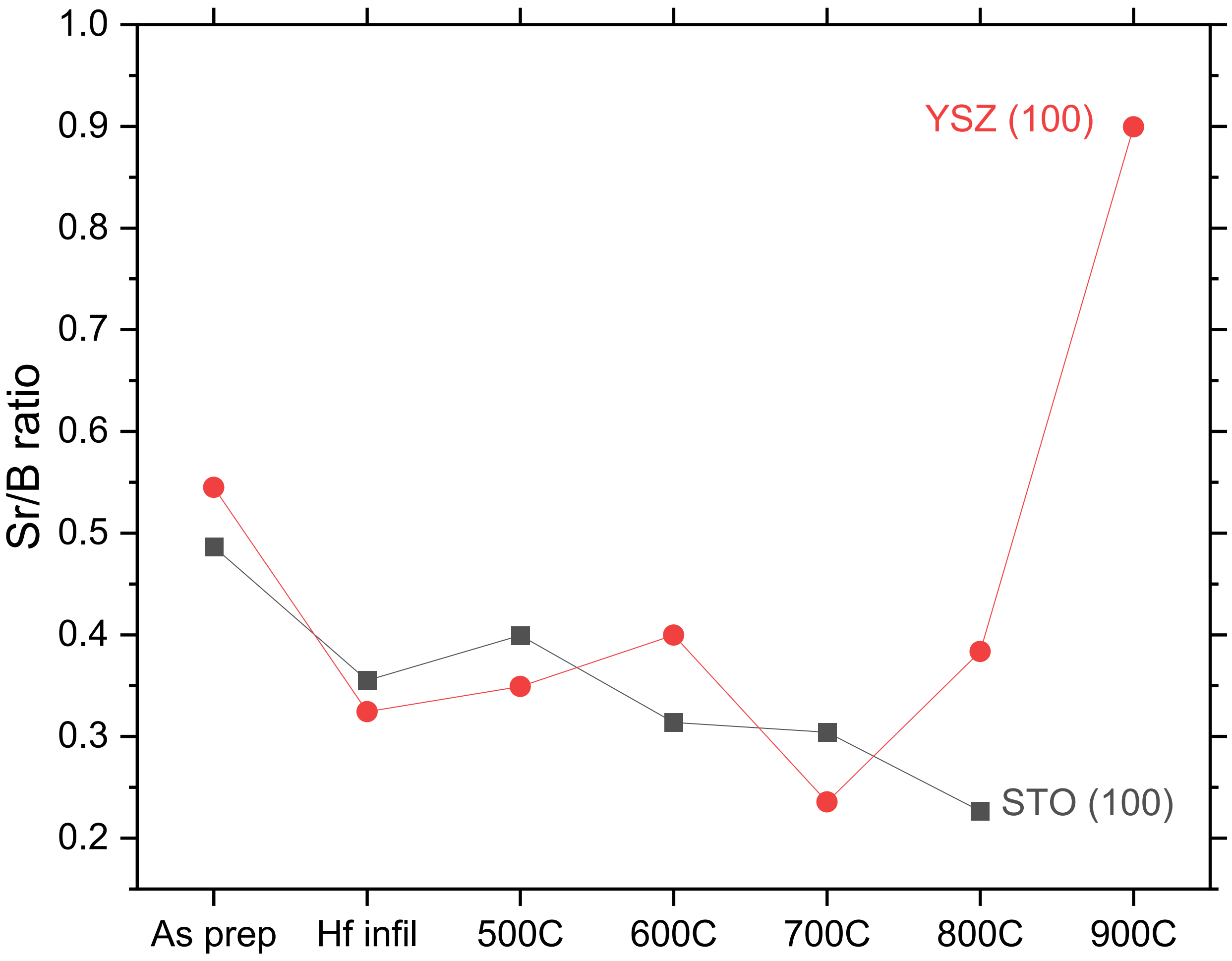
Comparison – YSZ100 (Hf) vs. YSZ110 (no Hf)



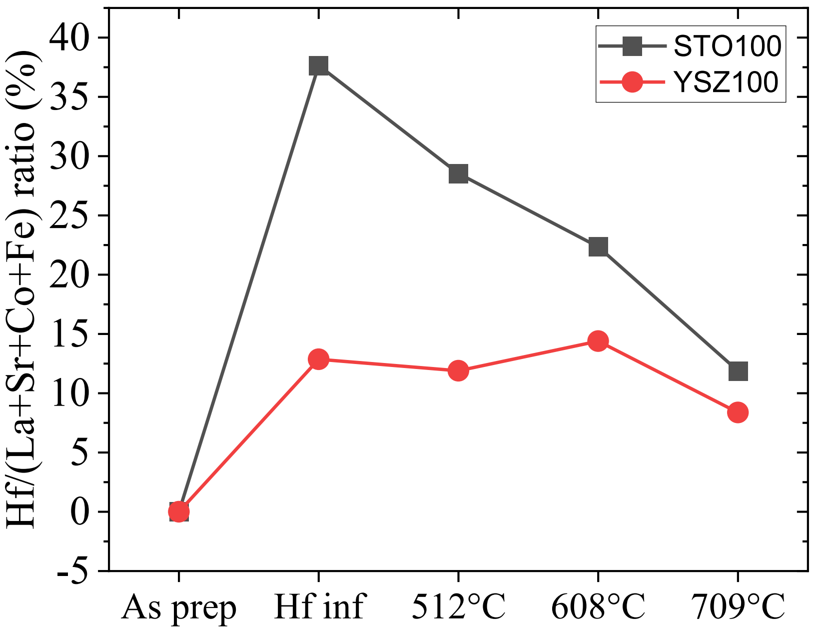


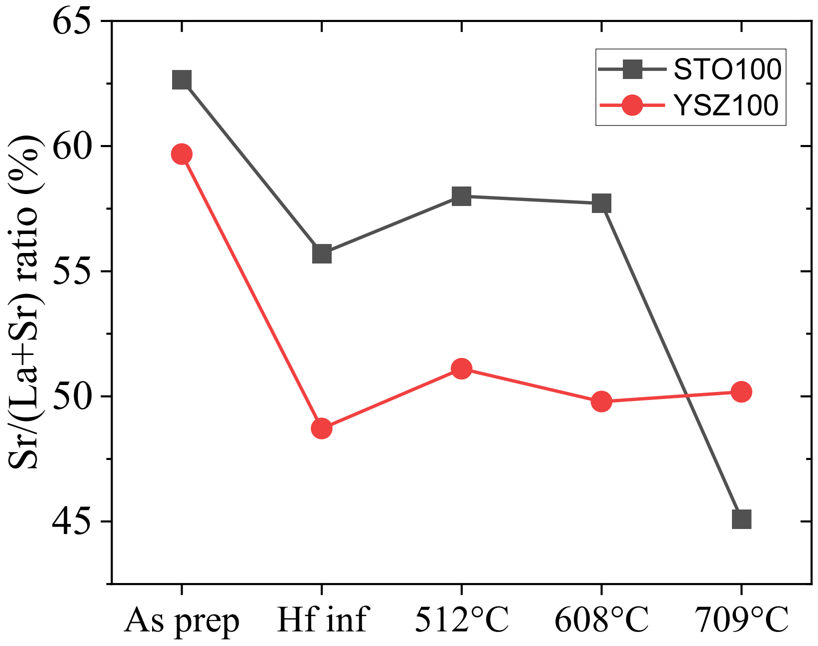
Comparison – poly vs epi Hf infiltrated

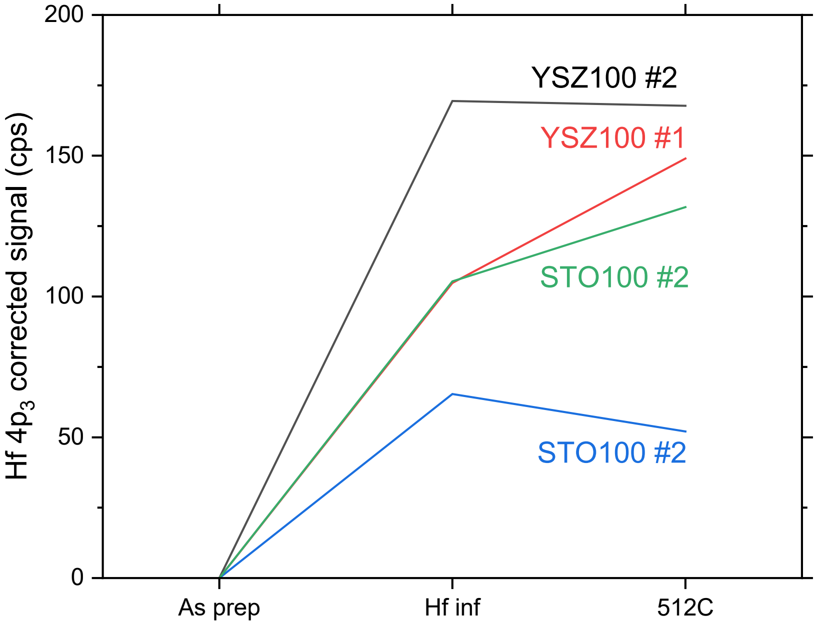




June 2023 – Hf deposition uncontrolled





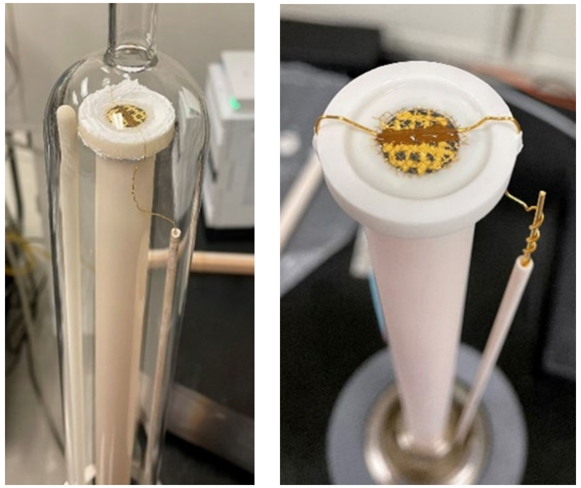


- Both solution infiltration (left, middle) and ALD (right) do not deposit equivalent amounts of Hf on the surfaces
  - Higher rate of diffusion could be simply driven by higher concentration gradient – difficult to isolate stability with such discrepancies
- Next step: deposit HfO_2 via PLD on clean surface in the chamber

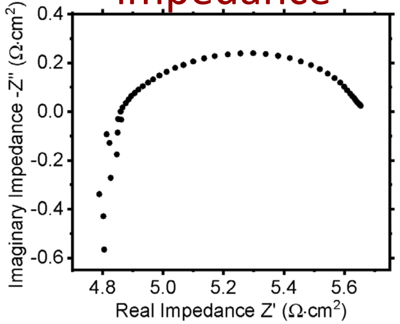
Electrochemical Characterization of Oxygen Electrodes at MIT

Double Chamber Electrochemical Setup

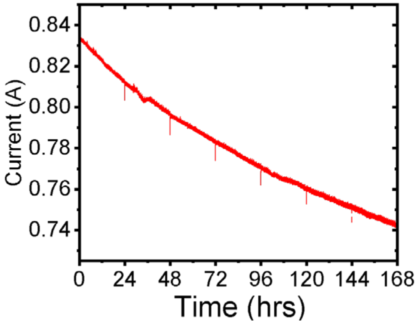
- I-V measurements
- Electrochemical impedance spectroscopy (EIS)



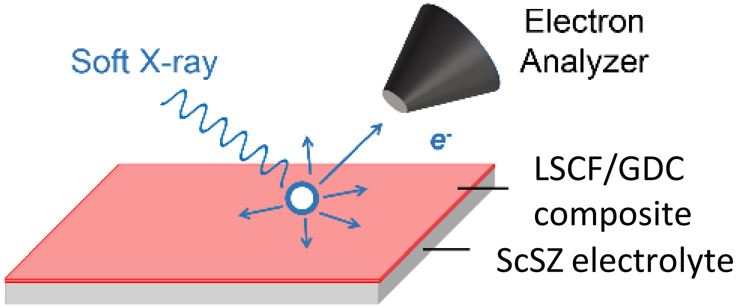
Charge transfer impedance



Temporal stability of current

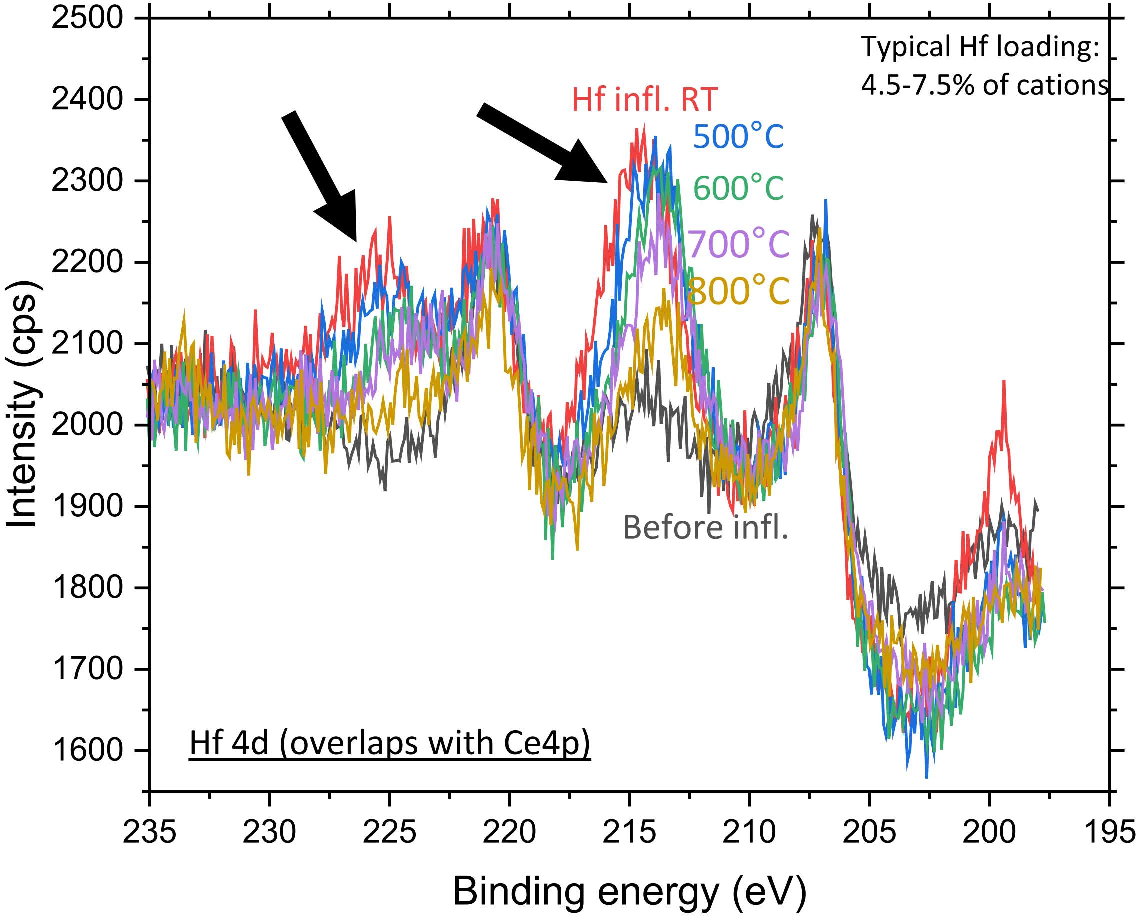


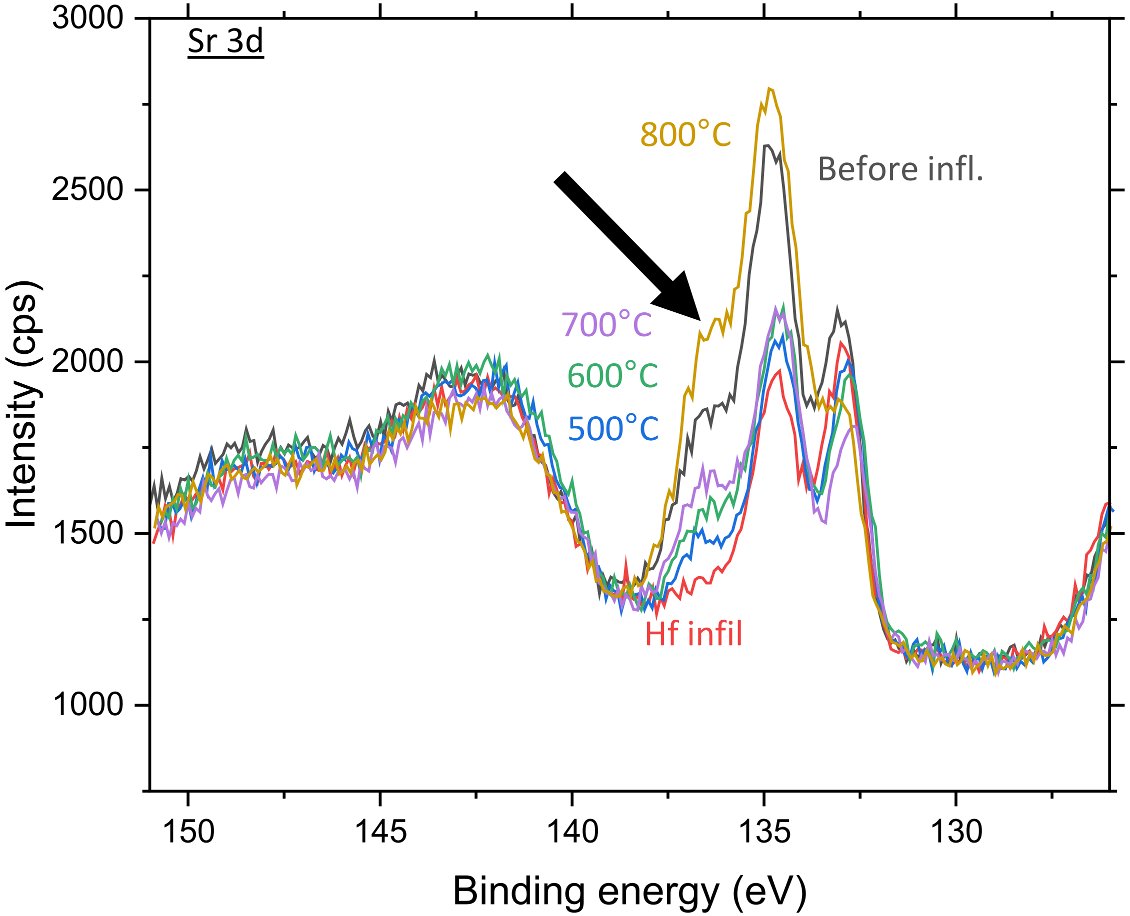
Surface chemical characterization:
XPS, AES, EDX



Structural characterization:
XRD, SEM, TEM

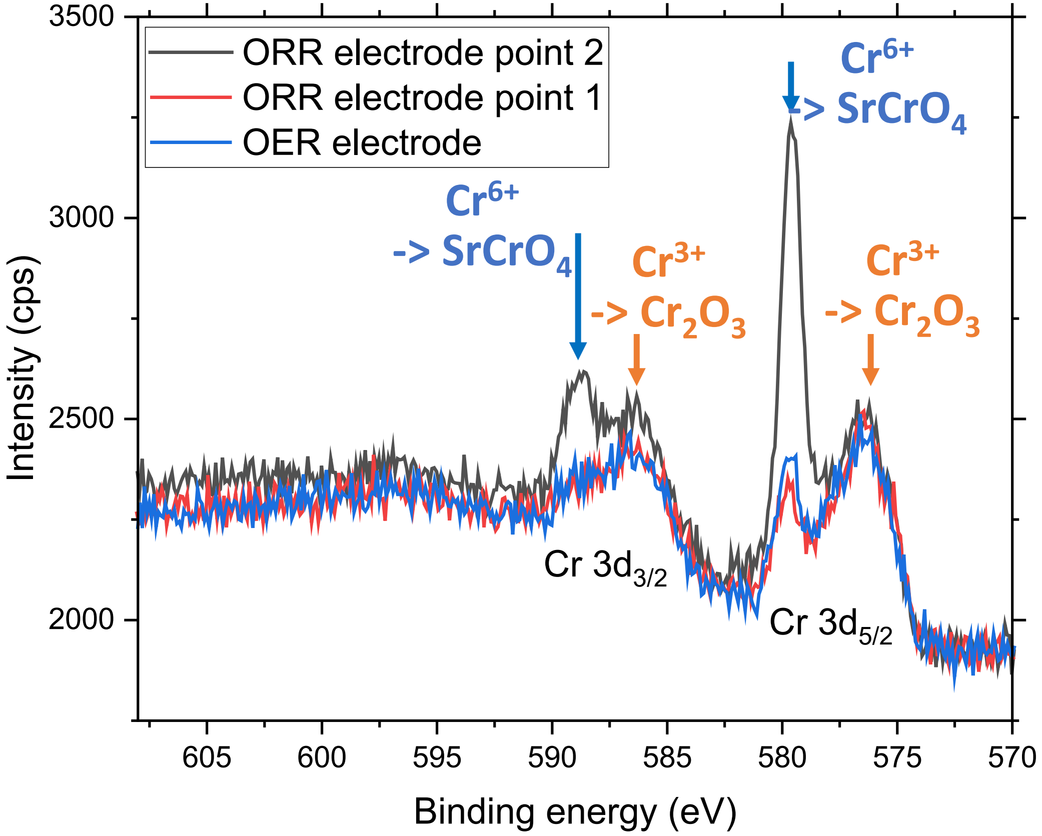
XPS post-mortem analysis – dissolution of dopants at T > 600°C

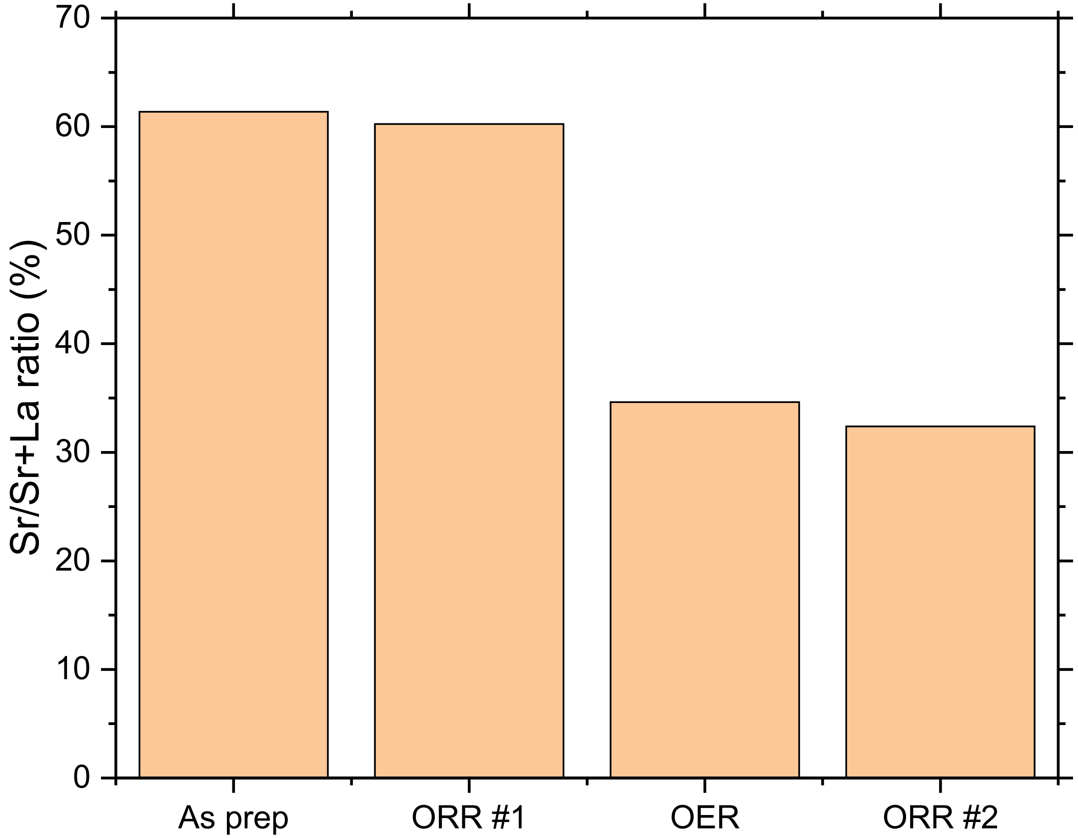




- ☐ Infiltrated Hf/Zr species exhibit dissolution away from surface at T > 700°C)
- ☐ Surface Sr 3d component constant to 500/600/700°C, much larger increase at 800°C (upon near complete dissolution of Hf).

Cr poisoning initial tests – evidence of $SrCrO_4$ and Cr_2O_3 formation





- ☐ XPS shows evidence of $SrCrO_4$ formation – localized, near where mesh touched cell
- ☐ ORR#1, OER – **Cr_2O_3** deposition correlated with increased Sr content
- ☐ ORR#2 - **$SrCrO_4$** deposition correlated with less Sr (Sr clustering into chromate particles?)

XPS analysis based on: Phys. Chem. Chem. Phys., 2015, 17, 4870

Surface infiltrant cation diffusion – lattice vs. grain boundary

Epitaxial thin films

$La_{0.8}Sr_{0.2}Co_{0.8}Fe_{0.2}O_{3-\delta}$ (100)

$SrTiO_3$ (100)

Polycrystalline thin films

$La_{0.8}Sr_{0.2}Co_{0.8}Fe_{0.2}O_{3-\delta}$

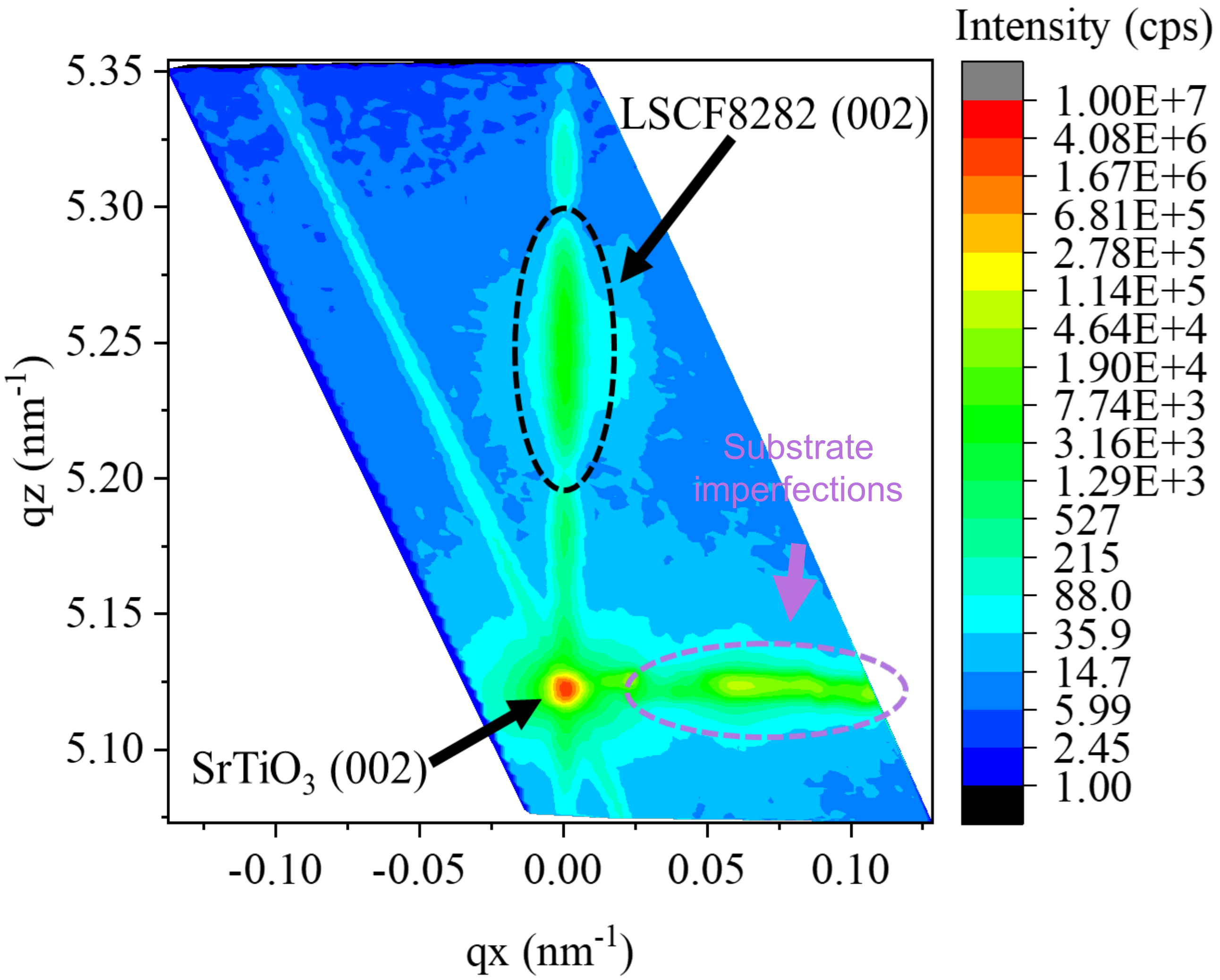
$(Y_2O_3)_{0.08}(ZrO_2)_{0.92}$ (100)

$La_{0.8}Sr_{0.2}Co_{0.8}Fe_{0.2}O_{3-\delta}$

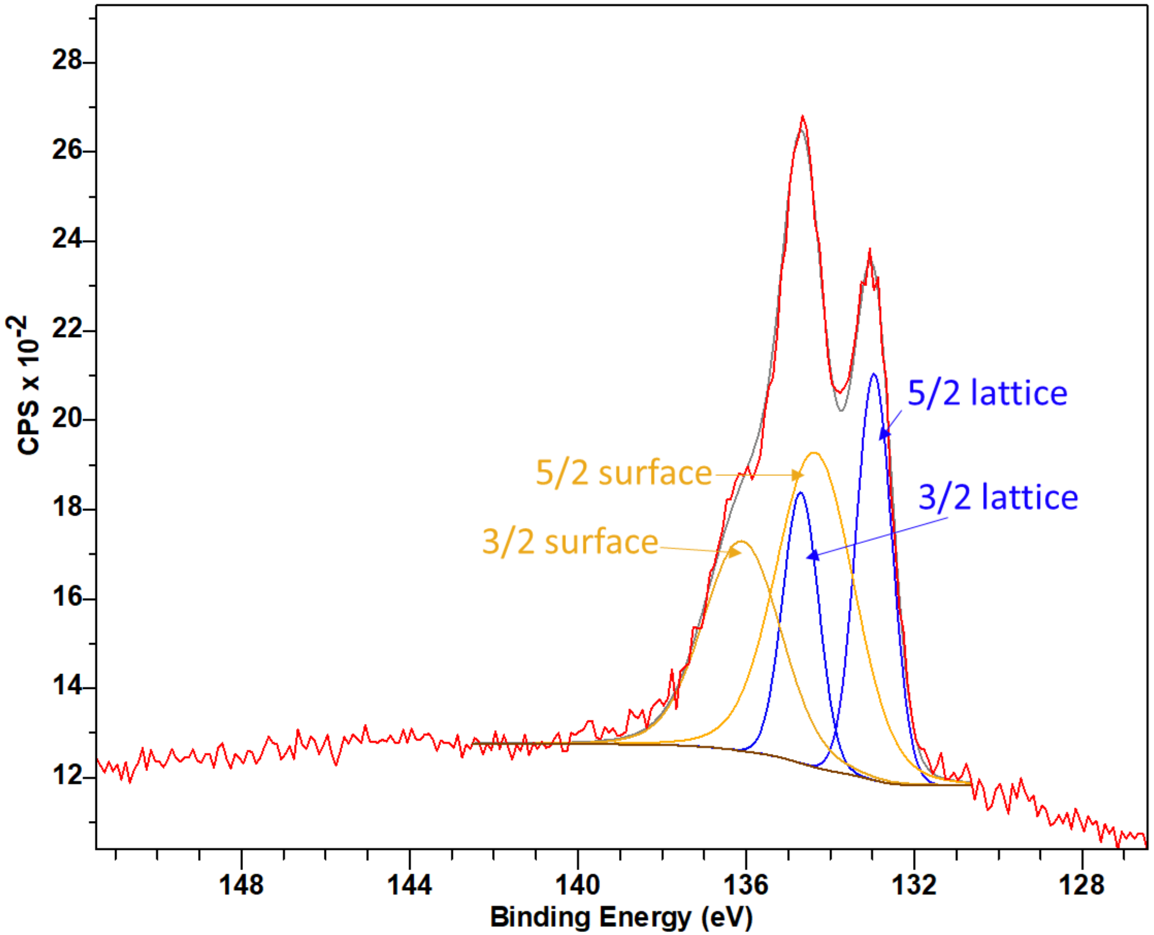
$(Y_2O_3)_{0.08}(ZrO_2)_{0.92}$ (110)

Thin film characterisation - STO

RSM around 002 reflection on STO(100)

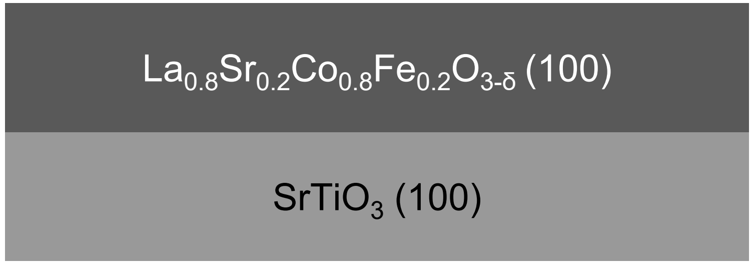


Sr 3d region of LSCF8282 (as prepared) on STO(100)



Surface infiltrated cation diffusion – lattice vs. grain boundary

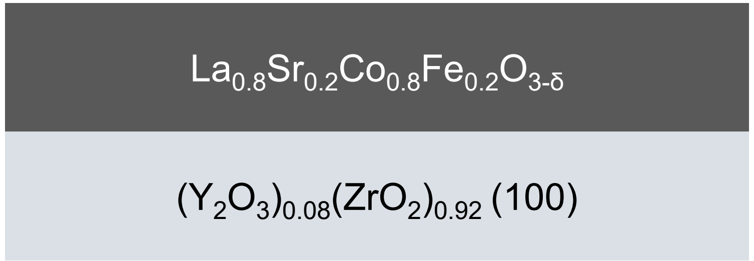
Epitaxial thin films



$La_{0.8}Sr_{0.2}Co_{0.8}Fe_{0.2}O_{3-\delta}$ (100)

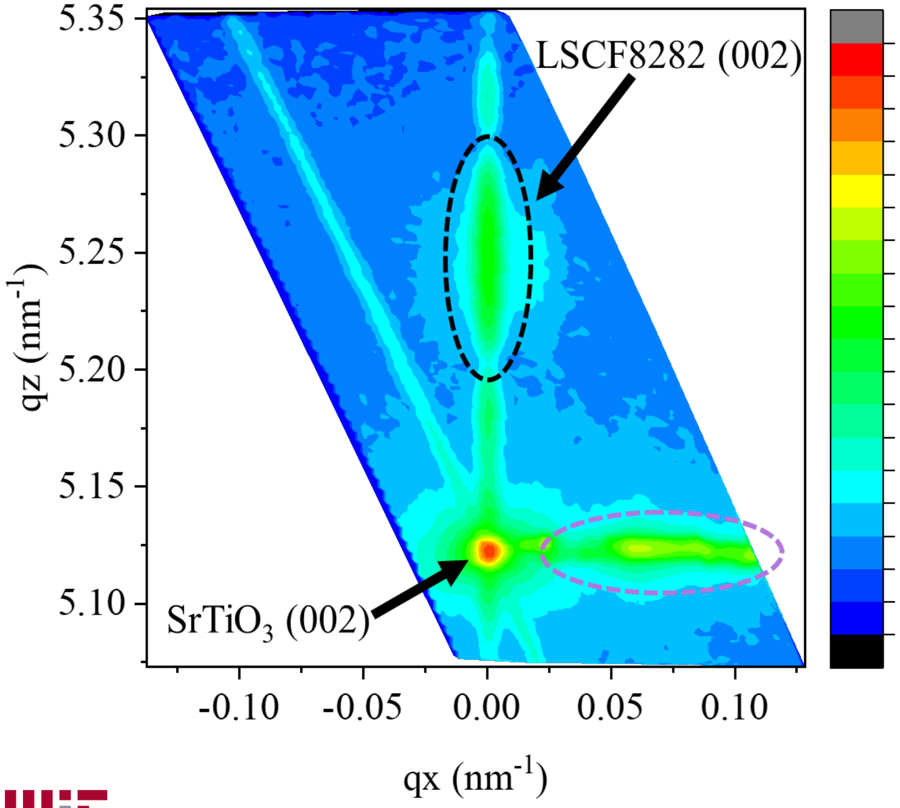
$SrTiO_3$ (100)

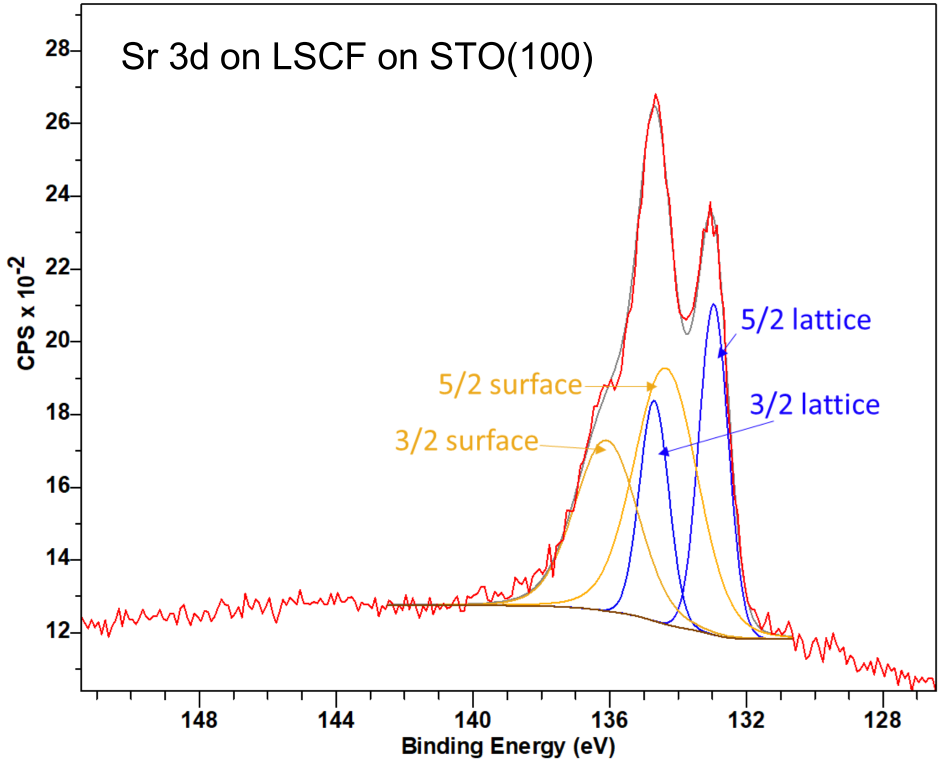
Polycrystalline thin films



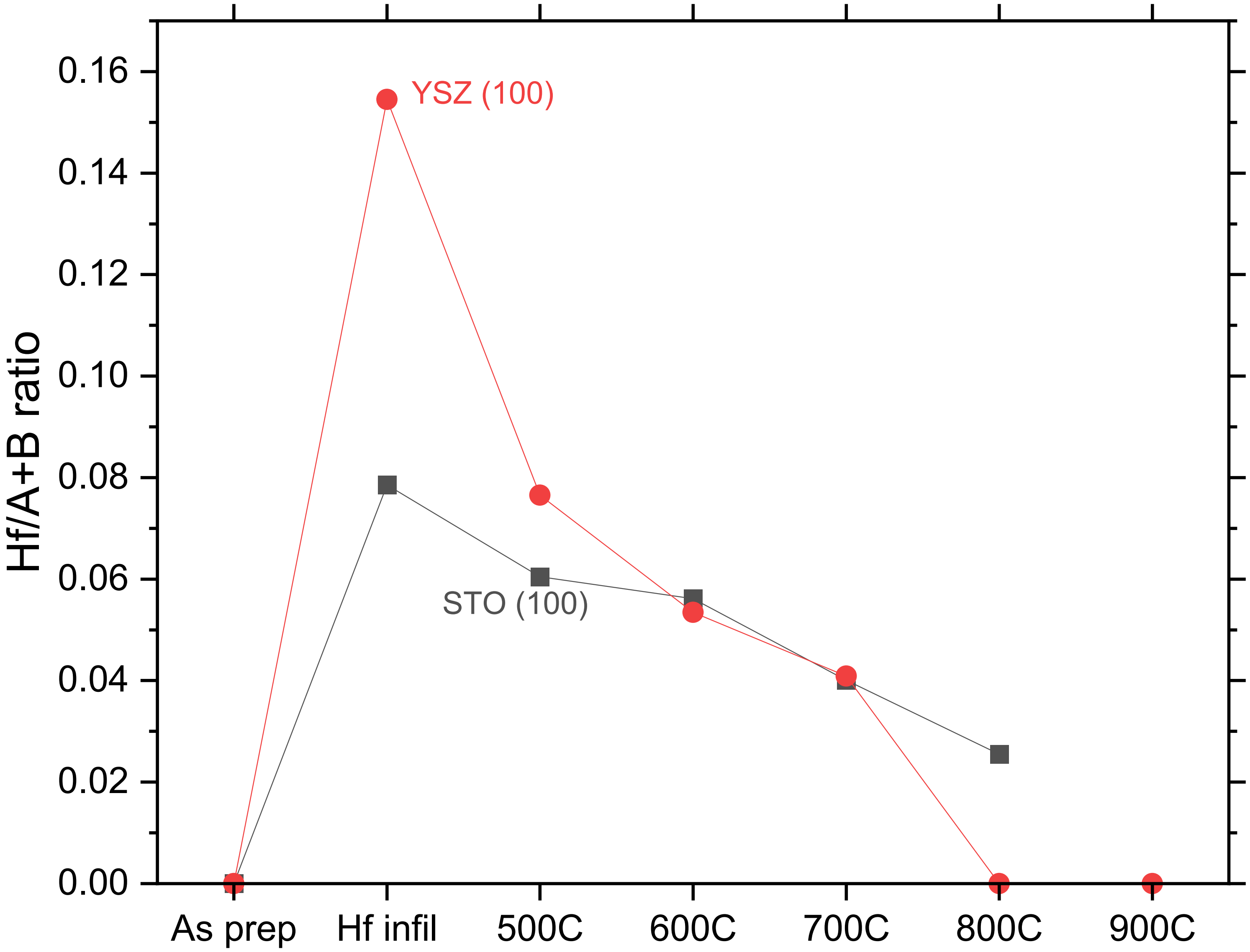
$La_{0.8}Sr_{0.2}Co_{0.8}Fe_{0.2}O_{3-\delta}$

$(Y_2O_3)_{0.08}(ZrO_2)_{0.92}$ (100)

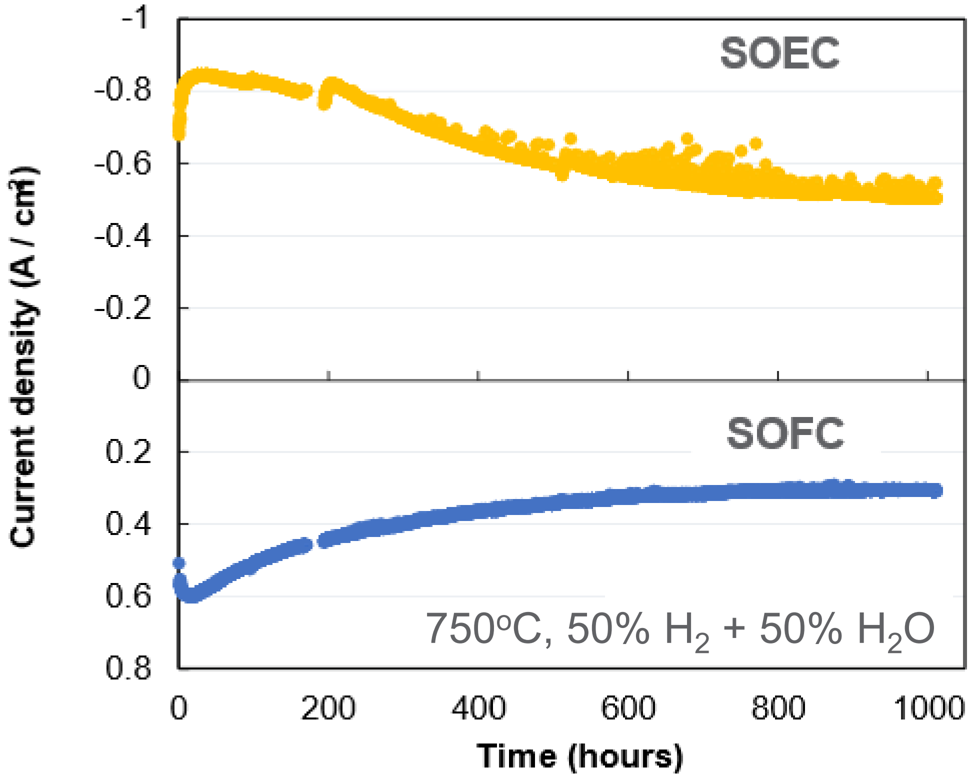


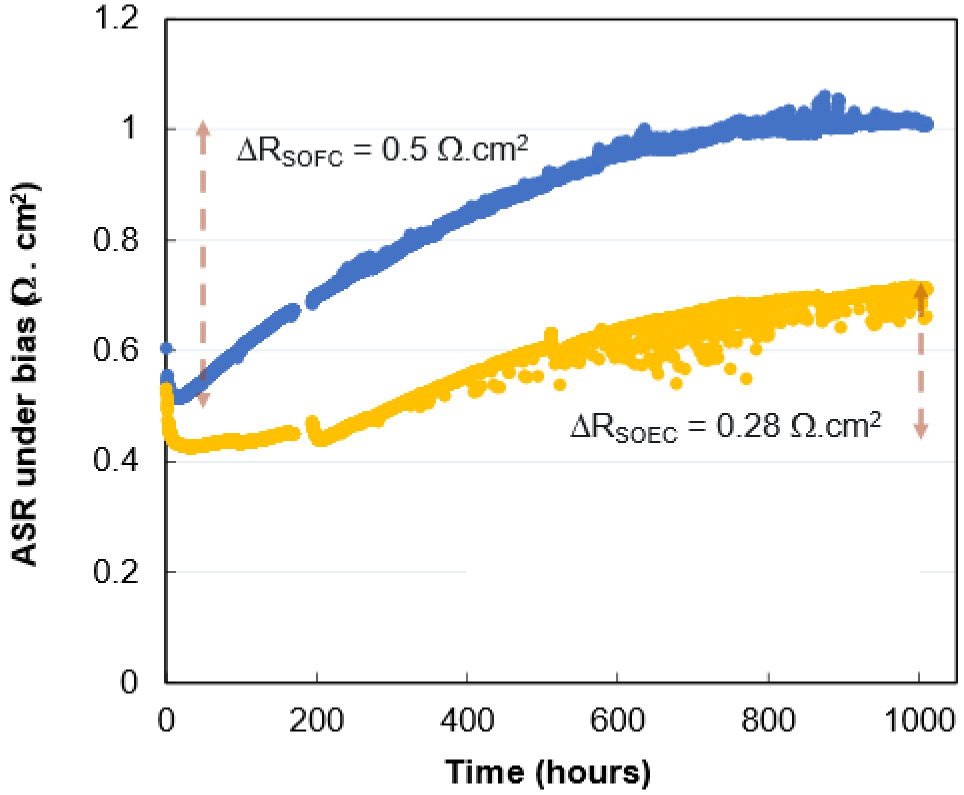


Comparison – poly vs epi Hf infiltrated



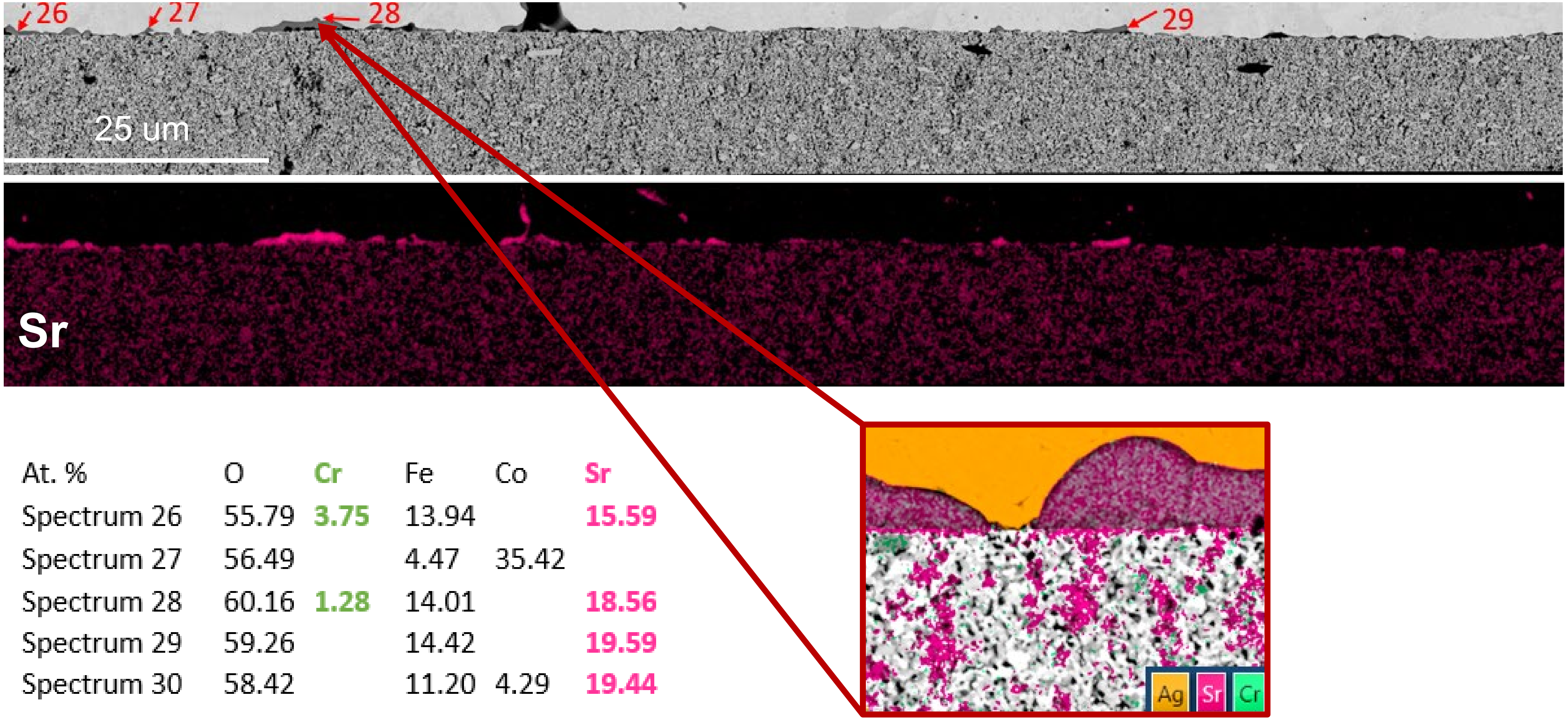
Uncoated SS Interconnect was used as Cr source





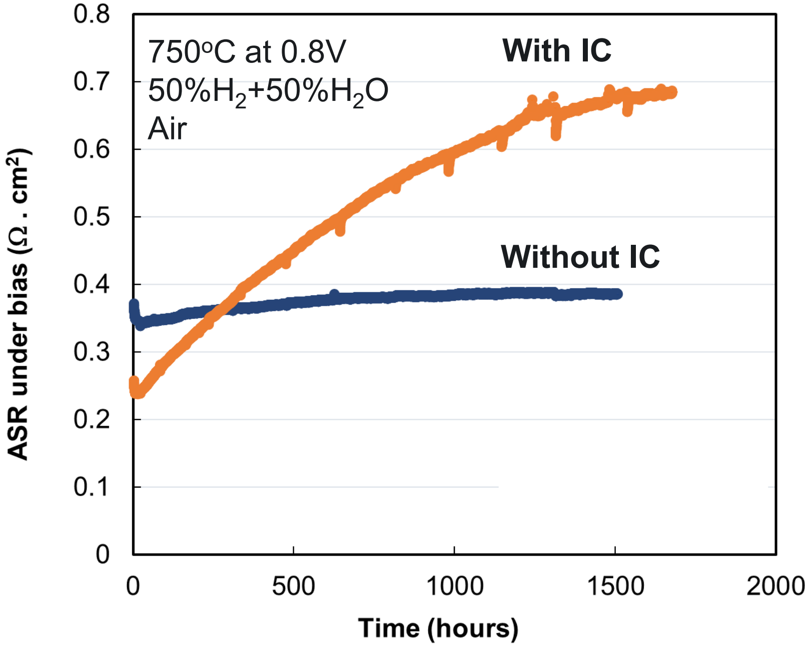
- In this case a 3%H_2O was used during SOFC operation
- Similar degradation rates are observed

Electrode Surface is Sr Enriched, Shows Cr Presence after 1000 hrs

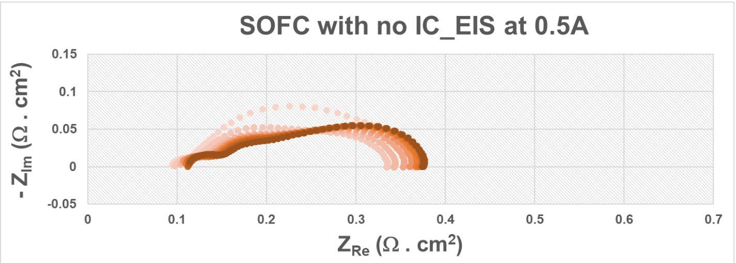


At. %	O	Cr	Fe	Co	Sr
Spectrum 26	55.79	3.75	13.94		15.59
Spectrum 27	56.49		4.47	35.42	
Spectrum 28	60.16	1.28	14.01		18.56
Spectrum 29	59.26		14.42		19.59
Spectrum 30	58.42		11.20	4.29	19.44

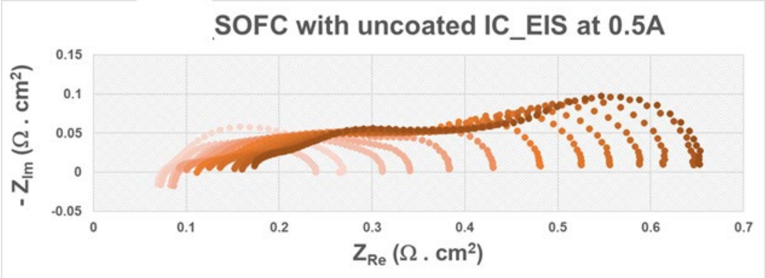
SOFC Testing With and Without Uncoated Interconnect



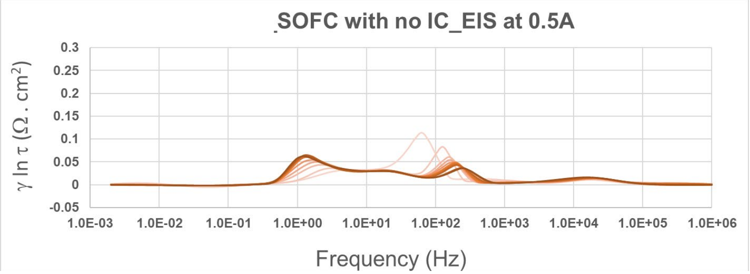
SOFC with no IC_EIS at 0.5A



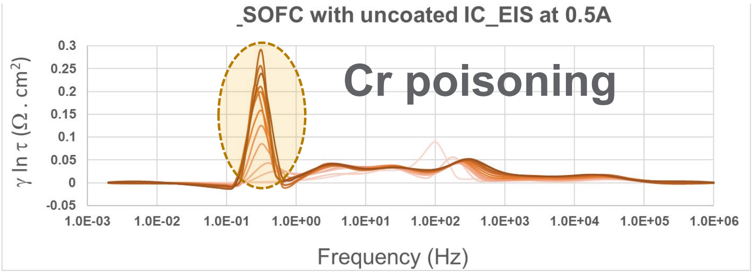
SOFC with uncoated IC_EIS at 0.5A

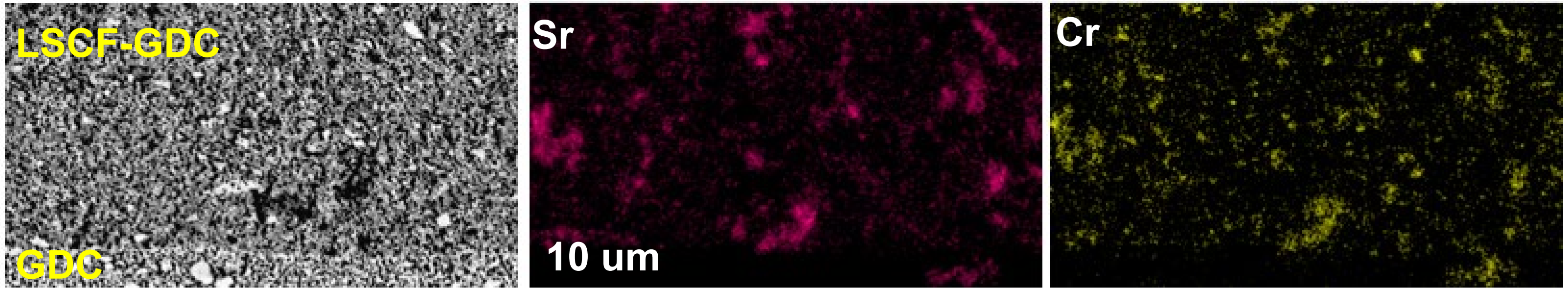


SOFC with no IC_EIS at 0.5A



SOFC with uncoated IC_EIS at 0.5A

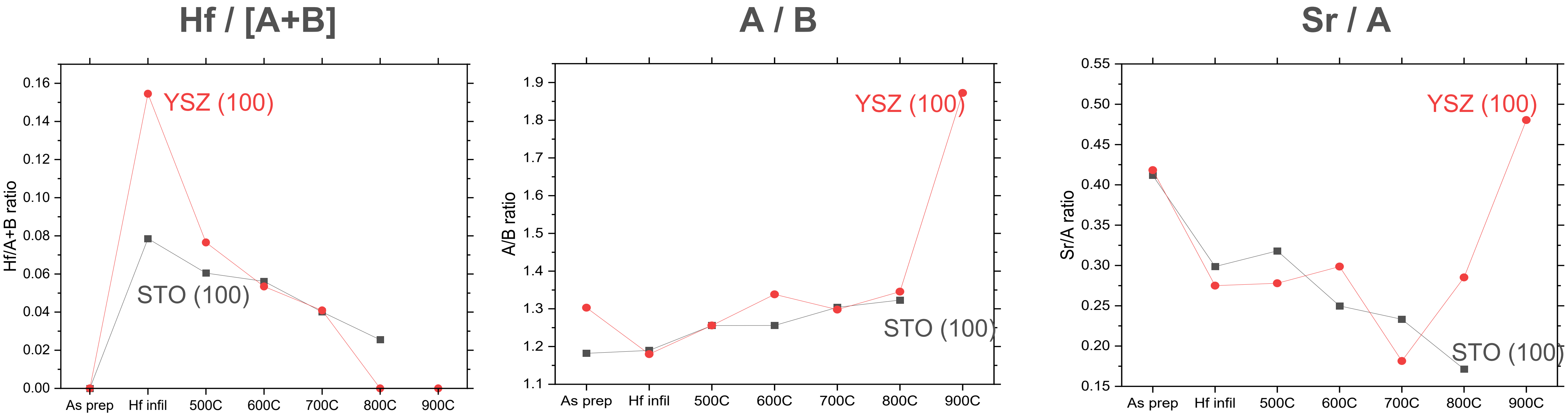




Low frequency process is related to oxygen adsorption or oxygen exchange

Ohmic resistance significantly increased compared to test without interconnect

Surface chemistry on epitaxial vs. polycrystal LSCF with Hf

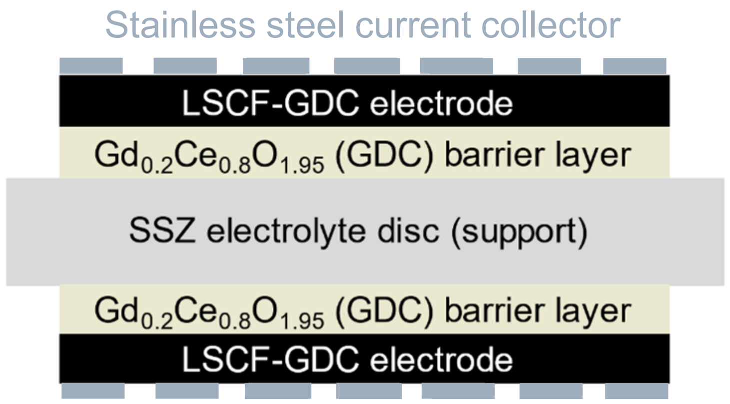

Hf / [A+B]

A / B

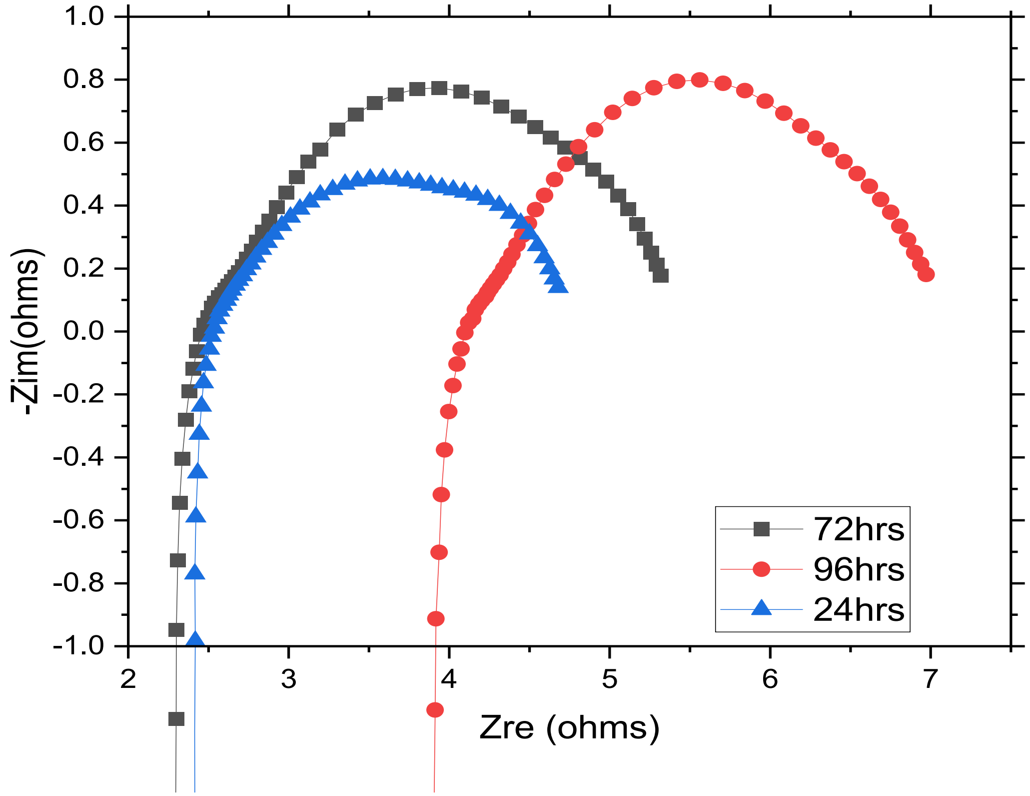
Sr / A

- Very similar results on epitaxial and polycrystal LSCF films, indicating the surface Hf can diffuse into the lattice, and upper limit of stability is 700 ^{o}C.

Cr poisoning initial tests –

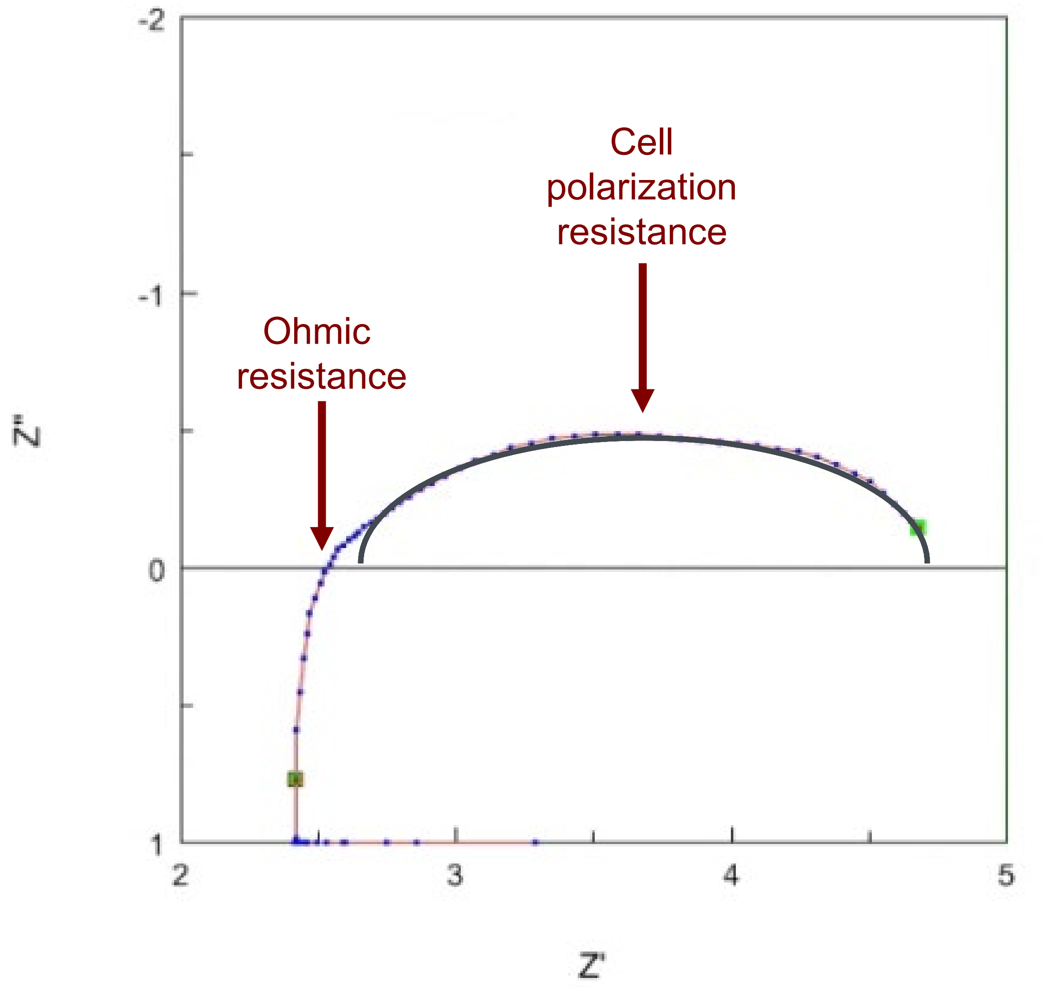


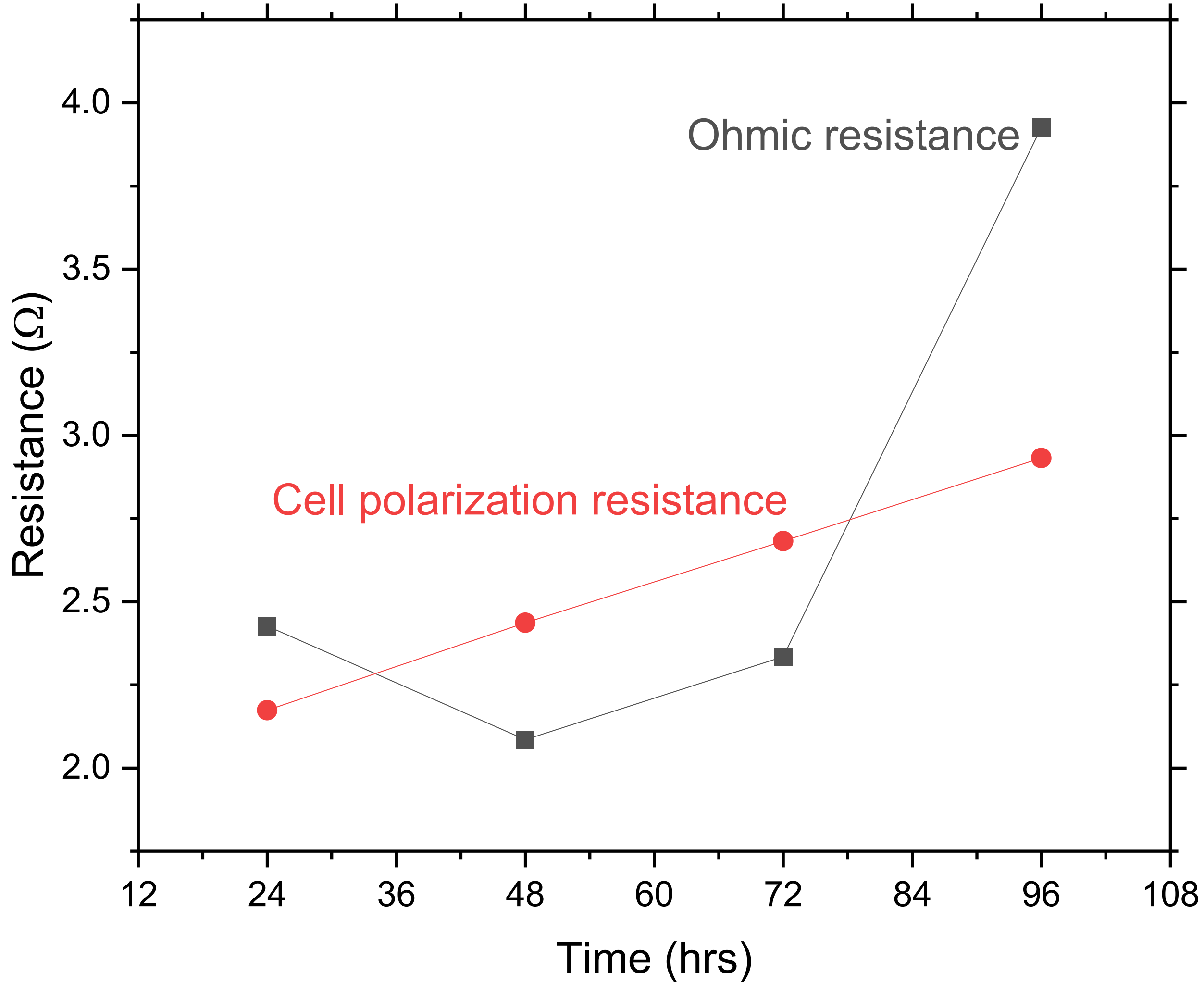
LSCF: $(La_{0.6}Sr_{0.4})_{0.95}Co_{0.2}Fe_{0.8}O_3$



- Constant current measurements at 600mA
- Cell potential shows steady increase with time –
  - degradation of electrode, or of SS current collectors?

Polarization resistance of the electrodes continuously degrades





- Cell polarization resistance shows steady increase with time – possibly due to Cr deposition

Hf infiltration process, and next steps to penetrate deeper into porous LSCF electrodes

Infiltration steps:

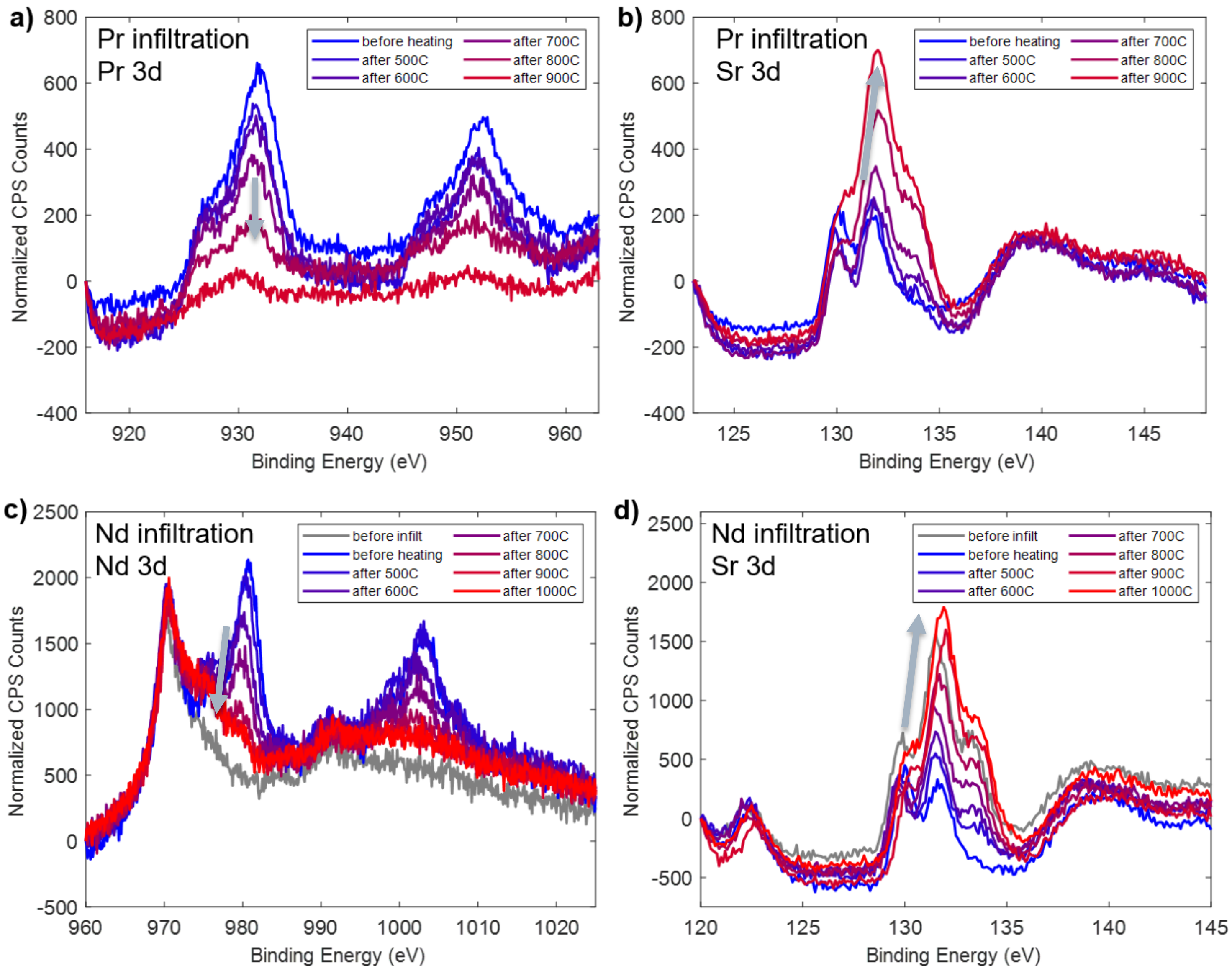
- ☐ 0.08 mol/L Hf solution is prepared by mixing Hf(IV)Cl4 in water at room temperature.
- ☐ The symmetric cells are submerged in the solution, or
- ☐ The Hf solution is drop cast on the oxygen electrode of the full cell
- ☐ Samples are rinsed with water and IPA followed by annealing at 250 °C to remove chloride

Next steps:

- ☐ Changing and combining infiltration parameters to increase the penetration depth of Hf into porous electrodes.

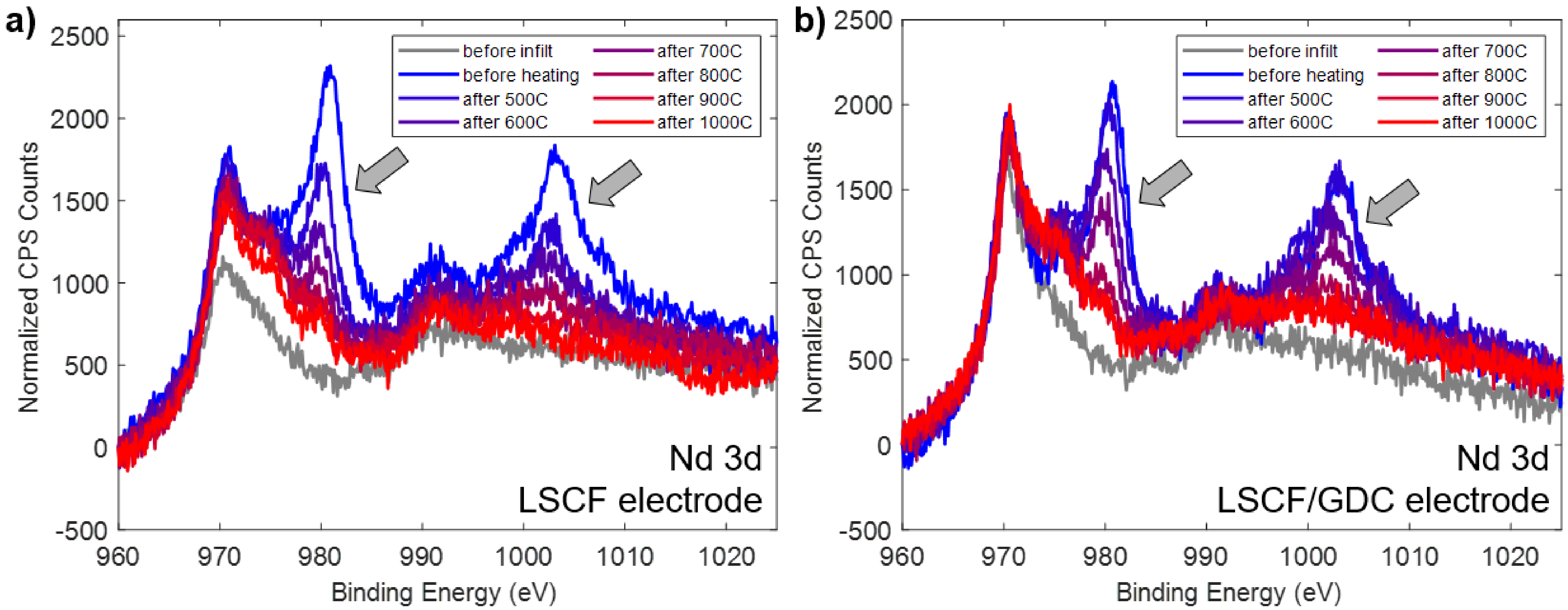
Temp.	Cycle number	Duration (minutes)
40 ^{o}C	1	3
40 ^{o}C	**2**	3
50 ^{o}C	1	3
50 ^{o}C	**2**	3
40 ^{o}C	1	**6**
50 ^{o}C	1	**6**

Larger lanthanide dopants also suppress Sr, but diffuse, too.



- Lanthanide cations (Pr, Nd) are of interest as hard-to-reduce cations of a considerably larger size than transition metal cations.
- Pr- and Nd- infiltrated LSCF/GDC electrodes demonstrate substantial diffusion of Nd and Pr into the electrode bulk (away from surface) above 800℃.
- Sr segregates more strongly with increasing temperature for both Pr- and Nd- infiltration.

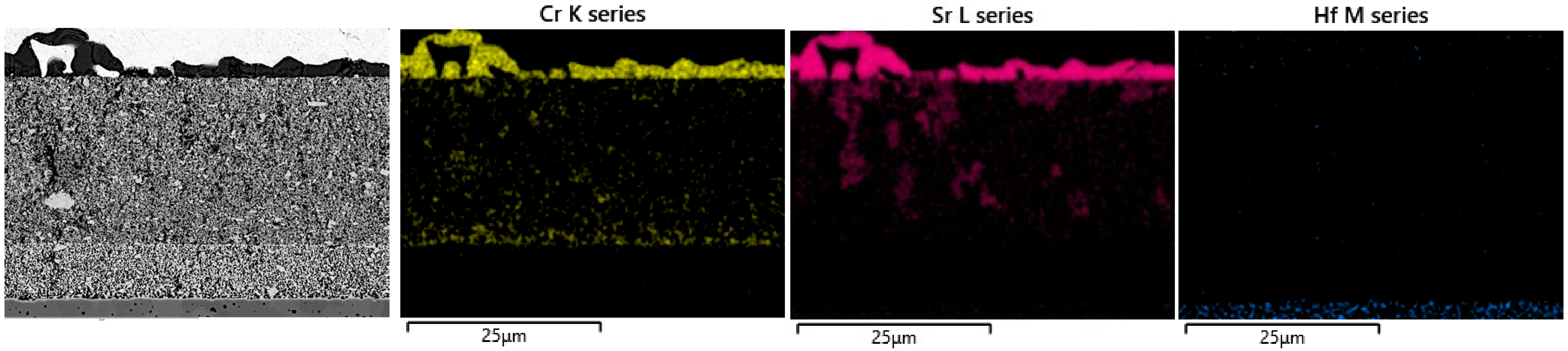
Dopant dissolution not dominated by GDC lattice diffusion.



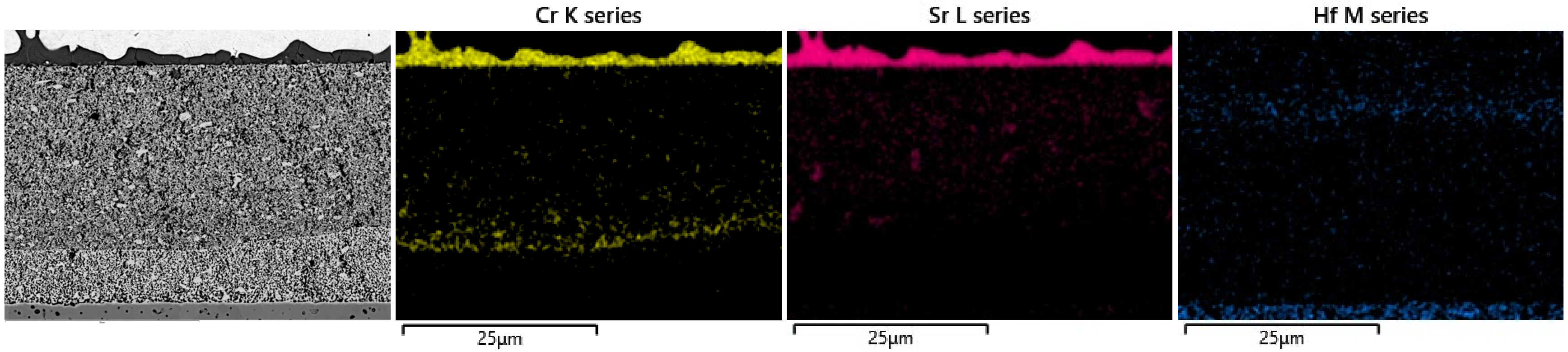
- In both the pure LSCF and LSCF/GDC electrode samples, the Nd 3d XPS signal appears largely depleted above 800℃
- Suggests that surface Nd cation diffusion is not likely dominated by diffusion through GDC particles in the electrode

SOFC: Surface $SrCrO_3$ formed; Cr Moves to the Active Interface; No Hf-Sr or Hf-Cr Interaction

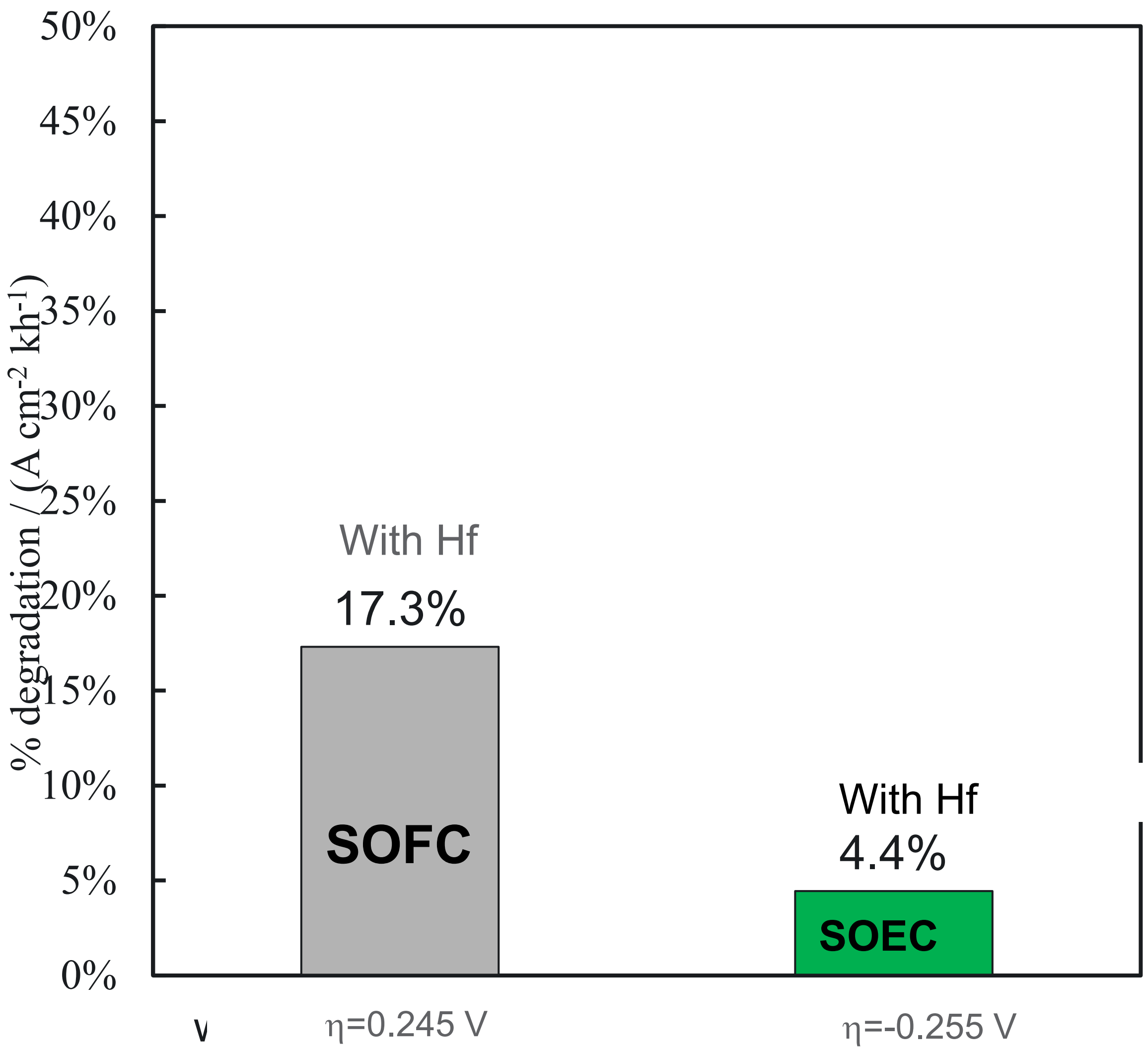
SOFC – no Hf

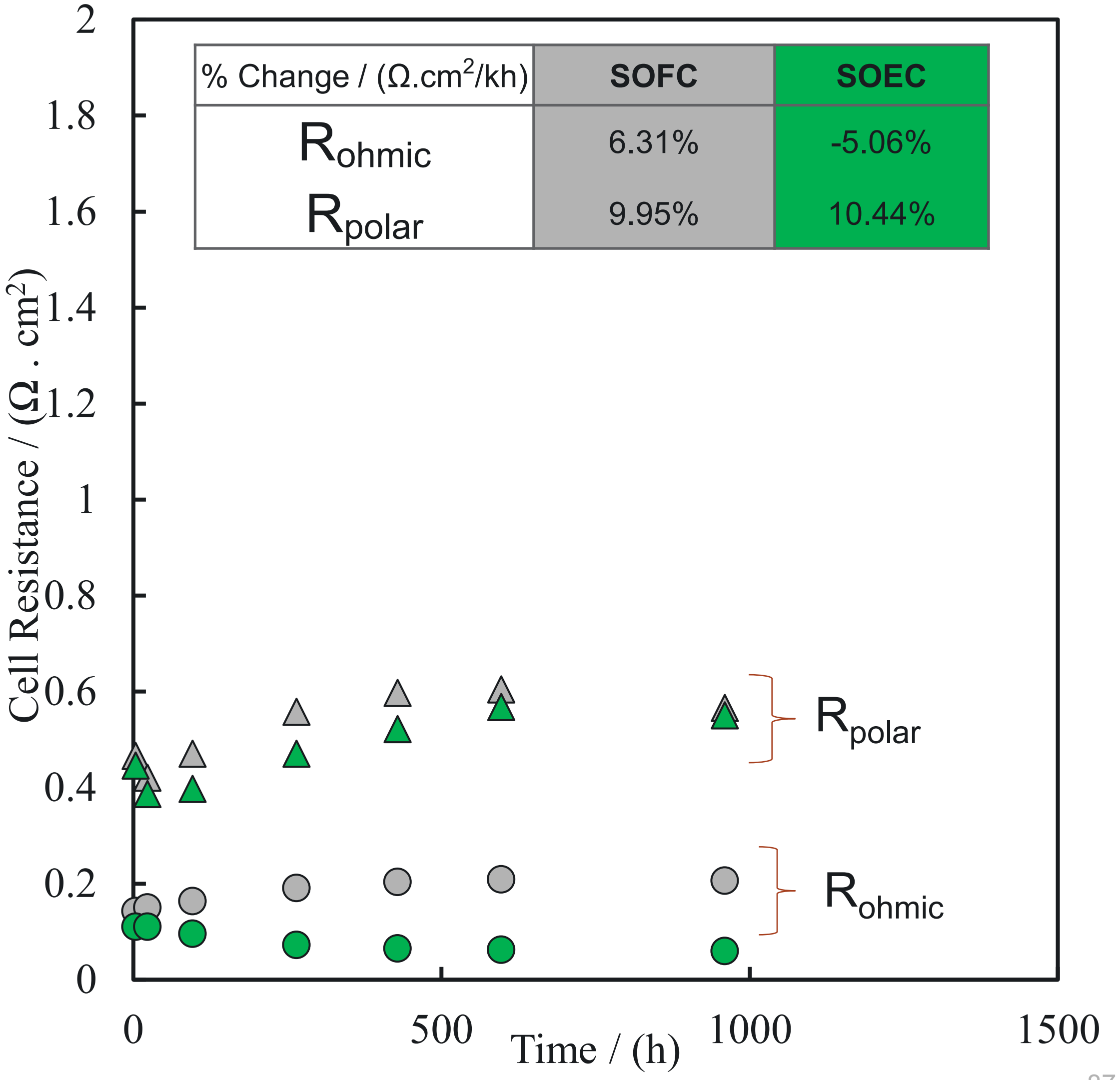


SOFC – Hf infiltrated



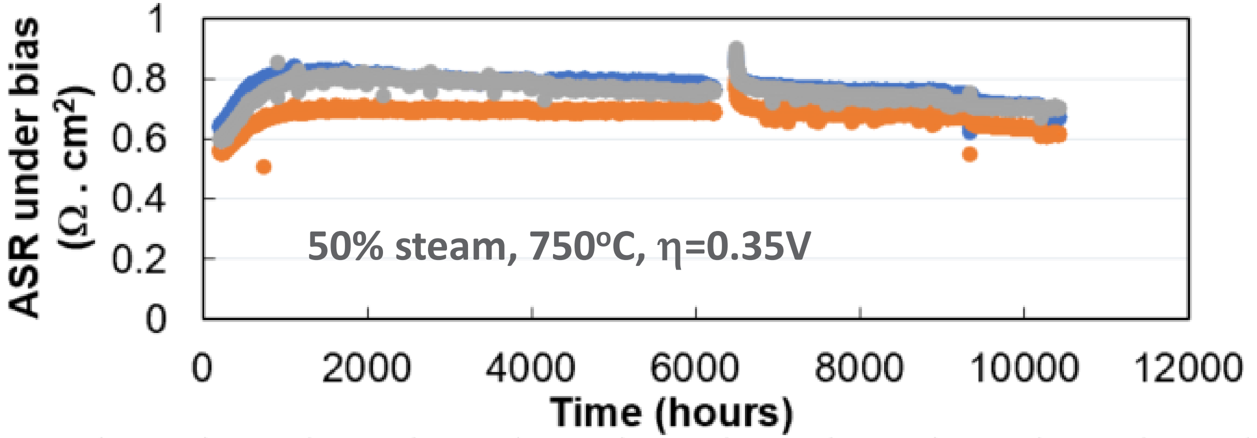
Cr Poisoning: Increased Degradation is Observed in SOFC vs SOEC at Similar Overpotentials

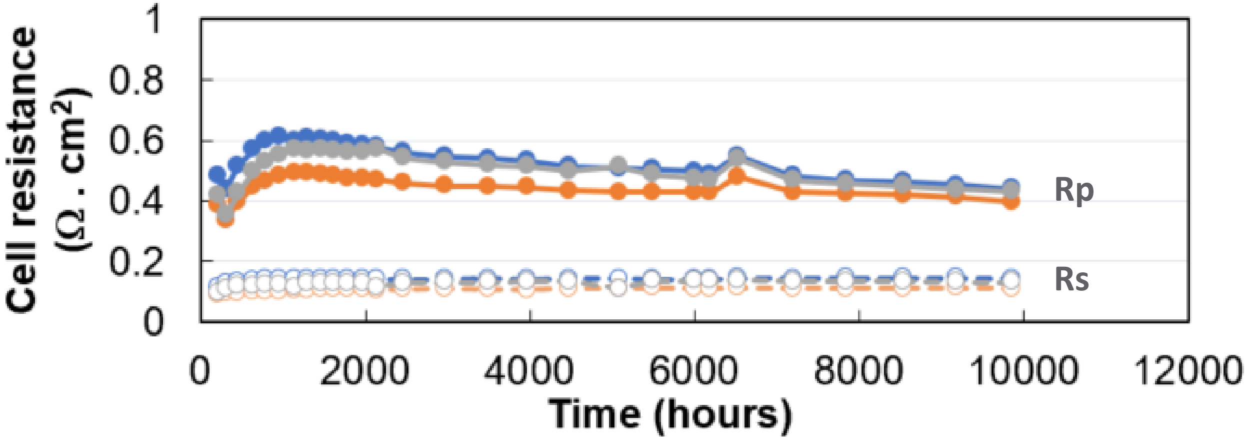




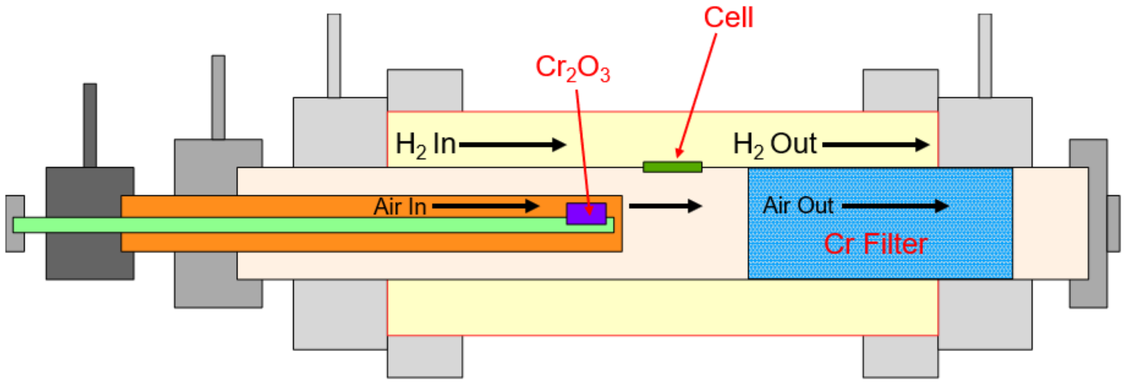
% Change / ($\Omega.cm^2$/kh)	SOFC	SOEC
R_{ohmic}	6.31%	-5.06%
R_{polar}	9.95%	10.44%

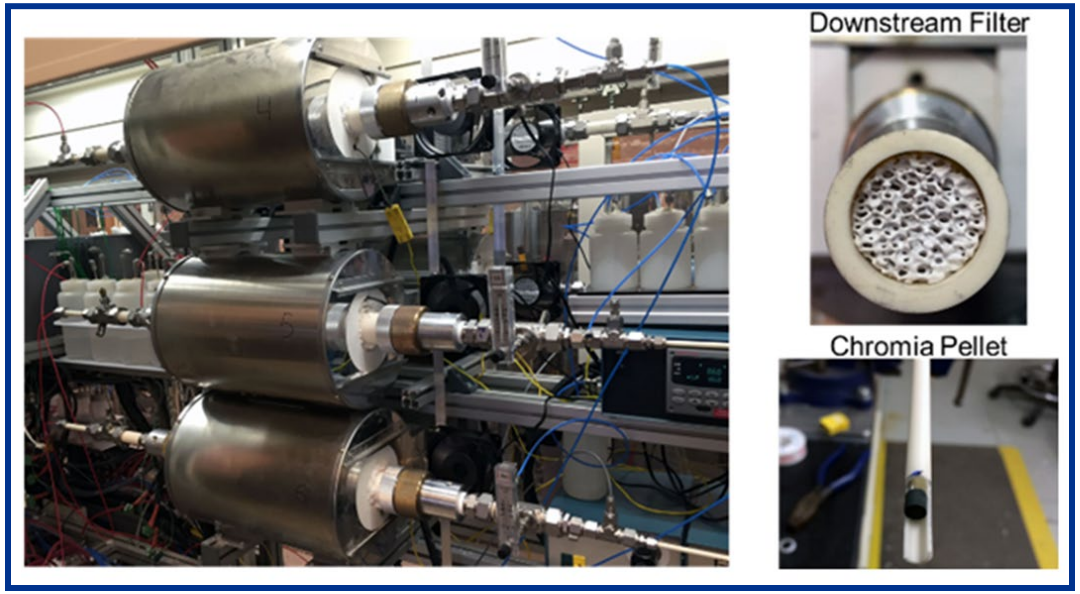
Baseline Testing of LSCF Electrodes in SOEC Mode

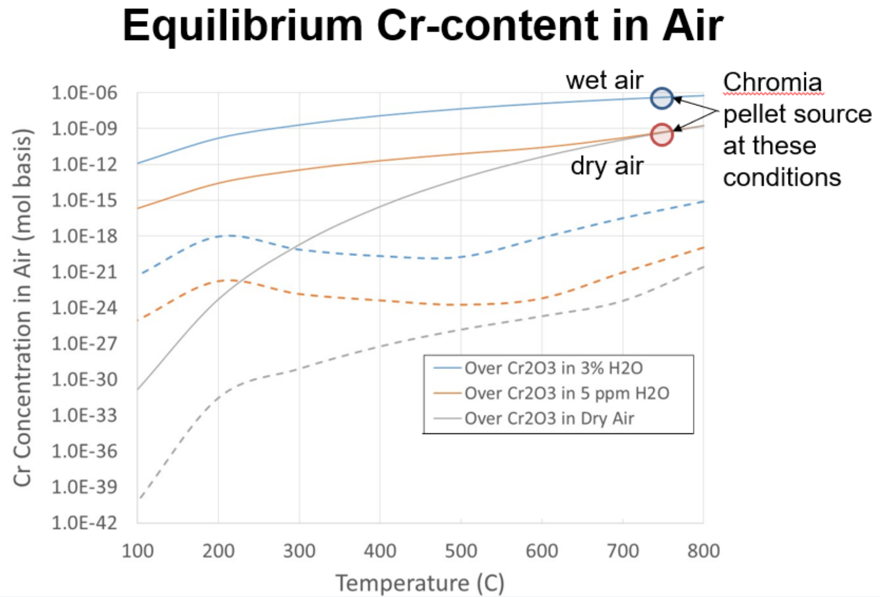




PNNL team established controlled Cr poisoning testing capabilities

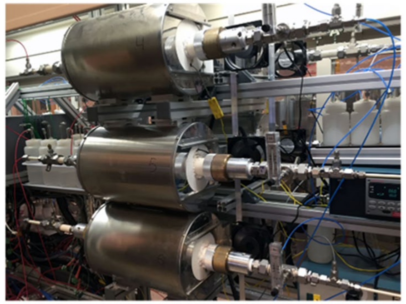




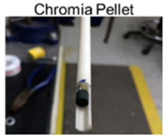


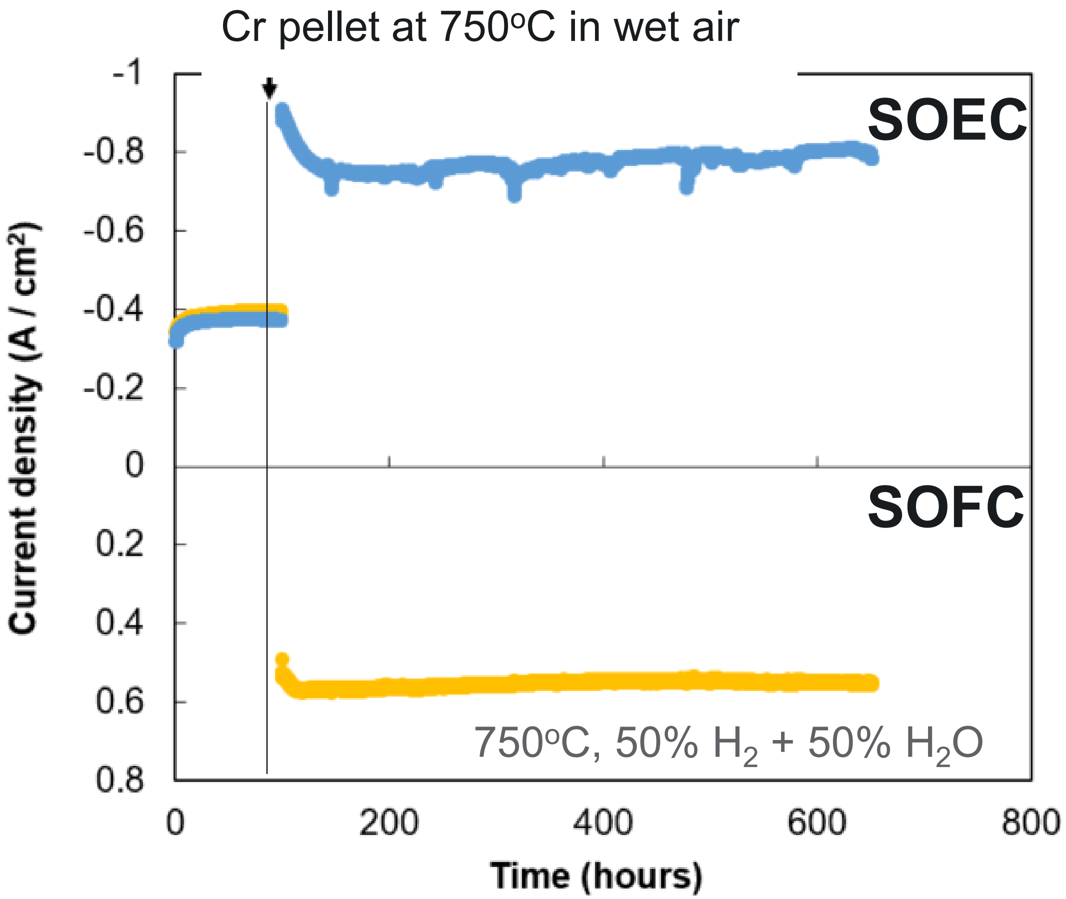
Cell: Ni/YSZ | YSZ | GDC – LSCF/GDC
$T=750^{o}C$
$H_2/H_2O=50/50$

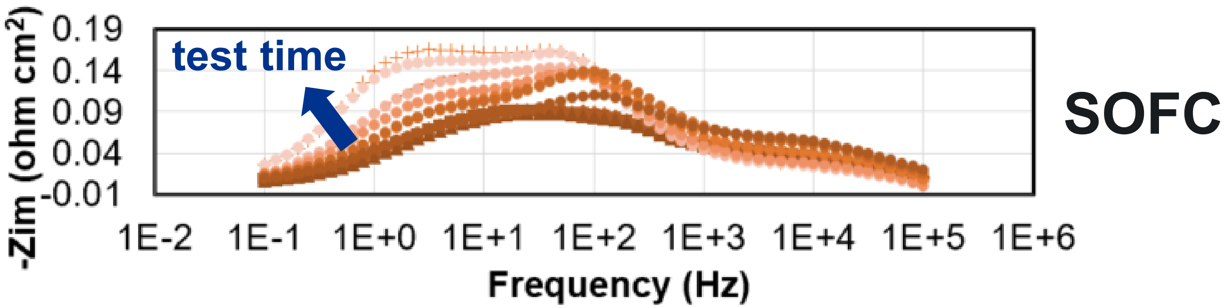
Cr Poisoning of SOFC and SOEC, ~1 ppm

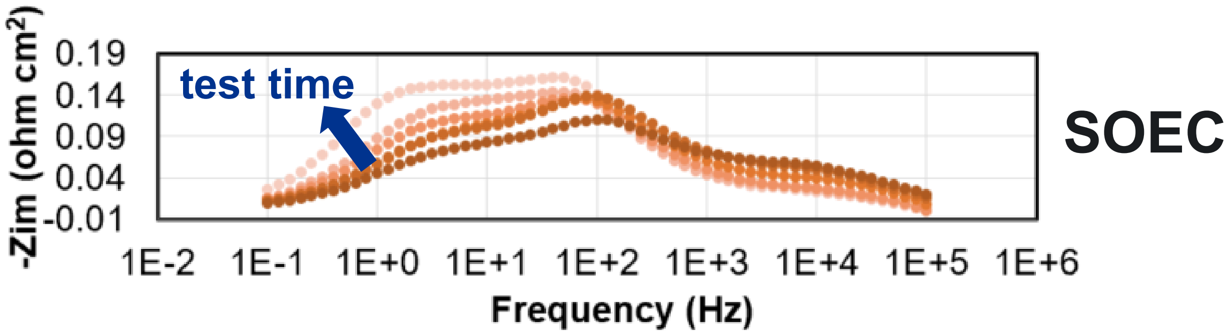



Downstream Filter


Chromia Pellet

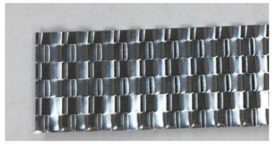

Cr pellet at 750°C in wet air



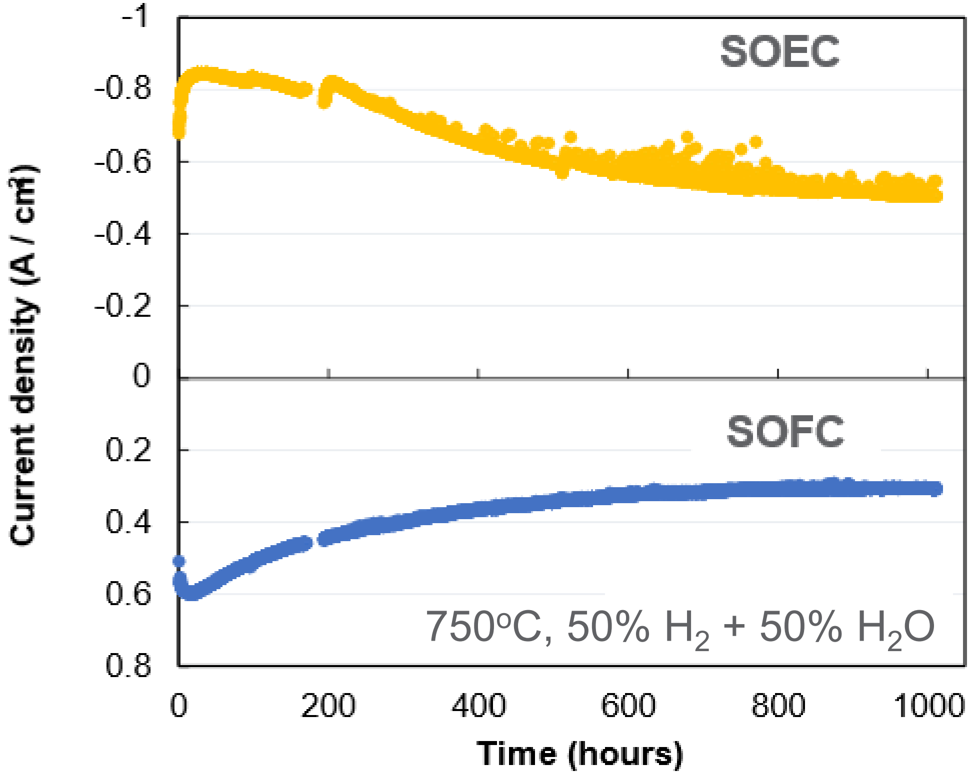


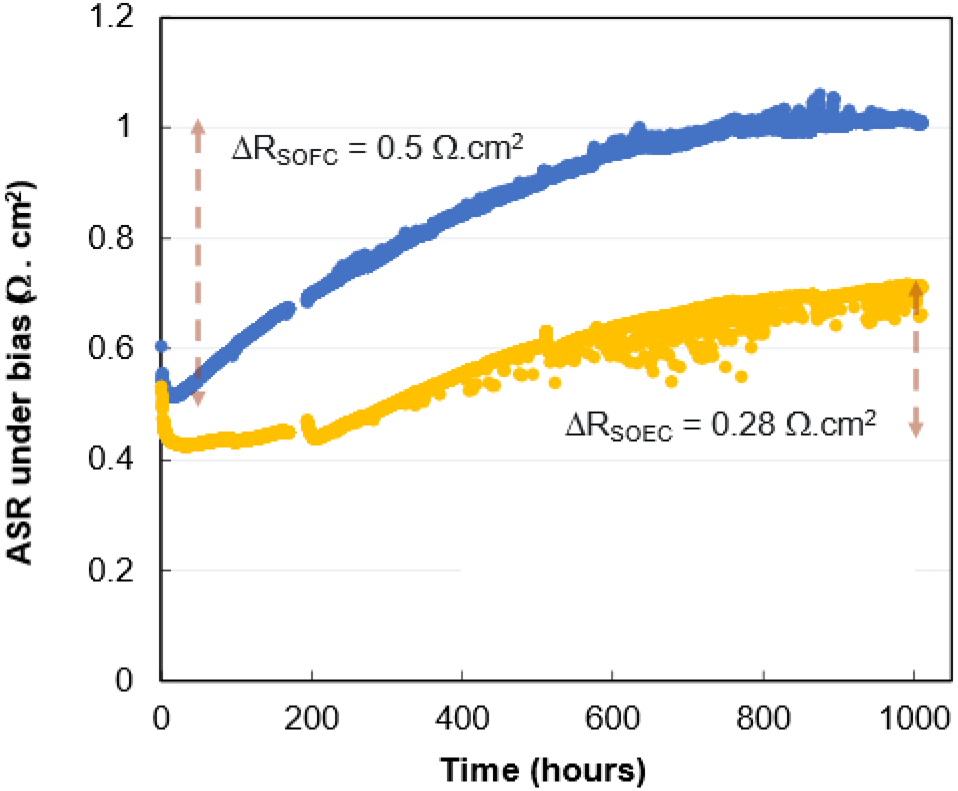
Low frequency process is increasing during exposure to Cr in SOFC and SOEC

Cr Poisoning of SOFC and SOEC: Higher Levels (uncoated interconnect)



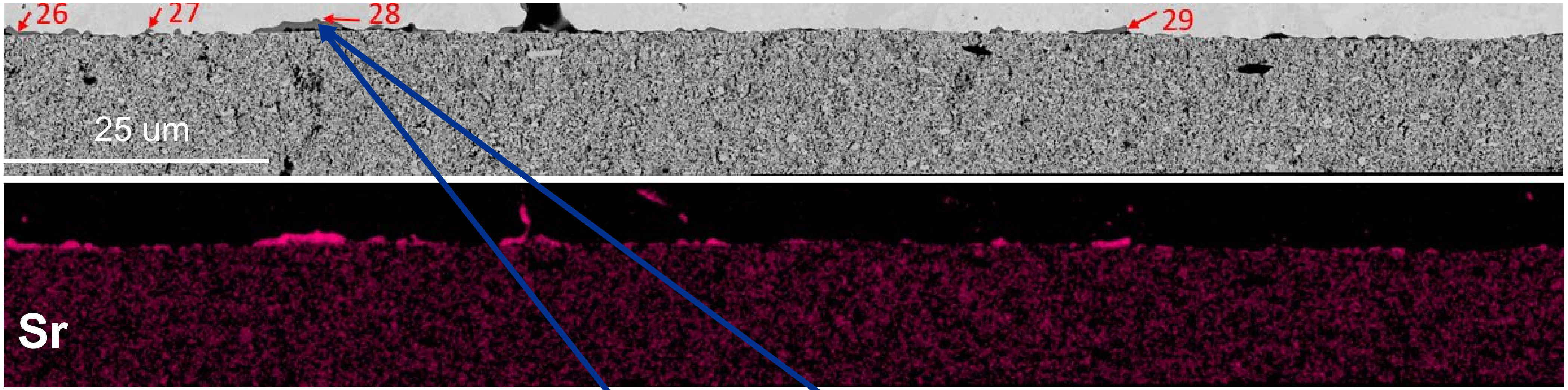
439SS: 17-19% Cr

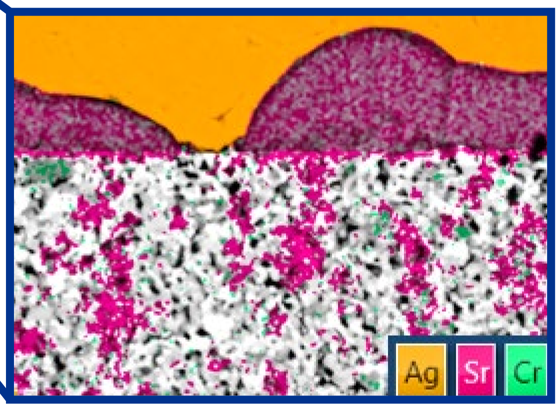




- In this case a 3%H_2O was used during SOFC operation
- Similar degradation rates are observed

Electrode Surface is Sr Enriched, Shows Cr-Sr-O Presence after 1000 hrs





At. %	O	Cr	Fe	Co	Sr
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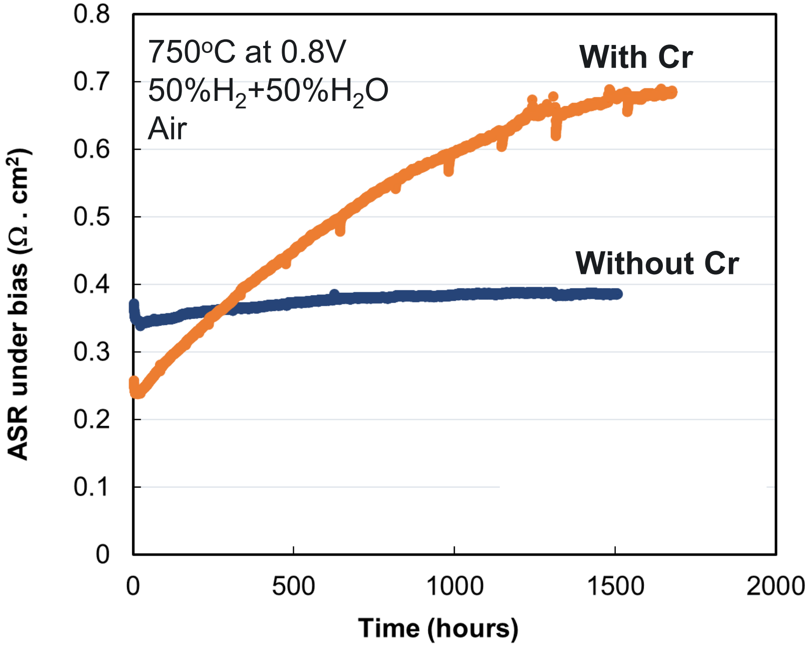
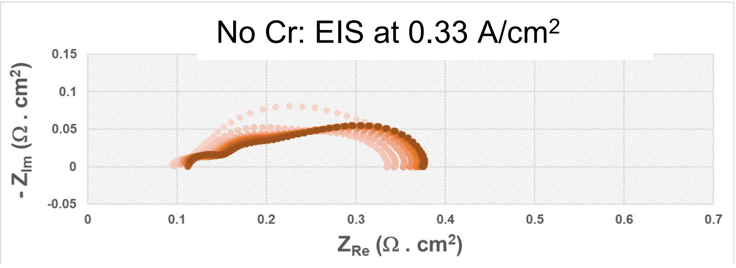
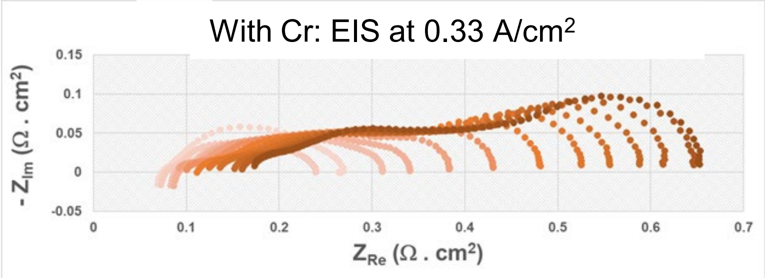
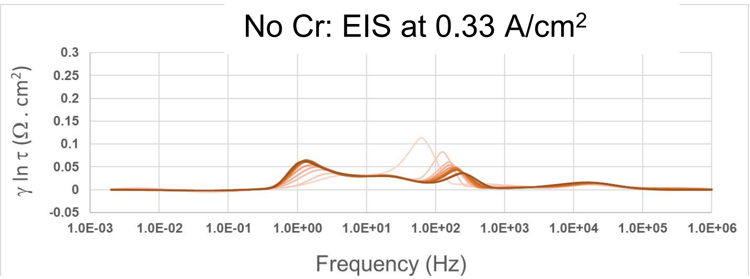
Spectrum 28	60.16	1.28	14.01		18.56
Spectrum 29	59.26		14.42		19.59
Spectrum 30	58.42		11.20	4.29	19.44

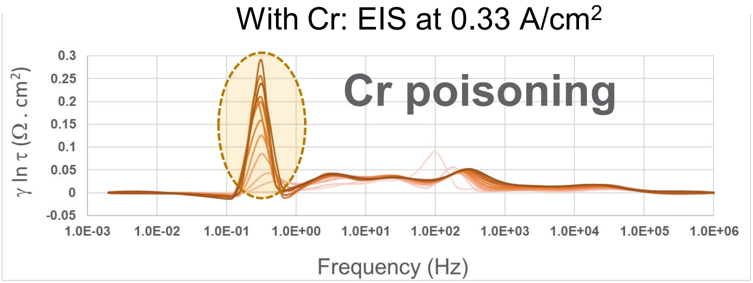
Effect of Cr Poisoning on SOFC Operation for 1700 hr at 750°C



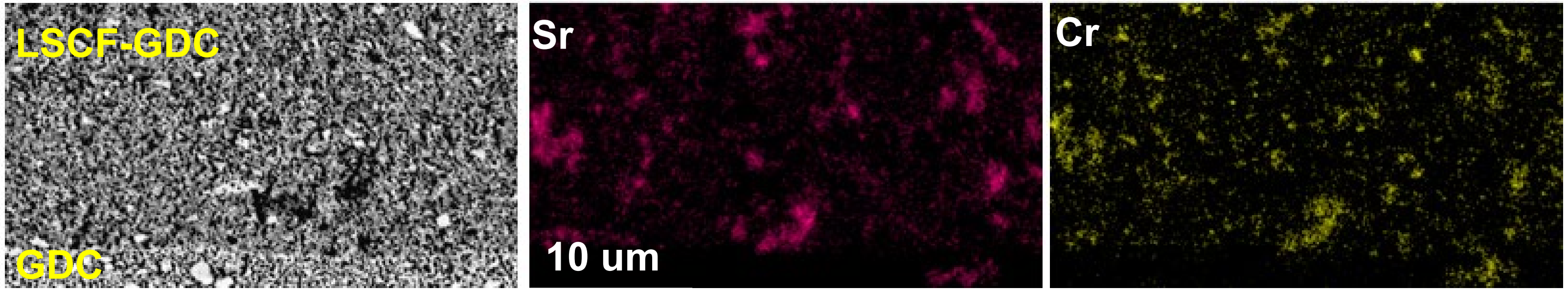








SEM/EDS Analysis: Cr is associated with Sr in bulk electrode as well



EIS and DRT analysis:

- Ohmic resistance significantly increased compared to test without interconnect
- Low frequency process is related to oxygen adsorption or oxygen exchange

Effect of Hf Infiltration

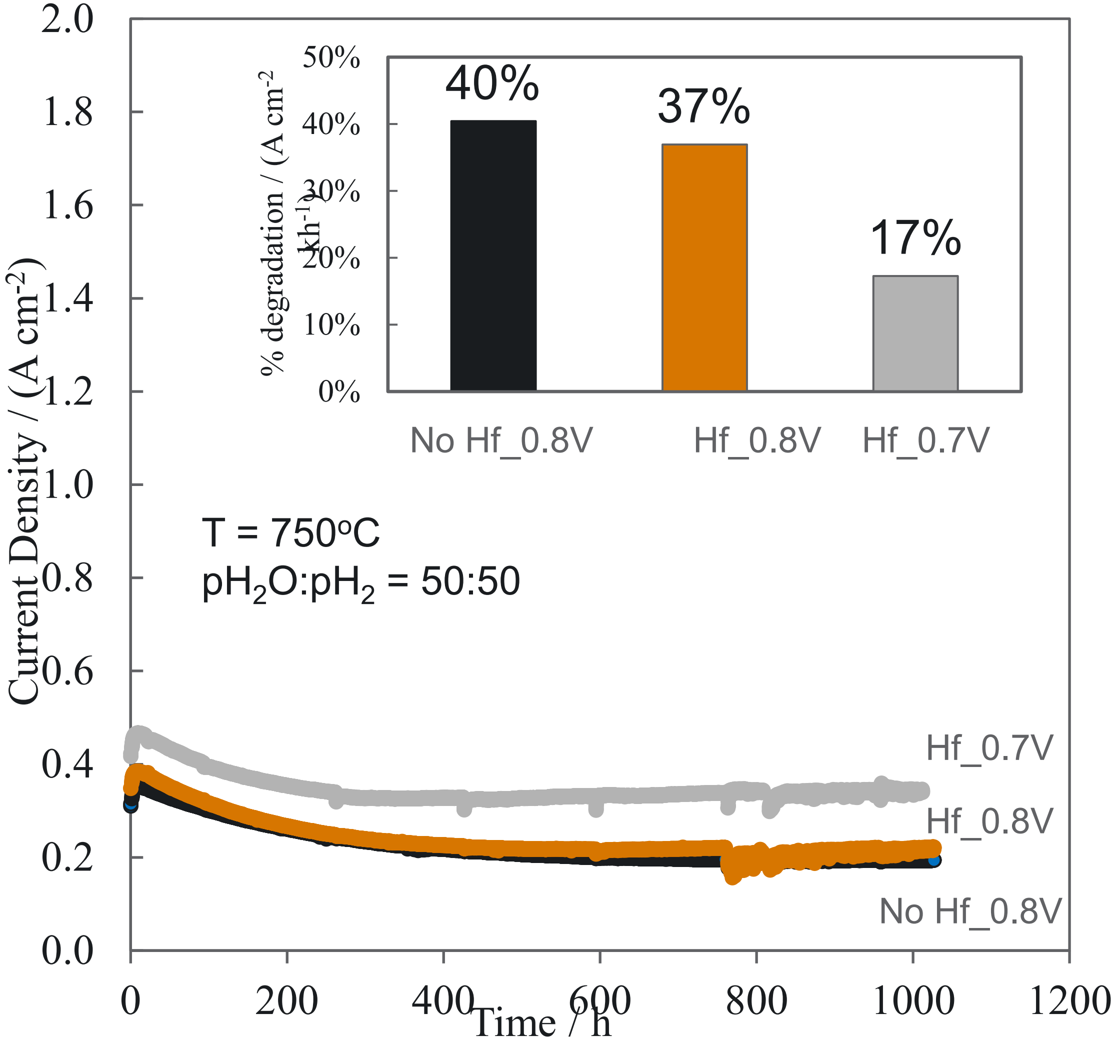
- Infiltrated and tested 4 button cells with $HfCl_4$ using provided recipe against 2 standards without Hf
- Conducted a series of chrome poisoning experiments in both SOFC and SOEC modes of operation
- Conducted post-test SEM analysis to analyze the interaction between hafnium and chrome
- Initiated sulfur exposure studies on the degradation of LSCF-GDC in SOEC mode
- Conducted post-test SEM analysis of LSCF after exposure to sulfur

Infiltration Recipe from MIT:

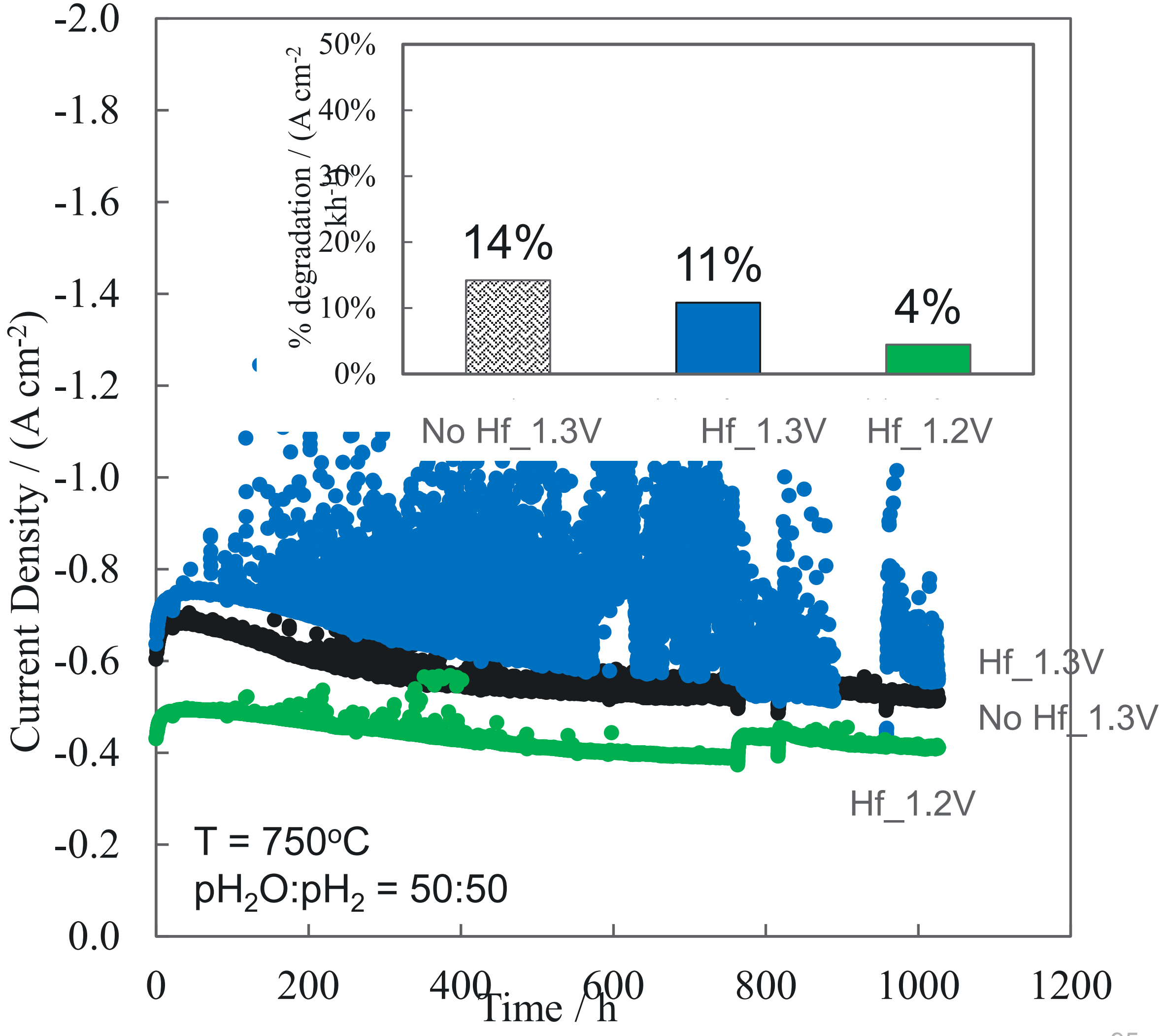
1. Prepare 2 beakers for post-infiltration rinsing (1x 50mL IPA, 1x 50mL DI water)
2. Add 30mL of DI and a stir bar to a beaker.
3. Weigh 0.24g $Hf(IV)Cl_4$ and add it to the beaker (do in fume hood)
4. Stir for 2 minutes (until all salt dissolves)
5. Using a pipette, cover air electrode in solution for 5 minutes
   (Original instructions suggested the submersion of the cell into the solution; it is assumed this was planned for symmetrical cells and adjusted accordingly for full cell testing)
6. After, dip the cell into IPA for 5 seconds then into DI for 5 seconds (rinse excess from the service)
7. Dry sample with Nitrogen
8. Anneal sample in a covered alumina boat @ 250 °C for 15 minutes.

Cr Poisoning: Hafnium Infiltration Shows Little to No Effect on Long Term Cell Degradation

SOFC

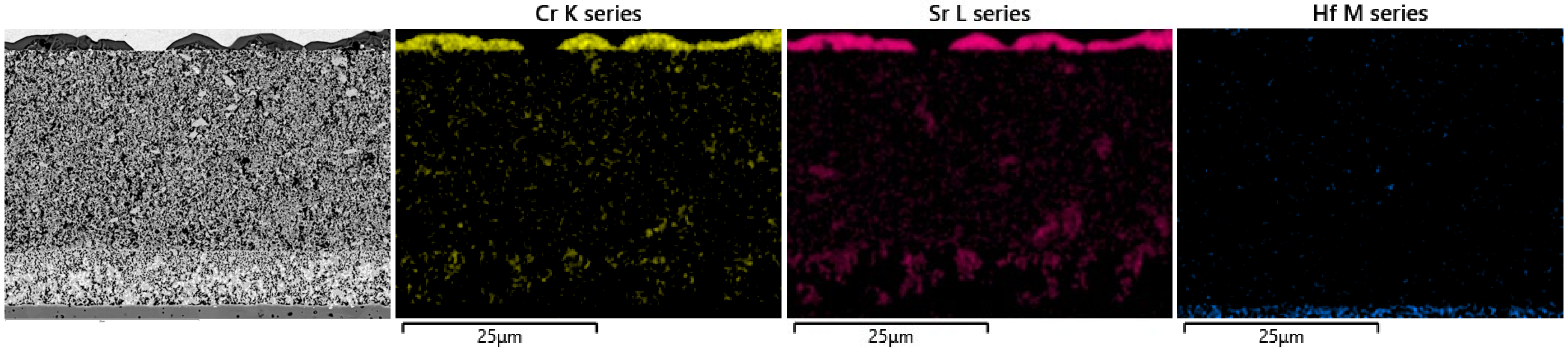


SOEC

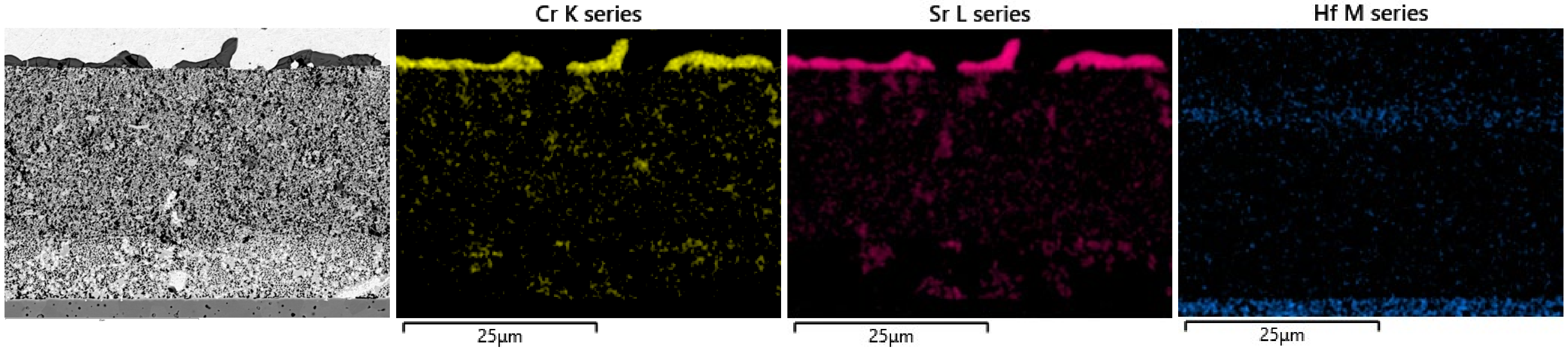


SOEC: Surface $SrCrO_3$ formed; Cr is Distributed in Electrode; No Hf-Sr or Hf-Cr Interaction

SOEC – no Hf



SOFC – Hf infiltrated

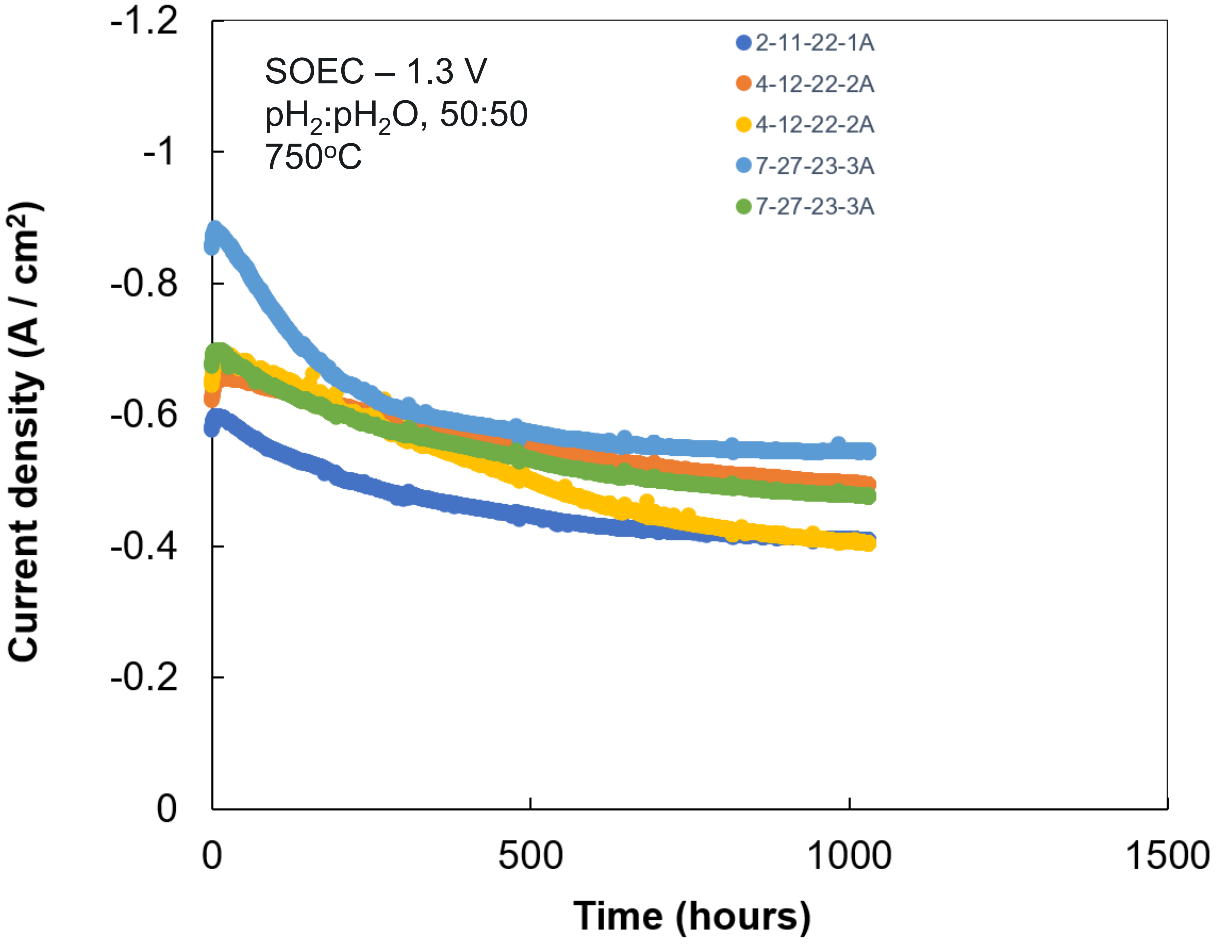


Hf Infiltration: Next Steps – to increase infiltrate penetration deeper into the electrodes

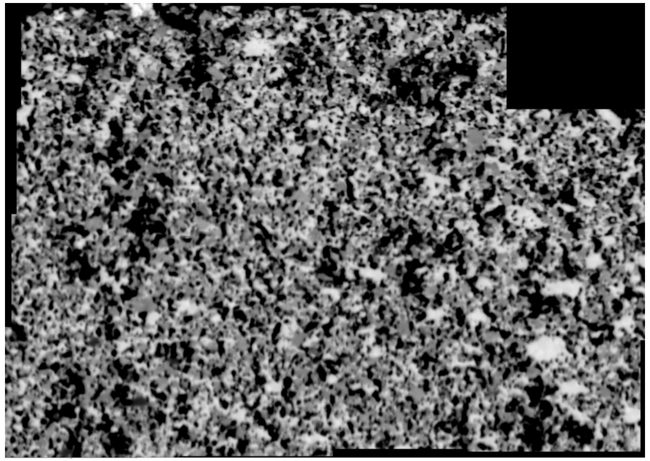
- Repeated Hf infiltration using more 4x concentrated solutions
- Increased infiltration time
- Calcined coupons to assess Hf distribution in LSCF
- Exposed coupons to Cr for potential interaction
- Sent to SEM analysis

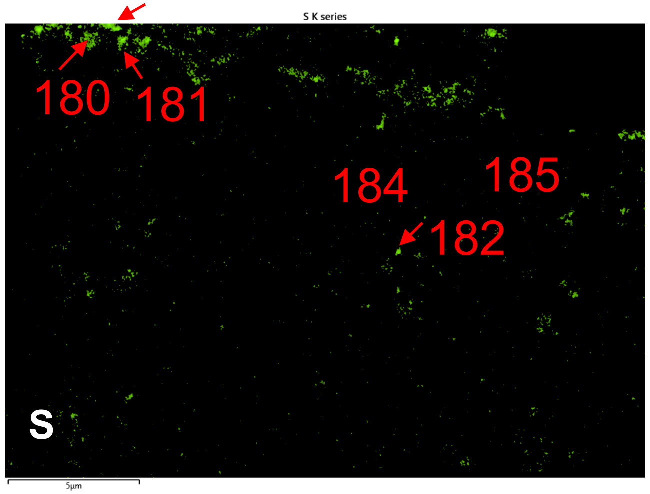
1000 hr Sulfur Exposure Test at 750°C: LSCF-GDC

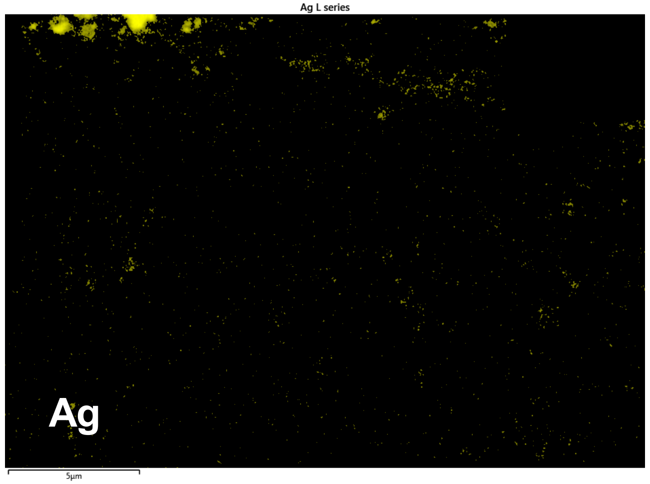
- 6x standard 8Ni-YSZ, GDC:LSCF cells
- Tested at 750oC for 1000hr
- Submitted for post-test SEM analysis

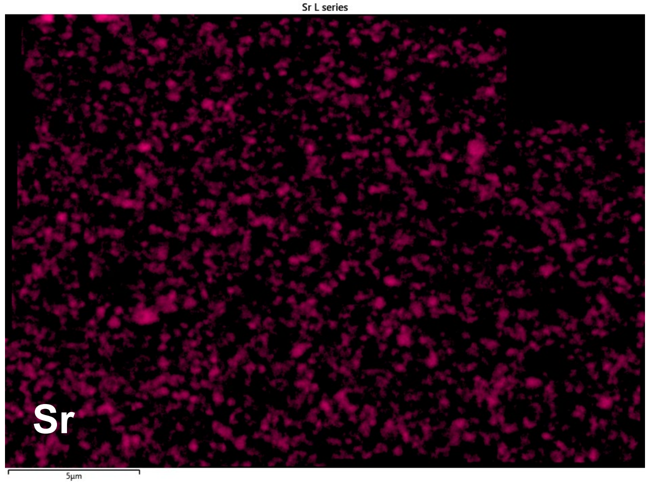


Previous Findings: Sr-S-O was Detected in LSCF after 5000 hours of SOEC Operation at 750°C



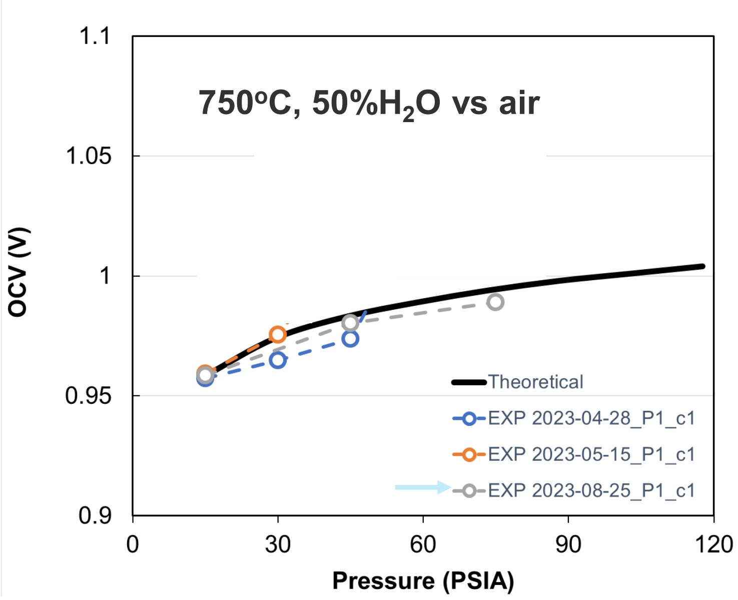


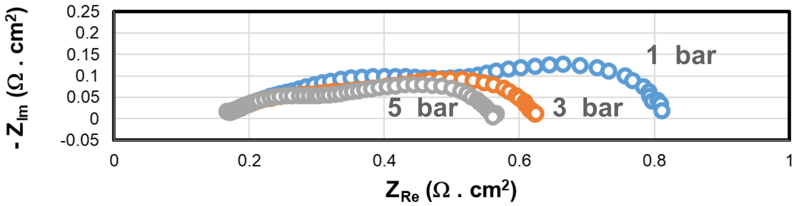


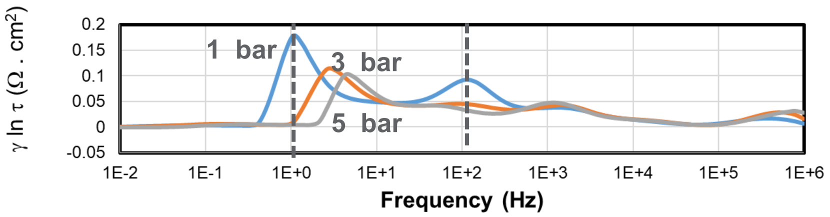


Label	O	P	S	Fe	Co	Sr	Ag	La	Ce	Gd
Spectrum 179	52.06	5.52	3.43	3.37	0.47	15.19	3.79	9.24	5.89	1.04
Spectrum 180	42.41		2.7	11.57	2.18	4.32	6.69	19.13	9.11	1.87
Spectrum 181	42.15		7.19	8.38	2.35	3.48	7.23	15.57	10.51	3.13
Spectrum 182	45.58		5.09	10.46	4.44	10.35		12.92	10.03	1.13
Spectrum 184	47.88	0.41	3.5	5.98	0.96	1.74	3.88	13.45	19.04	3.17
Spectrum 185	44.71		2.92	8.34	2.78	3.79	2.83	18.53	13.73	2.37

SOEC Performance Increases with Pressure







Pressurized button cell

- Low frequency (mass transport) and mid-frequency (diffusion) processes are suppressed
- Troubleshooting seals for long-term tests