Numerical Study to Optimize the Microstructure of an LSM/YSZ Backbone for Nanoparticle Infiltration

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Introduction

➢ Background and Motivation

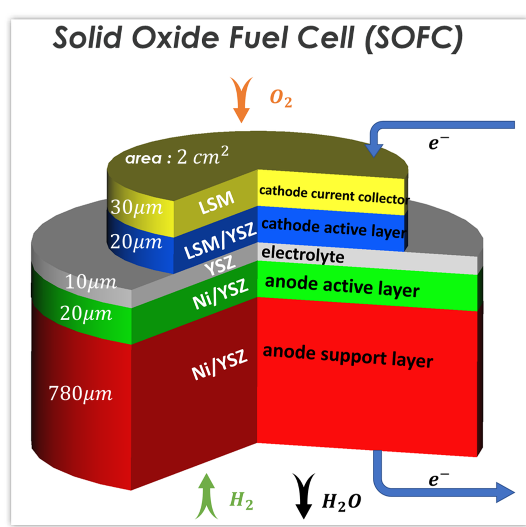
- The performance of solid oxide fuel cells (SOFCs) can be improved by introducing nanoparticles onto the electrode backbone to influence both the microstructural properties and electrochemical activity of the electrode.
- However, the amount of improvement is in some way limited by the properties of the baseline backbone.

➢ Purpose of the Study

- This study focuses on the optimization of the microstructure of a Lanthanum Strontium Manganite (LSM)/Yttria-Stabilized Zirconia (YSZ) backbone for nanoparticle infiltration via an in-house-developed multiphysics simulation.

Multiphysics Modeling

➢ Governing equations



- Charge conservation (electron-conducting phase)

$$a_{int}C_{DL}\frac{\partial}{\partial t}(\varphi_e-\varphi_l) + \nabla\cdot(-\sigma_e\nabla\varphi_e) = i_F$$

double layer capacitor — charge transport — electrochemical reactions

- Species transport

$$\varepsilon\frac{\partial\phi}{\partial t} = \nabla\cdot\left(D_\phi^{eff}\nabla\phi\right) - S_\phi$$

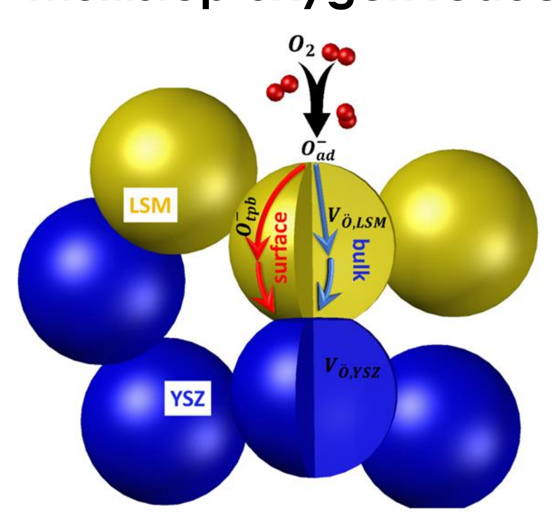
concentration change — diffusion — consumption/production rate

➢ Electrochemical model

- **Butler-Volmer model** (anode)

$$i_{F,a} = i_{0,a}(P_{H_2}^{\infty})^a(P_{H_2O}^{\infty})^b\left\{\frac{P_{H_2}}{P_{H_2}^{\infty}}\exp\left[\frac{(1-\alpha)nF\eta_a}{RT}\right] - \frac{P_{H_2O}}{P_{H_2O}^{\infty}}\exp\left[-\frac{\alpha nF\eta_a}{RT}\right]\right\}$$

- **Multistep oxygen reduction reaction (ORR) model** (cathode)



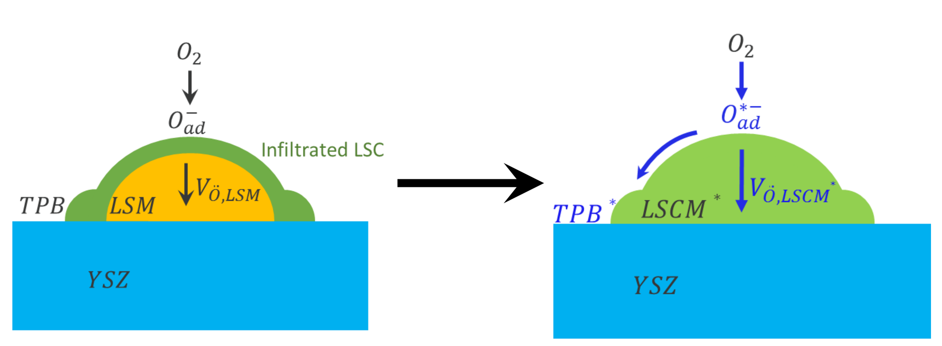
surface adsorption and dissociation (ads): $1/2O_2 + S + e^- \leftrightarrow O_{ad}^-$

surface charge transfer (sct): $O_{ad}^- + V_{O,YSZ}^{\cdot\cdot} + e^- \leftrightarrow O_{O,YSZ}^x + S$

incorporation (inc): $O_{ad}^- + V_{O,LSM}^{\cdot\cdot} + e^- \leftrightarrow O_{O,LSM}^x + S$

bulk charge transfer (bct): $O_{O,LSM}^x + V_{O,YSZ}^{\cdot\cdot} \leftrightarrow O_{O,YSZ}^x + V_{O,LSM}^{\cdot\cdot}$

➢ Nanoparticle infiltration model



Inter-Diffusion model

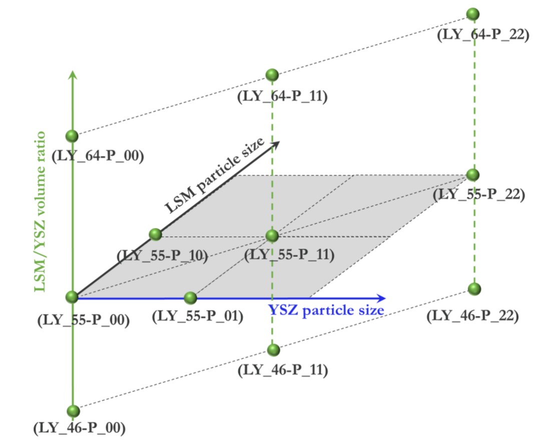
- backbones are modified to another material (e.g., LSCM) due to the inter-diffusion between LSC infiltrates and LSM backbone

References

- T. Yang et al., *Journal of The Electrochemical Society*, 166(8), pp F448-F457, 2019.
- T. Yang et al., *International Journal of Hydrogen Energy*, 43(32), pp 15445-15456, 2018.
- T. Yang et al., *Physical Chemistry Chemical Physics*, 19(45), pp 30464 – 30472, 2017.
- T. Yang et al., *International Journal of Electrochemical Science*, 12, pp 6801-6828, 2017.

Results

➢ Baseline LSM/YSZ backbones

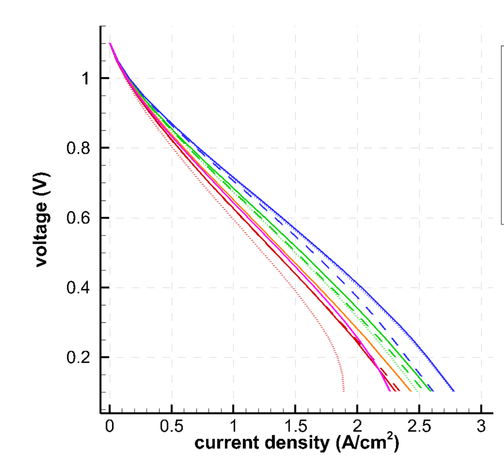


ex) LY_46-P_01

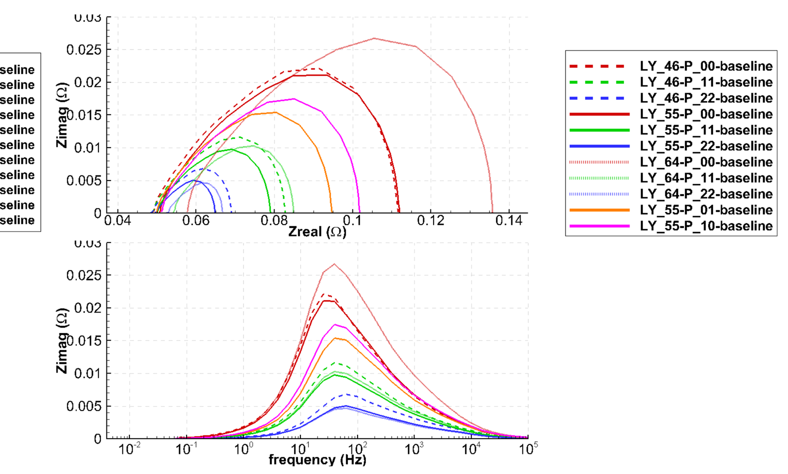
volume fraction: $V_{LSM}:V_{YSZ} = 40\%:60\%$

grain size: $\beta_{LSM} = 0$, $\beta_{YSZ} = 1$

$r_{LSM} = 0.67^{\beta_{LSM}}\cdot r_{LSM,ref}$ $r_{YSZ} = 0.5^{\beta_{YSZ}}\cdot r_{YSZ,ref}$

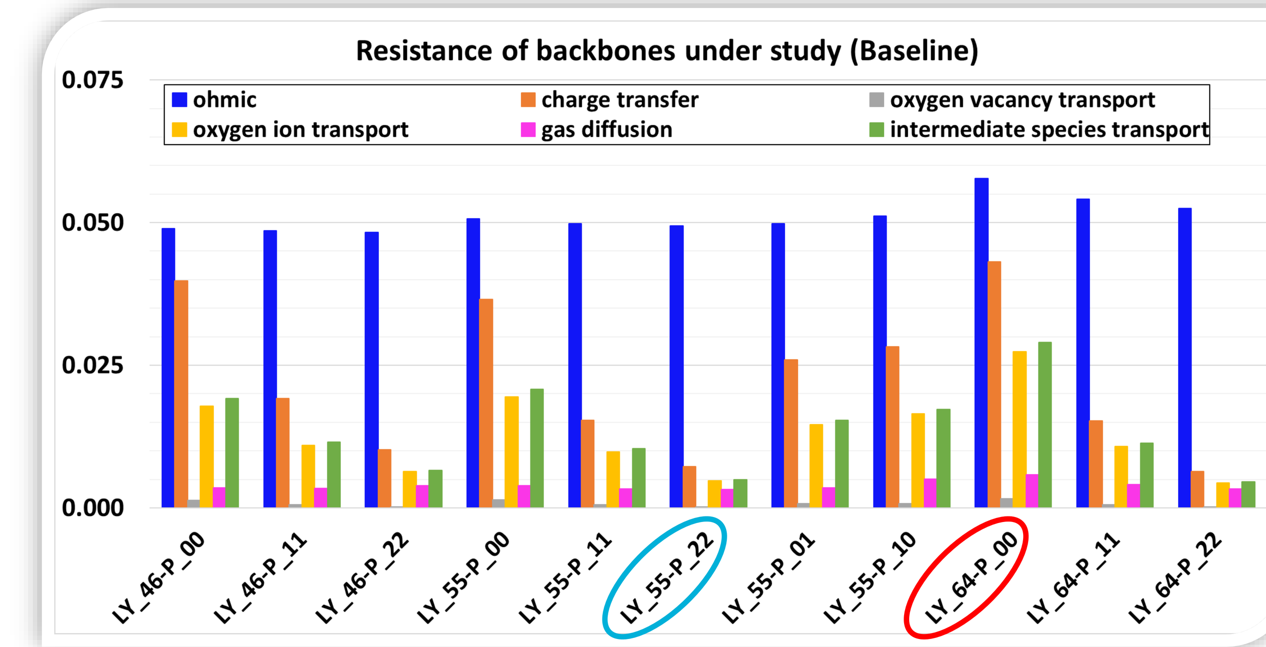


V-I curves



Impedance behavior

Simulated cell performance with baseline backbones



lowest R_{total}: LY_55-P_22   highest R_{total}: LY_64-P_00

Resistance components associated with different physical processes

➢ LSM/YSZ infiltrated with nano-LSC

ads^*: $1/2O_2 + S_I + e^- \underset{k_{ads}^*}{\overset{k_{ads}^*}{\longleftrightarrow}} O_{ad,I}^-$

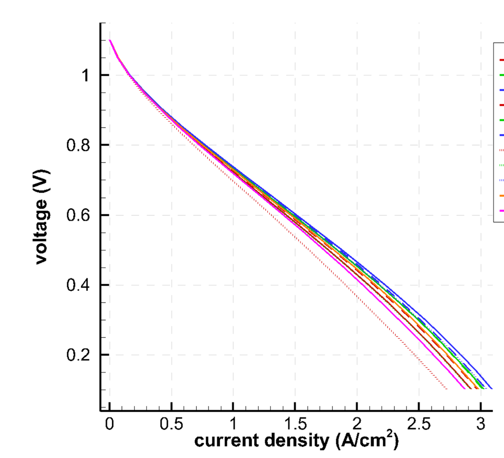
sct: $O_{ad,I}^- + V_{O,YSZ}^{\cdot\cdot} + e^- \underset{k_{sct}}{\overset{k_{sct}}{\longleftrightarrow}} O_{O,YSZ}^x + S$

inc^*: $O_{ad,I}^- + V_{O,I}^{\cdot\cdot} + e^- \underset{k_{inc}^*}{\overset{k_{inc}^*}{\longleftrightarrow}} O_{O,I}^x + S$

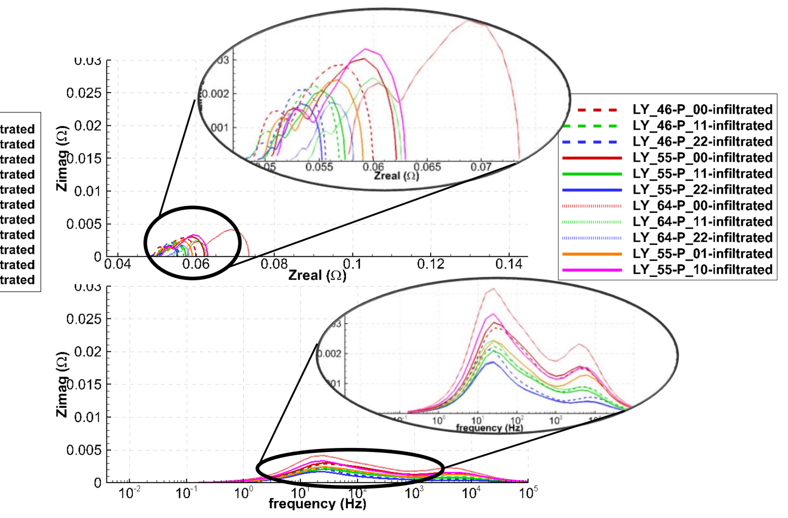
$diff$: $O_{O,I}^x \longleftrightarrow O_{O,LSM}^x$

bct: $O_{O,LSM}^x + V_{O,YSZ}^{\cdot\cdot} \underset{k_{B4}}{\overset{k_{B4}}{\longleftrightarrow}} O_{O,YSZ}^x + V_{O,LSM}^{\cdot\cdot}$

Parameters changed by infiltration

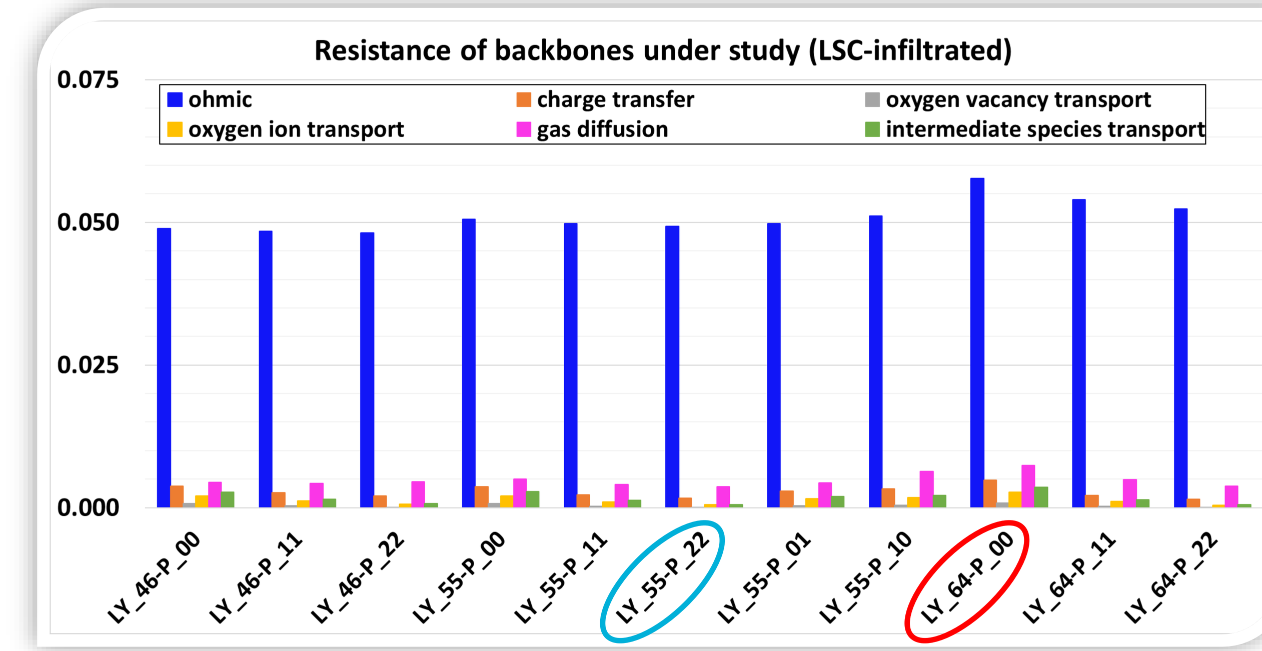


V-I curves



Impedance behavior

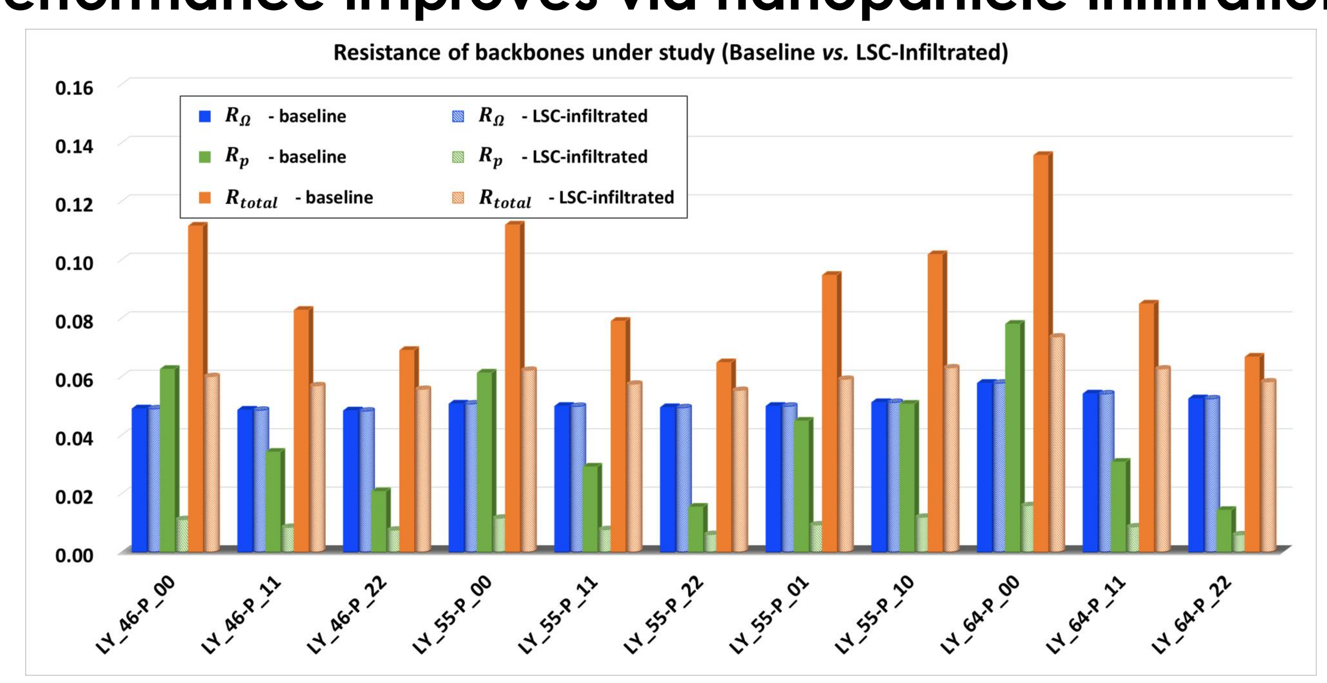
Simulated cell performance with baseline backbones



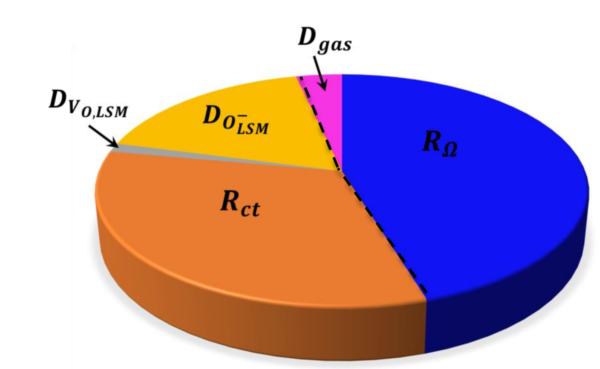
lowest R_{total}: LY_55-P_22   highest R_{total}: LY_64-P_00

Resistance components associated with different physical processes

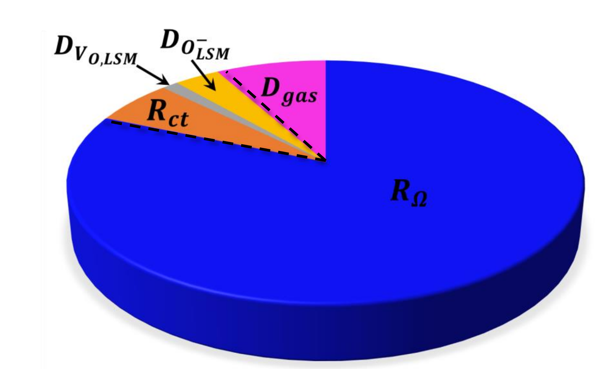
➢ Performance improves via nanoparticle infiltration



Polarization and ohmic resistance change due to nano-LSC infiltration into LSM/YSZ composite cathode



Baseline backbone



LSC-infiltrated backbone

- After infiltration, the charge transfer resistance, especially the surface adsorption and dissociation step, is significantly reduced, while the concentration resistance is slightly increased.
- Ohmic resistance dominates the resistance of infiltrated backbone

Summary

- Multiphysics simulations with multistep ORR mechanism were developed and calibrated to investigate the performance of LSM/YSZ composite backbones before and after nanoparticle infiltration.
- For baseline backbones and infiltrated backbones, the backbone LY_55-P_22 (V_LSM:V_YSZ=50%:50% with finest grain size) shows the best performance, while the backbone LY_64-P_00 (V_LSM:V_YSZ=60%:40% with coarsest grain size) shows the largest resistance.
- The infiltration mainly promotes the charge transfer, especially the surface adsorption and dissociation step, of cathode. After infiltration, the ohmic resistance becomes more dominant over polarization resistance.

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