

Supplementary Information

Mitigating Ni Migration in Solid Oxide Electrolysis by Altering Ni-YSZ Electrode Composition

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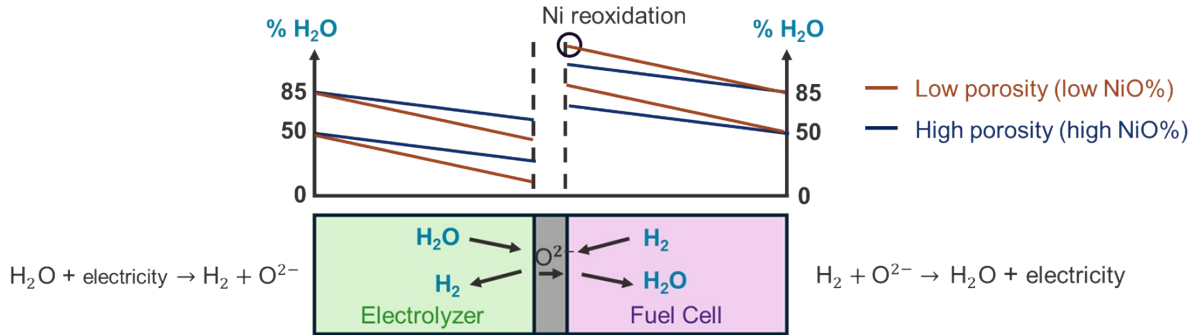
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Supplementary Text

Note that the SOFC electrodes sometimes delaminated along the electrode-electrolyte interface during or after the life tests under these conditions, especially for more extreme conditions – lower electrode porosity or higher current density. This is attributed to an increase in H_2O/H_2 ratio above the inlet 85%/15% value due to gas diffusion limitations, reaching a maximum at the SOFC electrode-electrolyte interface where Ni oxidation, and hence delamination, occurs.



Supplementary Figures

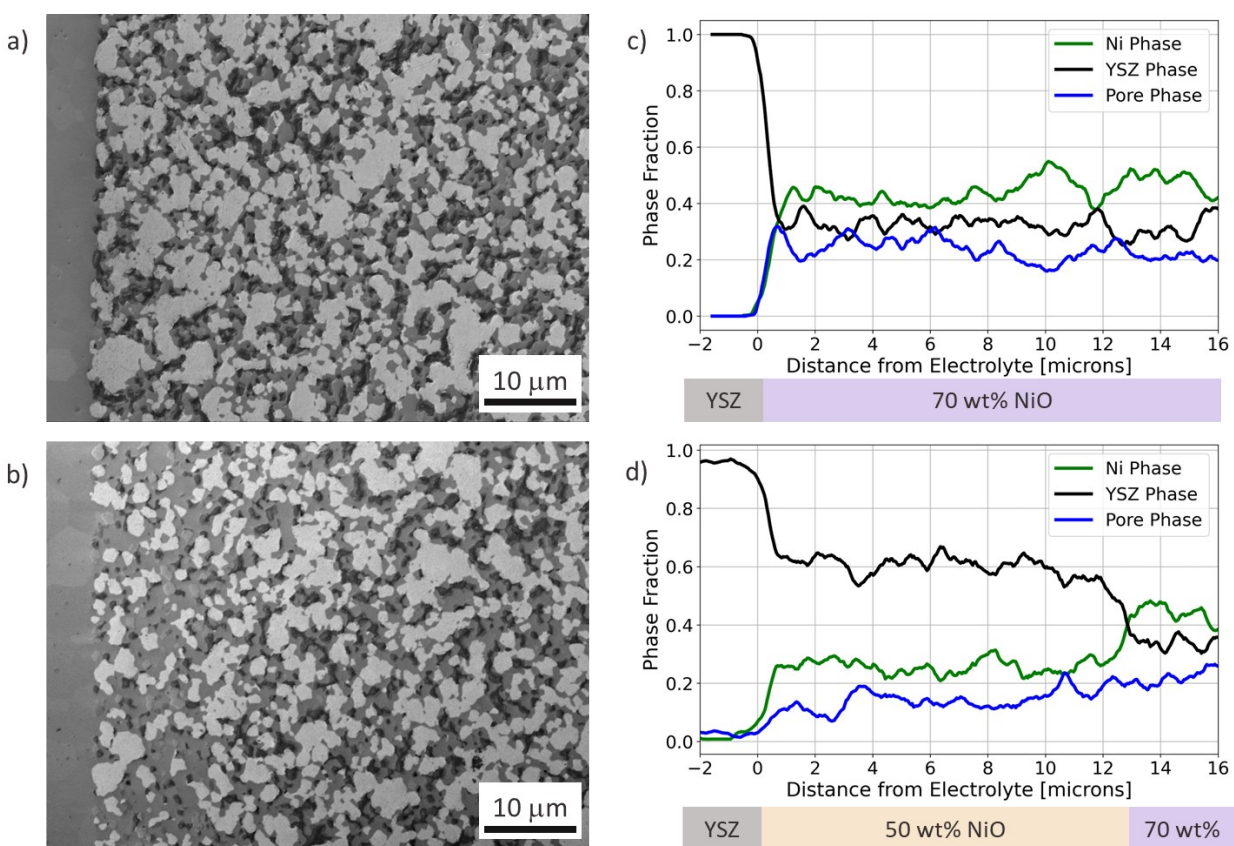


Figure S1 Cross sectional SEM images of pristine SOEC electrodes as reduced 100 hours before cell operation of (a) 70|70 and (b) and 70|50/70 cell. Ni has a lighter grey contrast, pores are black, and YSZ is a darker grey. Also shown are phase fractions versus distance taken from 2D image analysis for the 70|70 (c) and 70|50/70 (d) cells.

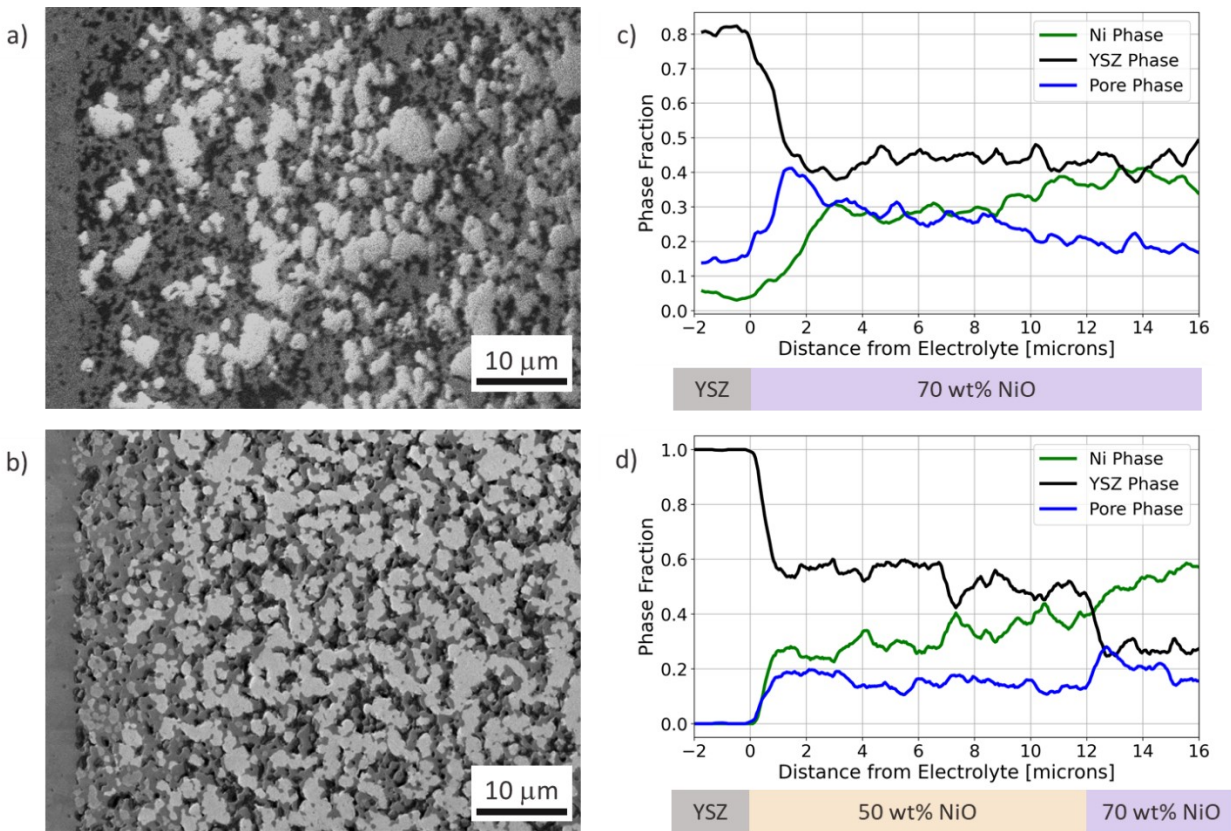


Figure S2 Cross sectional SEM images showing another sample of 70|70 (a) and 70|50/70 (b) cells after 500 h life testing at -1 A/cm^2 current density at 800°C in $85\% \text{ H}_2\text{O} - 15\% \text{ H}_2$. Ni has a lighter grey contrast, pores are black, and YSZ is a darker grey. Also shown are phase fractions versus distance taken from 2D image analysis for the 70|70 (c) and 70|50/70 (d) cells.

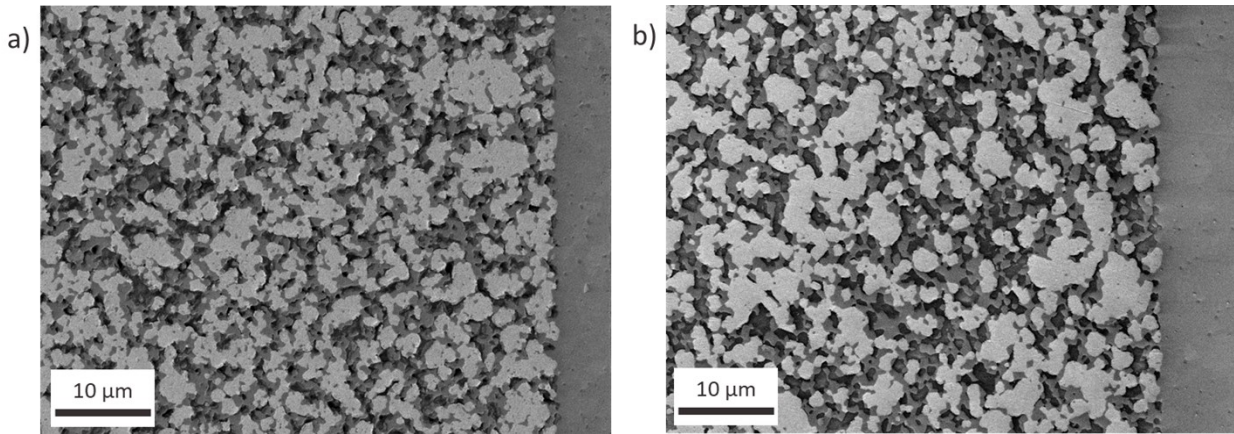


Figure S3 Cross sectional SEM images of SOFC electrodes after 500 h life testing at -1 A/cm^2 current density at 800°C in $85\% \text{ H}_2\text{O} - 15\% \text{ H}_2$ from (a) 70|70 and (b) and 70|50/70 cell.

Total Resistance = Inductance + Electrolyte Ionic + High Frequency + Transmission Line + Warburg

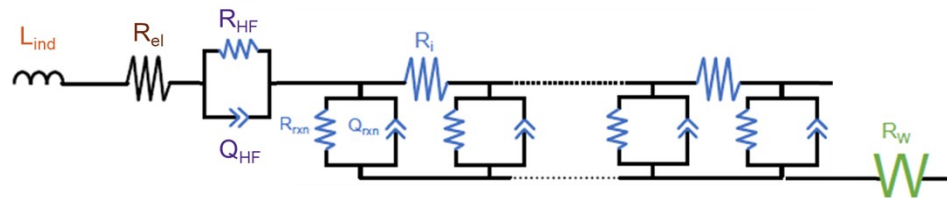


Figure S4 Equivalent circuit model that includes in series an inductance (L_{ind}) associated with test setup wiring, an electrolyte ionic resistance (R_{el}), a high frequency response (HF), a transmission line model element (TLM), and a Warburg gas diffusion element (R_w).

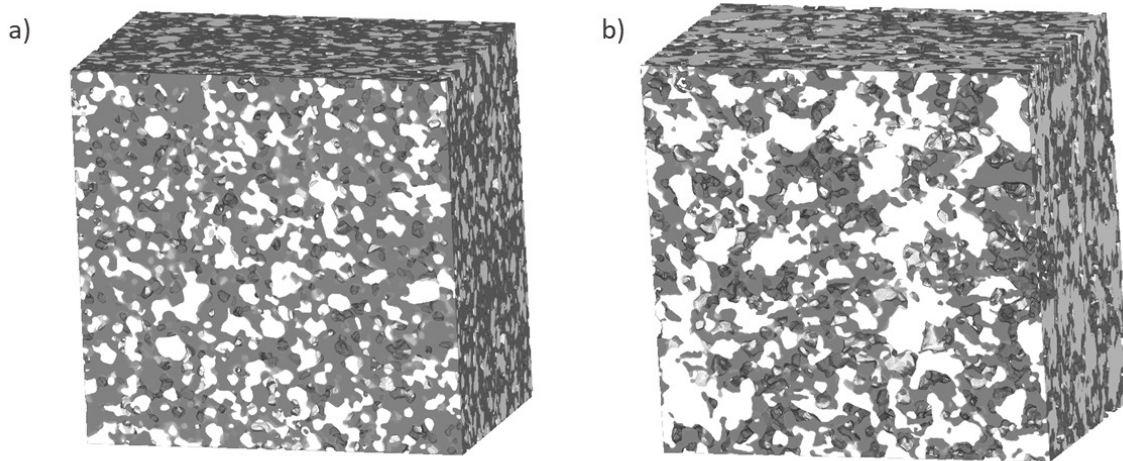


Figure S5 3D microstructure reconstruction of (a) 50 wt% NiO and (b) 70 wt% NiO characterized on Plasma FIB-SEM with size $27 \times 28 \times 27 \mu\text{m}^3$. The white, grey and black phases represent nickel, YSZ, and pore, respectively.

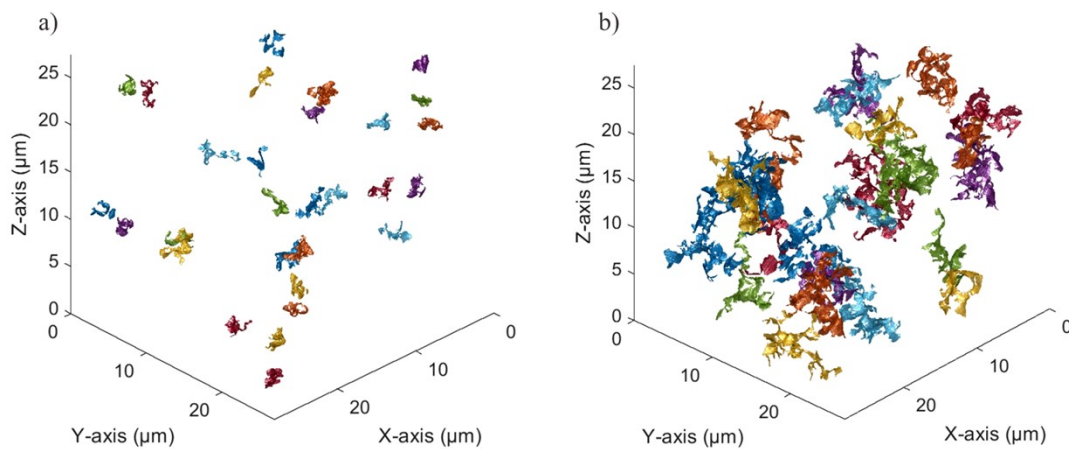


Figure S6 3D volume of Ni-pore interfaces of (a) 50 wt% NiO and (b) 70 wt% NiO electrodes. Only the 30 largest interfaces are shown for clarity, and the different colors indicate different isolated Ni-pore interfaces.

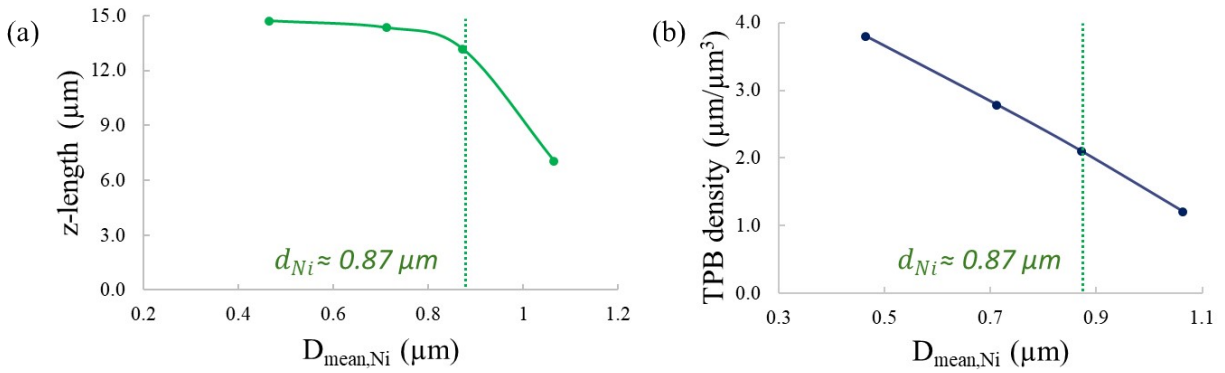


Figure S7 Evolution of the average z-length of Ni-Pore interfaces (a) and TPB lengths density (b) as a function of the Ni mean diameter (d_{Ni}). Microstructural properties were calculated on 3D artificial microstructures ($15 \mu\text{m}^3$ cubes) generated with Ni = 37.5 vol%, YSZ = 37.5 vol%, porosity = 25 vol%, and $d_{\text{YSZ}} = 0.68 \mu\text{m}$.

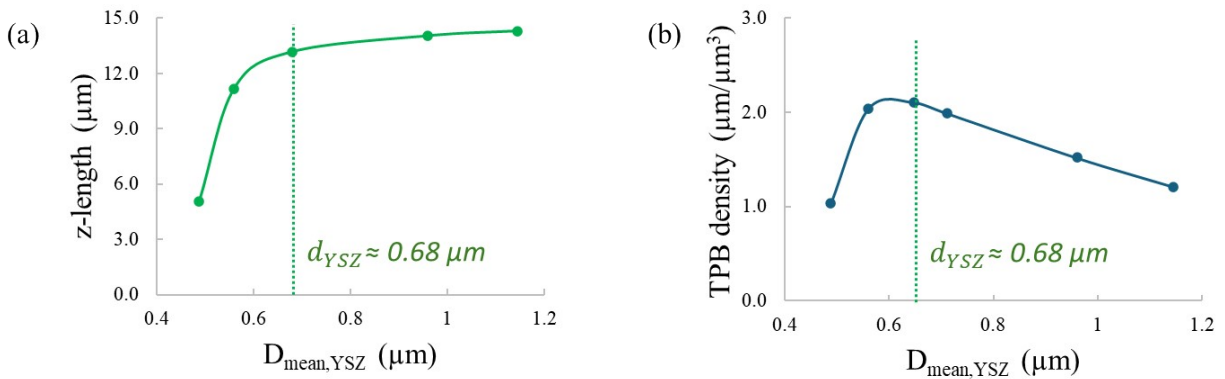


Figure S8 Evolution of the average z-length of Ni-Pore interfaces (a) and TPB lengths density (b) as a function of the YSZ mean diameter (d_{YSZ}). Microstructural properties were calculated on 3D artificial microstructures ($15 \mu\text{m}^3$ cubes) generated with Ni = 37.5 vol%, YSZ = 37.5 vol%, porosity = 25 vol%, and $d_{\text{Ni}} = 0.87 \mu\text{m}$.

Supplementary Table

Table S1 Microstructural properties of the 3D artificial microstructures

NiO:YSZ	45:55	50:50	55:45	60:40	65:35	70:30	75:25	[%wt]
Pore	16.5	20.1	21.8	23.1	27.0	28.6	30.8	[%vol]
YSZ	58.1	52.4	47.6	43.1	37.2	32.8	27.5	[%vol]
Ni	25.3	27.5	30.6	33.8	35.8	38.6	41.7	[%vol]
d_{pore}	0.45	0.50	0.52	0.57	0.64	0.68	0.72	[μm]
d_{YSZ}	0.74	0.74	0.70	0.71	0.70	0.69	0.68	[μm]
d_{Ni}	0.74	0.79	0.83	0.89	0.96	1.00	1.06	[μm]
TPB density	1.65	1.88	1.88	1.92	1.78	1.73	1.64	[μm^{-2}]
z-length	1.7	3.0	4.4	9.1	13.4	14.0	14.6	[μm]

Table S2 Ni-Pore interface size distribution for Ni-YSZ electrodes with different initial NiO wt%

Ni-Pore Interface Size [μm]		
NiO-YSZ	50-50	70-30
<1	18416	9478
1 to 2	1819	1562
2 to 3	132	402
3 to 4	8	133
4 to 5	0	43
≥ 5	0	41