

Supporting information:

Boosting performance and durability of Ni/YSZ cathode via decoration with ultrafine CGO electro-catalyst for hydrogen production at high current densities

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Initial characterizations:

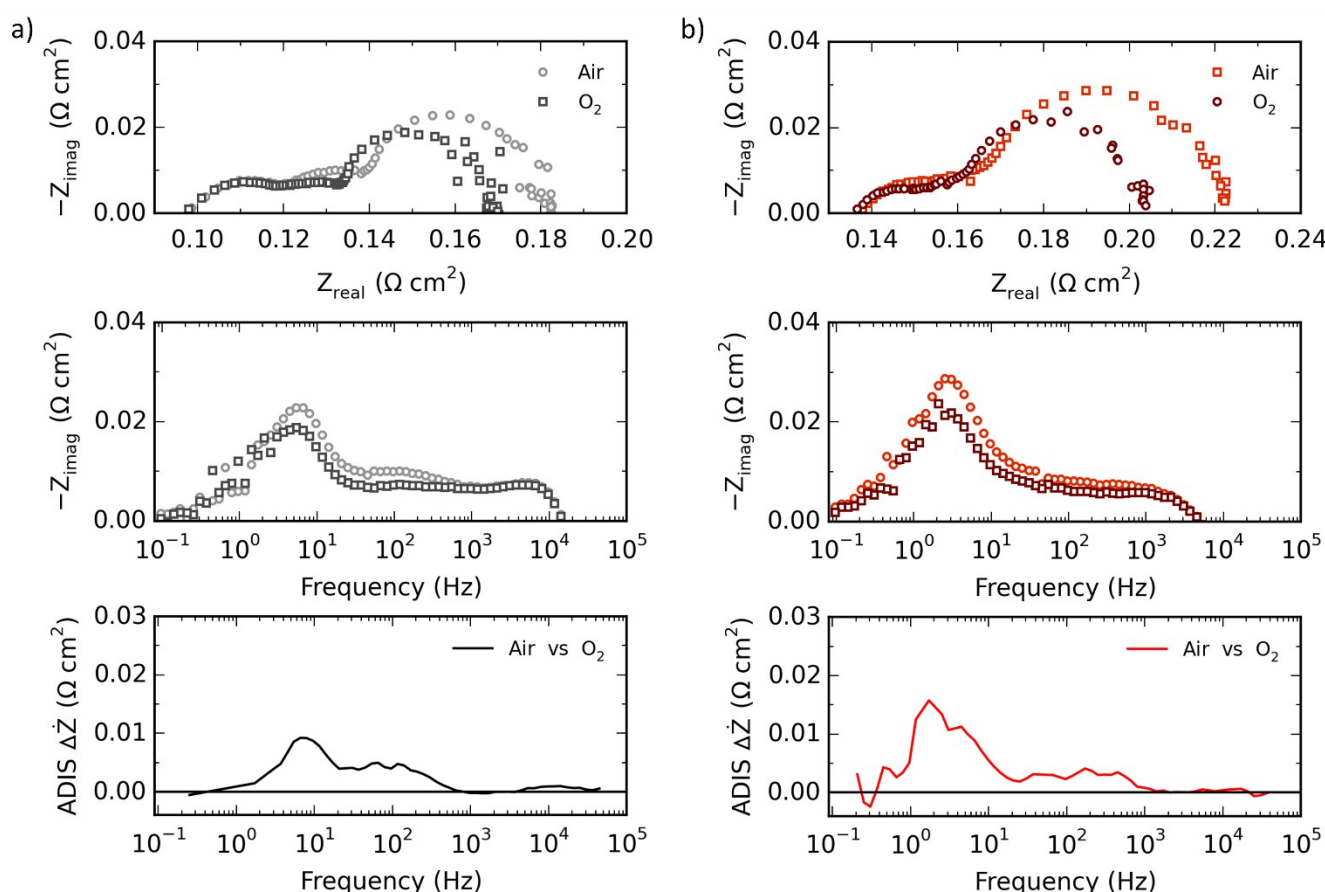


Figure S1: EIS of a) Cell A, and b) Cell B1 for oxygen electrode gas shift with corresponding ADIS plot where EIS measured by applying air was taken as a reference.

Durability tests:

Table S1: Cell voltages and resistances measured during the long-term durability test for Cell A with the bare Ni/YSZ electrode and Cell B1 with the CGO infiltrated Ni/YSZ electrode.

	Cell A			Cell B1		
Time (h)	0	60	750	0	500	1000
Voltage (V)	1.265	1.365	1.789	1.218	1.251	1.332
R_s ($m\Omega\text{ cm}^2$)	77.4	100.6	368.0	130.5	121.0	125.7
R_p ($m\Omega\text{ cm}^2$)	383.7	407.8	439.8	208.0	268.0	431.7

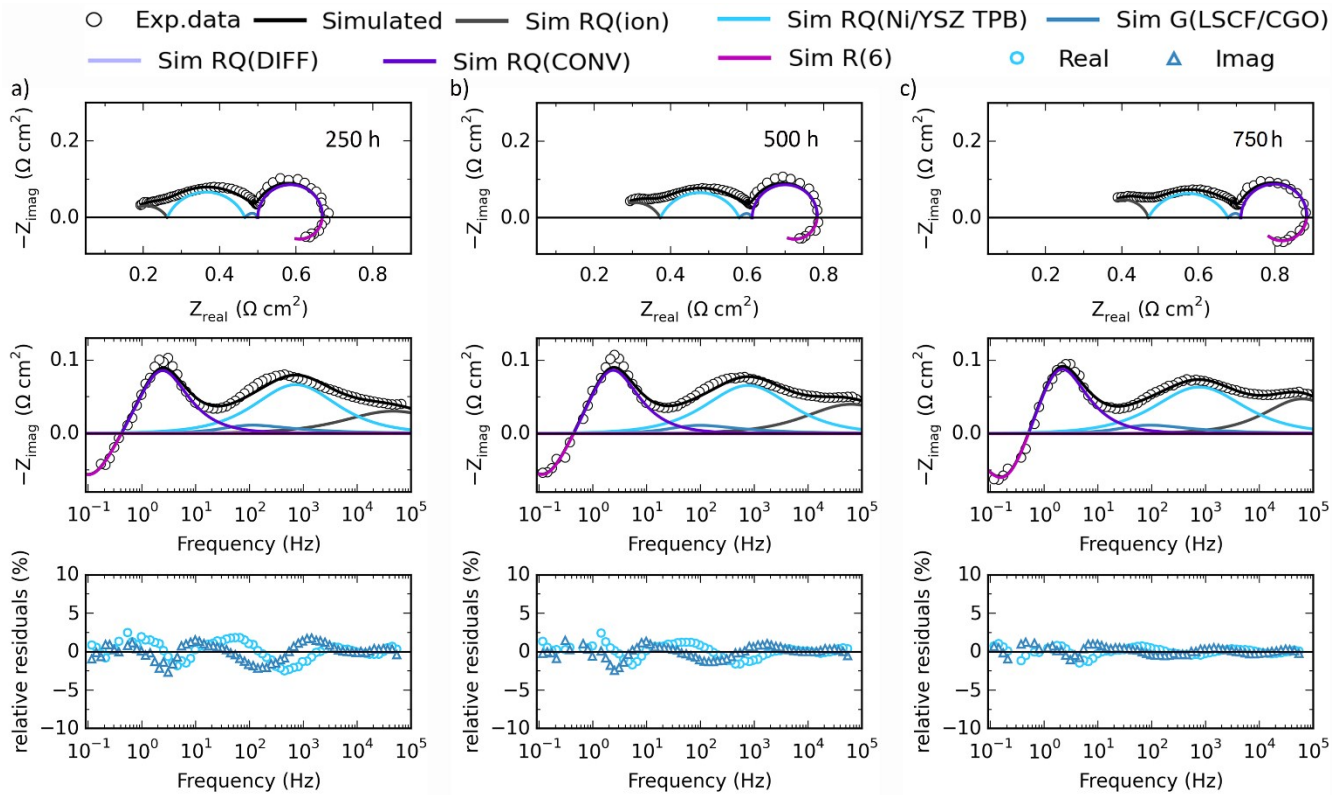


Figure S2: Results of fitting equivalent circuit models to the impedance spectra for Cell A during long-term steam electrolysis at -1.25 A/cm^2 , $800\text{ }^\circ\text{C}$ and 70% steam conversion: a) 250h, b) 500h, and c) 750h

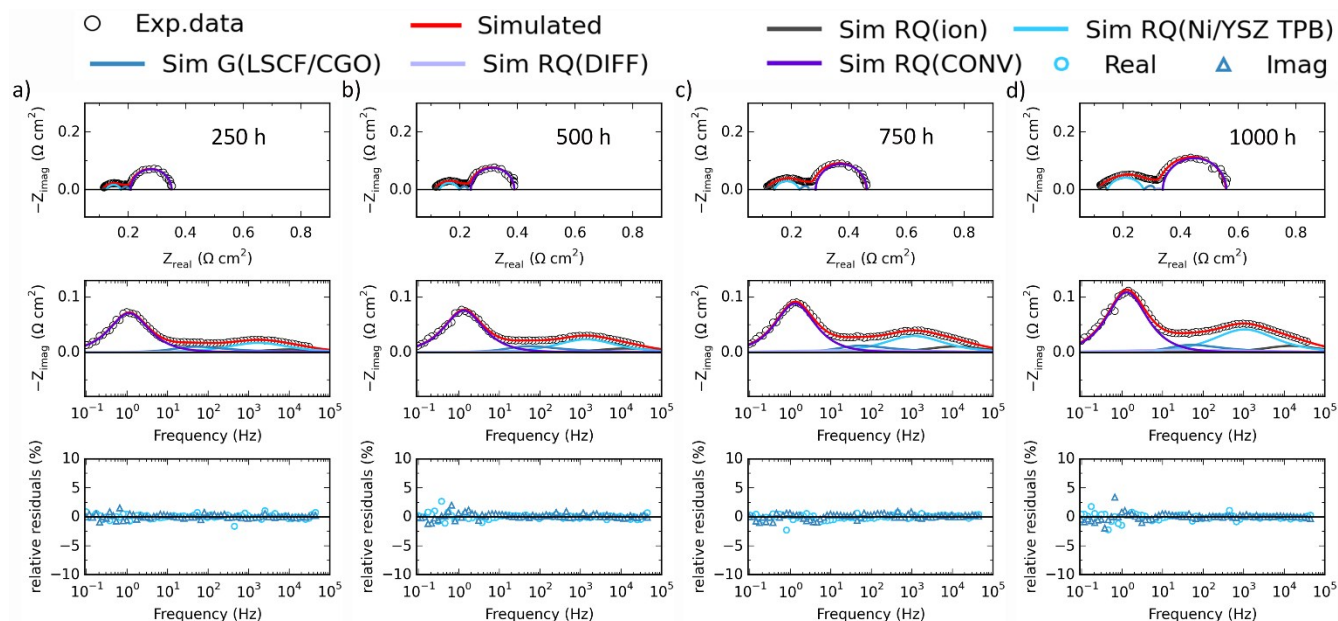


Figure S3: Results of fitting equivalent circuit models to the impedance spectra for Cell B1 during long-term steam electrolysis at -1.25 A/cm^2 , 800°C and 70% steam conversion: a) 250h, b) 500h, c) 750h, and d) 1000h

Table S2: Results of fitting equivalent circuit models to the impedance spectra for Cell A and Cell B1 during long-term steam electrolysis at -1.25 A/cm^2 , 800°C and 70% steam conversion (results from Figure S2 and Figure S3). The estimated uncertainty for the simulated resistances is $\sim \pm 7 \text{ m}\Omega \text{ cm}^2$.

R ($\Omega \text{ cm}^2$)	Cell A				Cell B1				
	0h	250h	500h	750h	0h	250h	500h	750h	1000h
R_s	0.067	0.154	0.240	0.338	0.117	0.111	0.110	0.113	0.113
R_{ion}	0.013	0.109	0.132	0.132	0.009	0.014	0.019	0.029	0.031
$R_{\text{Ni/YSZ TPB}}$	0.187	0.203	0.206	0.208	0.038	0.051	0.069	0.088	0.128
$R_{\text{LSCF/CGO}}$	0.026	0.031	0.031	0.032	0.027	0.028	0.031	0.034	0.038
R_{DIFF}	0.003	0.002	0.003	0.002	0.006	0.006	0.010	0.023	0.027
R_{CONV}	0.164	0.171	0.173	0.176	0.145	0.147	0.151	0.179	0.219
R_p	0.393	0.516	0.545	0.550	0.225	0.246	0.280	0.353	0.443

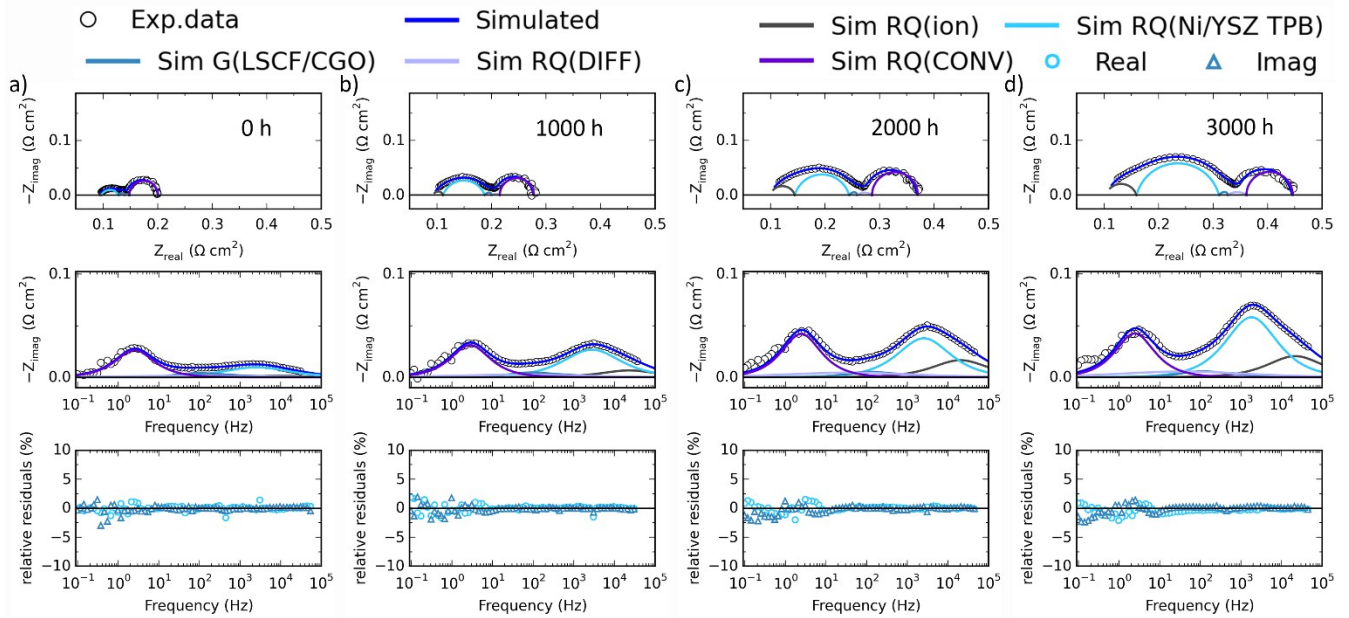


Figure S4: Results of fitting equivalent circuit models to the impedance spectra for Cell B2 during long-term test under -1.25 A/cm^2 (11test104) a) 0 h and b) 1000 h, c) 2000 h, d) 3000 h (under -1.25 A/cm^2)

Table S3: Results of fitting equivalent circuit models to the impedance spectra for Cell B2 during long-term steam electrolysis at -1.25 A/cm^2 , 800°C and 42% steam conversion (results from Figure S4). The estimated uncertainty for the simulated resistances is $\sim \pm 6 \text{ m}\Omega \text{ cm}^2$.

R ($\Omega \text{ cm}^2$)	Cell B2						
	0h	250h	500h	750h	1000h	2000h	3000h
R_s	0.093	0.089	0.090	0.091	0.093	0.099	0.103
R_{ion}	0.004	0.005	0.010	0.015	0.018	0.045	0.056
$R_{\text{Ni/YSZ TPB}}$	0.032	0.044	0.050	0.060	0.076	0.100	0.151
$R_{\text{LSCF/CGO}}$	0.011	0.012	0.013	0.013	0.013	0.015	0.016
R_{DIFF}	0.009	0.012	0.013	0.015	0.015	0.027	0.034
R_{CONV}	0.053	0.051	0.052	0.055	0.062	0.084	0.085
R_p	0.109	0.124	0.138	0.158	0.184	0.271	0.342

Microstructure of reference cells:

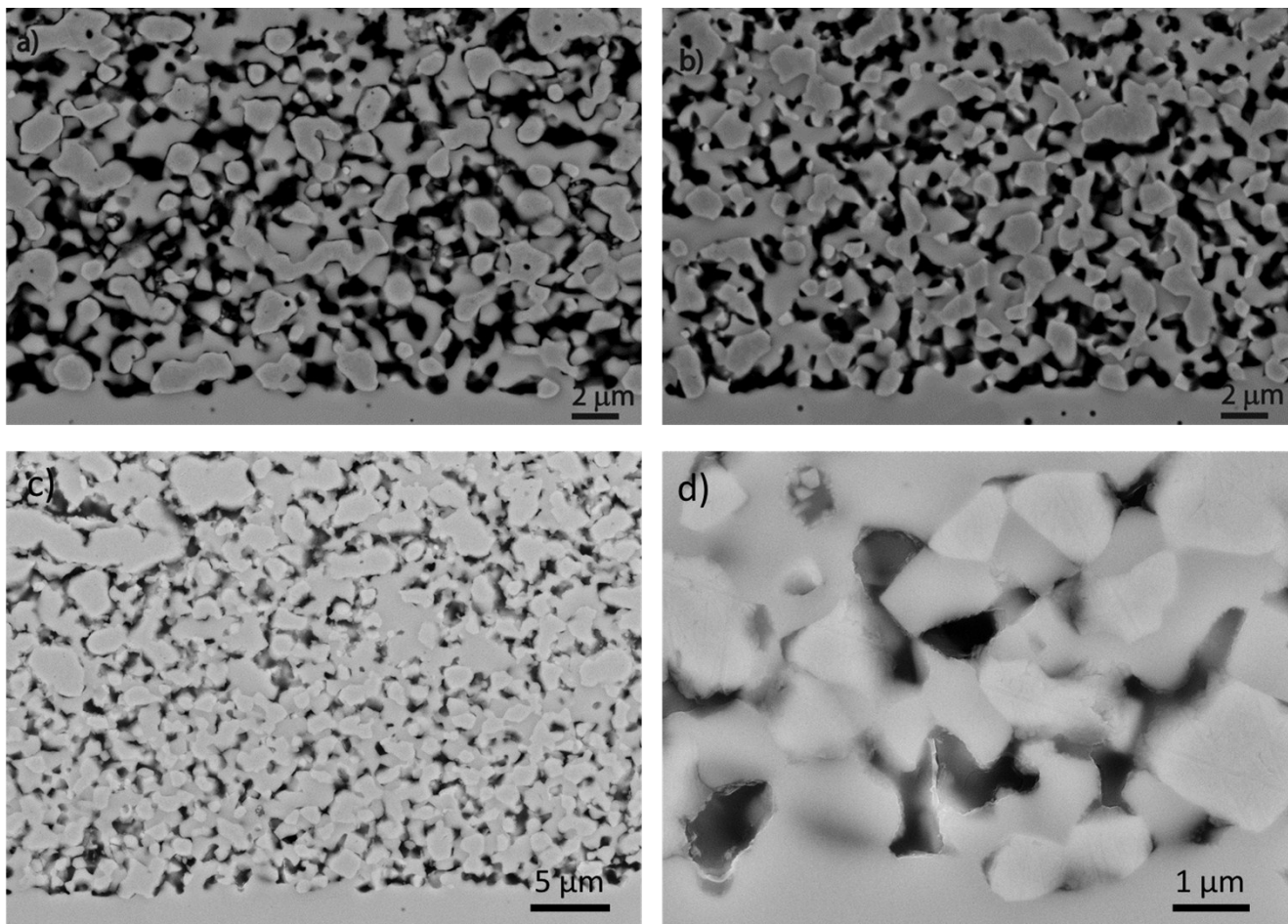


Figure S5: a) Microstructure of Cell A after long term electrolysis test, b) Microstructure of polished cross-section of reduced Ni/YSZ backbone or a cell after short test. (c,d) Microstructure of Cell B1 after long term electrolysis test.

Parallel study on CGO particles growth in reducing atmosphere:

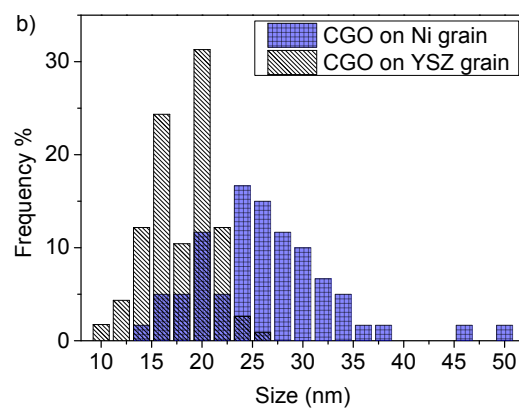
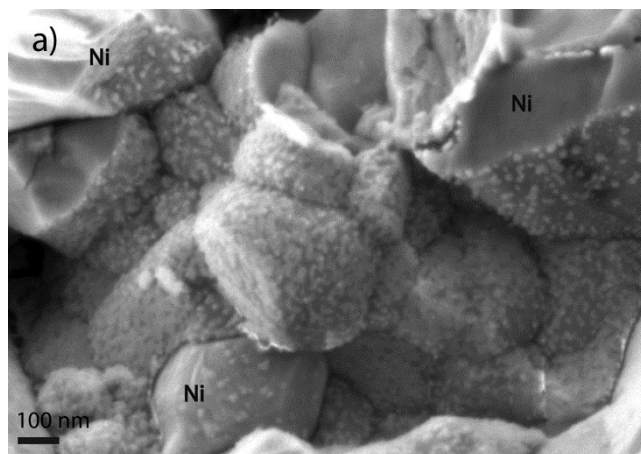


Figure S6: a) Microstructure of CGO infiltrated particles after 600 h at 550 °C in wet H_2/N_2 atmosphere, b) with particles size distribution measured from more than 100 CGO particles on the Ni grains and on the YSZ grains.