Supporting Information

Bottom-Up Redox State Engineering of Ni/GDC Thin-Film Anodes for Carbon Tolerant Syngas-Fueled SOFCs

Inyoung Jeong, [1†] *Daniel Gil,* [1] *Myung Seok Lee,*[1] *Yujae Jang,*[1] *Hyungjun Kim,*[1,2*] *and Suk Won Cha*[1*]

[1]Department of Mechanical Engineering, Seoul National University, 1 Gwanak-ro, Gwanak-gu, Seoul 08826, Republic of Korea

[2]Department of Materials Science and Engineering, Northwestern University, Evanston, Illinois 60208, United States

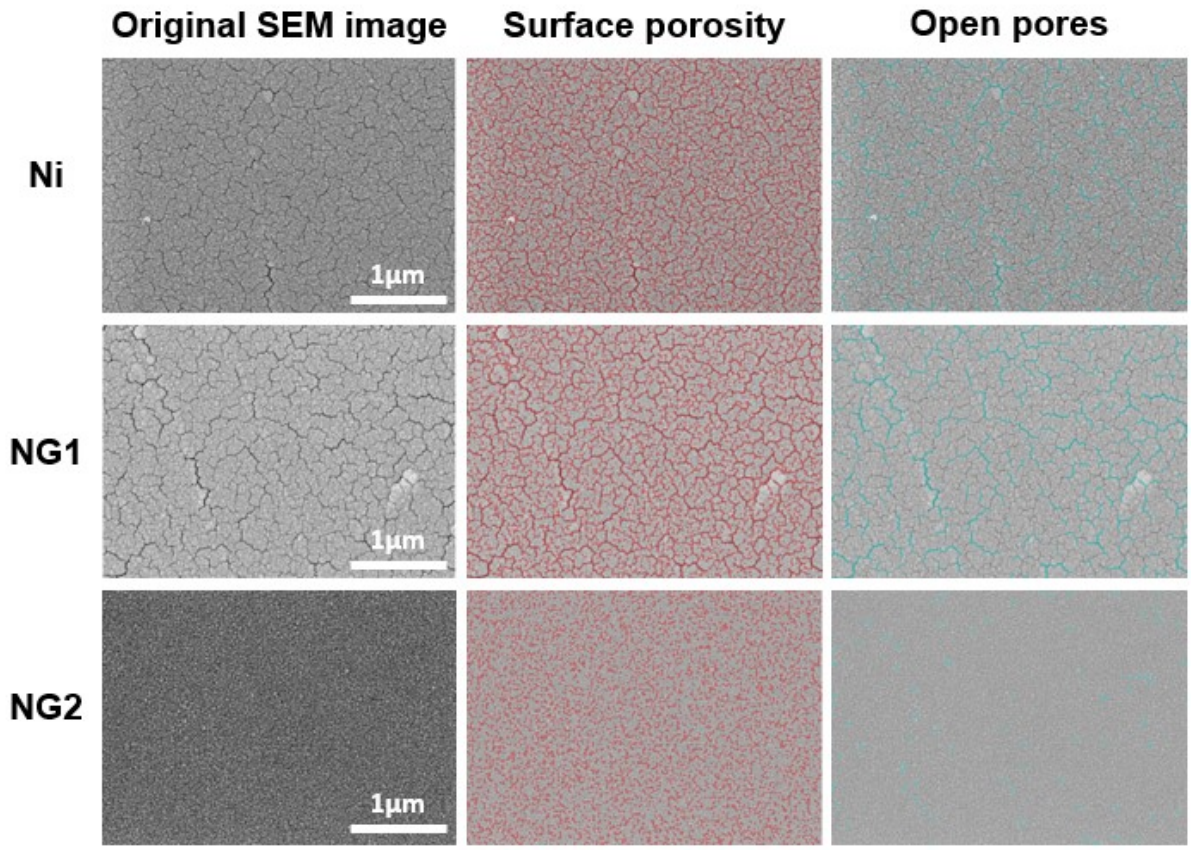

Figure S1. Quantitative analysis of the apparent surface porosity of the as-fabricated Ni, NG1, and NG2 anodes based on top-view FESEM images. For each anode, the original SEM image, the segmented open-pore regions used to calculate the total surface porosity, and the segmented coarse open-pore regions are shown. The estimated surface porosities were 29.57%, 29.29%, and 24.27% for Cells Ni, NG1, and NG2, respectively, while the corresponding coarse open-pore fractions were 5.21%, 6.47%, and 0.94%.

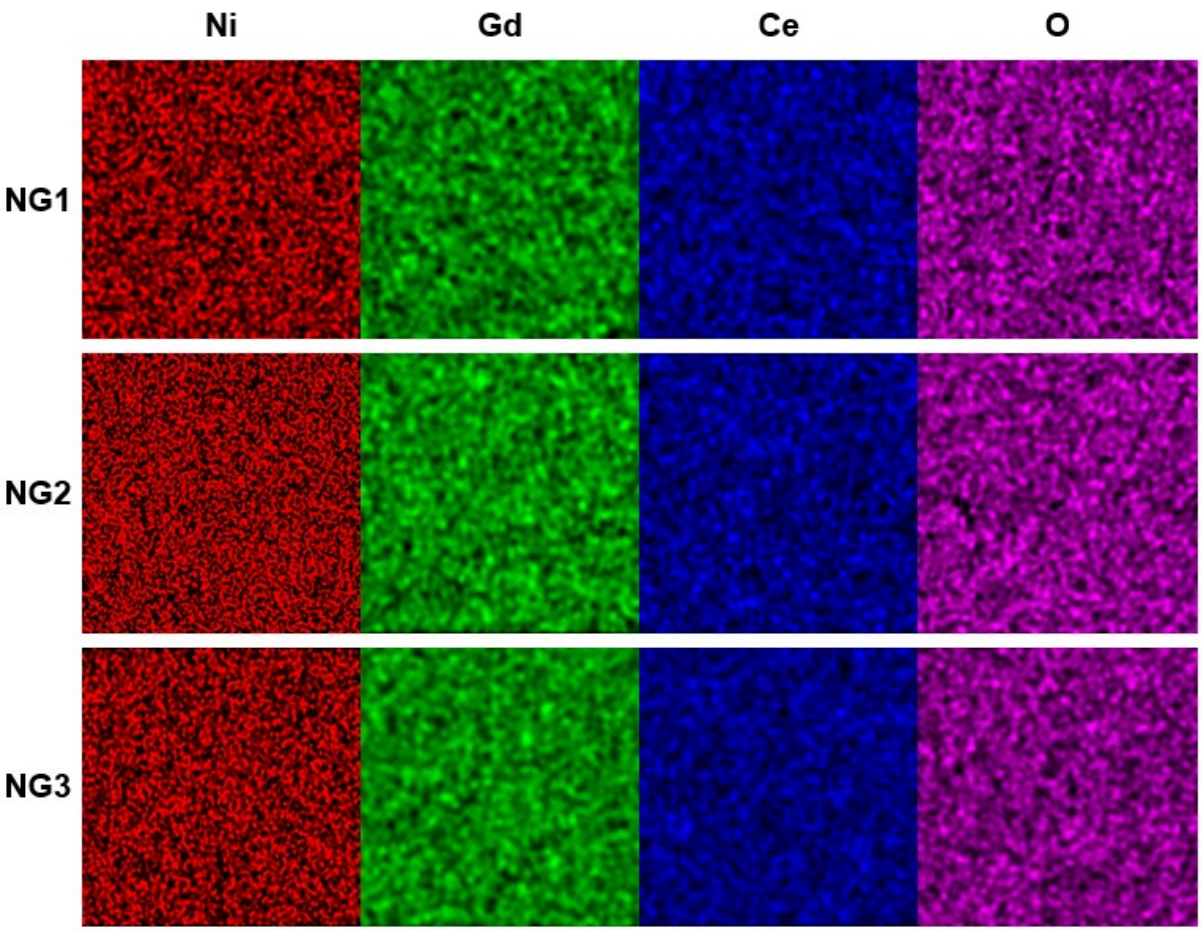

Figure S2. FESEM-EDS characterization of as-deposited Ni–GDC thin films

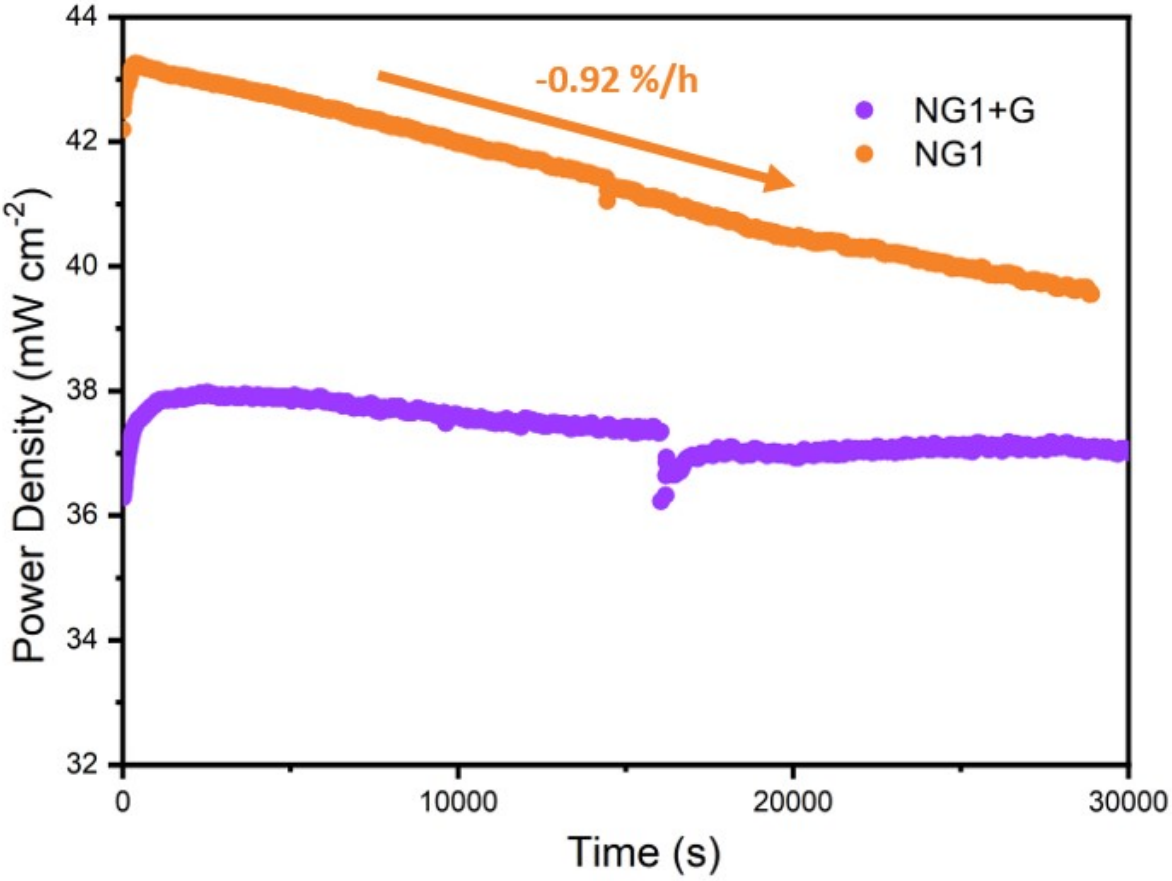

Figure S3. Long-term galvanostatic stability test of NG1 and NG1+G cells operated at 0.8 A for 9 hours under syngas conditions.

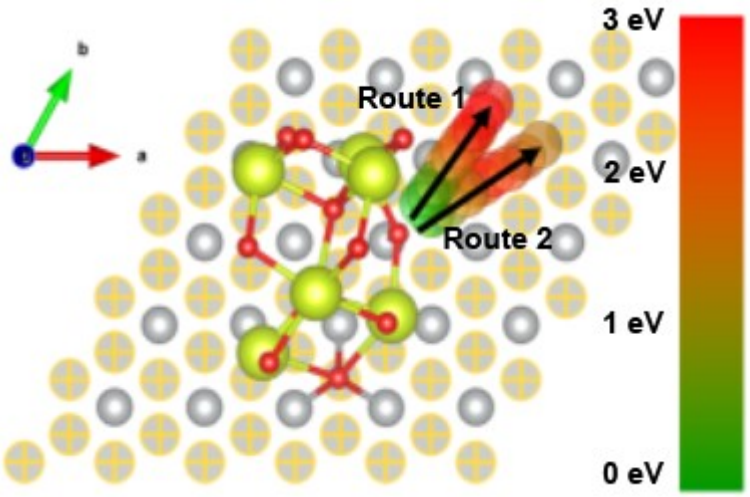

Figure S4. Energy landscape for CO adsorption at different sites on the Ni-ceria oxide interface, comparing Route 1 (fcc site) and Route 2 (bridge site). The color gradient represents adsorption energy, with lower energy (green) indicating more favorable adsorption.

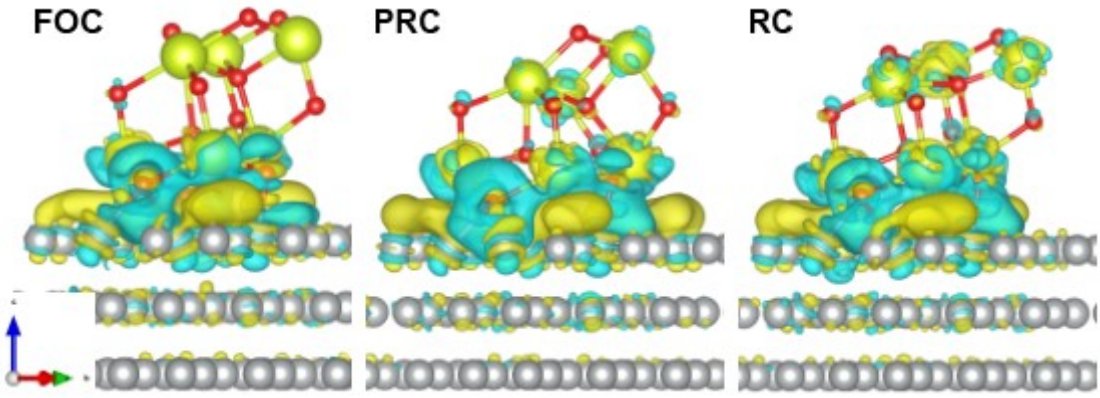

Figure S5. Energy density difference maps of the Ni-ceria oxide interfaces for the fully oxidized (FOC), partially reduced (PRC), and reduced (RC) ceria models. Yellow and cyan regions represent electron accumulation and depletion, respectively.

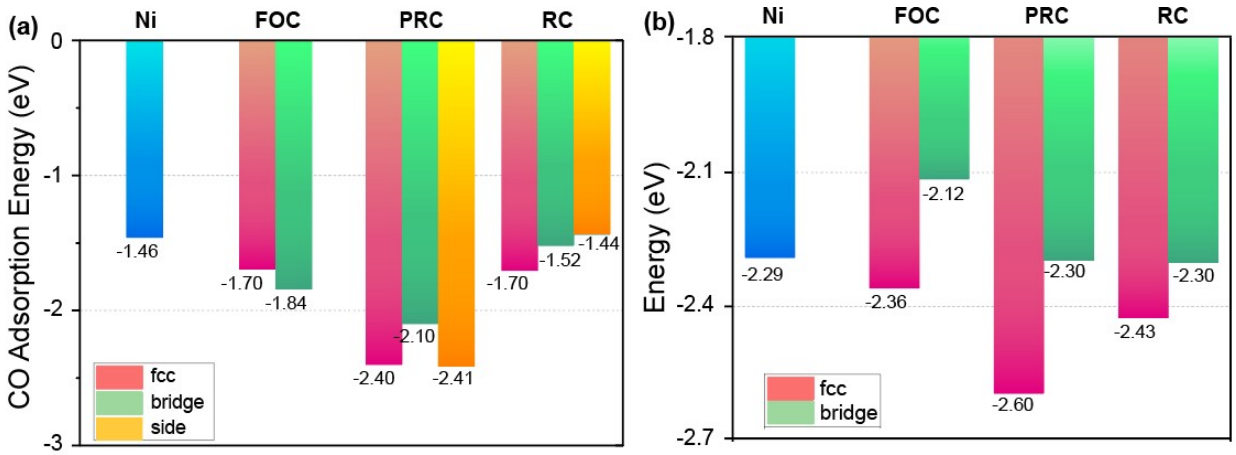

Figure S6. (a) CO adsorption energies on Ni and CeO_2 cluster models (FOC, PRC, RC) at different adsorption sites (fcc, bridge, side). (b) Energy differences for the reverse Boudouard reaction calculated at fcc and bridge sites on the same surfaces.

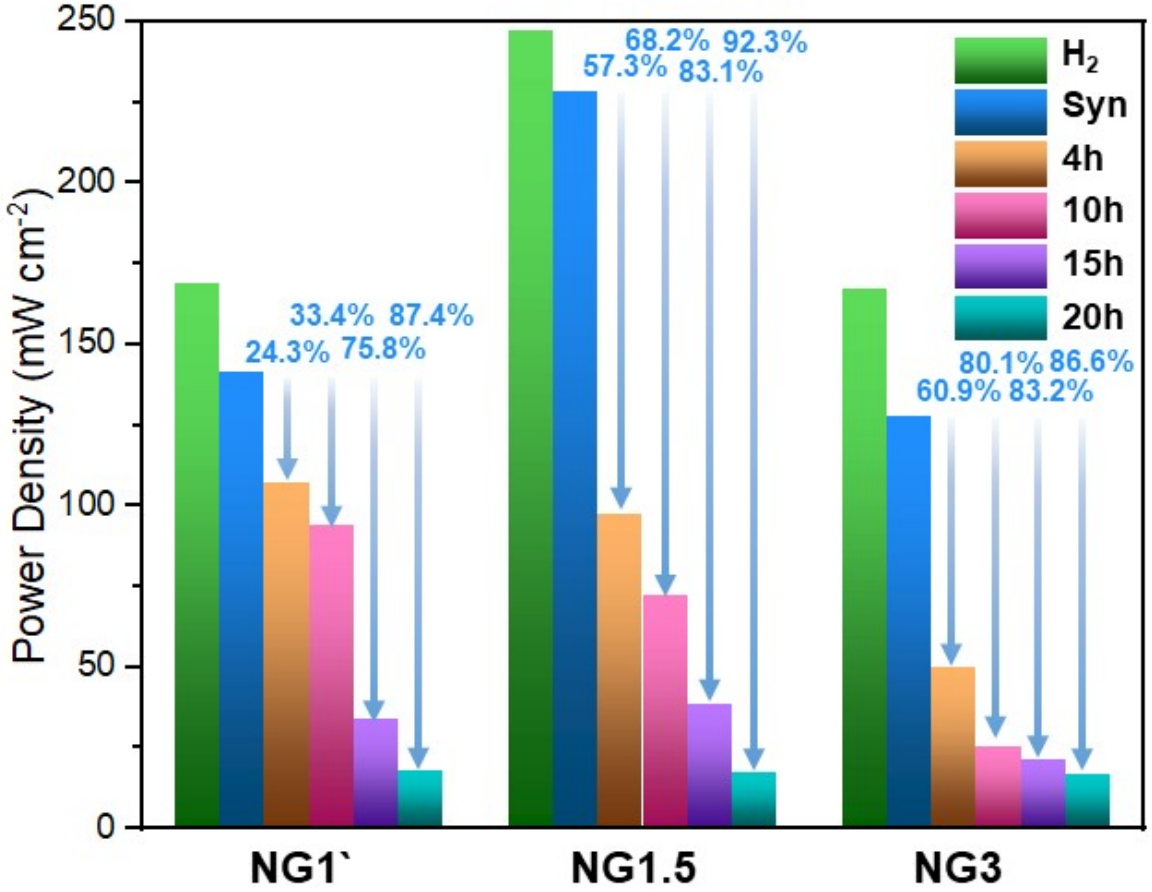

Figure S7. Power densities of NG1, NG1.5, and NG3 measured in H_2 and syngas, together with their evolution during extended 20 h syngas operation.

Table S1. EDS-derived elemental composition of the as-deposited Ni–GDC witness films

Elements(atomic. %)	Cell Ni	Cell NG1	Cell NG1.5	Cell NG2	Cell NG3
Ni	100.0	97.0	95.0	91.5	73.3
Gd	0	0.7	1.1	1.9	5.9
Ce	0	2.3	3.9	6.6	20.8