

Supporting Information

Fabrication and Evaluation of 5x5 cm²-sized Single Cells

The 5x5 cm²-sized cells were fabricated via tape-casting and a co-firing technique, as reported in our previous investigation. The NiO-GDC / Sn doped NiO-GDC anodes and GDC electrolyte tapes were cast to 100 μm and 16μm, respectively. Next, a laminator with 10 sheets of anode tape and a sheet of electrolyte tape was co-fired at 1350 °C. Finally, a LSCF-GDC cathode was screen-printed to obtain a 16 cm² active area and sintered at 1000 °C.

The cell performances and the carbon deposition for 5x5 cm²-sized cells can be affected from the flow paths and depths of housing. Even if the deeper flow path or different flow path can prevent the carbon deposition when supplying of dry CH₄, it also affected on the distribution and fuel utilization.

The cell performances for Ni-GDC and Sn-doped Ni-GDC single cells were conducted with the 2 mm depth and parallel flow path of SUS316L housing.

The 5x5cm² sized cells were also evaluated for confirming the carbon deposition in the area of lack of oxygen ions and heterogeneous distribution of CH₄ fuel. The 5x5 cm² Ni-GDC cell and Sn-doped Ni-GDC cell showed maximum powers of 7.51 W and 8.92 W with hydrogen, and 6.57 W and 7.95 W with dry methane, respectively. The 5x5 cm² SNG single cell showed incredibly over 50 hours of stability without carbon deposition, while the voltage of 5x5 cm² NG single cell dropped rapidly in short term of operation due to the cell fracture caused from the carbon deposition (**Fig. S3**). The carbon deposition occurred on the edge of 5x5 cm² NG single cell due to the lack of oxygen ions from the cathode and the inconsistent distribution of methane (**Fig. S4**).

Supporting Figures

Fig. S1. The average pore size distributions of the (a) Ni-GDC anode (dot) and Sn doped Ni-GDC (line), and (b) Ni/GDC-GDC (dot) and Sn doped Ni/GDC-GDC anode (line).

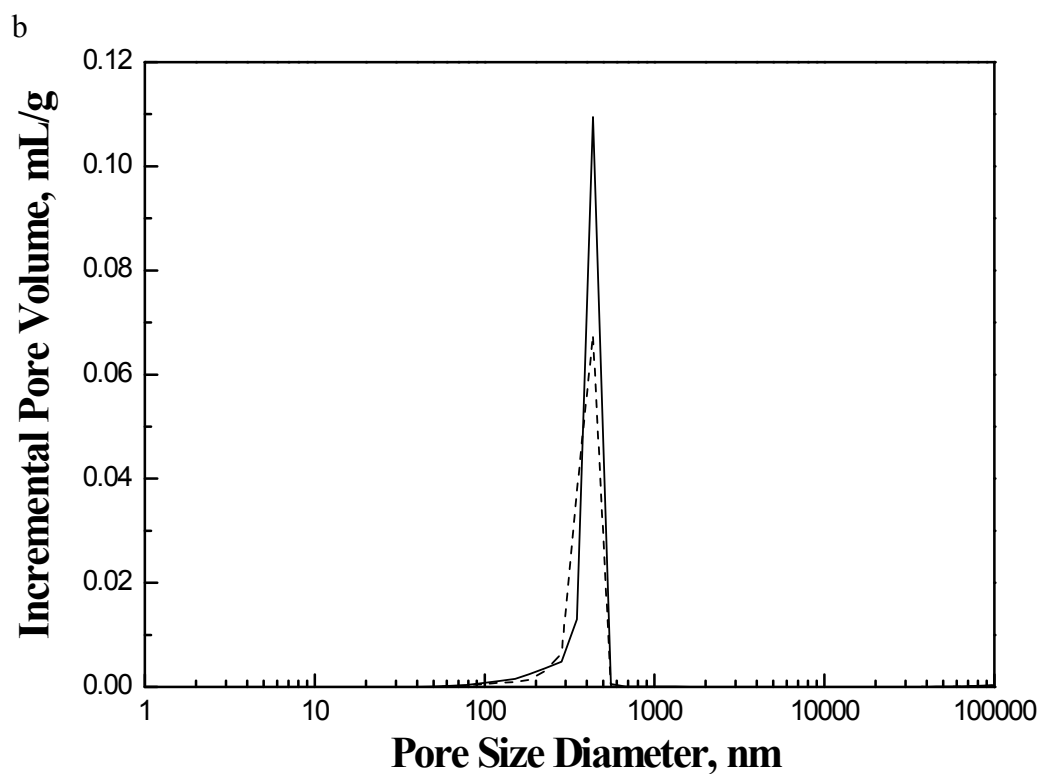
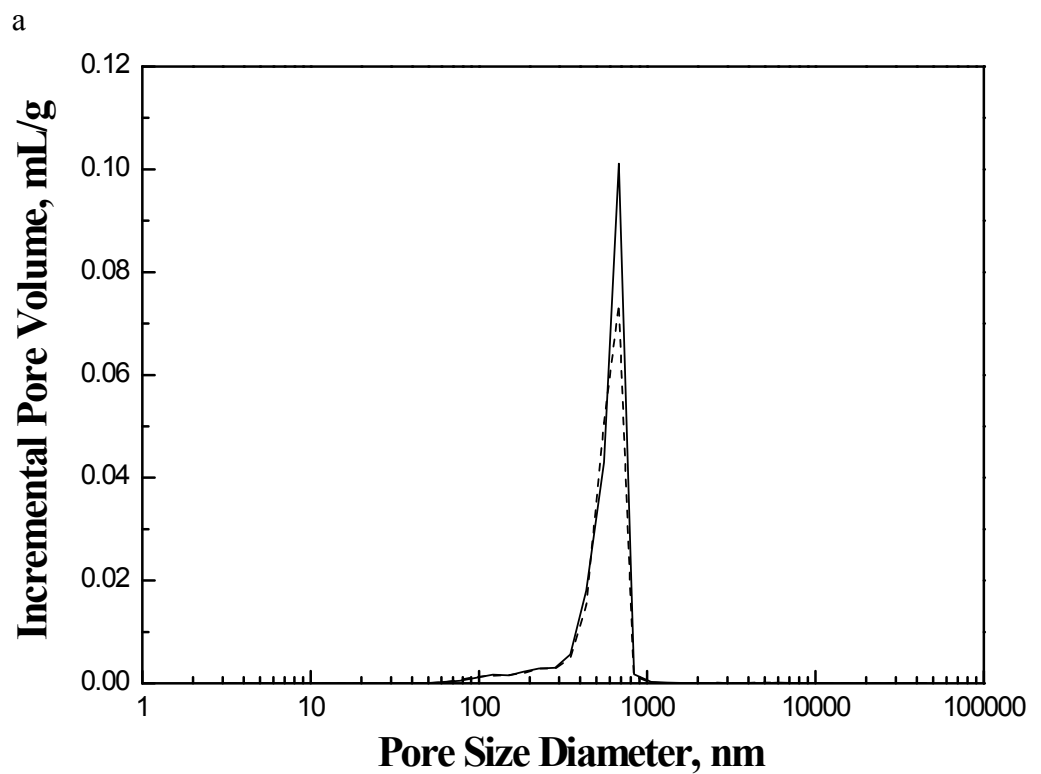
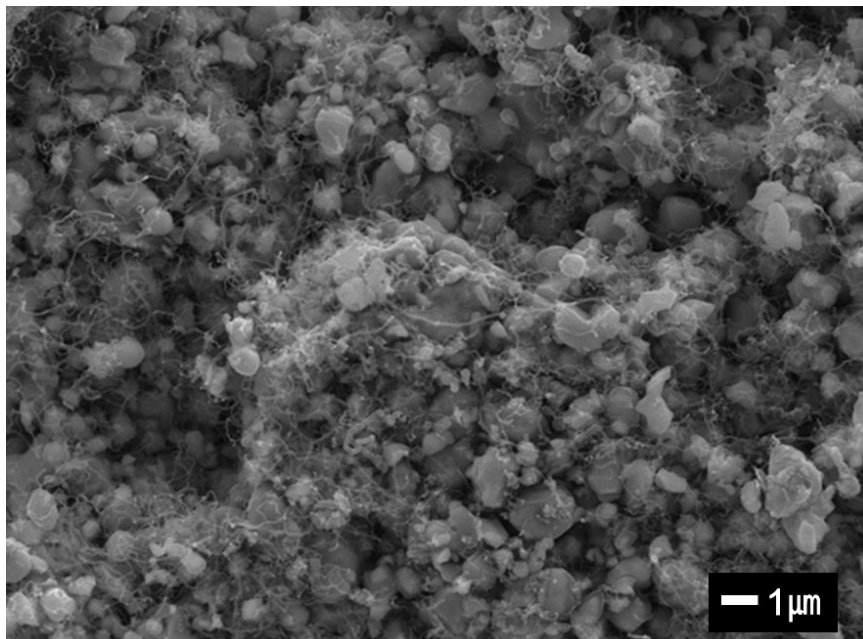


Fig. S2. Cross-sectional SEM images from (a) the Ni-GDC and (b) 0.5 wt% Sn-doped Ni-GDC single cells after their long-term stability tests with dry methane.

a



b

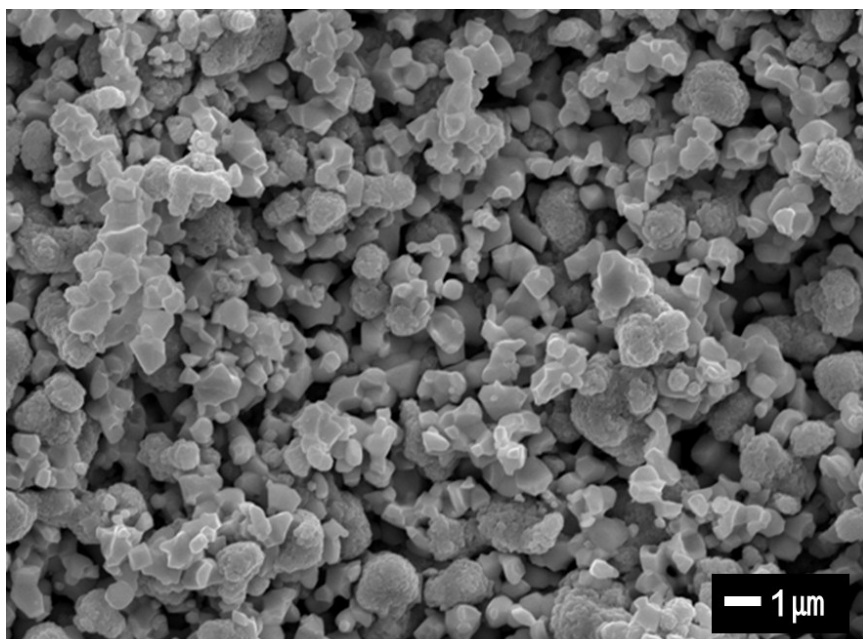


Fig S3. (a) I-V curves and (b) stability test of 5x5cm² sized Ni -GDC (closed) and 0.5 wt% Sn-doped Ni-GDC (opened) anode-supported single cells with H₂ (blue) and CH₄ (red) at 650 °C.

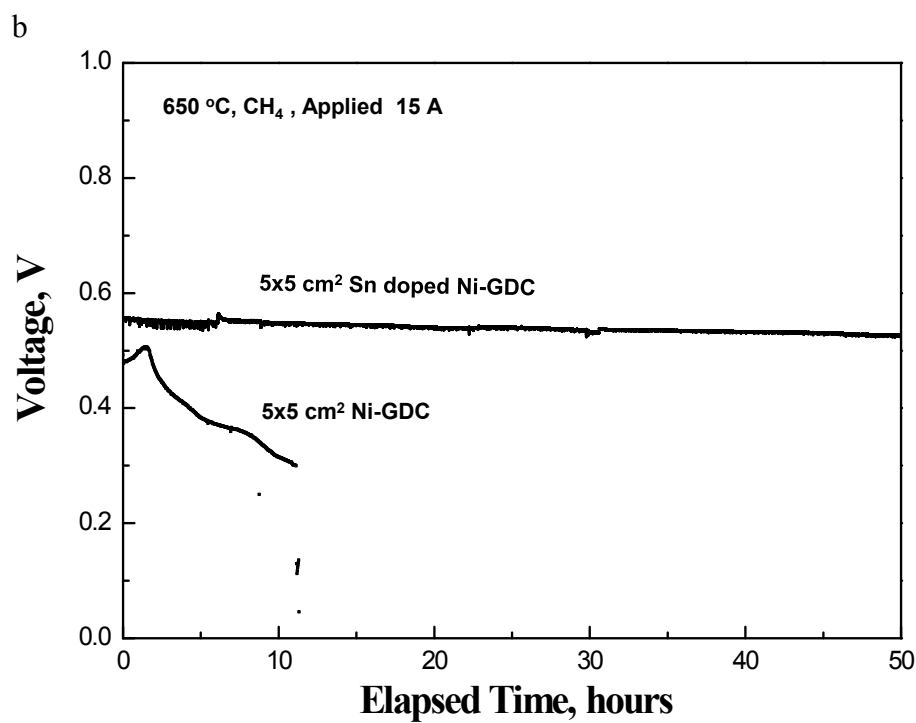
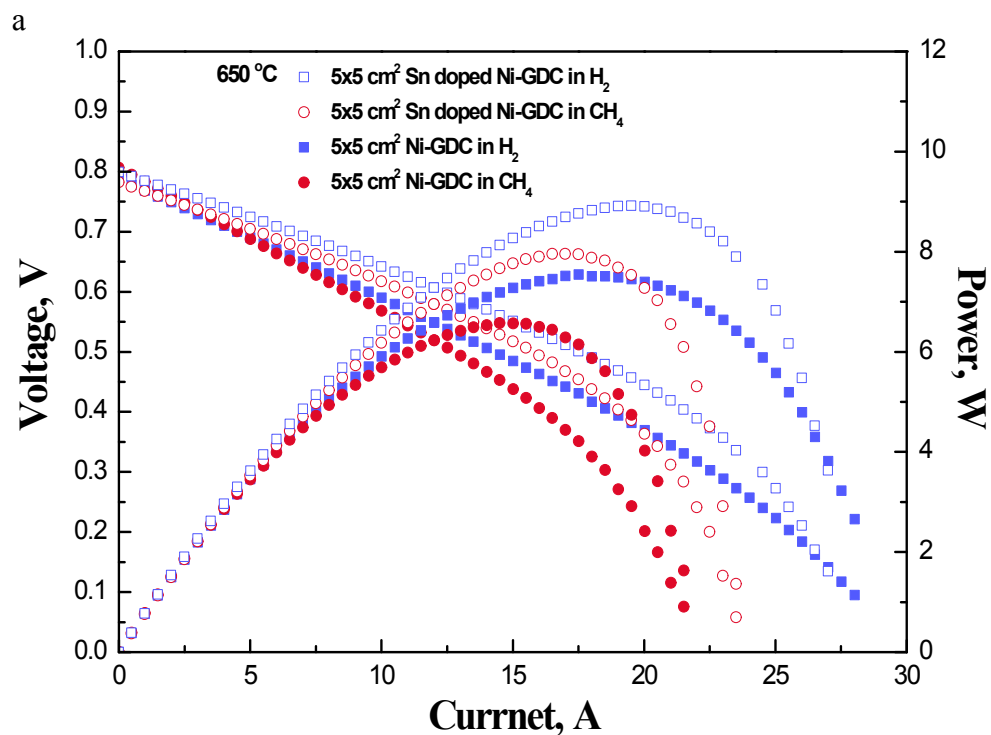


Fig. S4. Digital images of 5x5 cm² sized (a-b) Ni –GDC and (c-d) Sn doped Ni-GDC single cell with SUS316L housing after long-term operating with dry methane at 650 °C.

