Supplementary Information

Co-electrolysis of H_2O and CO_2 in a solid oxide electrolysis cell with hierarchically structured porous electrodes

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1. SOEC single cell fabricated by dry-pressing and screening-printing method

The traditional button cell with Ni-YSZ hydrogen electrode, YSZ electrolyte and LSM-YSZ oxygen electrode fabricated by dry-pressing, dip-coating and screen printing method, respectively, has also been studied for H_2O electrolysis and H_2O-CO_2 co-electrolysis (Fig. S1) [10]. Three different gas compositions were used as feeding gas, e.g. 30 vol.% H_2-70 vol.% H_2O, 50 vol.% H_2-50 vol.% H_2O, 25 vol.% H_2-50 vol.% H_2O-25 vol.% CO_2 (Fig. S2). When 50 vol.% H_2O-50 vol.% H_2 was used as feeding gas, the voltage of the cell fabricated by the traditional method was only 0.30 A cm^{-2} (Fig. S2).

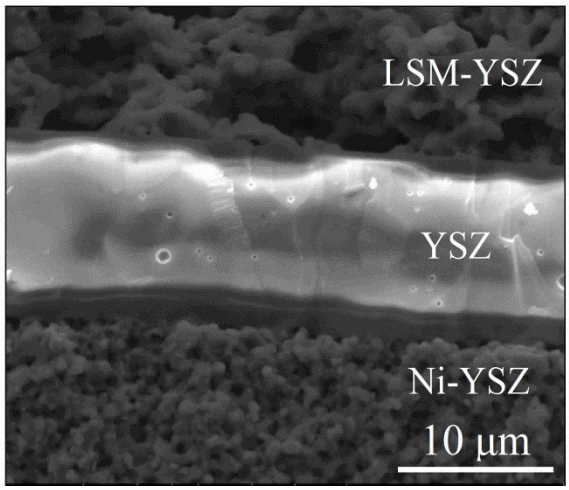

Fig. S1. Cross sectional SEM micrographs of LSM-YSZ/YSZ/Ni-YSZ single cell fabricated by traditional dry-pressing and screening-printing method.

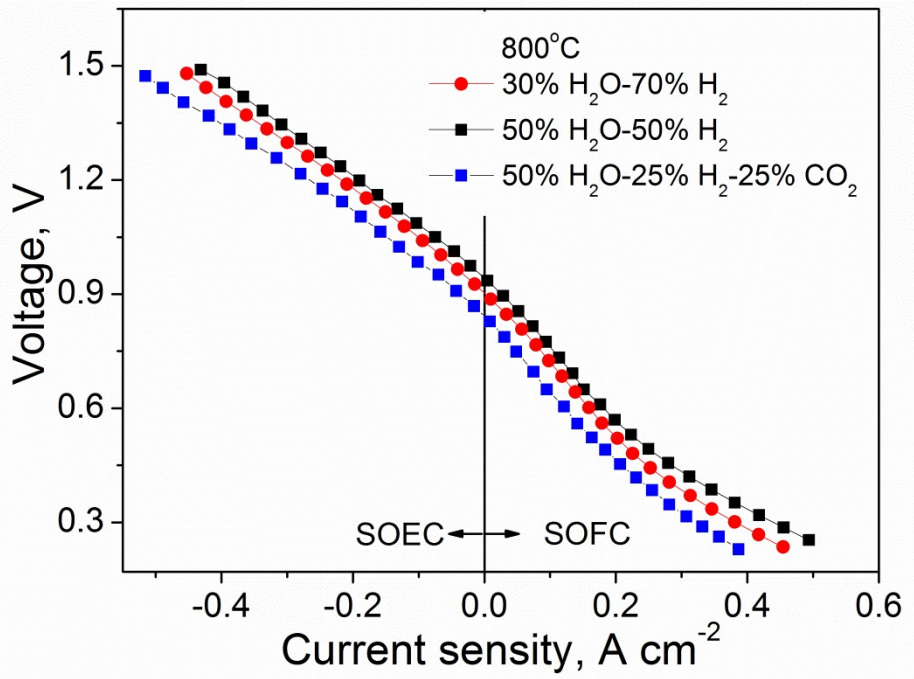

Fig. S2. DC potential-current density curves of LSM-YSZ/YSZ/Ni-YSZ cell operated in 50 vol.% H_2O-50 vol.% H_2 at 800°C.

2. SOEC single cell fabricated by dry-pressing and infiltration method

The button cell with Ni-SDC-YSZ hydrogen electrode, YSZ electrolyte and LSM-YSZ oxygen electrode fabricated by dry-pressing, dip-coating and infiltration method, respectively, has also been studied for H_2O electrolysis and H_2O-CO_2 co-electrolysis (Fig. S3) [12]. Three different gas compositions were used as feeding gas, e.g. 30 vol.% H_2-70 vol.% H_2O, 50 vol.% H_2-50 vol.% H_2O, 25 vol.% H_2-50 vol.% H_2O-25 vol.% CO_2 (Fig. S4). When 50 vol.% H_2O-50 vol.% H_2 was used as feeding gas, the cell performance was greatly enhanced, and a high current density of 0.51 A cm^{-2} was obtained under 1.3 applied voltage in 50 vol.% H_2O-50 vol.% H_2 (Fig. S4).

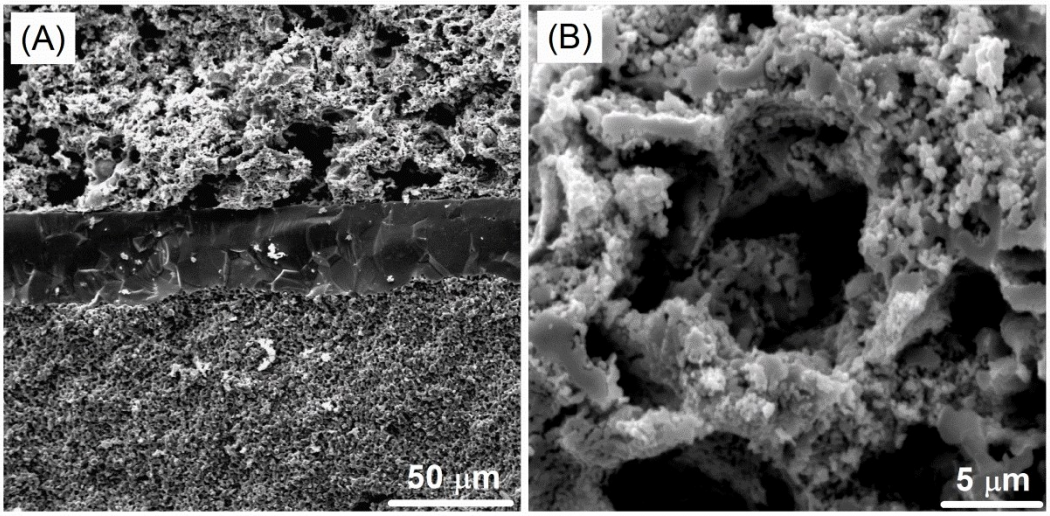

Fig. S3. (A) Cross-sectional SEM image of Ni-SDC-YSZ/YSZ/LSM-SDC-YSZ SOEC after electrolysis test, and (B) SEM images of LSM-SDC infiltrated LSM-SDC-YSZ oxygen electrode after electrolysis test.

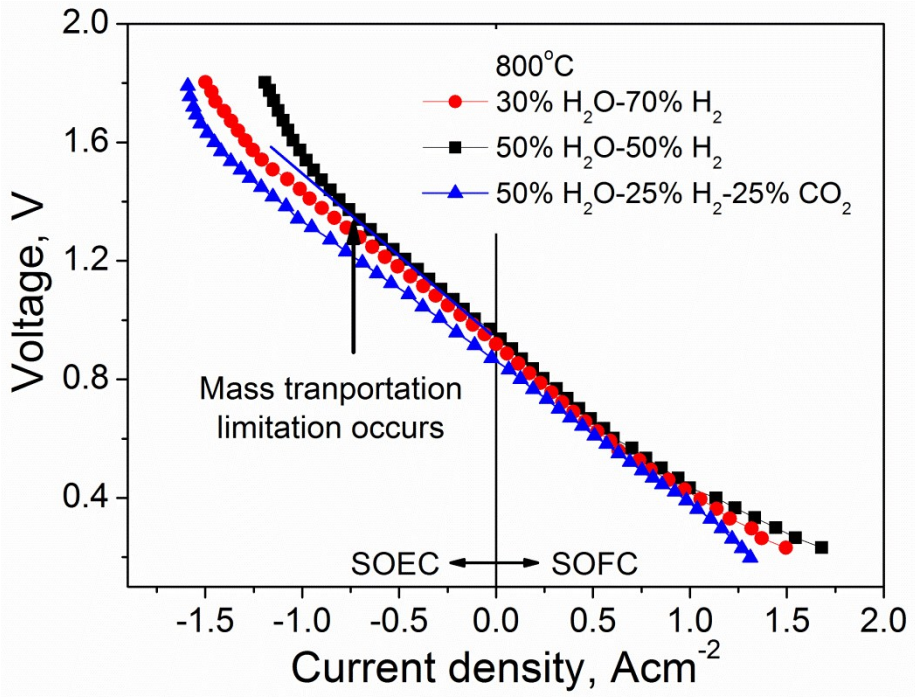

Fig. S4. DC potential-current density curves of Ni-SDC-YSZ/YSZ/LSM-SDC-YSZ single cells fabricated by dry-pressing and infiltration method operated in 50 vol.% H_2O-50 vol.% H_2 at 800°C.

3. Micro-tubular SOEC single cell fabricated by phase-inversion and infiltration method

The micro-tubular single cell with Ni-SDC-YSZ hydrogen electrode, YSZ electrolyte and LSM-SDC-YSZ oxygen electrode fabricated by phase-inversion, dip-coating and infiltration method, respectively, has also been studied for H_2O electrolysis and H_2O-CO_2 co-electrolysis (Fig. S5) [13]. Three different gas compositions were used as feeding gas, e.g. 30 vol.% H_2-70 vol.% H_2O, 50 vol.% H_2-50 vol.% H_2O, 25 vol.% H_2-50 vol.% H_2O-25 vol.% CO_2 (Fig. S6). When 50 vol.% H_2O-50 vol.% H_2 was used as feeding gas, the cell performance was greatly enhanced, and a high current density of 0.51 A cm^{-2} was obtained under 1.3 applied voltage in 50 vol.% H_2O-50 vol.% H_2 (Fig. S4).

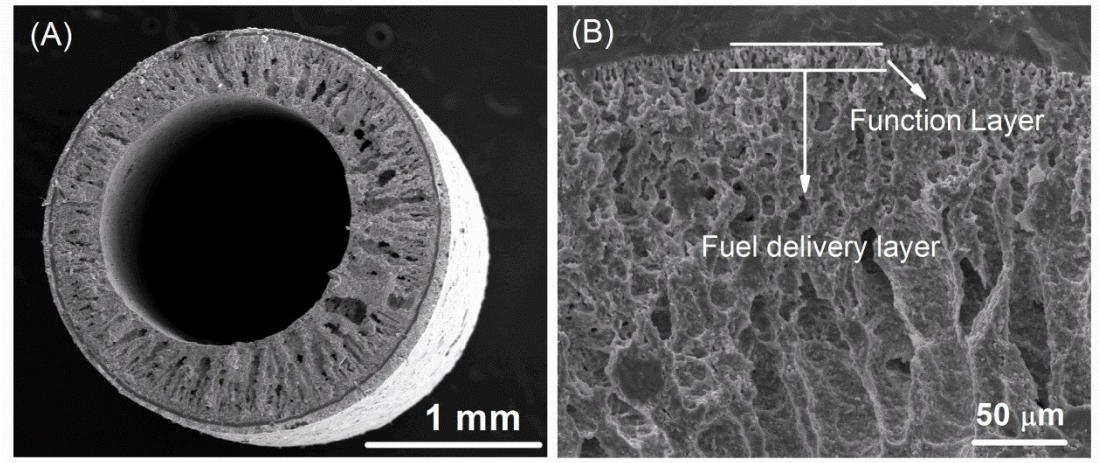

Fig. S5. (A) Overall view of the hydrogen electrode-supported Micro-tubular SOEC, (B) cross-sectional fracture surface micrograph of the hydrogen electrode-supported MT-SOC, indicating the microstructure of the hydrogen electrode fuel delivery layer and function layer.

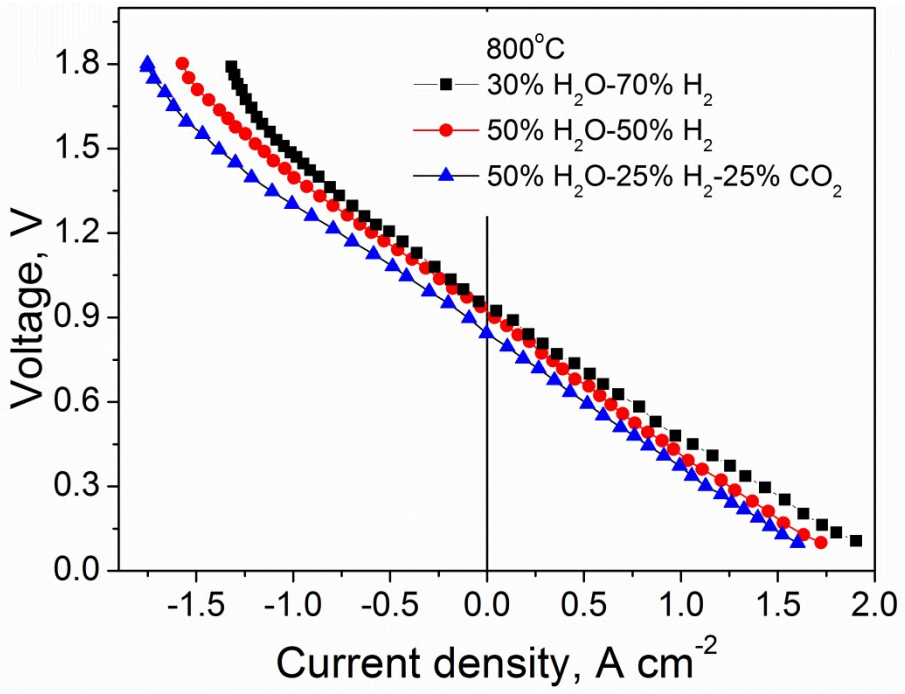

Fig. S6. DC potential-current density curves of Ni-SDC-YSZ/YSZ/LSM-SDC-YSZ single cells fabricated by phase-inversion and infiltration method operated in 50 vol.% H_2O-50 vol.% H_2 and 80 vol.% H_2O-20 vol.% H_2 at 900°C, respectively.

4. SOEC single cell fabricated by freeze-drying tape-casting and infiltration method

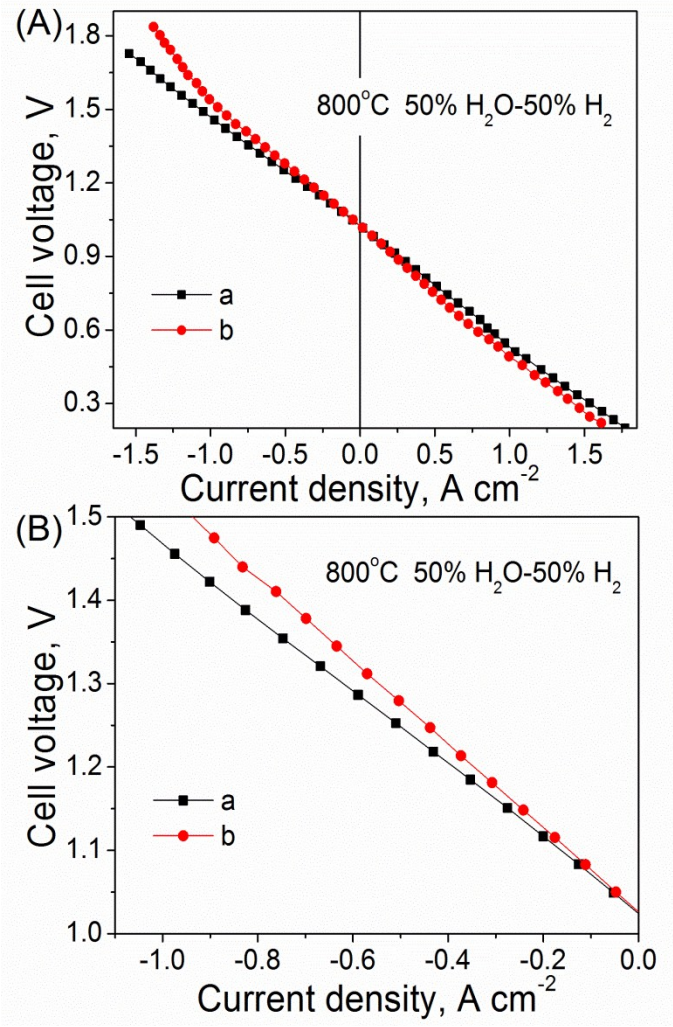

Fig. S7. DC potential-current density curves of Ni-SDC-YSZ/YSZ/LSM-SDC-YSZ single cells in fuel cell mode and co-electrolysis mode (A) or in co-electrolysis mode (from open circuit voltage (OCV) to 1.5 V) (B), fabricated by freeze-drying tape-casting and infiltration method (a) or traditional dry-pressing and infiltration method (b) in 50 vol.% H_2O-50 vol.% H_2 at 800°C, respectively.

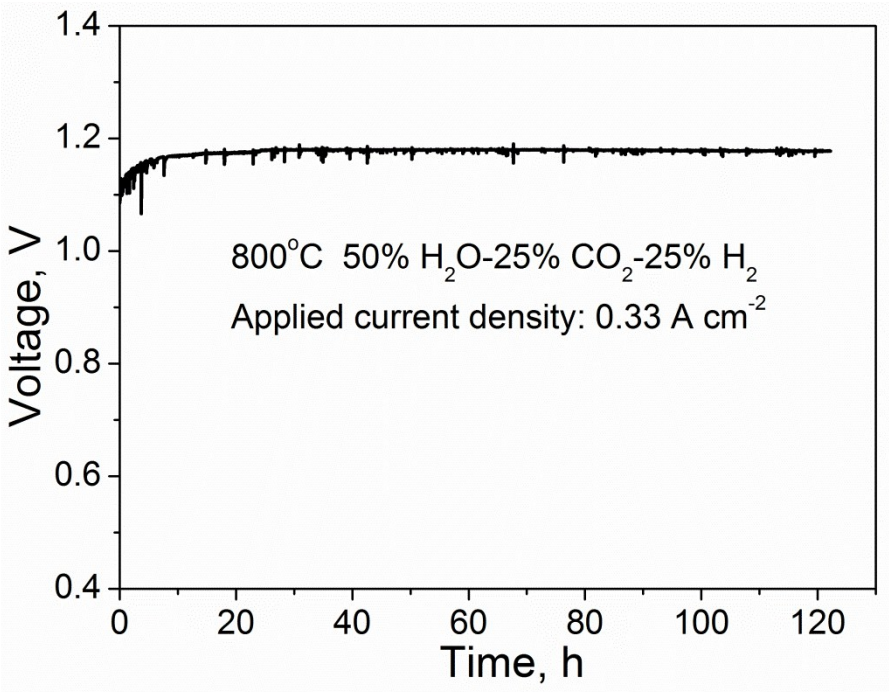

Fig. S8. A 0.33 A cm^{-2} constant current co-electrolysis testing of Ni-SDC-YSZ/YSZ/LSM-SDC-YSZ cell fabricated by Freeze-drying Tape-casting and infiltration method in 50 vol.% H_2O-25 vol.% CO_2-25 vol.% H_2 at 800°C.