Highly efficient electrolysis of pure CO_2 with symmetrical nanotructured perovskite electrodes

Yihang Li[a], Zhongliang Zhan[b], Changrong Xia[a*]

[a] CAS Key Laboratory of Materials for Energy Conversion, Department of Materials Science and Engineering & Collaborative Innovation Center of Suzhou Nano Science and Technology, University of Science and Technology of China, No. 96 Jinzhai Road, Hefei, Anhui Province, 230026, P. R. China.

*Tel: +86-551-63607475; Fax: +86-551-63601696; E-mail: xiacr@ustc.edu.cn

[b] Shanghai Institute of Ceramics, Chinese Academy of Sciences, 1295 Dingxi Road, Shanghai 200050, P. R.China

Experimental

Cell fabrication

The tri-layer LSGM (Fuel Cell Materials Company) ceramics, where a dense LSGM was sandwiched between two porous LSGM, were fabricated by laminating three tape-casted ceramic tapes with 40 *wt.*% starch as pore former，followed by sintering at 1450 ^{o}C for 5 h. SFM nanoparticles (20 wt.% relative to the LSGM scaffold) were then deposited into the porous LSGM layers by infiltration process under vacuum circumstance using SFM aqueous solution containing $Sr(NO_3)_2$, $Fe(NO_3)_3$•$9H_2O$, $(NH_4)_6Mo_7O_{24}$•$4H_2O$, citric acid and glycine. The details of infiltration process has been described in our recent work.[1] Finally, the cell was heated at 850 ^{o}C for 5 h to obtain crystallized SFM. The Au paste (Sino-Platinum Metals Co., Ltd.) was painted onto the electrode surfaces and heated at 600 ^{o}C for 1 h to ensure current collection. All the chemicals were from Sinopharm Chemical Reagent Co., Ltd.

Cell characterization

The phase structure of the infiltrated material was analyzed using X-ray diffration (XRD, Cu Kα radiation, λ=1.5418 Å) with the scanning rate of 10° per minute. The microstructures and morphologies were revealed using a scanning electron microscope (SEM, JEOL JSM-6700 F) and a high-resolution transmission electron microscope (HRTEM, JEOL JSM-2010). The single cell was sealed to an alumina ceramic tube with glass paste in a vertical furnace. The fuel electrode was feed with CO_2 (99.999%, Nanjing special gas Factory Co., Ltd.) at a rate of 40 mL min^{-1} using a mass flow controller (D08-2F, Qixing Huachuang Co., Ltd.) while the oxygen electrode was exposed to ambient air. The current-voltage characteristics were measured from 0 to 1.6 V with a scanning rate of 10 mV s^{-1} (Solartron 1287). The AC impedance measurements were carried out with a frequency response analyzer (Solartron 1260), typically from 10^6 Hz to 10^{-2} Hz with an amplitude of 10 mV.

The outlet CO_2 and CO contents were determined using online gas chromatography (FULI, GC9790II, China) with thermal conductivity detector. The post-test fuel electrode was analyzed using Raman spectroscopy (Raman, Lab RAM HR800, 1000-2000 cm^{-1}).

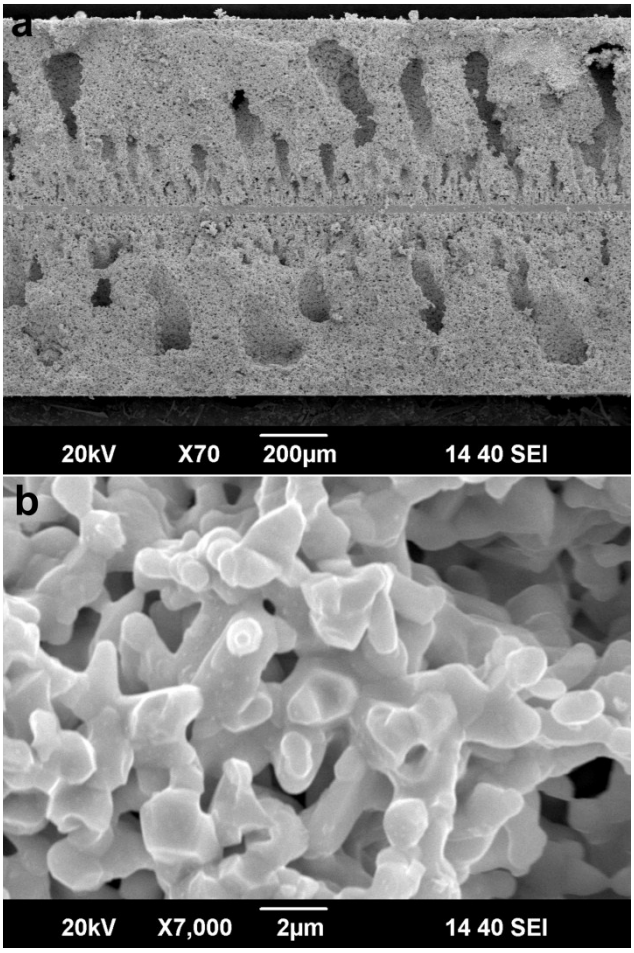

Figure S1 (a) Cross-section SEM image of LSGM mainframe consisting of a dense thin electrolyte and two porous scaffolds; (b) High-resolution SEM image of the porous part showing all the LSGM particles are connected to form a conduction network.

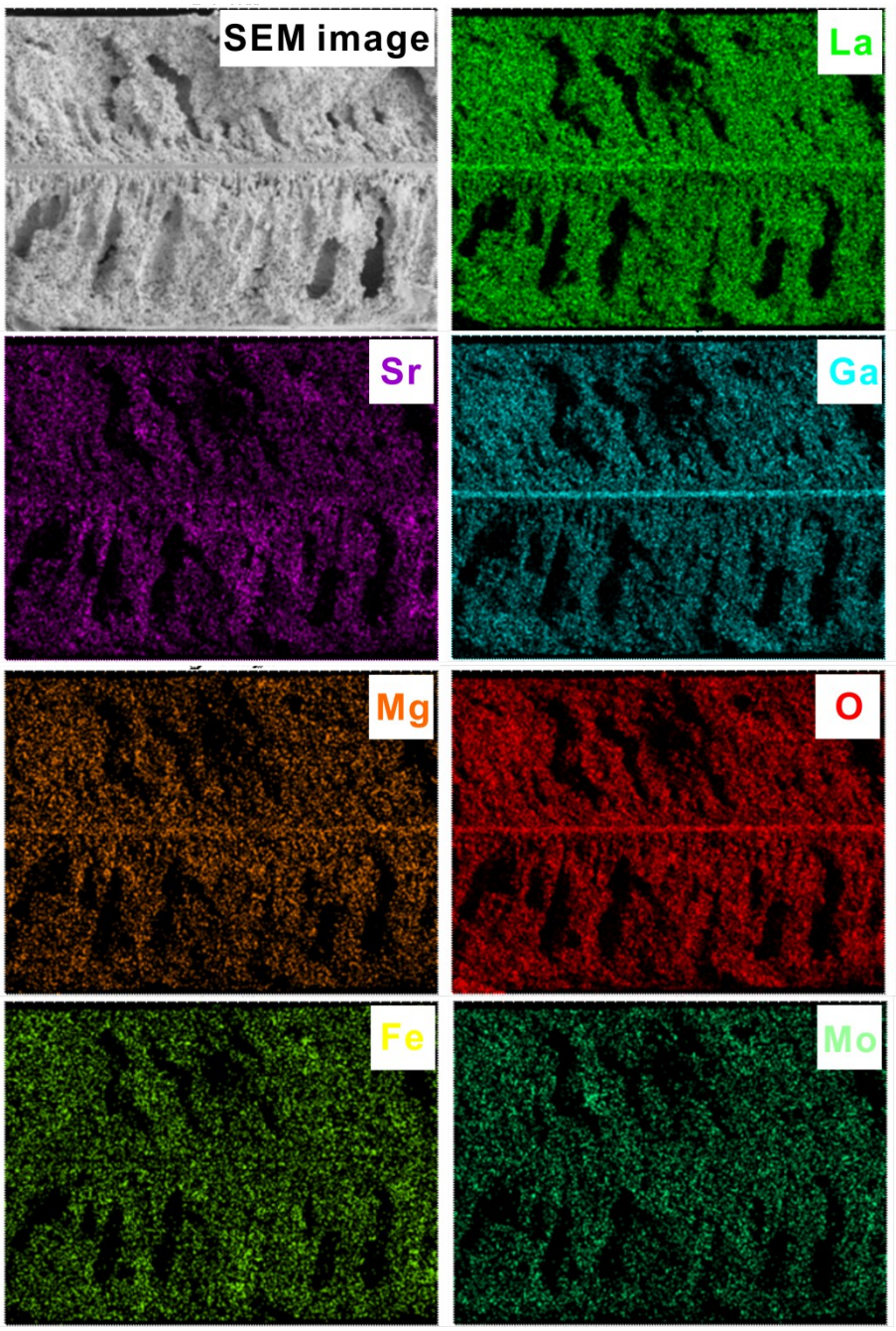

Figure S2 The EDS elemental mapping of LSGM and SFM, showing the distribution of SFM phase cross the thickness of the cell.

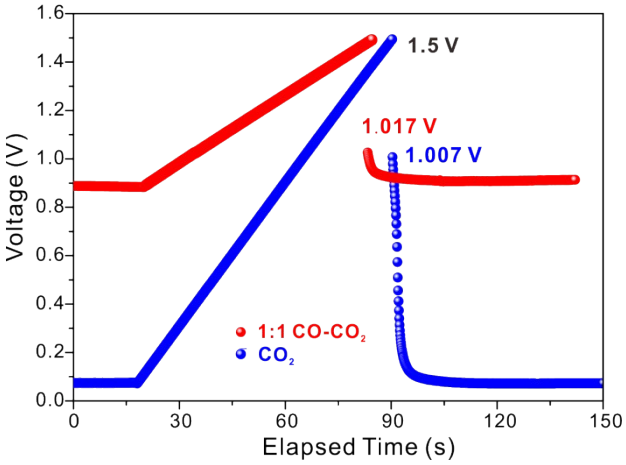

Figure S3 The records of cell voltage when pure CO_2 or 1:1 CO-CO_2 was fed to the fuel electrode

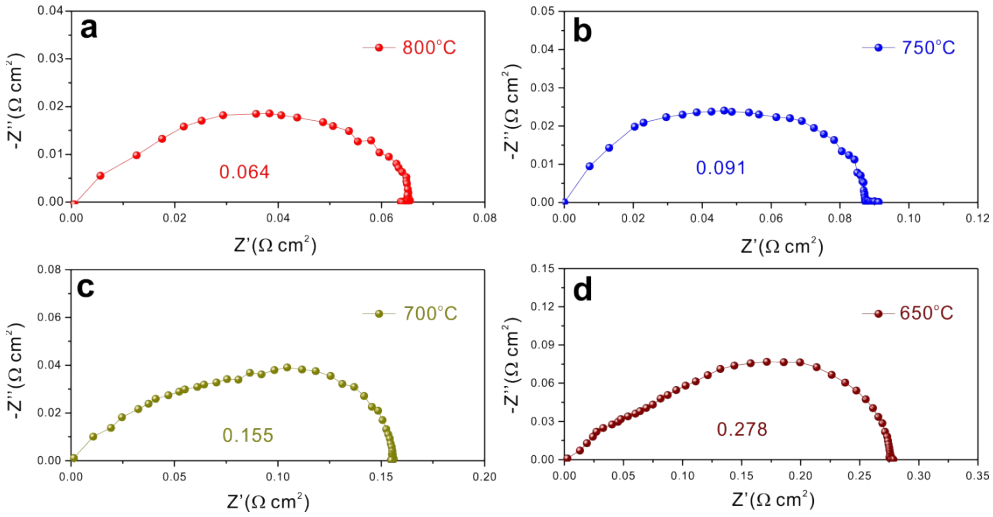

Figure S4 Area specific resistances of infiltrated SFM-LSGM electrode through as-prepared symmetrical cell in ambient air at 650-800°C

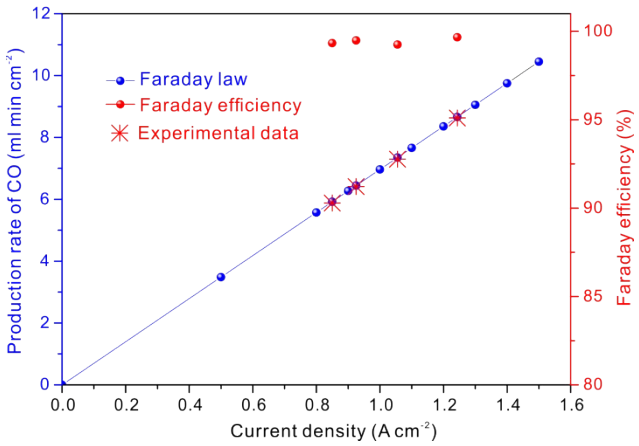

Figure S5 The production rate of CO and corresponding Faraday efficiency under a series of applied voltages from 1.2 to 1.5 V

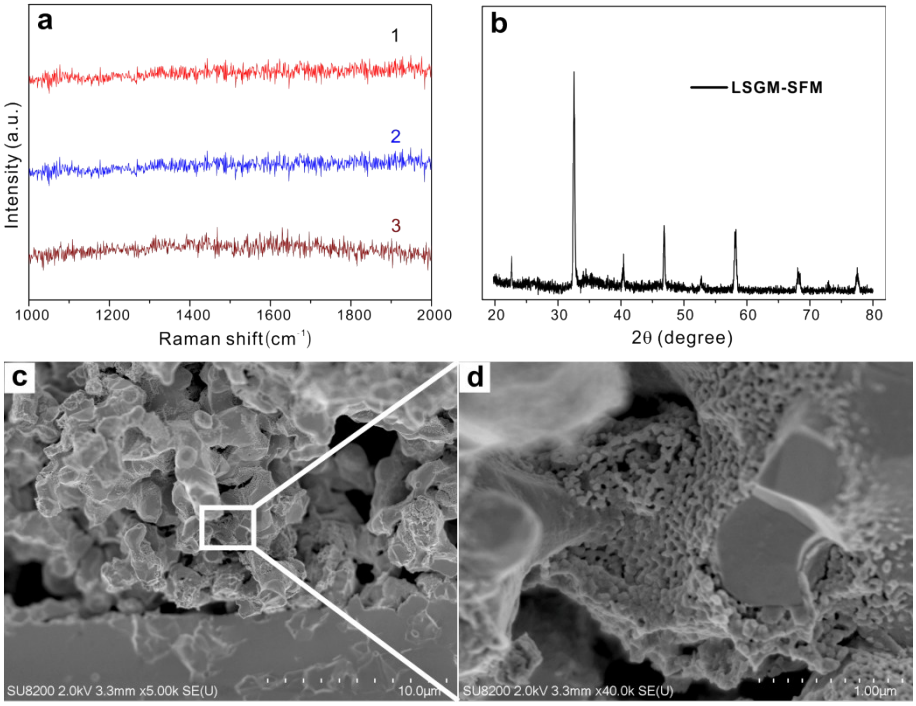

Figure S6 Raman spectrums (a), XRD pattern (b) and SEM microstructures (c and d) for the fuel electrode after 53-hour durability test.

Table S1 The fitting results of EIS data under Voc conditions

Conditions	Ro (Ω cm^2)	Rp (Ω cm^2)	R_1 (Ω cm^2)	R_2 (Ω cm^2)	R_3 (Ω cm^2)	R_4 (Ω cm^2)
650	0.327	1.222	0.058	0.084	0.955	0.125
700	0. 230	0.648	0.055	0.075	0.518	-
750	0.186	0.413	0.044	0.043	0.326	-
800	0.151	0.226	0.036	0.020	0.170	-

Table S2 The fitting results of EIS data under the bias conditions

Conditions	Ro (Ω cm^2)	Rp (Ω cm^2)	R_1 (Ω cm^2)	R_1' (Ω cm^2)	R_2 (Ω cm^2)	R_3 (Ω cm^2)
1.2 V	0.150	0.185	0.018	0.006	0.020	0.141
1.3 V	0.149	0.142	0.010	0.005	0.017	0.110
1.4 V	0.149	0.114	0.008	0.003	0.015	0.090
1.5 V	0.149	0.103	0.007	0.002	0.013	0.081

References

1 Y. Li, P. Li, B. Hu and C. Xia, *J. Mater. Chem. A*, 2016, **4**, 9236.