

**In-situ Construction of Hetero-structured Perovskite Composites with  
Exsolved Fe and Cu Metallic Nanoparticles as Efficient CO<sub>2</sub>  
Reduction Electrocatalyst for High Performance Solid Oxide  
Electrolysis Cells**

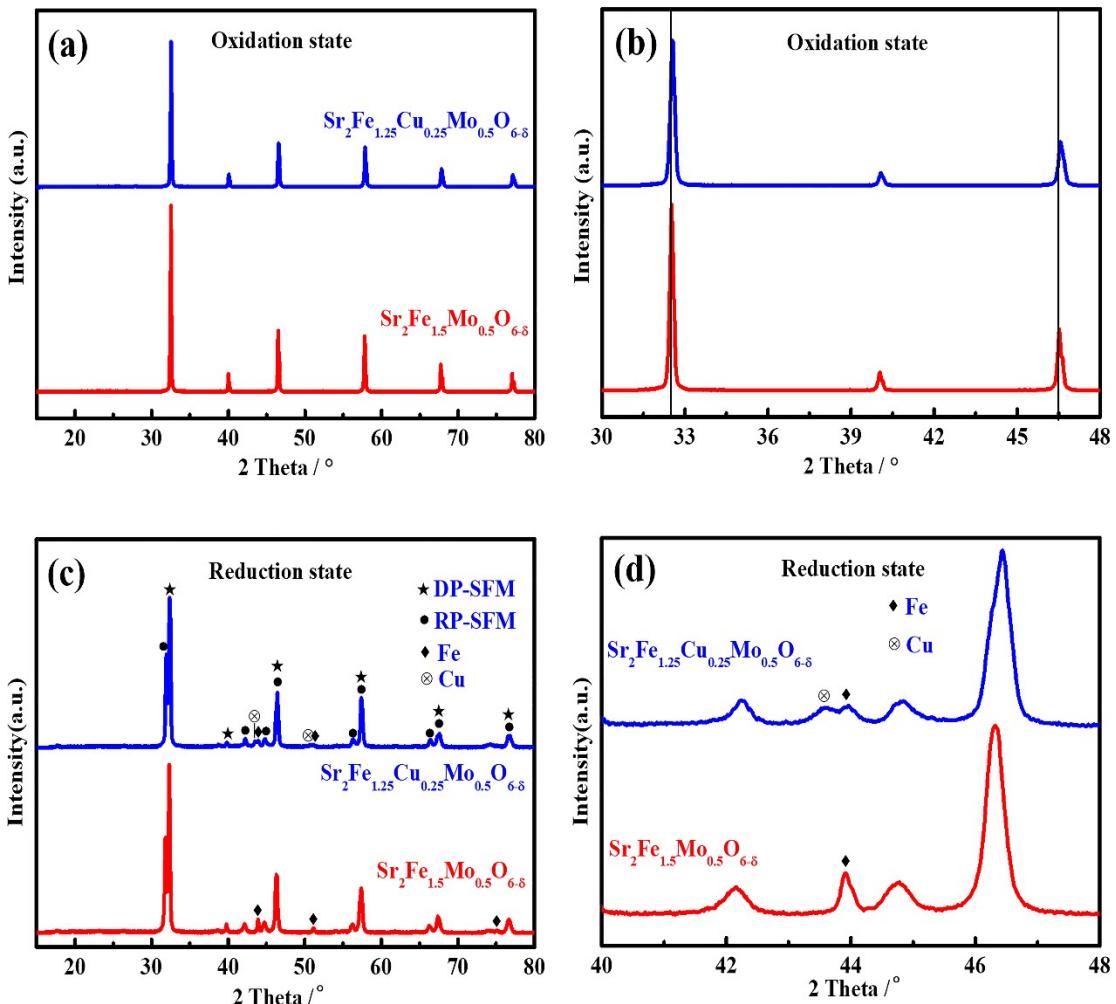
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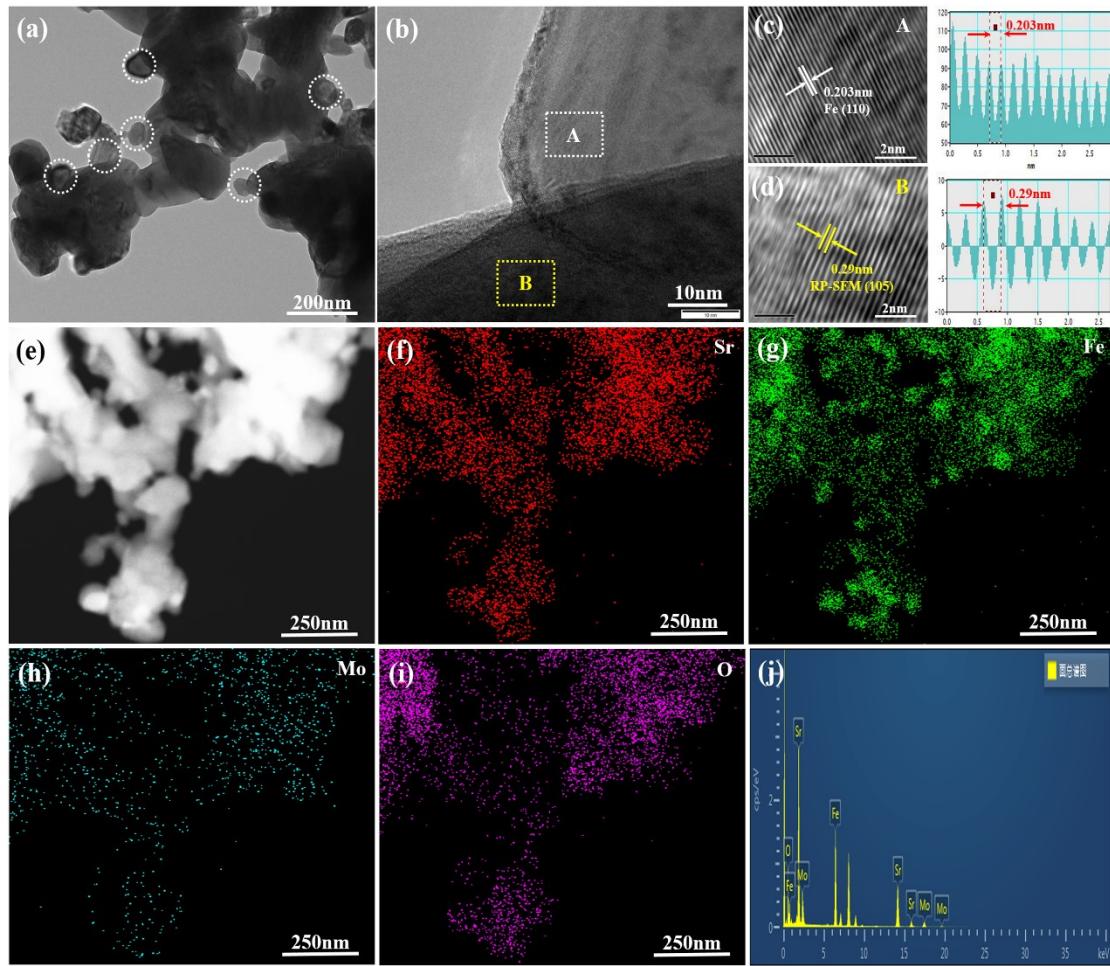
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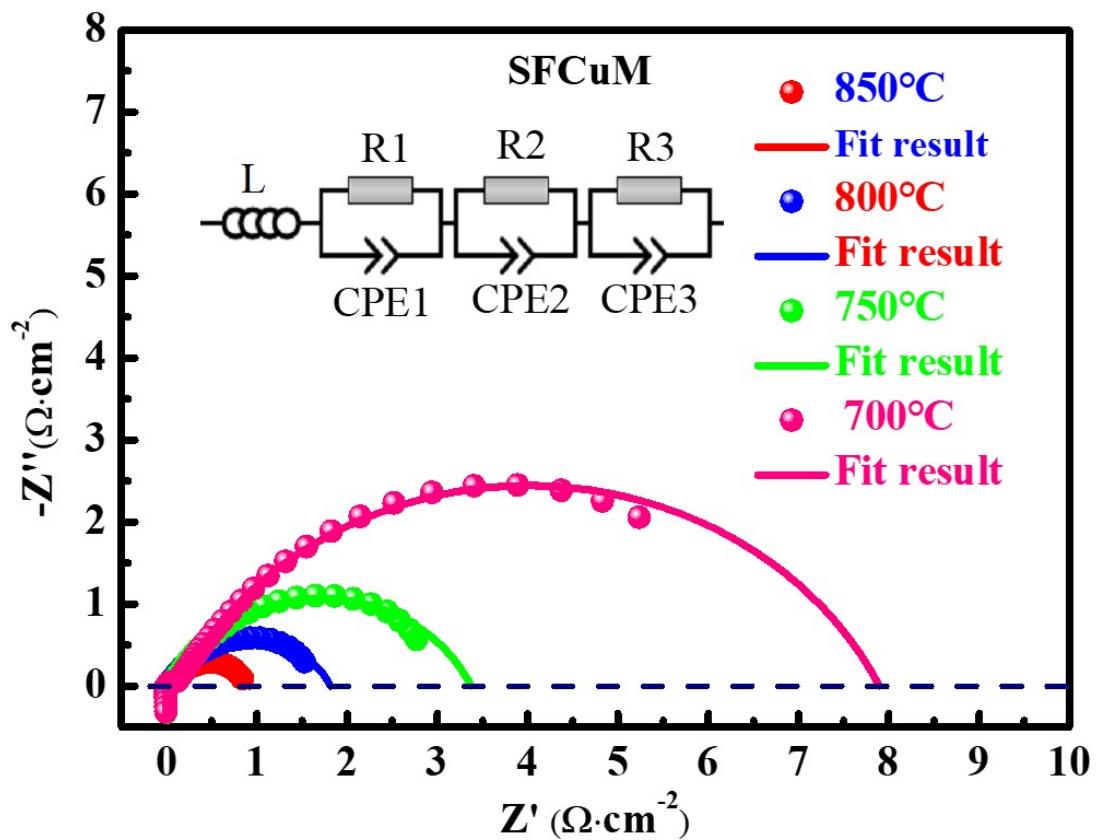
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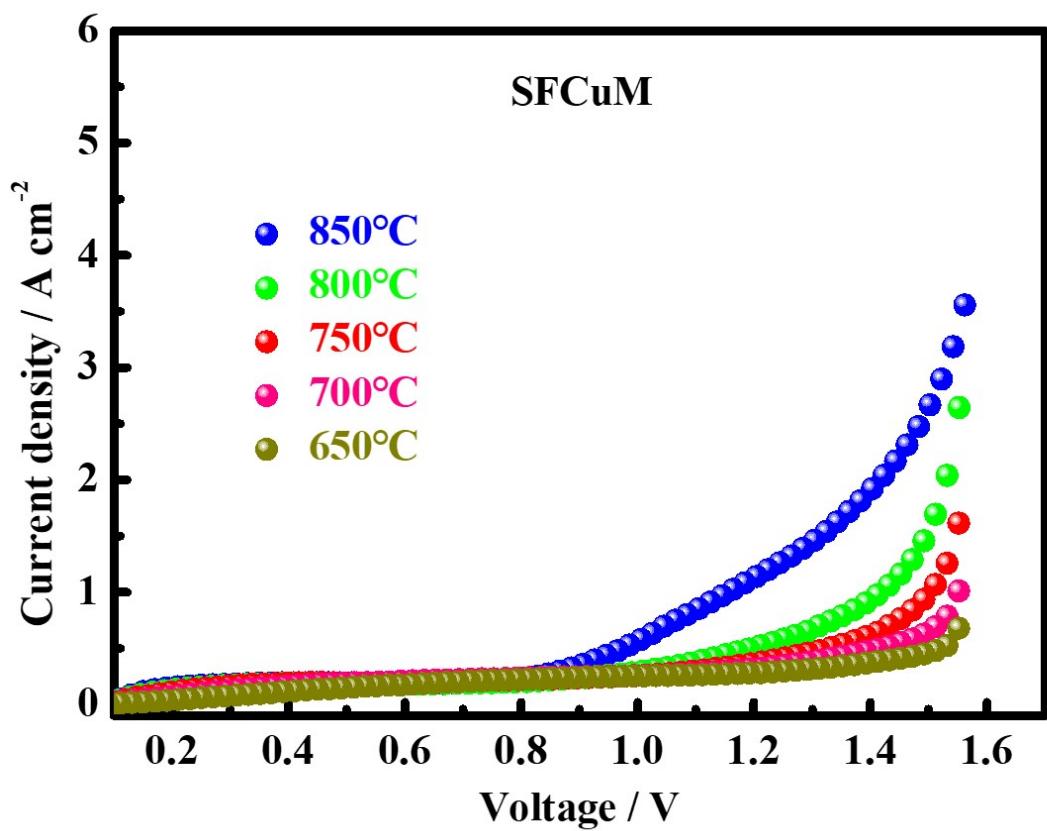
**Fig. S1.** (a) As-synthesized  $\text{Sr}_2\text{Fe}_{1.5}\text{Mo}_{0.5}\text{O}_{6-\sigma}$  (SFM) and  $\text{Sr}_2\text{Fe}_{1.25}\text{Cu}_{0.25}\text{Mo}_{0.5}\text{O}_{6-\sigma}$  (SFCuM) powders; (b) magnification of (a); (c) reduced  $\text{Sr}_2\text{Fe}_{1.5}\text{Mo}_{0.5}\text{O}_{6-\sigma}$  (SFM) and  $\text{Sr}_2\text{Fe}_{1.25}\text{Cu}_{0.25}\text{Mo}_{0.5}\text{O}_{6-\sigma}$  (SFCuM) powders; (d) magnification of (c).



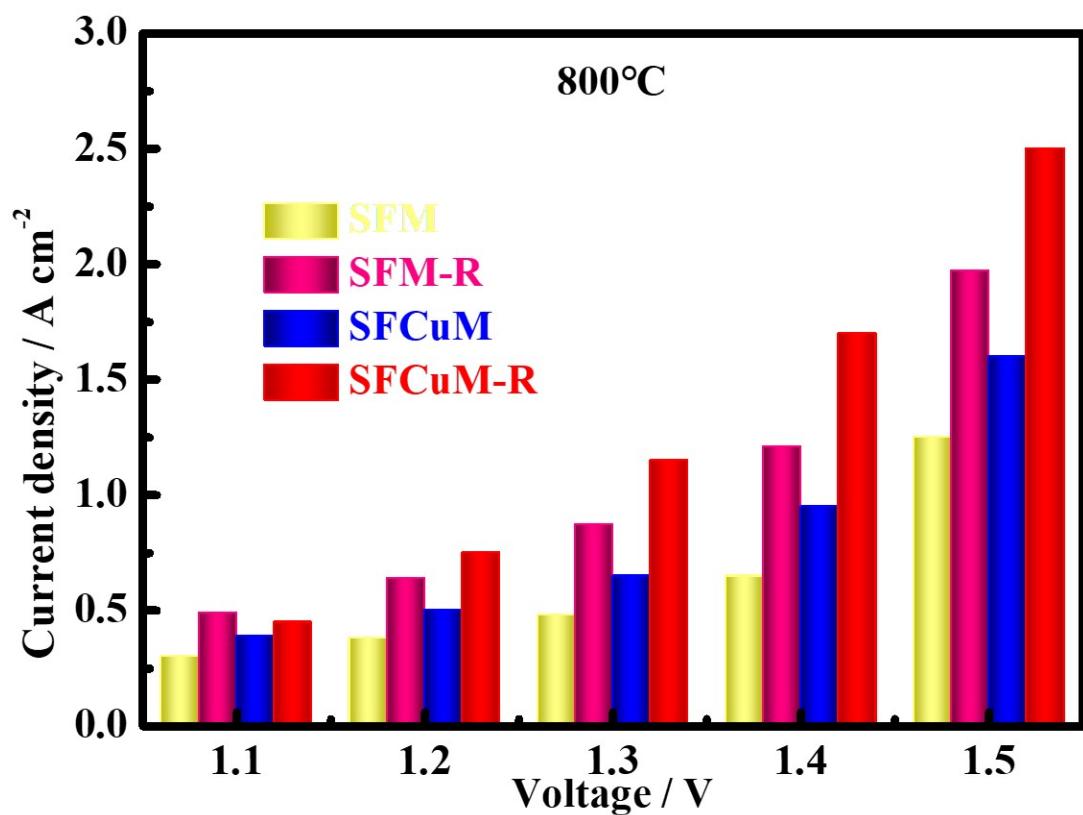
**Fig. S2.** (a-d) HR-TEM micrographs of the reduced  $\text{Sr}_2\text{Fe}_{1.5}\text{Mo}_{0.5}\text{O}_{6-\sigma}$  (SFM) particles; (e-i) TEM-EDS elemental mappings of the reduced SFM particles, (j) amount of each element in SFM.



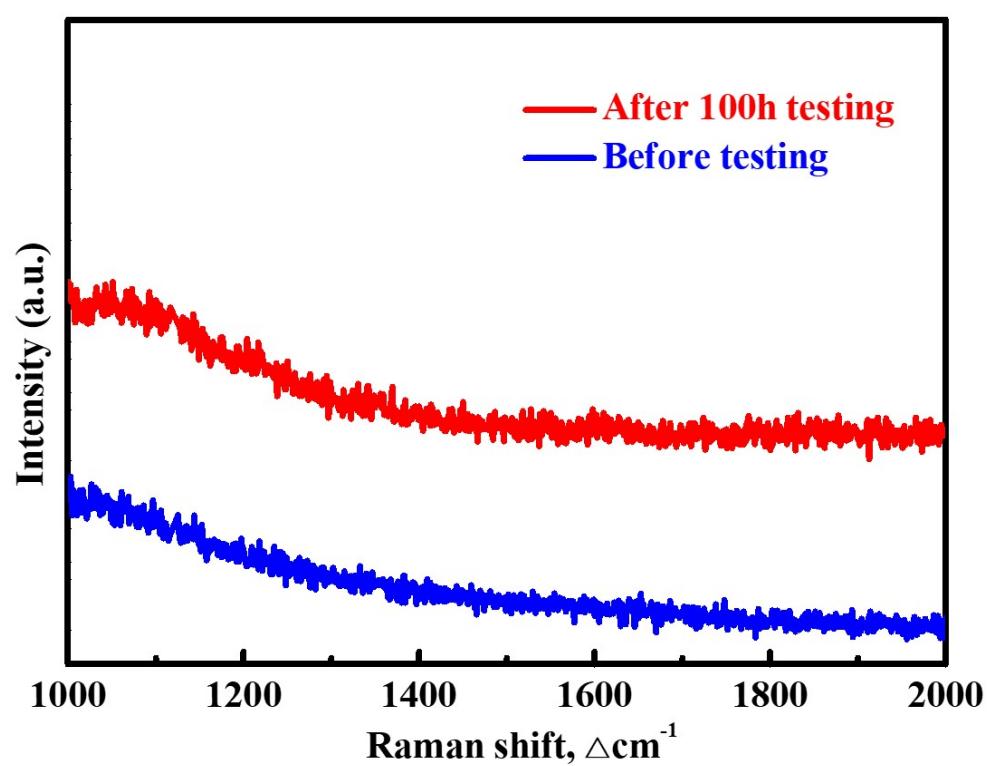
**Fig. S3.** EIS of LSGM electrolyte-supported symmetrical cells with the configuration SFCuM/LDC/LSGM/LDC/SFCuM under a 1:1 CO–CO<sub>2</sub> atmosphere at different operating temperatures.



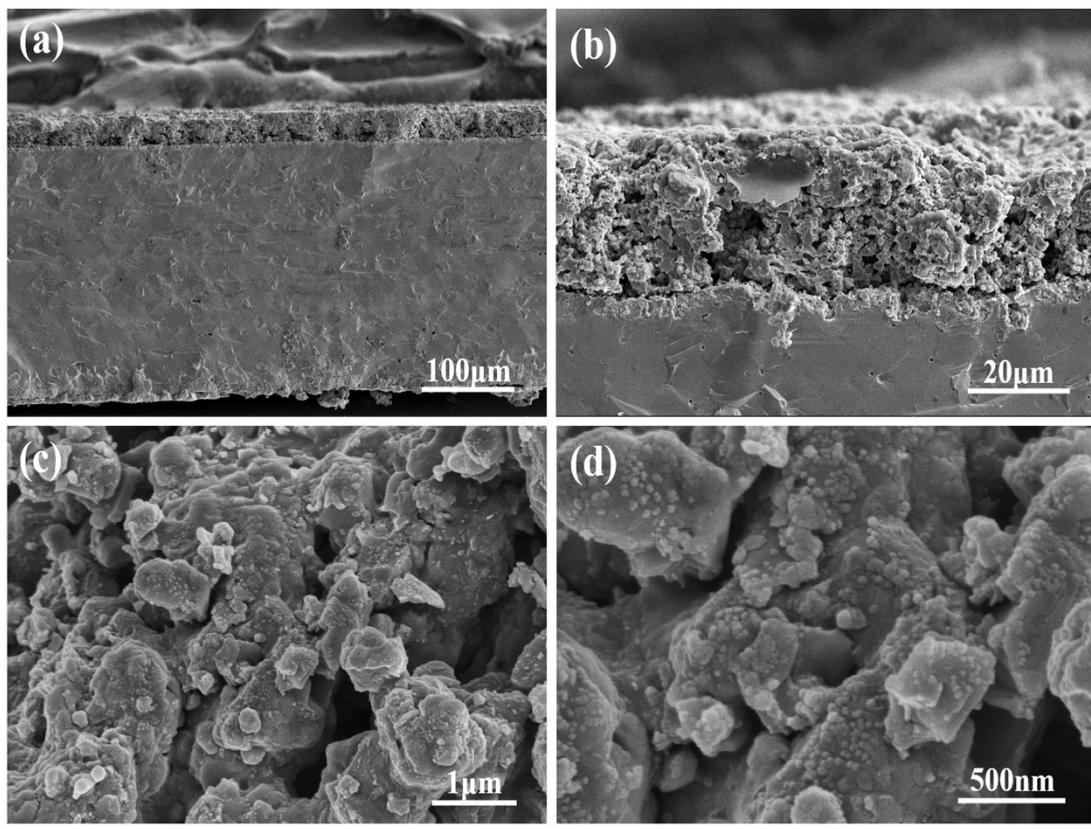
**Figure S4.** Current density of LSGM electrolyte-supported single cells with the configuration SFCuM/LDC/LSGM/LSCF-SDC under pure CO<sub>2</sub> atmosphere at different operating temperatures.



**Figure S5.** Current density comparison of the LSGM-electrolyte supported single cell with various cathodes at the operating temperature of 800°C.



**Figure S6.** Raman spectroscopy of the SFCuM-R cathode before and after long-term stability testing.



**Figure S7.** Cross section images of LSGM electrolyte supported single cell with SFCuM-R as the cathode and LSCF-SDC as the anode after 100h long-term stability testing.

**Table S1** Superficial C and O stoichiometry of the  $\text{Sr}_2\text{Fe}_{1.25}\text{Cu}_{0.25}\text{Mo}_{0.5}\text{O}_{6-\delta}$  electrode  
based on XPS peak fitting results.

Superficial elements / Atomic %	$\text{CO}_3^{2-}/\text{C-C}$	$\text{O}_{\text{ad}}/\text{CO}_3^{2-}$	$\text{O}_{\text{ad}}/\text{O}_{\text{latt}}$
Oxidizing	0.56	3.63	2.40
Reducing	0.58	3.75	4.54

**Table S2** Superficial elements stoichiometry of the  $\text{Sr}_2\text{Fe}_{1.25}\text{Cu}_{0.25}\text{Mo}_{0.5}\text{O}_{6-\delta}$  electrode

based on XPS results

Superficial elements /Atomic %	Sr	Fe	Mo	Cu	O	C	Sr/(Fe+Mo+Cu)	$\text{CO}_3^{2-}$ /Sr
Oxidizing 4	11.5	2.25	2.81	2.44	47.99	32.97	1.54	0.76
Reducing 8	11.0	1.88	1.82	2.03	47.31	35.87	1.93	0.84

**Table S3.** Current density comparison for CO<sub>2</sub> electrolysis obtained at 1.5 V and 800 °C with diverse cathodes.

Fuel electrodes	Electrolyte	Anode	Feeding gas	Current density	Refs.
La <sub>0.6</sub> Sr <sub>0.4</sub> Fe <sub>0.8</sub> Ni <sub>0.2</sub> O <sub>3-δ</sub>	YSZ	LSCF-SDC	CO <sub>2</sub> -30% CO	0.75	<sup>1</sup>
Ce-La <sub>0.7</sub> Sr <sub>0.3</sub> Cr <sub>0.5</sub> Fe <sub>0.5</sub> O <sub>3-δ</sub>	YSZ	LSCF	CO <sub>2</sub> -30% CO	0.9	<sup>2</sup>
Sr <sub>2</sub> Fe <sub>1.5</sub> Mo <sub>0.5</sub> O <sub>6-δ</sub> F <sub>0.1</sub>	LSGM	LSGM-SDC	100% CO <sub>2</sub>	1.36	<sup>3</sup>
NiFe@La <sub>0.6</sub> Sr <sub>0.4</sub> Fe <sub>0.8</sub> Mn <sub>0.2</sub> O <sub>3</sub>	LSGM	BLC	CO <sub>2</sub> -1% CO	1.70	<sup>4</sup>
CoFe@(Pr <sub>0.4</sub> Sr <sub>0.6</sub> ) <sub>3</sub> (Fe <sub>0.85</sub> Mo <sub>0.15</sub> ) <sub>2</sub> O <sub>7</sub>	YSZ	LSCF-SDC	CO <sub>2</sub> -30% CO	1.01	<sup>5</sup>
Sr <sub>2</sub> Fe <sub>1.4</sub> Mn <sub>0.1</sub> Mo <sub>0.5</sub> O <sub>6-δ</sub> -SDC	LSGM	LSCF-SDC	100% CO <sub>2</sub>	1.35	<sup>6</sup>
Co@La <sub>1.2</sub> Sr <sub>0.8</sub> Co <sub>0.4</sub> Mn <sub>0.6</sub> O <sub>4</sub> -GDC	LSGM	LSCF-GDC	CO <sub>2</sub> -30% CO	0.75	<sup>7</sup>
FeNi <sub>3</sub> @Sr <sub>2</sub> Fe <sub>1.5</sub> Mo <sub>0.5</sub> O <sub>6-δ</sub>	LSGM	LSM-GDC	CO <sub>2</sub> -5% N <sub>2</sub>	0.90	<sup>8</sup>
Sr <sub>2</sub> Fe <sub>1.25</sub> Cu <sub>0.25</sub> Mo <sub>0.5</sub> O <sub>6-δ</sub>	LSGM	LSCF-SDC	100% CO <sub>2</sub>	1.60	This work
Sr <sub>2</sub> Fe <sub>1.25</sub> Cu <sub>0.25</sub> Mo <sub>0.5</sub> O <sub>6-δ</sub> -R	LSGM	LSCF-SDC	100% CO <sub>2</sub>	2.5	This work

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