

## Supplementary Materials:

Redox performances and optimizations of chemical composition of lanthanum-strontium-manganese-based perovskite oxide for two-step thermochemical CO<sub>2</sub> splitting

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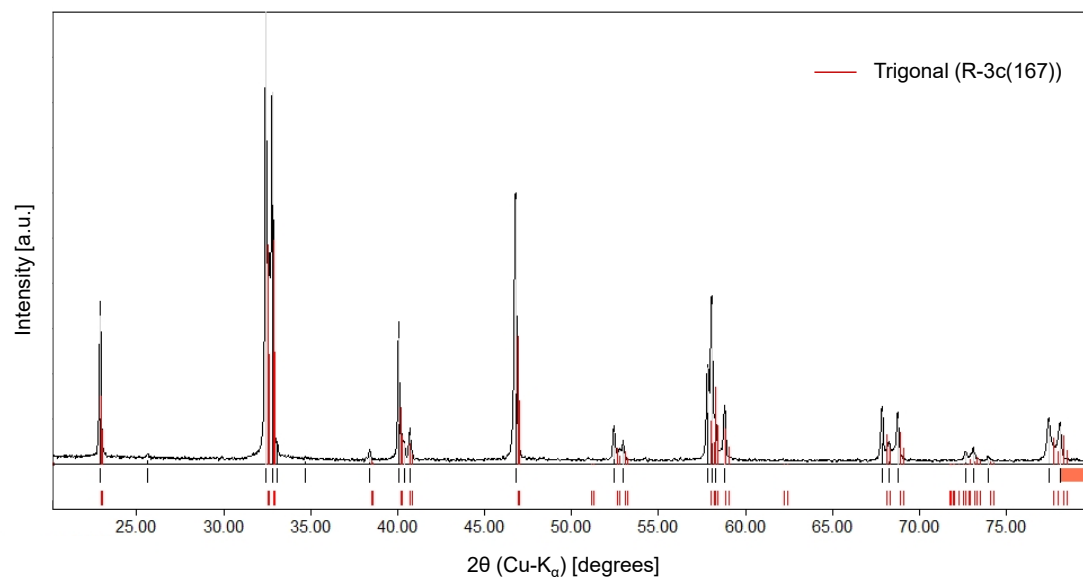


Figure S1. Powder XRD pattern of the as-prepared sample ( $x = 0$ ,  $\text{La}_{1-x}\text{Sr}_x\text{Mn}_{0.8}\text{Ni}_{0.2}\text{O}_3$ ).

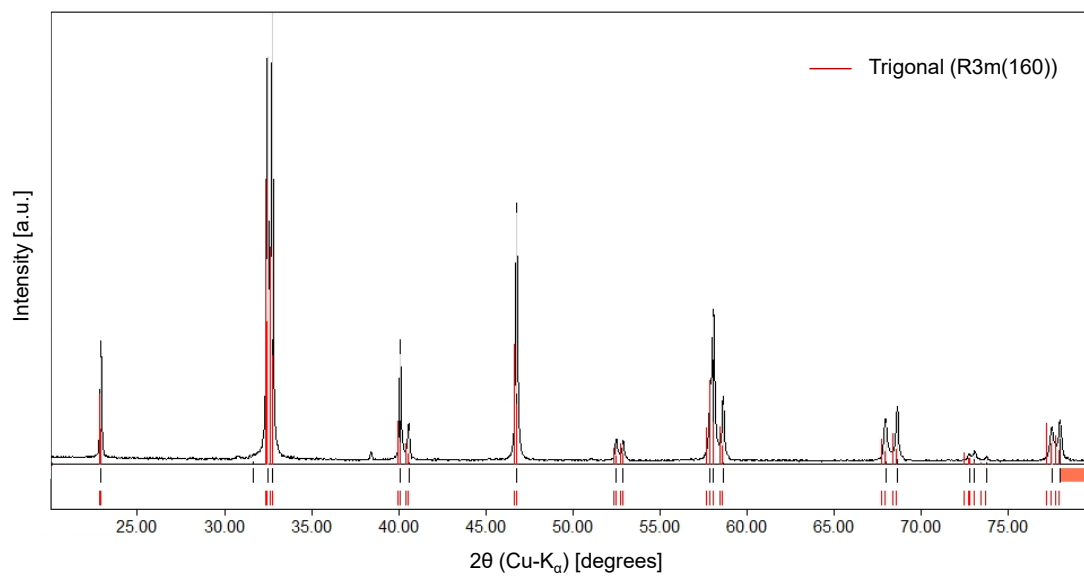


Figure S2. Powder XRD pattern of the as-prepared sample ( $x = 0.2$ ,  $\text{La}_{1-x}\text{Sr}_x\text{Mn}_{0.8}\text{Ni}_{0.2}\text{O}_3$ ).

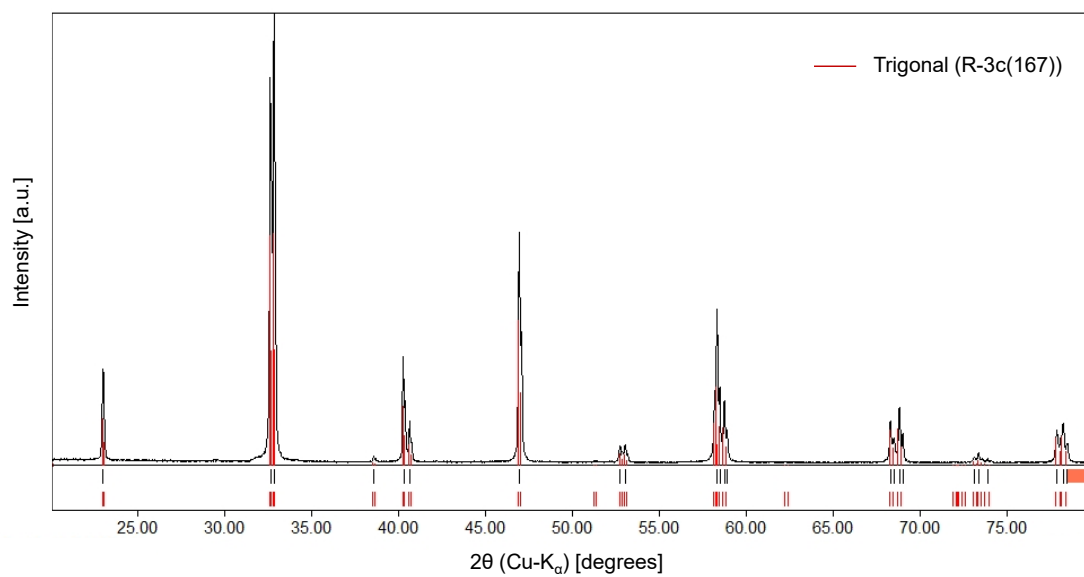


Figure S3. Powder XRD pattern of the as-prepared sample ( $x = 0.3$ ,  $\text{La}_{1-x}\text{Sr}_x\text{Mn}_{0.8}\text{Ni}_{0.2}\text{O}_3$ ).

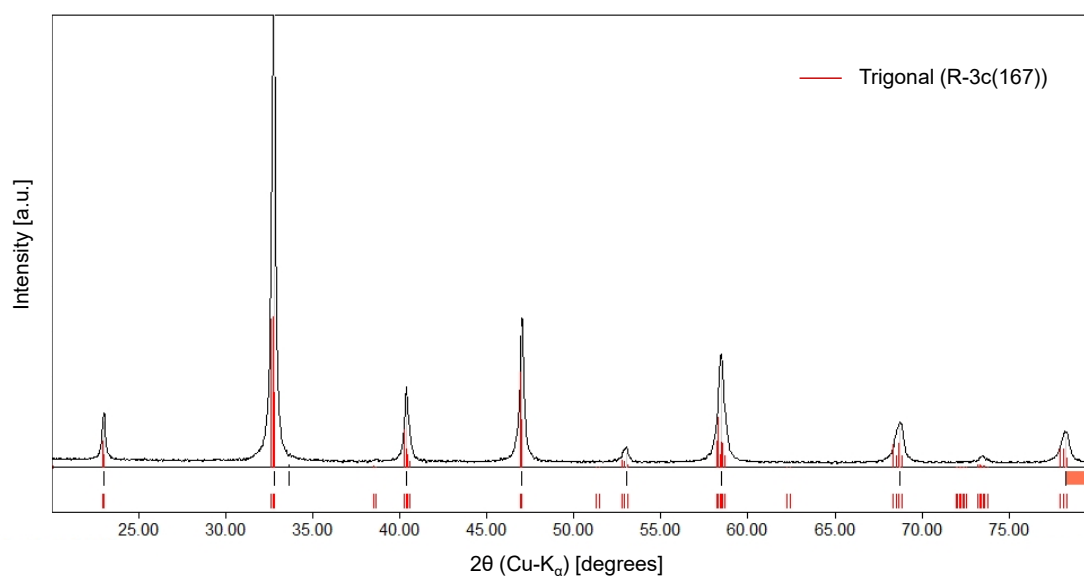


Figure S4. Powder XRD pattern of the as-prepared sample ( $x = 0.4$ ,  $\text{La}_{1-x}\text{Sr}_x\text{Mn}_{0.8}\text{Ni}_{0.2}\text{O}_3$ ).

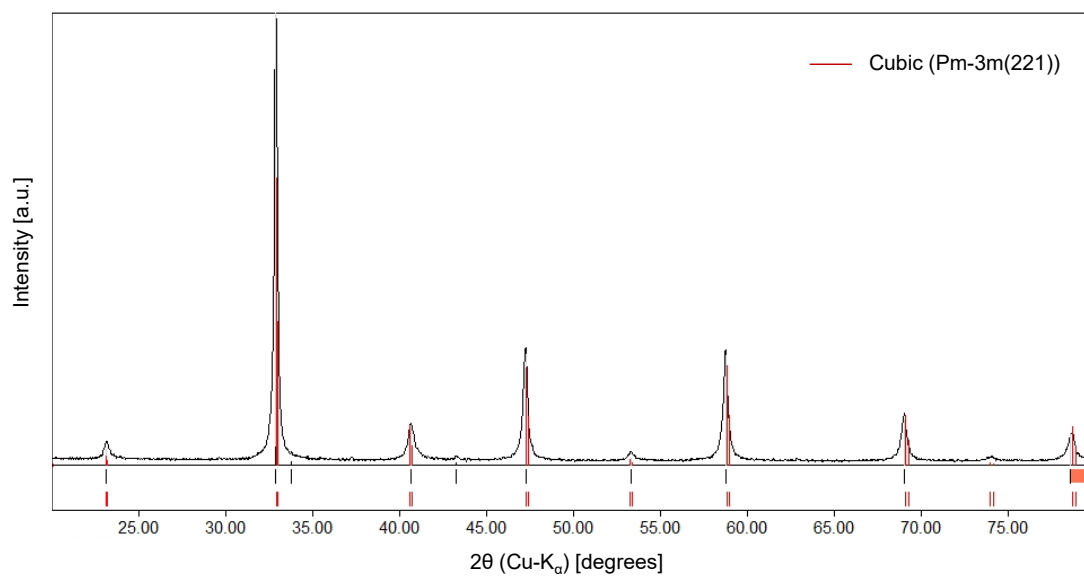


Figure S5. Powder XRD pattern of the as-prepared sample ( $x = 0.6$ ,  $\text{La}_{1-x}\text{Sr}_x\text{Mn}_{0.8}\text{Ni}_{0.2}\text{O}_3$ ).

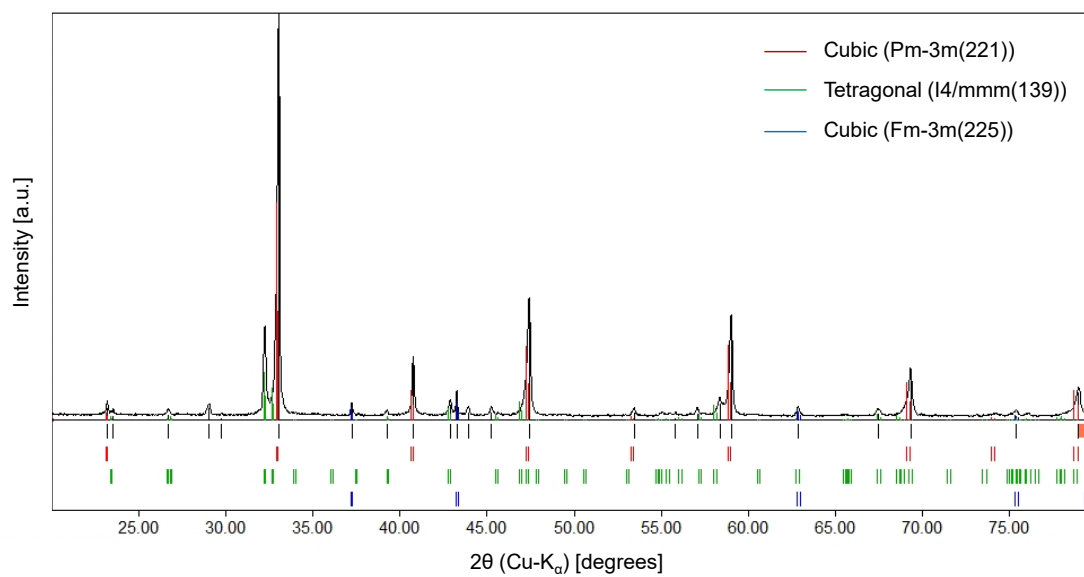


Figure S6. Powder XRD pattern of the as-prepared sample ( $x = 0.8$ ,  $\text{La}_{1-x}\text{Sr}_x\text{Mn}_{0.8}\text{Ni}_{0.2}\text{O}_3$ ).

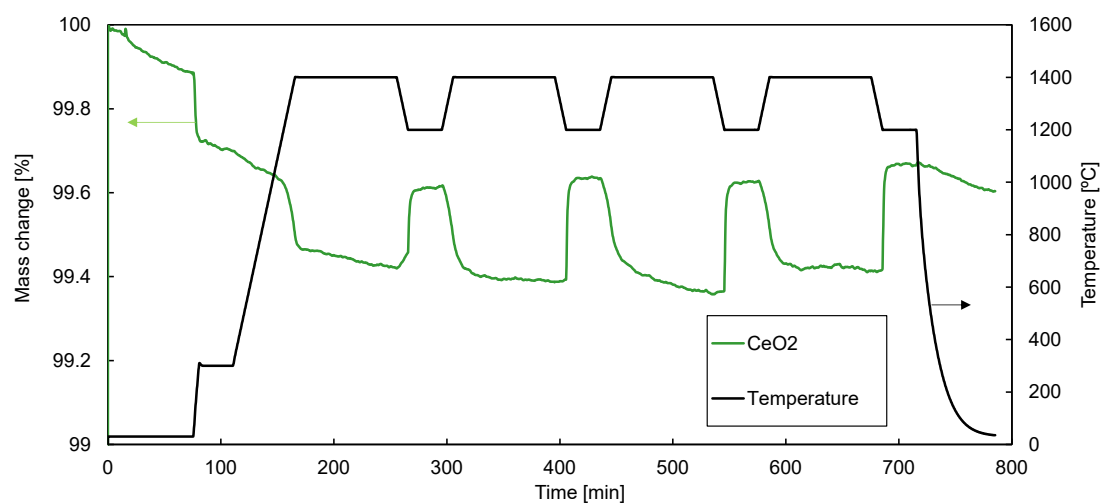


Figure S7. Thermochemical cycling test of the ceria sample ( $\text{CeO}_2$ ).



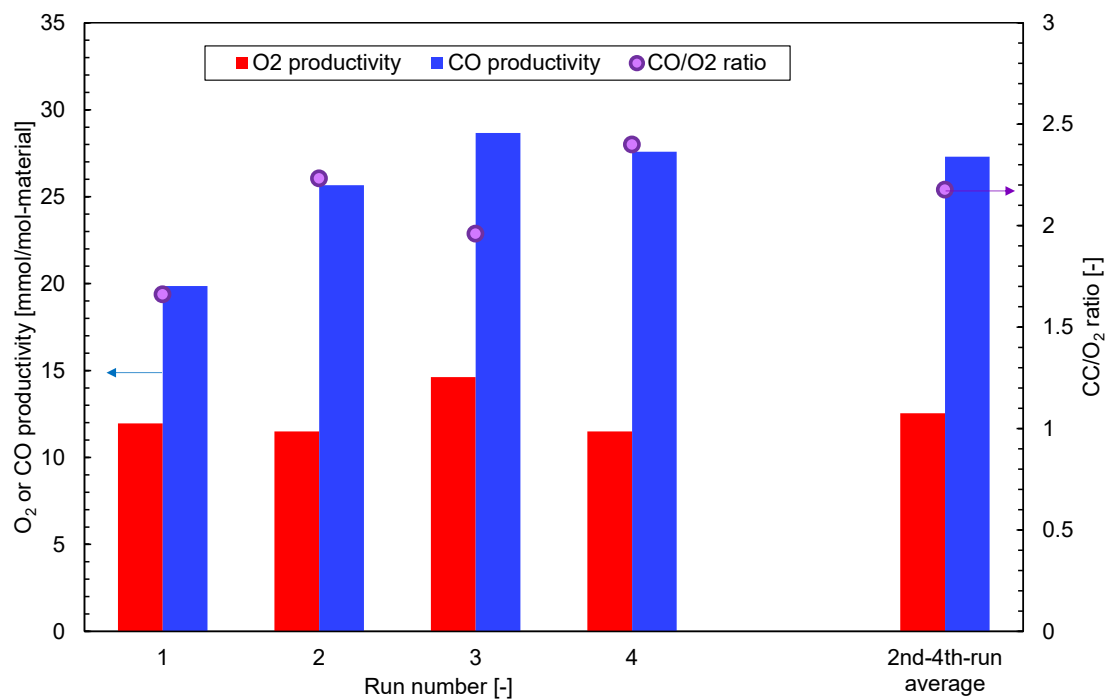


Figure S8. O<sub>2</sub> and CO productivities, and CO/O<sub>2</sub> ratio for thermochemical cycling using the ceria sample (CeO<sub>2</sub>).

Table S1. Comparison of O<sub>2</sub> productivity, average CO productivity, cycle-averaged CO production rate, and reaction conditions of LSMCo0.35, LSMNi0.20, LSMMg0.125, CeO<sub>2</sub>, and LSM perovskites reported in the literature.

Materials	Reaction equipment	Run number [-]	TR temp. [°C]	average O <sub>2</sub> productivity [ $\mu$ mol /g]	CS temp. [°C]	average CO productivity [ $\mu$ mol /g]	CS duration [min]	CO <sub>2</sub> conc. [-]	total flow rate at CS step [mL/min]	total feeding amount of CO <sub>2</sub> [ $\mu$ mol]	Cycle-averaged CO production rate [ $\mu$ mol/(min·g)]	Reference
La <sub>0.7</sub> Sr <sub>0.3</sub> Mn <sub>0.65</sub> Co <sub>0.35</sub> O <sub>3</sub> (LSMCo0.35)	TGA	2	1400	90	1200	533	30	0.500	100	$6.05 \times 10^4$	17.8	Present study
La <sub>0.7</sub> Sr <sub>0.3</sub> Mn <sub>0.8</sub> Ni <sub>0.3</sub> O <sub>3</sub> (LSMNi0.20)	TGA	2	1400	90	1200	456	30	0.500	100	$6.05 \times 10^4$	15.2	Present study
La <sub>0.7</sub> Sr <sub>0.3</sub> Mn <sub>0.875</sub> Mg <sub>0.125</sub> O <sub>3</sub> (LSMMg 0.125)	TGA	2	1400	90	1200	387	30	0.500	100	$6.05 \times 10^4$	12.9	Present study
CeO <sub>2</sub>	TGA	2	1400	90	1200	158	30	0.500	100	$6.05 \times 10^4$	5.27	Present study
La <sub>0.7</sub> Sr <sub>0.3</sub> Mn <sub>0.9</sub> Mg <sub>0.1</sub> O <sub>3</sub>	TGA	3	1400	90	1200	354	30	0.500	100	$6.05 \times 10^4$	11.8	[53]
La <sub>0.7</sub> Sr <sub>0.3</sub> Mn <sub>0.9</sub> Ni <sub>0.1</sub> O <sub>3</sub>	TGA	3	1400	90	1200	351	30	0.500	100	$6.05 \times 10^4$	11.7	[53]
La <sub>0.7</sub> Sr <sub>0.3</sub> Mn <sub>0.9</sub> Co <sub>0.1</sub> O <sub>3</sub>	TGA	3	1400	90	1200	347	30	0.500	100	$6.05 \times 10^4$	11.6	[53]
La <sub>0.7</sub> Sr <sub>0.3</sub> MnO <sub>3</sub>	TGA	3	1400	90	1200	248	30	0.500	100	$6.05 \times 10^4$	8.27	[53]
La <sub>0.5</sub> Sr <sub>0.5</sub> Mn <sub>0.95</sub> Sc <sub>0.05</sub> O <sub>3</sub>	TGA	3	1400	45	1100	506	45	0.400	40	$6.05 \times 10^4$	11.2	[39]
La <sub>0.5</sub> Sr <sub>0.5</sub>	TGA	3	1400	45	1100	460	45	0.400	40	$2.90 \times$	10.2	[39]

Mn <sub>0.75</sub> Ga <sub>0.25</sub> O <sub>3</sub>										10 <sup>4</sup>		
La <sub>0.65</sub> Sr <sub>0.35</sub> MnO <sub>3</sub>	TGA	2	1400	45	1050	194	60	0.500	20	$2.42 \times 10^4$	3.23	[29]
La <sub>0.5</sub> Sr <sub>0.5</sub> MnO <sub>3</sub>	TGA	2	1400	45	1050	242	60	0.500	20	$2.42 \times 10^4$	4.03	[29]
La <sub>0.6</sub> Sr <sub>0.4</sub> Mn <sub>0.5</sub> Co <sub>0.5</sub> O <sub>3</sub>	TGA	2	1300	45	1050	139	45	0.500	20	$1.81 \times 10^4$	3.09	[29]
LaMn <sub>0.5</sub> Ni <sub>0.5</sub> O <sub>3</sub>	TGA	2	1400	45	1050	105	45	0.500	20	$1.81 \times 10^4$	2.33	[29]
La <sub>0.7</sub> Sr <sub>0.3</sub> Mn <sub>0.9</sub> Cr <sub>0.1</sub> O <sub>3</sub>	fixed bed	3	1350	30	1200	215	50	0.840	75	$1.27 \times 10^5$	4.30	[47]
LaCo <sub>0.7</sub> Zr <sub>0.3</sub> O <sub>3</sub>	TGA	3	1300	20	800	224	60	0.500	80	$9.68 \times 10^4$	3.73	[41]
La <sub>0.7</sub> Sr <sub>0.3</sub> MnO <sub>3</sub>	TGA	9	1400	60	1000	209	30	0.500	100	$6.05 \times 10^4$	6.97	[33]
La <sub>0.6</sub> Sr <sub>0.4</sub> MnO <sub>3</sub>	TGA	9	1400	60	1000	295	30	0.500	100	$6.05 \times 10^4$	9.83	[33]
La <sub>0.3</sub> Sr <sub>0.7</sub> MnO <sub>3</sub>	TGA	9	1400	60	1000	342	30	0.500	100	$6.05 \times 10^4$	11.4	[33]
La <sub>0.6</sub> Sr <sub>0.4</sub> FeO <sub>3</sub>	TGA	1	1350	20	1000	251	50	1.000	100	$2.02 \times 10^4$	5.02	[45]
La <sub>0.6</sub> Sr <sub>0.4</sub> MnO <sub>3</sub>	TGA	1	1350	20	1000	469	50	1.000	100	$2.02 \times 10^4$	9.38	[45]
La <sub>0.6</sub> Sr <sub>0.4</sub> Mn <sub>0.8</sub> Fe <sub>0.2</sub> O <sub>3</sub>	TGA	1	1350	20	1000	330	50	1.000	100	$2.02 \times 10^4$	6.60	[45]
La <sub>0.5</sub> Sr <sub>0.5</sub> Mn <sub>0.83</sub> Mg <sub>0.17</sub> O <sub>3</sub>	TGA	2	1400	45	1050	208	60	0.500	20	$2.42 \times 10^4$	3.47	[31]
La <sub>0.5</sub> Sr <sub>0.5</sub> Mn <sub>0.75</sub> Al <sub>0.25</sub> O <sub>3</sub>	TGA	2	1400	45	1050	221	60	0.500	20	$2.42 \times 10^4$	3.68	[31]
La <sub>0.5</sub> Sr <sub>0.5</sub> Mn <sub>0.9</sub> Mg <sub>0.1</sub> O <sub>3</sub>	TGA	2	1400	45	1050	215	60	0.500	20	$2.42 \times 10^4$	3.58	[32]
La <sub>0.6</sub> Sr <sub>0.4</sub> Mn <sub>0.6</sub> Al <sub>0.4</sub> O <sub>3</sub>	stagn- ation flow reactor	1	1350	-	1000	294	-	-	-	-	-	[36]

<div>(La<sub>0.8</sub>Sr<sub>0.2</sub>)</div> <div>(Mn<sub>0.2</sub>Fe<sub>0.2</sub></div> <div>Co<sub>0.4</sub>Al<sub>0.2</sub>)O<sub>3</sub> </div>	TGA	2	1400	45	1050	85	60	0.500	20	<div>2.42 ×</div> <div>10<sup>4</sup></div>	1.42	[67]
<div>(La<sub>0.5</sub>Sr<sub>0.5</sub>)</div> <div>Mn<sub>0.9</sub>Mg<sub>0.1</sub>O<sub>3</sub> </div>	TGA	2	1400	45	1050	248	60	0.500	20	<div>2.42 ×</div> <div>10<sup>4</sup></div>	4.13	[67]