

Electronic Supplementary Material (ESI) for Catalysis Science & Technology.  
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## Supporting Information for:

### **One-Pot Synthesis of Highly Mesoporous Ni/MgAl<sub>2</sub>O<sub>4</sub> Spinel Catalyst for Efficient Steam-Methane Reforming: Influence of Inert Annealing**

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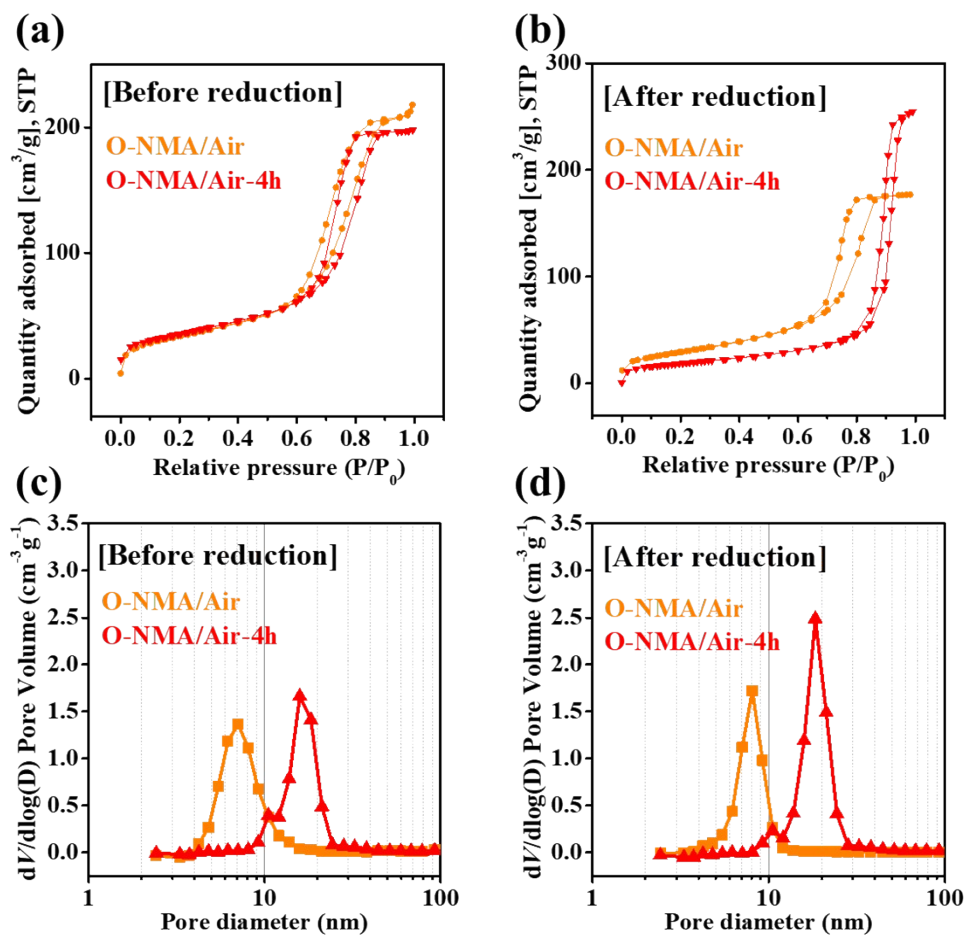
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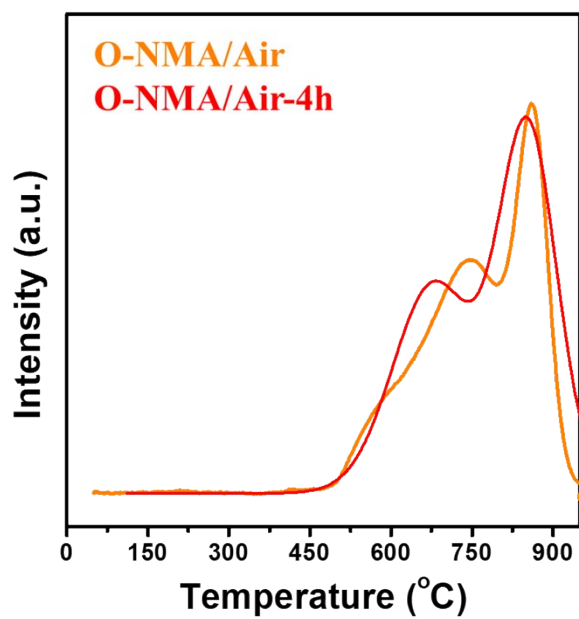
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**Fig. S1 – N<sub>2</sub> adsorption/desorption isotherms (a, b) and BJH pore-size distributions (c, d) of mesoporous Ni/MgAl<sub>2</sub>O<sub>4</sub> catalysts subjected to calcination in air for 2.0 h and 4.0 h, before (a, c) and after reduction (b, d).**



**Fig. S2 – H<sub>2</sub>-TPR of mesoporous Ni/MgAl<sub>2</sub>O<sub>4</sub> catalysts subjected to calcination under air for 2.0 h and 4.0 h.**

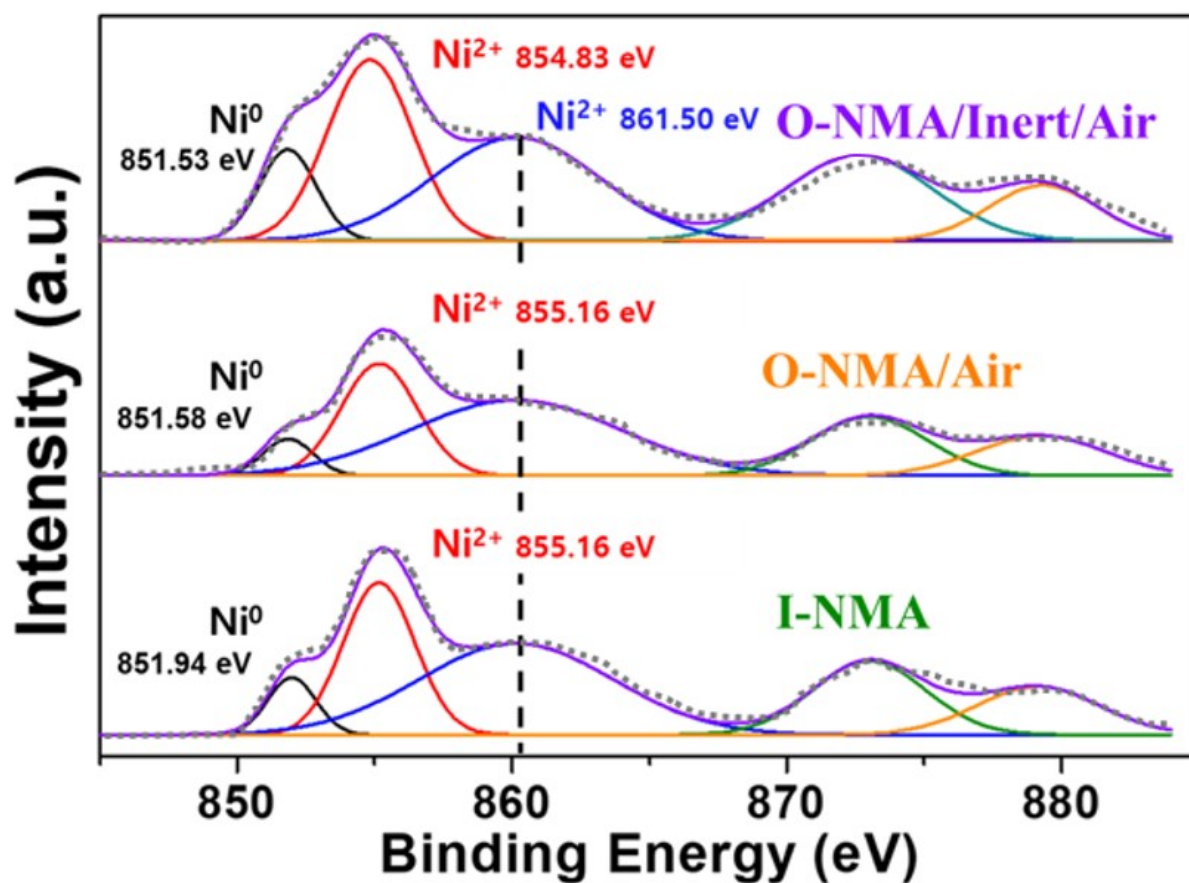


Fig. S3 – Fitted high-resolution XPS spectra of the Ni 2p in the studied catalysts.

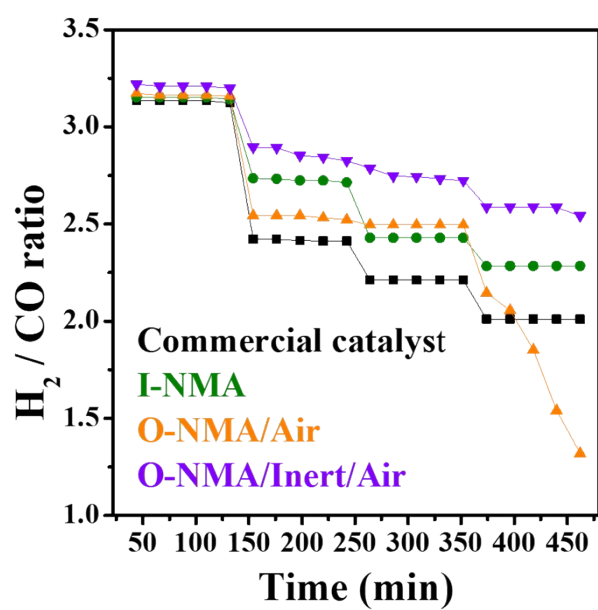


Fig. S4– H<sub>2</sub>/CO ratios of mesoporous Ni/MgAl<sub>2</sub>O<sub>4</sub> catalysts and commercial catalyst at different GHSVs ranging from 10,000 to 40,000 mL g<sub>cat</sub><sup>-1</sup> h<sup>-1</sup>.

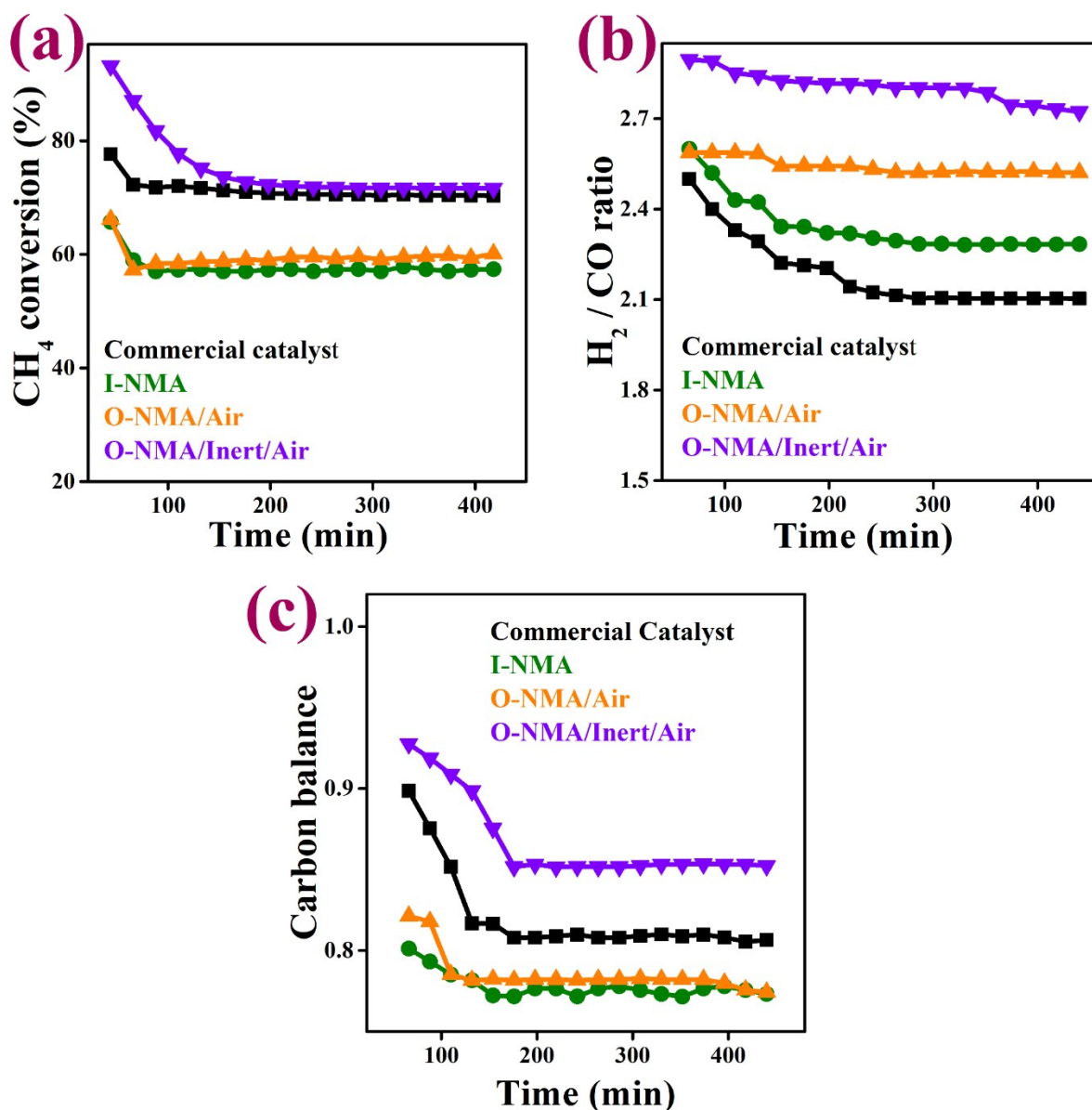


Fig. S5 – CH<sub>4</sub> conversions (a), H<sub>2</sub>/CO ratio (b), and carbon balance (c) of mesoporous Ni/MgAl<sub>2</sub>O<sub>4</sub> catalysts and commercial catalyst at H<sub>2</sub>O/CH<sub>4</sub>= 1, T= 700 °C, P= 1.0 atm., and GHSV= 10,000 mL g<sub>cat</sub><sup>-1</sup> h<sup>-1</sup> for 420 min.

**Table S1: BET surface area, pore volume, and pore diameter of the studied catalysts.**

<b>Catalyst</b>	<b>Calcined samples</b>			<b>Reduced samples</b>		
	<b>BET Surface Area (m<sup>2</sup>/g)</b>	<b>Pore Volume (cm<sup>3</sup>/g)</b>	<b>Pore diameter (nm)</b>	<b>BET Surface Area (m<sup>2</sup>/g)</b>	<b>Pore Volume (cm<sup>3</sup>/g)</b>	<b>Pore diameter (nm)</b>
<b>I-NMA</b>	156.17	0.4185	7.65	98.93	0.2454	17.63
<b>O-NMA/Air</b>	211.18	0.4723	8.01	104.37	0.2735	8.48
<b>O-NMA/Inert/Air</b>	253.08	0.5814	6.58	122.49	0.3433	9.36
<b>O-NMA/Air<sub>4h</sub></b>	187.49	0.4327	17.08	83.45	0.2517	19.88

**Table S2: ICP-OES data of the studied catalysts.**

Catalyst	Ni (wt%)	Mg (wt%)	Al (wt%)
I-NMA	18.1	7.7	34.2
O-NMA/Air	18.3	7.8	33.8
O-NMA/Inert/Air	18.2	7.6	34.4
O-NMA/Air <sub>4h</sub>	18.3	7.8	34.1

**Table S3: Binding energies of Ni species in the reduced catalysts.**

Catalyst	Binding energy (eV)	
	Ni <sup>0</sup>	Ni <sup>2+</sup>
I-NMA	851.94	855.16
O-NMA/Air	851.58	855.16
O-NMA/Inert/Air	851.53	854.83



**Table S4. Comparison of SMR catalytic activity and stability of our catalyst with that of similar composition reported in literature.**

Catalyst	Function	Ni wt%	Reaction Temperature (°C)	GHSV h <sup>-1</sup>	CH <sub>4</sub> conversion	Reference
O-NMA/Inert/Air	SMR	18	800	10,000	<b>97.62</b>	<b>This work</b>
O-NMA/Inert/Air	SMR	18	800	20,000	<b>92.05</b>	<b>This work</b>
O-NMA/Inert/Air	SMR	18	800	30,000	<b>88</b>	<b>This work</b>
Ni/MgAl <sub>2</sub> O <sub>4</sub>	SMR	10	750	9,600	97	1
NiAl <sub>2</sub> O <sub>4</sub>	SMR	31	800	12,000	97	2
NiAl <sub>2</sub> O <sub>4</sub>	SMR	29	700	12,000	91	2
NiAl <sub>2</sub> O <sub>4</sub>	SMR	15	650	38,400	27	3
NiCaAl <sub>2</sub> O <sub>4</sub>	SMR	10	850	32,000	73	4
NiAl <sub>2</sub> O <sub>4</sub>	SMR	10	600	32,000	84	5
Ni/MgAl <sub>2</sub> O <sub>4</sub>	SMR	15.3	600	48,500	48	6
15Ni/MgAl <sub>2</sub> O <sub>4</sub>	SMR	15	600	53,400	51	7
Ni/MgAl	SMR	15	700	3,000	75	8

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