## **Supporting Information**

## Photoreduction of Carbon Dioxide of Atmospheric Concentration with Water

to Methane over CoAl-Layered Double Hydroxide Nanosheets

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Figure S1. (a) TEM image of CoAl-LDH; (b) EDS profile of CoAl-LDH in selected area of

(a); (c-e) Elemental mappings of Co, Al, and O, respectively in selected area of (a).



Figure S2. UV-visible absorption spectrum of CoAl-LDH.



Figure S3. Time course of CO evolution under normal condition, without addition of CO<sub>2</sub>, Xe

lamp irradiation, or CoAl-LDH.



**Figure S4**. Time course of CH<sub>4</sub> evolution under normal condition, without addition of CO<sub>2</sub>, Xe lamp irradiation, or CoAl-LDH.



Figure S5. Time courses of CO and H<sub>2</sub> evolution for CoAl-LDH in the photocatalytic

conversion of CO<sub>2</sub> with water.



Figure S6. Time courses of  $CH_4$  evolution for CoAl-LDH and P25 in the photocatalytic conversion of  $CO_2$  with water.

Photocatalyst	Mass	cocatalyst	Reactant CO <sub>2</sub> Reducing agent		Light source	Wavelength	Temperature	Products	Amounts	ref
	[mg]					[nm]	[K]		[µmol/(g*h)]	
NiM-LDH (M=Al, Ga, In)	100		500 µmol	0.4 mL H <sub>2</sub> O	200 W Hg-Xe		RT	СО	1.9, 3.1, 3.6	[2]
MgM-LDH (M=Al, Ga, In)	100		500 µmol	0.4 mL H <sub>2</sub> O	200 W Hg-Xe		RT	СО	2.4, 2.6, 1.6	[2]
ZnM-LDH (M=Al, Ga, In)	100		500 µmol	0.4 mL H <sub>2</sub> O	200 W Hg-Xe		RT	СО	1.9, 0.7, 0.6	[2]
Zn <sub>3</sub> Al-LDH	100		180 µmol	1.7 mmol H <sub>2</sub>	500W Xe arc	200-1100	305-313	CO, CH <sub>3</sub> OH	0.62, 0.039	[3]
ZnCuAl-LDH	100		180 µmol	1.7 mmol H <sub>2</sub>	500W Xe arc	200-1100	305-313	CO, CH <sub>3</sub> OH	0.37, 0.13	[3]
ZnGa-LDH	100		180 µmol	1.7 mmol H <sub>2</sub>	500W Xe arc	200-1100	305-313	CO, CH <sub>3</sub> OH	0.08, 0.051	[3]
ZnCuCa-LDH	100		180 µmol	$1.7 \text{ mmol H}_2$	500W Xe arc	200-1100	305-313	CO, CH <sub>3</sub> OH	0.079, 0.17	[3]
ZnCr-LDH	50	Pt,Pd,Au		water vapour	200 W Hg-Xe	240-400	RT	CO	7.6, 4.7, 3.4	[4]
C <sub>3</sub> N <sub>4</sub> /MgAl-LDH	200	Pd	200 torr	water solution	500 W Hg-Xe		RT	$CO, CH_4$	0.2, 0.77	[5]
TiO <sub>2</sub>	150	Pd	suturated	1.5 mL H <sub>2</sub> O	500 W Hg-Xe	>310	RT	СО	0.35	[6]
Cu <sub>2</sub> O/TiO <sub>2</sub> nanosheets	20		40 mL	6 μL H <sub>2</sub> O	300 W Xe		RT	$CH_4$	2.78	[7]
PbS/TiO <sub>2</sub>		Cu		Saturated gas	300 W Xe	250-1800	RT	CO, CH <sub>4</sub>	0.82, 0.58	[8]
β-Ga <sub>2</sub> O <sub>3</sub>	100		150 µmol	50 µmol H <sub>2</sub>	200 W Hg-Xe			СО	0.76	[9]
NaNbO <sub>3</sub>	100	Pt	80 Kpa	$3 \text{ mL H}_2\text{O}$	300 W Xe	>300		$CH_4$	4.9	[10]
Zn <sub>2</sub> GeO <sub>4</sub> Nanoribbons	100	Pt,RuO <sub>2</sub>	230 mL	1 mL H <sub>2</sub> O	300W Xe arc			$CH_4$	6.5	[11]
ZnGa <sub>2</sub> O <sub>4</sub> /Zn <sub>2</sub> GeO <sub>4</sub>	100		230 mL	0.4 mL H <sub>2</sub> O	300W Xe arc	>200		$CH_4$	2.3	[12]
MgAl-LDO/TiO <sub>2</sub> cuboids	100			water	100W Hg	365	323	CO	1.5	[13]
CaLa <sub>4</sub> Ti <sub>4</sub> O <sub>15</sub>	300		15 mL/min	360 mL H <sub>2</sub> O	400W Hg	200-700	RT	CO	0.23	[14]
CaLa <sub>4</sub> Ti <sub>4</sub> O <sub>15</sub>	300	Ag	15 mL/min	360 mL H <sub>2</sub> O	400W Hg	200-700	RT	CO	0.07	[14]
BaLa <sub>4</sub> Ti <sub>4</sub> O <sub>15</sub>	300	NiO	15 mL/min	360 mL H <sub>2</sub> O	400W Hg	200-700	RT	CO	7.6	[14]
BaLa <sub>4</sub> Ti <sub>4</sub> O <sub>15</sub>	300	Cu	15 mL/min	360 mL H <sub>2</sub> O	400W Hg	200-700	RT	CO	2	[14]
SrLa <sub>4</sub> Ti <sub>4</sub> O <sub>15</sub>	300		15 mL/min	360 mL H <sub>2</sub> O	400W Hg	200-700	RT	CO	0.2	[14]
BiOCl	100		400 ppm	100 mL H <sub>2</sub> O	500 W Xe	200-1000	RT	CO	1.1	[15]
ZnAl-LDH nanosheets	100		100 mL	0.4 mL H <sub>2</sub> O	300W Xe arc	200-2500	RT	CO	7.6	[1]
HNb <sub>3</sub> O <sub>8</sub>	100		159 mL	Water vapor	350 W Xe	200-2500	318	$CH_4$	3.58	[16]
KNb <sub>3</sub> O <sub>8</sub>	100		159 mL	Water vapor	350 W Xe	200-2500	318	$CH_4$	1.71	[16]
P25/CoAl-LDH	50		1 bar	$5 \text{ mL H}_2\text{O}$	300W Xe	200-1200	RT	CO	2.21	[17]
CoAl-LDH/ P25	50		1 bar	$5 \text{ mL H}_2\text{O}$	300W Xe	200-1200	RT	CO	4.57	[18]
<b>CoAl-LDH nanosheets</b>	30		400 ppm	0.5 mL H <sub>2</sub> O	500 W Xe	200-1000		CH <sub>4</sub>	4.3	This
			(0.25 mL)							work

Table S1. LDHs and other types of photocatalysts towards CO<sub>2</sub> reduction into hydrocarbons and CO reported in the literature <sup>[1]</sup>



Figure S7. CO<sub>2</sub> adsorption capacities of several kinds of LDH and P25.



Figure S8. XRD patterns of CoAl-n (n=0, 0.25, 0.5, 2).



Figure S9. CO<sub>2</sub> adsorption capacities of CoAl-n (n=0, 2).



Figure S10. XRD patterns of ZnAl-LDH, NiAl-LDH, CoFe-LDH and CoCr-LDH.



Figure S11. UV-visible absorption spectra of CoAl-LDH, ZnAl-LDH, NiAl-LDH, CoFe-LDH and

CoCr-LDH.



Figure S12. FT-IR spectra of ZnAl-LDH, NiAl-LDH, CoFe-LDH and CoCr-LDH.



Figure S13. Time courses of  $CH_4$  evolution for several kinds of LDH in the photocatalytic conversion

of CO<sub>2</sub> with water.

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