Supporting Information

Solar-Driven Dehydrogenation and Dehydration of Formate to Syngas with Near-Zero CO_2 Emission

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Table S1. Photocatalytic syngas production via photocatalysis.

Photocatalyst	Photosensitizer	Reaction condition	CO and H_2 production rate	Remarks	Year	Ref.
Co_3O_4-CdS-100	N. A.	300W Xe lamp (AM 1.5G); 0.4M formate in NaOH aqueous solution (pH ~ 10)	CO: 1435.5 μmol g^{-1} h^{-1} H_2: 1848.3 μmol g^{-1} h^{-1}		2024	This work
2% Ag/TiO_2-SP	N. A.	150W solar simulator; MeOH aqueous solution bubbled with CO_2	CO: 103 μmol g^{-1} h^{-1} H_2: 220 μmol g^{-1} h^{-1}		2012	[1]
Co-ZIF-9	$[Ru(bpy)_3]Cl_2\cdot6H_2O$	Xe lamp (>420 nm); MeCN and TEOA aqueous solution bubbled with CO_2	CO: 41.8 μmol H_2: 29.9 μmol	Noble metal; Sacrificial agent;	2014	[2]
$Fe(CO)_3bpy$	$[Ru(bpy)_3]Cl_2$; $[Ir(ppy)_2(bpy)]PF_6$	Hg lamp (400-700 nm); NMP/TEOA aqueous solution bubbled with CO_2	CO: 35 μmol H_2: 42 μmol	Noble metal; Sacrificial agent;	2015	[3]
Meso. TiO_2	N. A.	200W UV lamp; Gas phase moisture CO_2	CO: 5.26 μmol g^{-1} h^{-1} H_2: 16.7 μmol g^{-1} h^{-1}		2015	[4]
TiO_2 fiber	N. A.	Four 6W UV lamps (365 nm); CO_2/H_2O mix gas	CO: 10.20 μmol g^{-1} h^{-1} H_2: 19.94 μmol g^{-1} h^{-1}		2016	[5]
TiO_2/ReP:CoP	(*E*)-2-Cyano-3-(50-(500-(p-(diphenylamino) phenyl)thiophen-	Three 60W LED lamps (>400 nm); DMF	CO: 77.3 μmol g^{-1} h^{-1} H_2: 22.1 μmol g^{-1} h^{-1}		2016	[6]

	200-yl)-thiophen-20-yl)-acrylic acid (Dye)	aqueous solution bubbled with CO_2				
Rh-Au@$SrTiO_3$	N. A.	300W Xe lamp (>400 nm); Gas phase CO_2 at 70 kPa	CO: 66.8 μmol g^{-1} h^{-1} H_2: 50.5 μmol g^{-1} h^{-1}		2016	[7]
Co_6–MOF	$[Ru(bpy)_3]Cl_2·6H_2O$	150W Xe lamp (420-780 nm); MeCN and TEOA aqueous solution bubbled with CO_2	CO: 39.6 μmol H_2: 28.13 μmol	Noble metal; Sacrificial agent;	2017	[8]
$Co(bpy)_2Cl_2$	$[Ru(bpy)_3]Cl_2$	300W Xe lamp (>420 nm); MeCN and TEOA aqueous solution bubbled with CO_2	CO: 62.3 μmol H_2: 69.9 μmol	Noble metal; Sacrificial agent;	2018	[9]
C-BMZIFs	$[Ru(bpy)_3]Cl_2·6H_2O$	100W LED light (420 nm); MeCN and TEOA aqueous solution bubbled with CO_2	CO: 6883 μmol g^{-1} h^{-1} H_2: 3600 μmol g^{-1} h^{-1}	Noble metal; Sacrificial agent;	2018	[10]
$MTC_{3.17P}$-MS	N. A.	300W Xe lamp (Solar); 0.1 M $KHCO_3$ and 0.1 M Na_2SO_3 aqueous solution bubbled with CO_2	CO: 80 μmol g^{-1} h^{-1} H_2: 160 μmol g^{-1} h^{-1}		2018	[11]
Pd/CoAl-7.57	$[Ru(bpy)_3]Cl_2·6H_2O$	300W Xe lamp (400-800 nm); MeCN and TEOA aqueous solution	CO: 1300 μmol g^{-1} h^{-1} H_2: 600 μmol g^{-1} h^{-1}	Noble metal; Sacrificial agent;	2019	[12]

		bubbled with CO_2 at 1.8 bar				
$(Co/Ru)_{2.4}$-UiO-67(bpydc)	$[Ru(bpy)_3]Cl_2·6H_2O$	450 nm LED lamp; MeCN and TEOA aqueous solution bubbled with CO_2	CO: 282.5 μmol g^{-1} h^{-1} H_2: 570.1 μmol g^{-1} h^{-1}	Noble metal; Sacrificial agent;	2019	[13]
$[Co_3(SiW_{12}O_{40})(H_2O)_3$-$(Htrz)_6Cl]$ ·Cl·$6H_2O$	$[Ru(bpy)_3]Cl_2·6H_2O$	300W Xe lamp (>420 nm); MeCN and TEOA aqueous solution bubbled with CO_2	CO: 6167 μmol g^{-1} h^{-1} H_2: 6066 μmol g^{-1} h^{-1}	Noble metal; Sacrificial agent;	2019	[14]
CoN_4-SiO_2	g-C_3N_4	LED (450 nm); MeCN and TEA solution bubbled with CO_2	CO: 398 μmol g^{-1} h^{-1} H_2: 804 μmol g^{-1} h^{-1}	Sacrificial agent;	2019	[15]
$Ag_{1.0}Au_{1.0}/TiO_2$	N. A.	Four 6W UV lamps (365 nm); CO_2/H_2O mix gas	CO: 0.15 μmol g^{-1} h^{-1} H_2: 0.29 μmol g^{-1} h^{-1}		2020	[16]
CdSNRs	Fe(III)-Salen	300W Xe lamp (>420 nm); 1.33 M formic acid aqueous solution;	CO: 71500 μmol g^{-1} h^{-1} H_2: 150000 μmol g^{-1} h^{-1}	CO_2 emission;	2020	[17]
$[Co_5(btz)_6(NO_3)_4(H_2O)_4]$	$[Ru(bpy)_3]Cl_2$	300W Xe lamp (>420 nm); MeCN and TEOA solution bubbled with CO_2	CO: 79.2 μmol H_2: 140.6 μmol	Noble metal; Sacrificial agent;	2020	[18]
CoAl-LDH/MoS_2	$[Ru(bpy)_3]Cl_2·6H_2O$	300W Xe lamp (>400 nm); MeCN and TEOA aqueous solution bubbled	CO: 8070 μmol g^{-1} h^{-1} H_2: 8415 μmol g^{-1} h^{-1}	Noble metal; Sacrificial agent;	2020	[19]

		with CO_2 at 1.8 bar				
Pt modified Re-Bpy-sp^{2}c-COF	N. A.	300W Xe lamp (>420 nm); MeCN and TEOA solution bubbled with CO_2	CO: from ~1000 to ~100 μmol g^{-1} h^{-1} H_2: from ~200 to ~1200 μmol g^{-1} h^{-1}	Noble metal; Sacrificial agent;	2020	[20]
$Fe_{0.5}Ni_{0.5}$-COFs	$[Ru(bpy)_3]Cl_2·6H_2O$	5W white LED (400-800 nm); MeCN and TEOA aqueous solution bubbled with CO_2	CO: ~1750 μmol g^{-1} h^{-1} H_2: ~2500 μmol g^{-1} h^{-1}	Noble metal; Sacrificial agent;	2020	[21]
CoO-Mo8 UNWs	$[Ru(bpy)_3]Cl_2·6H_2O$	300W Xe lamp (>400 nm); MeCN and TEOA aqueous solution bubbled with CO_2	CO: 4165 μmol g^{-1} h^{-1} H_2: 11555 μmol g^{-1} h^{-1}	Noble metal; Sacrificial agent;	2020	[22]
$Co_2[Co_{20}Mo_{16}P_{24}]$	$[Ru(bpy)_3]Cl_2·6H_2O$	Xe lamp; MeCN and TEOA aqueous solution bubbled with CO_2	CO: ~16600 μmol g^{-1} h^{-1} H_2: ~56000 μmol g^{-1} h^{-1}	Noble metal; Sacrificial agent;	2020	[23]
$[Co(H_2O)_6]$[Co-POM]	$[Ru(bpy)_3]Cl_2·6H_2O$	Xe lamp (>420 nm); MeCN and TEOA aqueous solution bubbled with CO_2	CO: 24000 μmol g^{-1} h^{-1} H_2: 13300 μmol g^{-1} h^{-1}	Noble metal; Sacrificial agent;	2020	[24]
Mn SAs	$[Ru(bpy)_3]Cl_2·6H_2O$	Xe lamp (>420 nm); MeCN and TEOA aqueous solution bubbled with CO_2	CO: 1470 μmol g^{-1} h^{-1} H_2: 1310 μmol g^{-1} h^{-1}	Noble metal; Sacrificial agent;	2020	[25]
Fe-SAs/N-C	$[Ru(bpy)_3]Cl_2·6H_2O$	5W white	CO: 4500	Noble	2020	[26]

		LED (400-800 nm); MeCN and TEOA aqueous solution bubbled with CO_2	μmol g^{-1} h^{-1} H_2: 4950 μmol g^{-1} h^{-1}	metal; Sacrificial agent;		
Cu_2O/MnO_x	N. A.	300W Xe lamp (>420 nm); 0.1 M $KHCO_3$ and 0.1 M Na_2SO_3 aqueous solution bubbled with CO_2	CO: 114.2 μmol g^{-1} h^{-1} H_2: 82.2 μmol g^{-1} h^{-1}		2020	[27]
CeO_2-LDH	$[Ru(bpy)_3]Cl_2{\cdot}6H_2O$	300W Xe lamp (visible light); MeCN and TEOA aqueous solution bubbled with CO_2 at 1.8 bar	CO: 5 μmol g^{-1} h^{-1} H_2: 52 μmol g^{-1} h^{-1}	Noble metal; Sacrificial agent;	2021	[28]
$Fe_{0.5}Ni_{0.5}$ MOFs	$[Ru(bpy)_3]Cl_2{\cdot}6H_2O$	5W white LED (400-1000 nm); MeCN and TEOA aqueous solution bubbled with CO_2	CO: 5000 μmol g^{-1} h^{-1} H_2: 5500 μmol g^{-1} h^{-1}	Noble metal; Sacrificial agent;	2021	[29]
CTF-TDPN	$[Co(bpy)_3]^{2+}$	300W Xe lamp (>420 nm); MeCN and TEOA aqueous solution bubbled with CO_2	CO: 200 μmol g^{-1} h^{-1} H_2: 140 μmol g^{-1} h^{-1}	Sacrificial agent;	2021	[30]
NVs-PCN	$Co(bpy)_3$	50W LED lamp (420 nm); MeCN and TEOA aqueous	CO: 400 μmol g^{-1} h^{-1} H_2: 100 μmol g^{-1} h^{-1}	Sacrificial agent;	2021	[31]

		solution bubbled with CO_2				
POP_2-Fe	$[Ru(bpy)_3]Cl_2 \cdot 6H_2O$	500W Xe lamp; DMF and TEOA solution bubbled with CO_2	CO: 3043 μmol g^{-1} h^{-1} H_2: 3753 μmol g^{-1} h^{-1}	Noble metal; Sacrificial agent;	2021	[32]
Pt/BP-OvMBWO	N. A.	300W Xe lamp; MeCN and TEOA aqueous solution bubbled with CO_2	CO: 20.5 μmol g^{-1} h^{-1} H_2: 16.8 μmol g^{-1} h^{-1}	Noble metal; Sacrificial agent;	2021	[33]
Ag/$LaFeO_3$	N. A.	300W Xe lamp; 0.5 M $NaHCO_3$ aqueous solution bubbled with CO_2	CO: 2.41 μmol g^{-1} h^{-1} H_2: 7.3 μmol g^{-1} h^{-1}		2021	[34]
CdS/EDA NW	N. A.	300W Xe lamp (>420 nm); MeCN and TEOA aqueous solution bubbled with CO_2	CO: 115.6 μmol g^{-1} h^{-1} H_2: 959.4 μmol g^{-1} h^{-1}	Sacrificial agent;	2022	[35]
CdS/TiO_2:Cu hollow spheres	N. A.	300W Xe lamp; Gas phase CO_2 + H_2S	CO: 781.3 μmol g^{-1} h^{-1} H_2: 5875.1 μmol g^{-1} h^{-1}	Sacrificial agent; Presence of H_2S;	2022	[36]
ReCo-NU1000	N. A.	Eight LED (450 nm); MeCN and BIH aqueous solution bubbled with CO_2	CO: 280 μmol g^{-1} h^{-1} H_2: 114 μmol g^{-1} h^{-1}	Sacrificial agent;	2023	[37]
CdS/CNT	Fe porphyrin complexes	300W Xe lamp (>420 nm); 1.33 M formic acid in MeCN aqueous	CO: 12616 μmol g^{-1} h^{-1} H_2: 20500 μmol g^{-1} h^{-1}	CO_2 emission;	2023	[38]

		solution				
CdS/W_2N_3	N. A.	300W Xe lamp (>420 nm); 6 M formic acid with proper NaOH aqueous solution (pH = 3.5)	CO: 103500 μmol g^{-1} h^{-1} H_2: 131000 μmol g^{-1} h^{-1}	CO_2 emission;	2023	[39]
Ni_xCo_{1-x}-GR	$[Ru(bpy)_3]Cl_2·6H_2O$	300W Xe lamp (>420 nm); MeCN and TEOA aqueous solution bubbled with CO_2	CO: 12526 to 2953 μmol g^{-1} h^{-1} H_2: 844 to 10027 μmol g^{-1} h^{-1}	Noble metal; Sacrificial agent;	2023	[40]
Janus PdZn-Co	$[Ru(bpy)_3]Cl_2·6H_2O$	300W Xe lamp (>420 nm); MeCN and TEOA aqueous solution bubbled with CO_2	CO: 20300 μmol g^{-1} h^{-1} H_2: 9900 μmol g^{-1} h^{-1}	Noble metal; Sacrificial agent;	2023	[41]
Co^{III}-PBA@Co^{II}-PBA	$[Ru(bpy)_3]Cl_2·6H_2O$	300W Xe lamp (>420 nm); MeCN and TEOA aqueous solution bubbled with CO_2	CO: 50560 μmol g^{-1} h^{-1} H_2: 41630 μmol g^{-1} h^{-1}	Noble metal; Sacrificial agent;	2023	[42]
$CdS/V_{0.1}W_{0.9}N_{1.5}$	N. A.	300W Xe lamp (AM 1.5G); 1 M formic acid solution	CO: 9867.5 μmol g^{-1} h^{-1} H_2: 46010 μmol g^{-1} h^{-1}	CO_2 emission;	2024	[43]

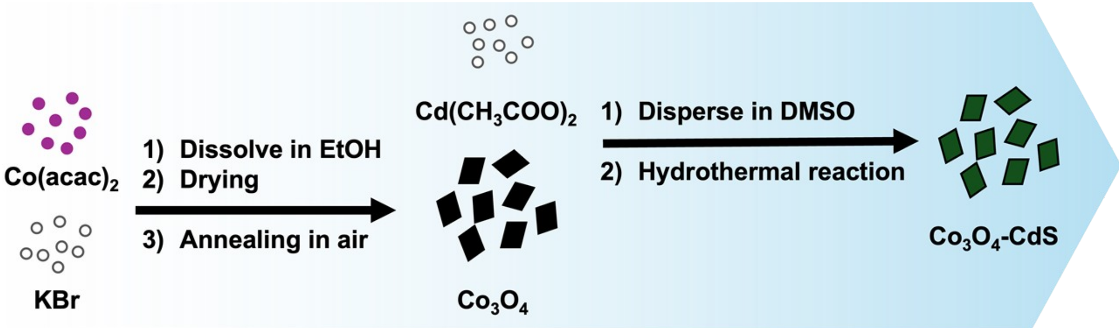

Fig. S1 Illustration of preparation method of Co_3O_4-CdS composites.

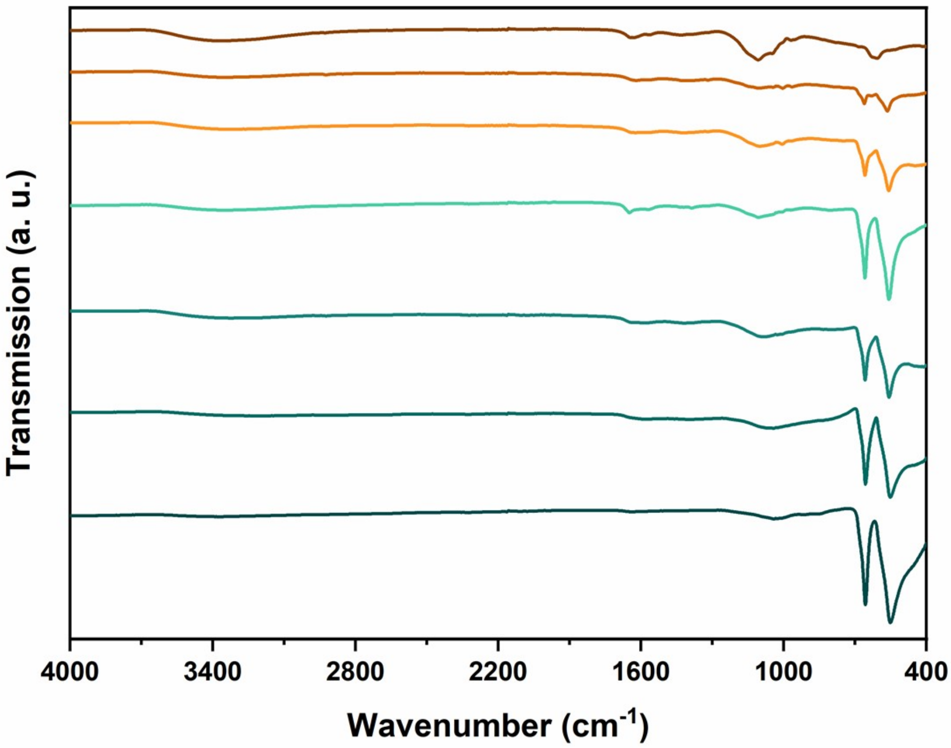

Fig. S2 FTIR spectra of Co_3O_4-CdS composites, from bottom to top: Co_3O_4, Co_3O_4-S, Co_3O_4-CdS-40, Co_3O_4-CdS-80, Co_3O_4-100, Co_3O_4-200, and CdS.

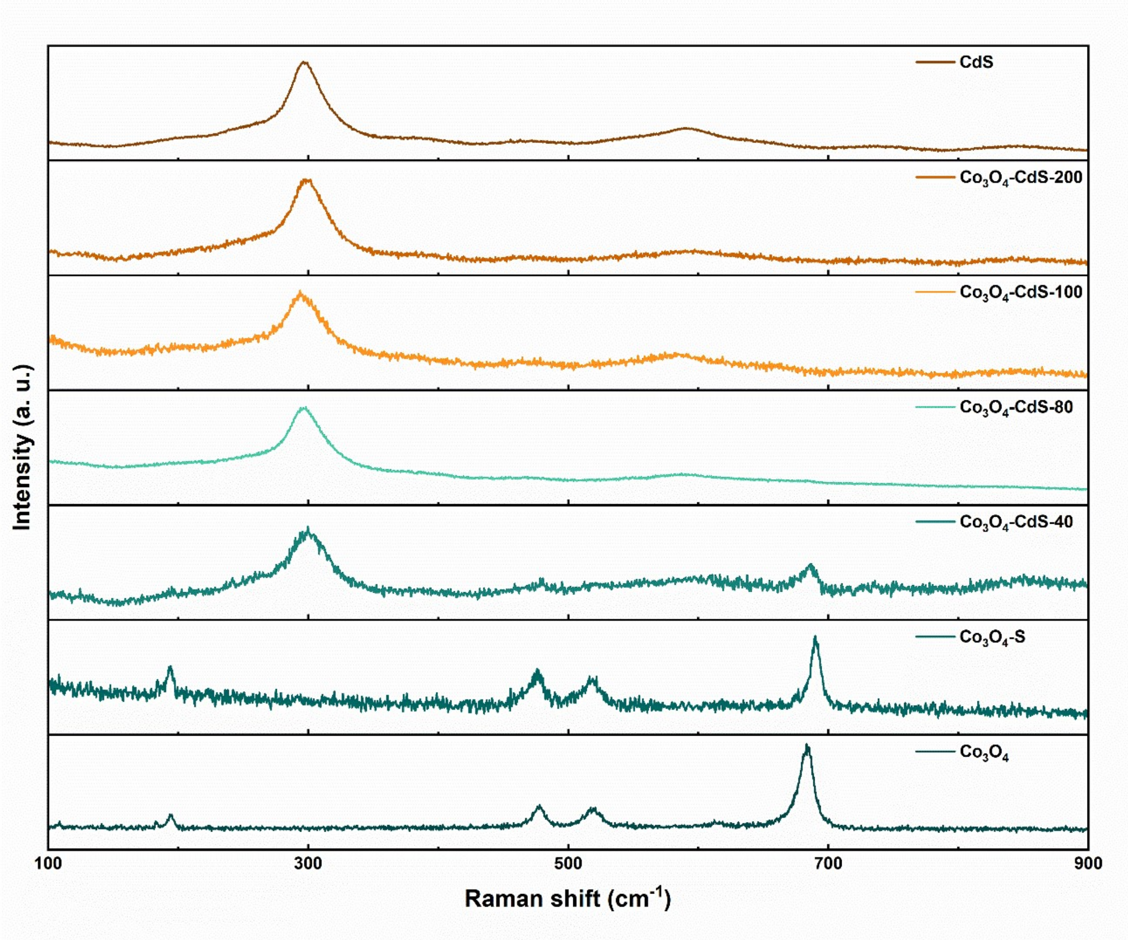

Fig. S3 Raman spectra of Co_3O_4-CdS composites.

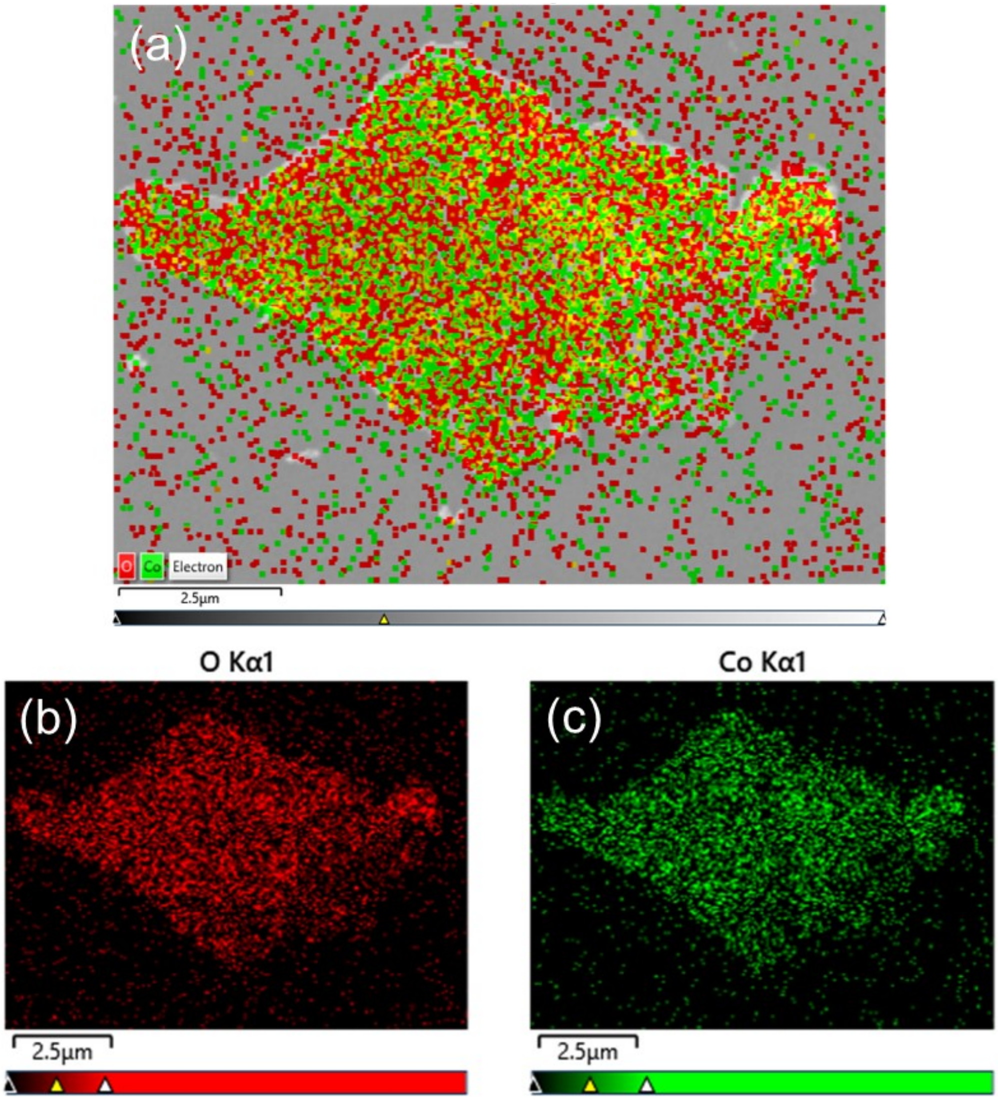

Fig. S4 (a-c) SEM elemental mapping of Co_3O_4.

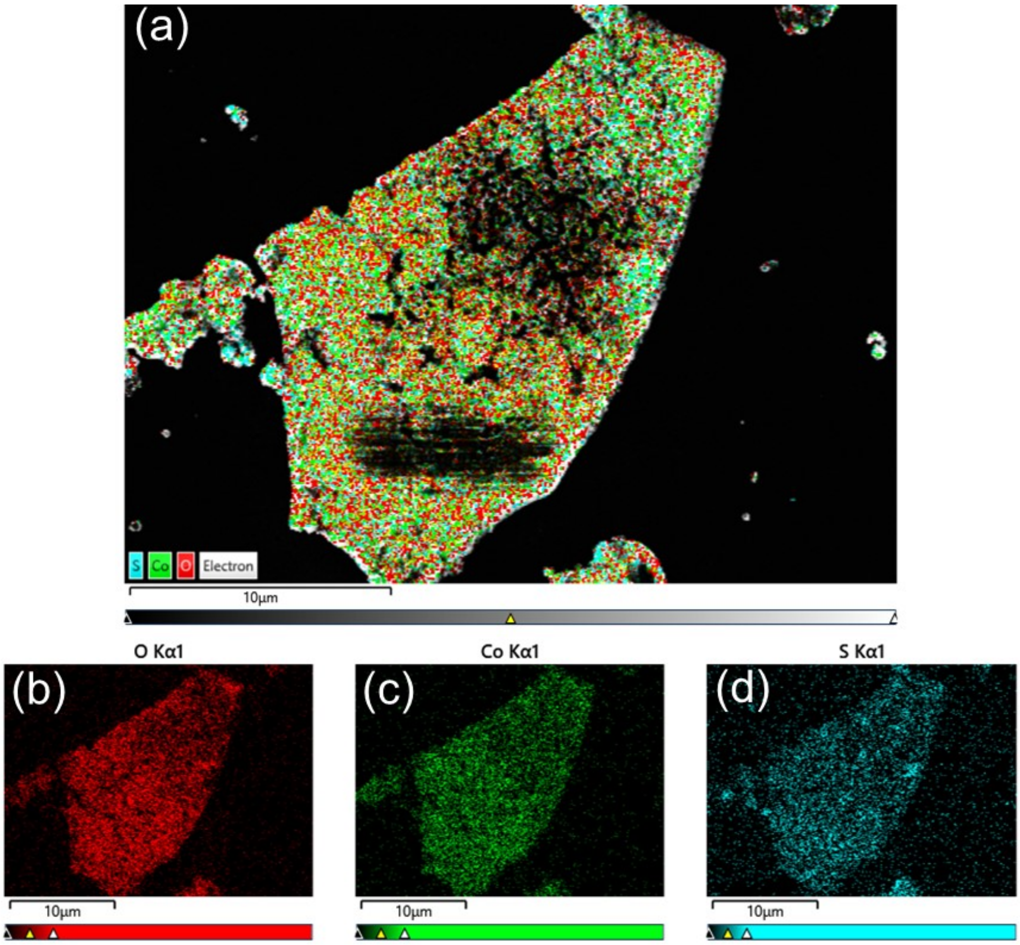

Fig. S5 (a-d) SEM elemental mapping of Co_3O_4-S.

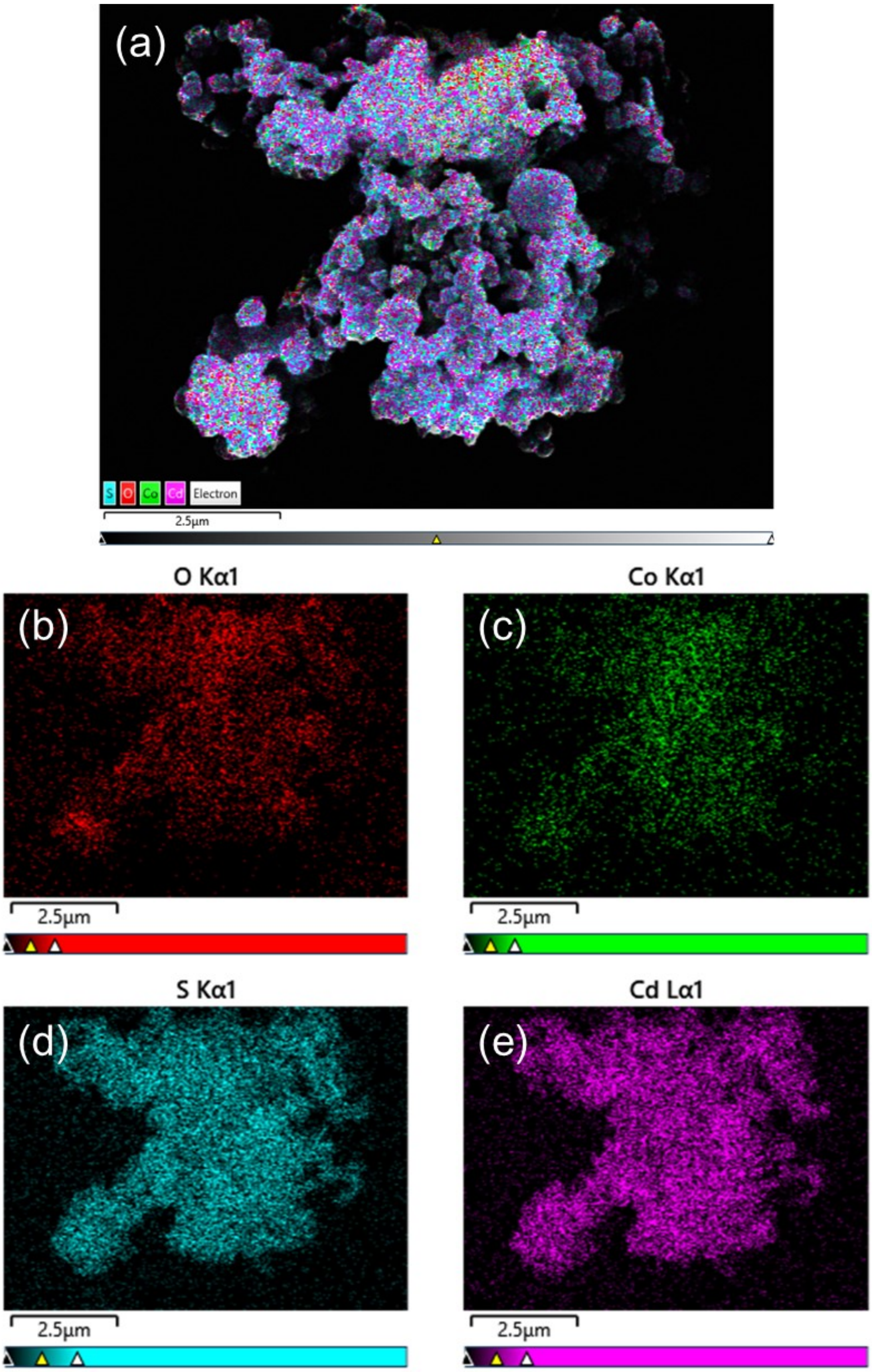

Fig. S6 (a-e) SEM elemental mapping of Co_3O_4-CdS-100.

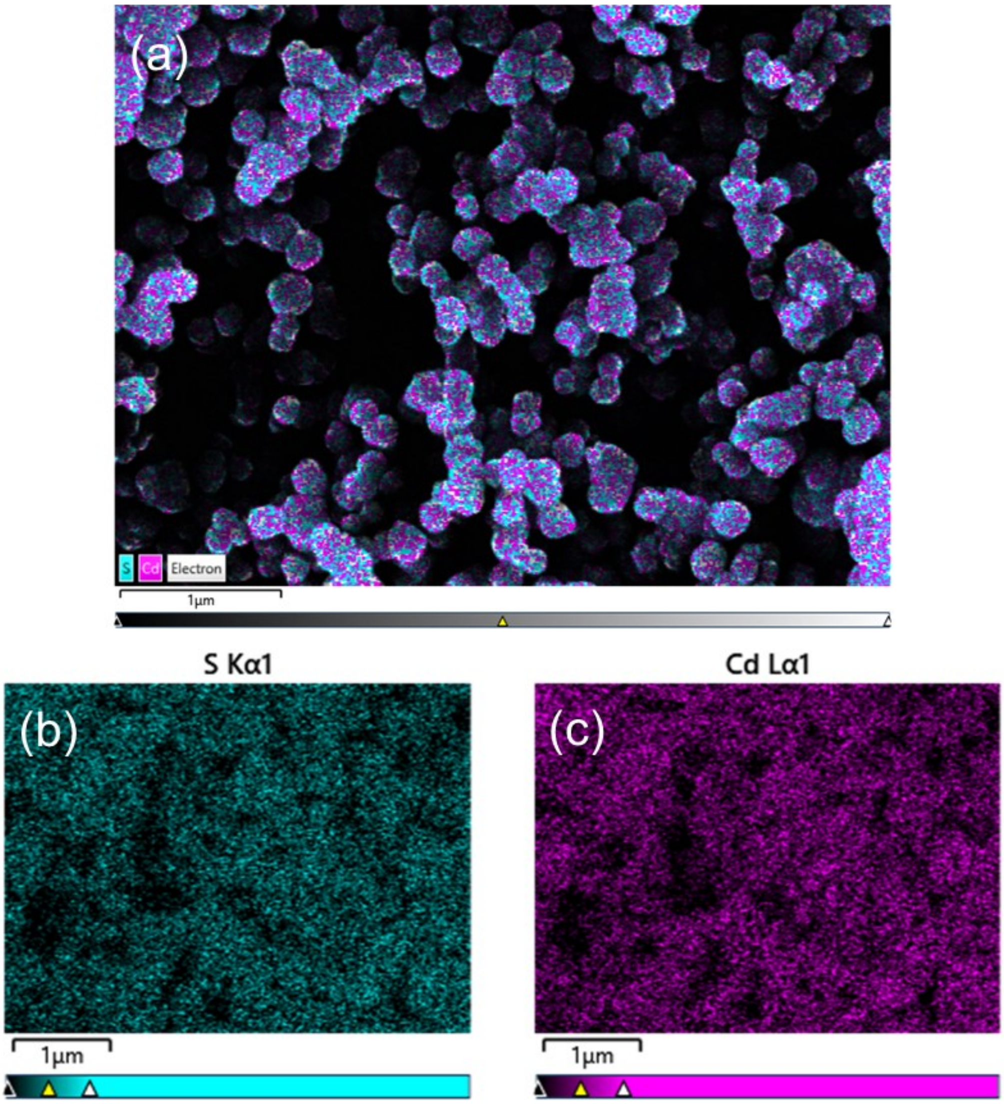

Fig. S7 (a-c) SEM elemental mapping of CdS.

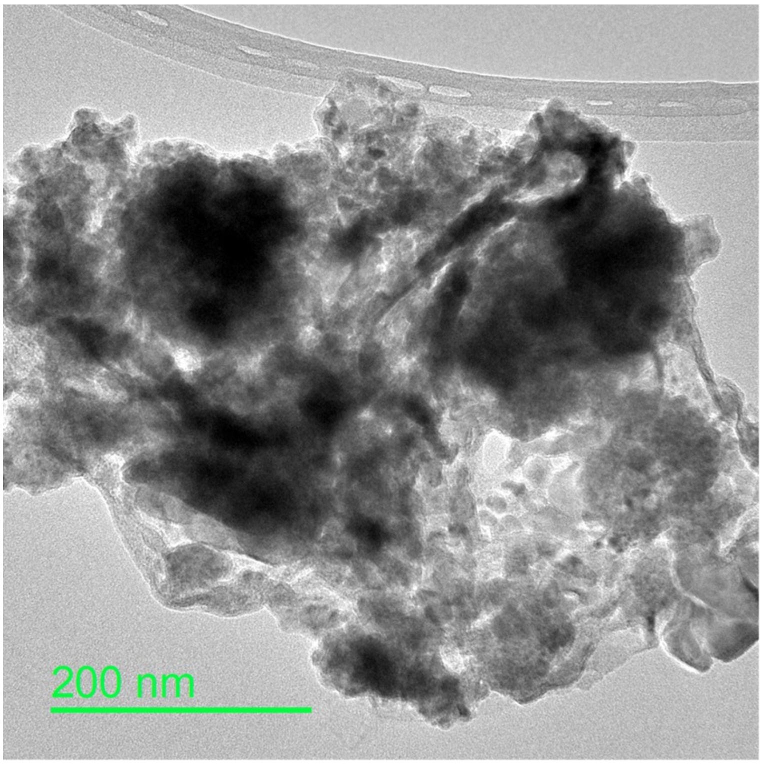

Fig. S8 TEM images of Co_3O_4-CdS-100.

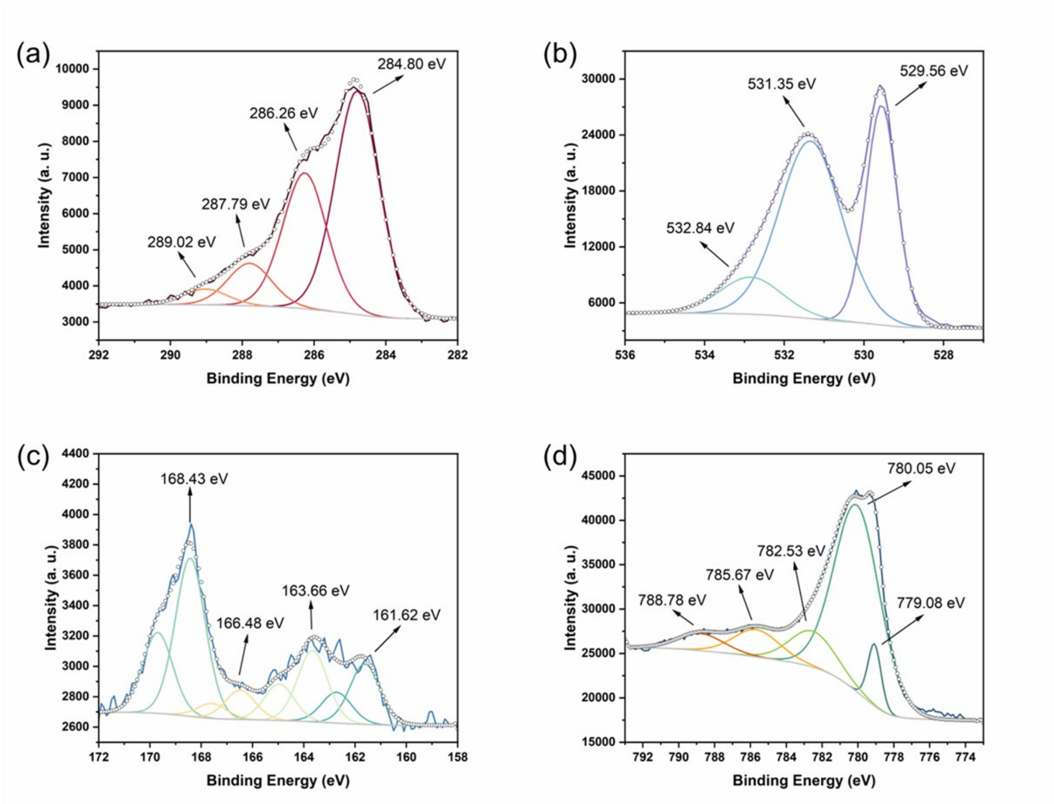

Fig. S9 XPS spectra of (a) C 1s, (b) O 1s, (c) S 2p, and (d) Co 2p of Co_3O_4-S.

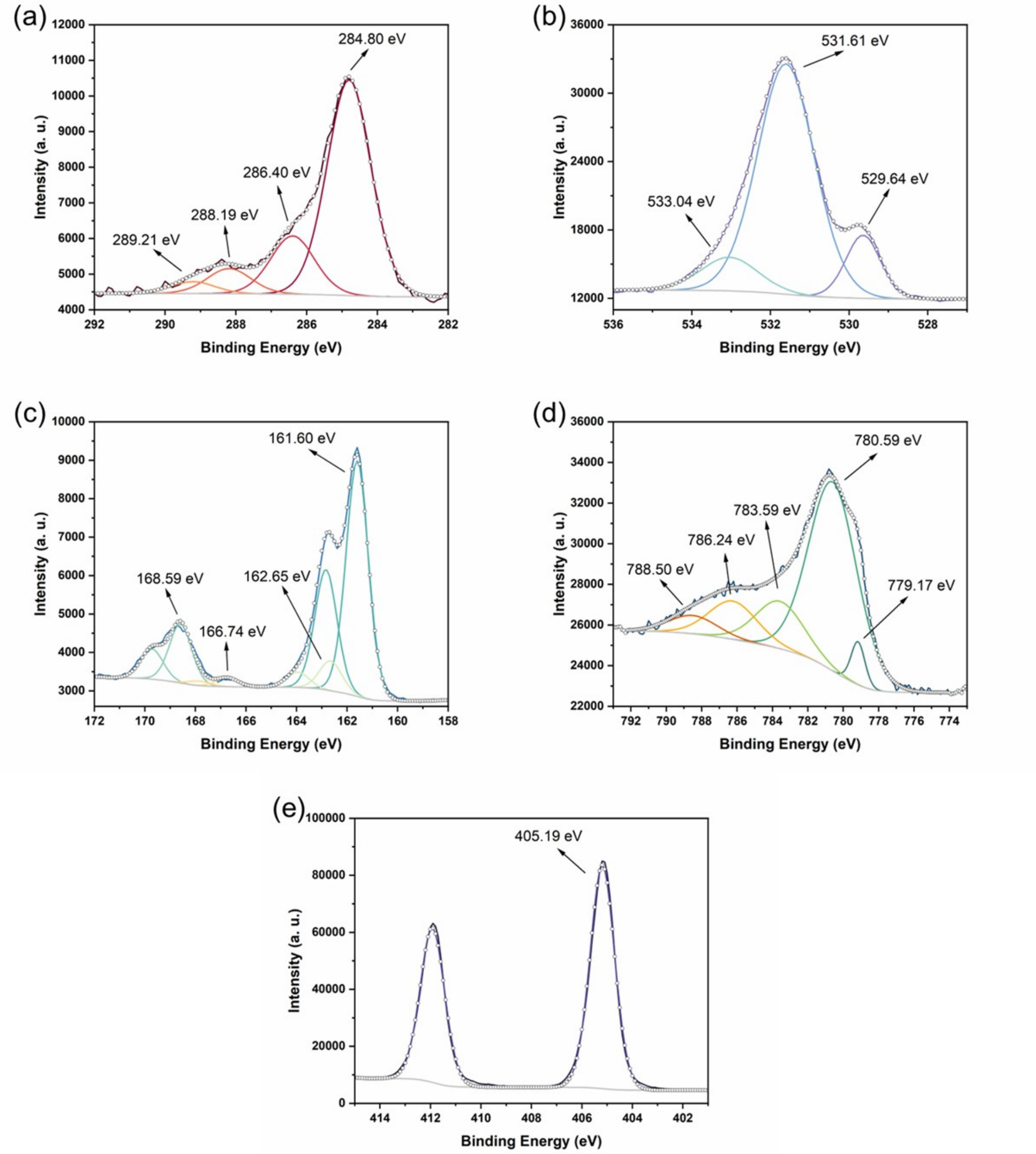

Fig. S10 XPS spectra of (a) C 1s, (b) O 1s, (c) S 2p, (d) Co 2p, and (e) Cd 3d of Co_3O_4-CdS-100.

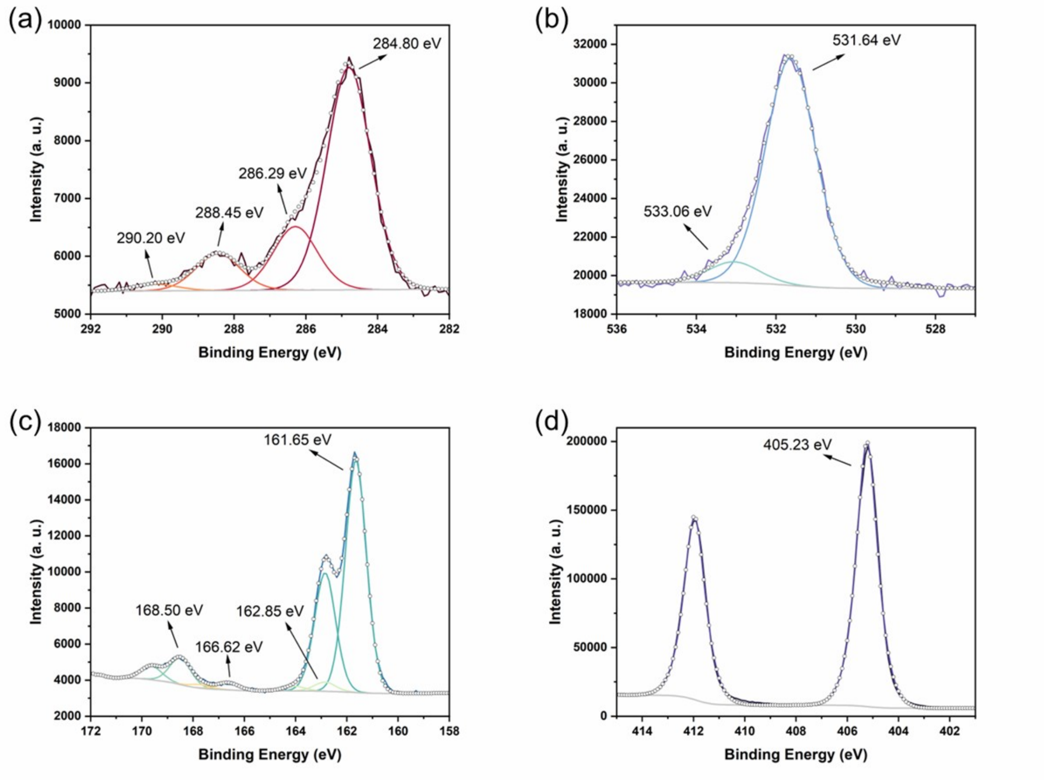

Fig. S11 XPS spectra of (a) C 1s, (b) O 1s, (c) S 2p, (d) Cd 3d of CdS.

Table S2. Summary of syngas production rates of different catalysts in the presence of 0.4 M formate.

Catalyst	pH	H_2 (μmol g^{-1} h^{-1})	CO (μmol g^{-1} h^{-1})
Co_3O_4-CdS-40	13	0	23.3
Co_3O_4-CdS-80	13	0	332.5
Co_3O_4-CdS-100	13	279.1	430.4
Co_3O_4-CdS-200	13	92.4	389.2

Table S3. Summary of syngas production rates of Co_3O_4-CdS-100 in the presence of 0.4 M formate at different pH conditions.

Catalyst	pH	H_2 (µmol g^{-1} h^{-1})	CO (µmol g^{-1} h^{-1})	CO_2 (µmol g^{-1} h^{-1})
Co_3O_4-CdS-100	4	9435.7	76.8	8857.0
Co_3O_4-CdS-100	7	3101.1	1300.2	37.4
Co_3O_4-CdS-100	10	1848.3	1435.5	4.2
Co_3O_4-CdS-100	11	708.4	779.9	2.5
Co_3O_4-CdS-100	12	274.1	590.5	0.8
Co_3O_4-CdS-100	13	279.1	430.4	0

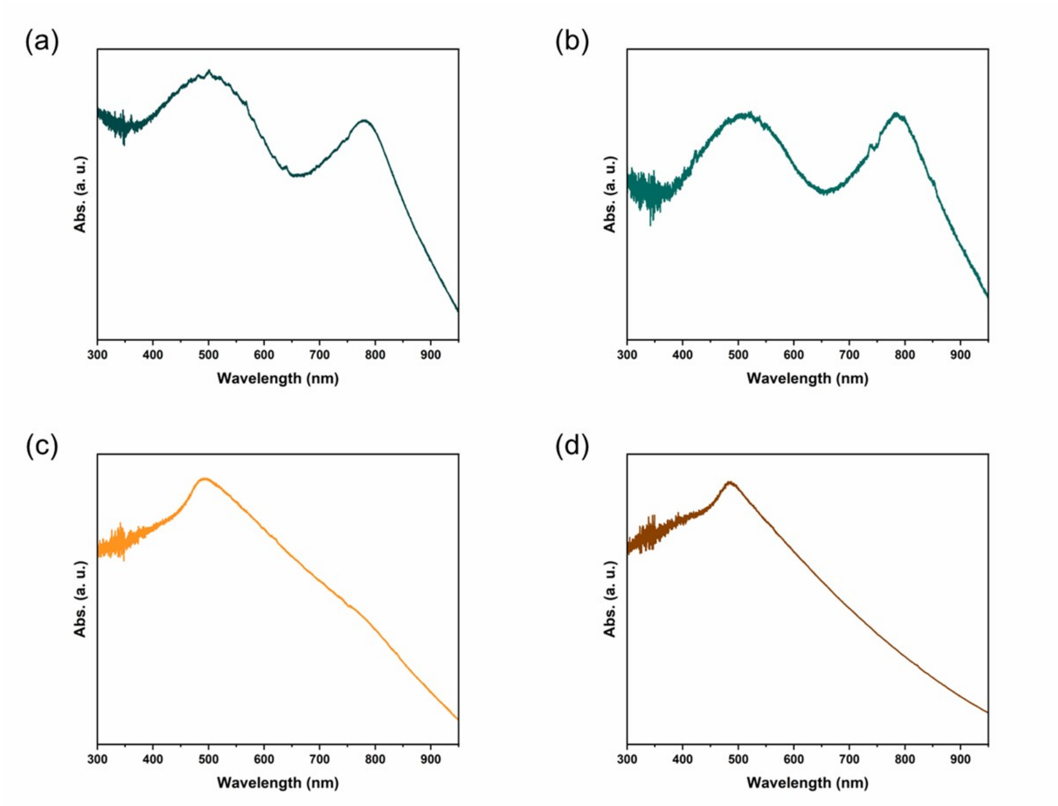

Fig. S12 UV-vis spectra of (a) Co_3O_4, (b) Co_3O_4-S, (c) Co_3O_4-CdS-100, and (d) CdS.

Table S4. Summary of TRPL results.

Sample	**t1 (ns)**	**Error bar (±) ns**	**t2 (ns)**	**Error bar (±) ns**	**Average Lifetime (ns)**	**Error bar (±) ns**
Co_3O_4-S (805 nm)	8.09	1.13	37.23	2.65	15.75	2.89
Co_3O_4-CdS-100 (510 nm)	11.28	0.39	47.10	0.88	18.85	0.96
CdS (508 nm)	11.00	0.37	46.33	0.88	17.68	0.95

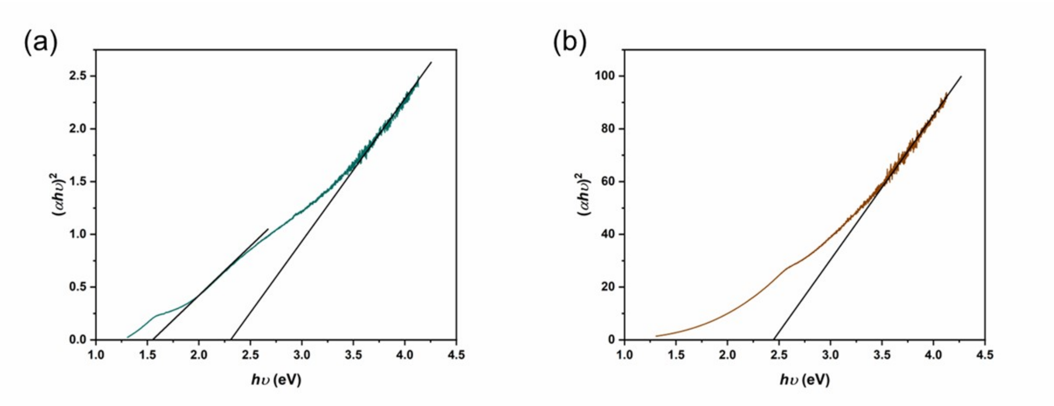

Fig. S13 Tauc plots of (a) Co_3O_4-S, and (b) CdS.

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