## The synergy of photodeposited CoNi co-catalysts for the photocatalytic performance of C<sub>3</sub>N<sub>4</sub>/CdS nanosheets: optimized Gibbs free energy and Co-S bridging bonds

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Precursors	CoCl <sub>2</sub> ·6H <sub>2</sub> O	NiCl <sub>2</sub> ·6H <sub>2</sub> O	NaH <sub>2</sub> PO <sub>2</sub> ·H <sub>2</sub> O	Illumination time
Samples	(mg)	(mg)	(mg)	(min)
CN/20CS-30Co <sub>0.67</sub> Ni <sub>0.33</sub>	104.58	52.29	48.97	30
CN/20CS-30Co <sub>0.5</sub> Ni <sub>0.5</sub>	78.44	78.44	48.97	30
CN/20CS-30Co <sub>0.33</sub> Ni <sub>0.67</sub>	52.29	104.58	48.97	30
CN/20CS-20Co <sub>0.33</sub> Ni <sub>0.67</sub>	52.29	104.58	48.97	20
CN/20CS-40Co <sub>0.33</sub> Ni <sub>0.67</sub>	52.29	104.58	48.97	40
CN/20CS-30Co		156.88	48.97	30
CN/20CS-30Ni	156.88		48.97	30

Table S1 Experimental parameters for loading different co-catalysts on CN/20CS.

Table S2 Composition of different electrolytes.

Reagent Group	CoCl <sub>2</sub> ·6H <sub>2</sub> O (mg)	NiCl <sub>2</sub> ·6H <sub>2</sub> O (mg)	NaH <sub>2</sub> PO <sub>2</sub> ·H <sub>2</sub> O (mg)	TEO A (mL)	H <sub>2</sub> O (mL)
А				8	40
В	52.29	104.58	48.97	8	40
С	156.88		48.97	8	40
D		156.88	48.97	8	40



Fig. S1 (a) SEM and (b) TEM images of  $C_3N_4$ , and (c) SEM and (d) TEM images of CdS. The insets in panel (a) and (c) are the EDX results of  $C_3N_4$  and CdS, respectively.



Fig. S2 (a-e) Element mapping images of CN/20CS-30Co and (f-j) CN/20CS-30Ni. The upper and lower insets in panel (a) are the detected sample and EDX images of CN/20CS-30Co, respectively. The upper and lower insets in panel (f) are the detected sample and EDX images of CN/20CS-30Ni, respectively.

Catalyst Content (Wt%)	CN/20CS-30Co	CN/20CS-30Co <sub>0.67</sub> Ni <sub>0.33</sub>	CN/20CS-30Co <sub>0.5</sub> Ni <sub>0.5</sub>	CN/20CS-30Co <sub>0.33</sub> Ni <sub>0.67</sub>	CN/20CS-30Ni
Со	1.30	0.46	0.34	0.22	
Ni		0.60	0.74	0.85	1.15
Total	1.30	1.06	1.08	1.07	1.15

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Fig. S3 Rough scan XPS spectra of CN/20CS and CN/20CS-30Co\_{0.33}Ni\_{0.67}.

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Catalyst Potential value (mV) Number	CdS	CdS-30Co <sub>0.33</sub> Ni <sub>0.67</sub>
1	-19.8.	-13.2
2	-21.7	-14.8
3	-20.6	-17.7
Average	-20.7	-15.2

Table S4 Zeta potential values of CdS and CdS-30Co<sub>0.33</sub>Ni<sub>0.67</sub>



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Table S5 Calculated  $E_{CB}$  and  $E_{VB}$  (vs. NHE) for  $C_3N_4$  and CdS.

Sample	E <sub>g</sub> /eV	E <sub>CB</sub> /eV	$E_{VB}/eV$
C <sub>3</sub> N <sub>4</sub>	2.60	-1.13	1.47
CdS	2.30	-0.70	1.60



Fig. S5 Time courses of photocatalytic hydrogen production of (a)  $C_3N_4$  and different CN/CS, and (b) those of CN/20CS-Co<sub>0.33</sub>Ni<sub>0.67</sub> for different photodeposition time.



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Fig. S7 The AQE of CN/20CS and CN/20CS- $30Co_{0.33}Ni_{0.76}$  under different monochromatic light excitation.



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