Supporting Information

In-Situ Surface Alkalinized g-C_3N_4 toward Enhancement of Photocatalytic H2 Evolution under Visible-Light Irradiation

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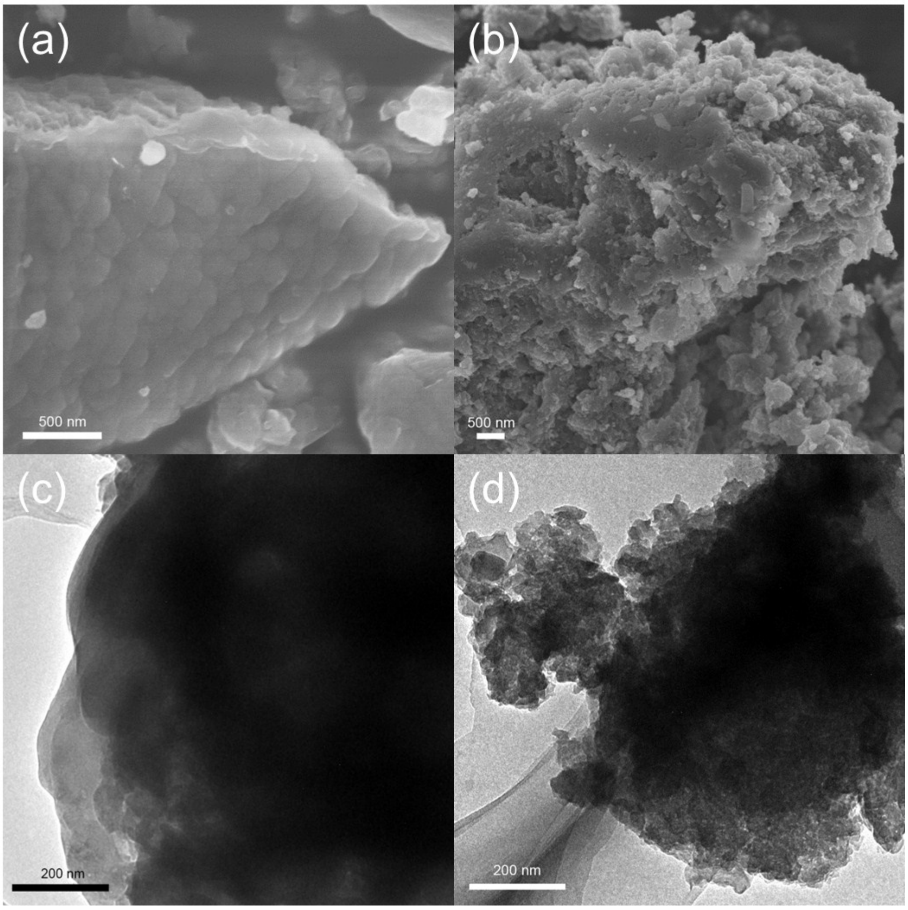

Figure S1. The SEM images of the pristine CN (a) and the CN-KCl/0.1gNH_4Cl (b) and TEM images of the pristine CN (c) and the CN-KCl/0.1gNH_4Cl (d).

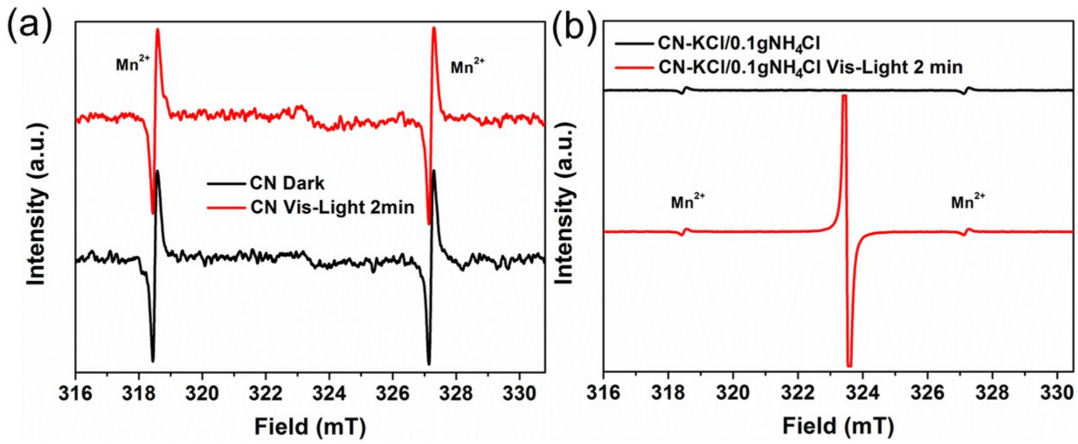

Figure S2. ESR spectra measured in the aqueous solution of the pristine CN (a) and CN-KCl/0.1gNH_4Cl (b) without DMPO exposed to visible light (420 < λ <800 nm).

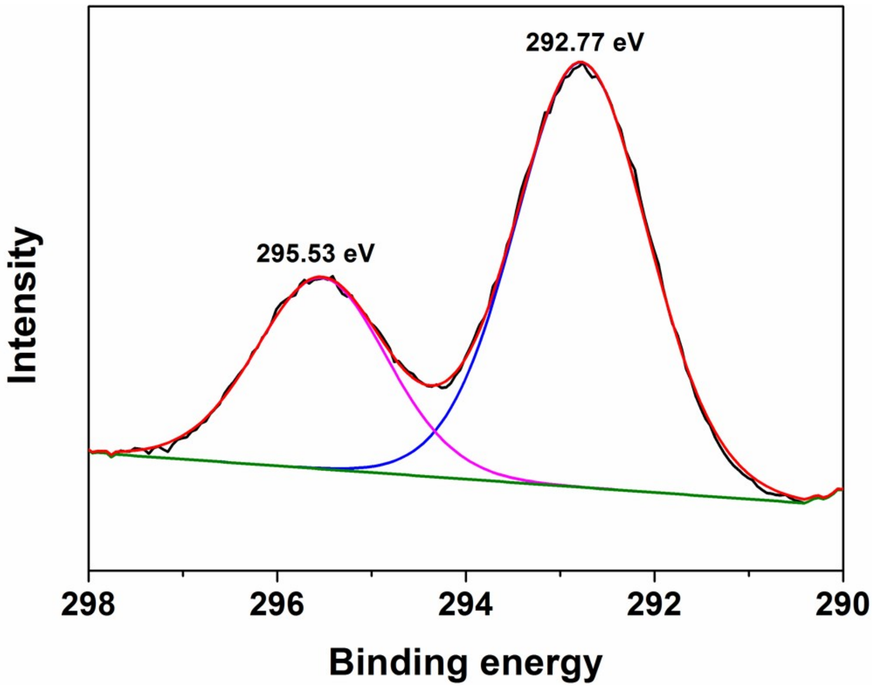

Figure S3. K 2p XPS spectra of the CN-KCl/0.1gNH_4Cl sample.

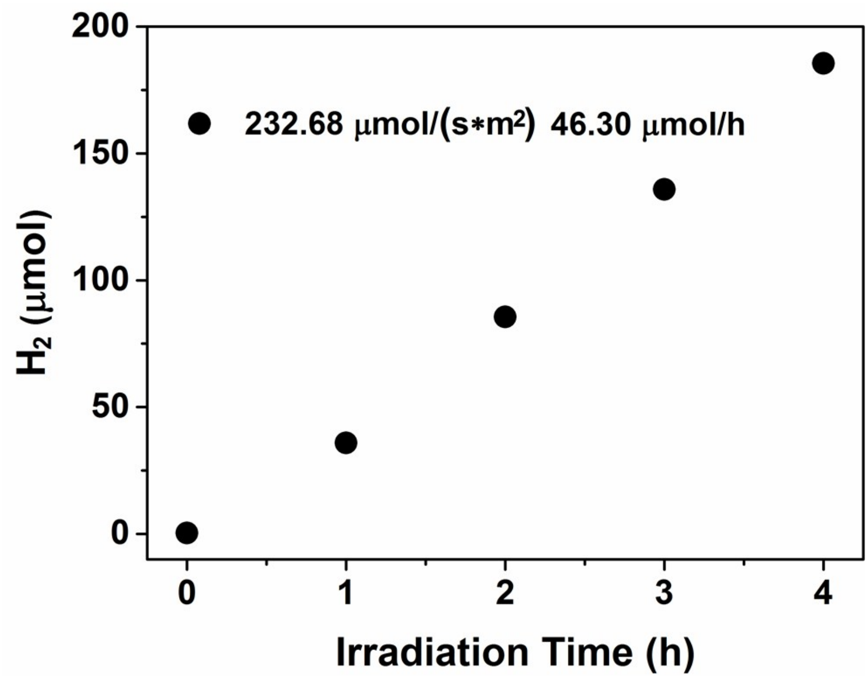

Figure S4. The photocatalytic activity of CN-KCl/0.1gNH_4Cl for AQY determination. A time course of H_2 production from water under 300 W Xenon lamp using a ~420 nm band-pass filter.

In one hour,

$N(H_2)$ = 46.30 μmol;

N(photon) = 232.68 μmol/(s*m^2)*19.36 cm^2*3600 s = 1621.68 μmol;

thus,

$$AQY (\%) = [2\times N(H_2)/N(photon)]\times 100\% = 2 \times 46.30 / 1621.68 \times 100\% = 5.7\%$$

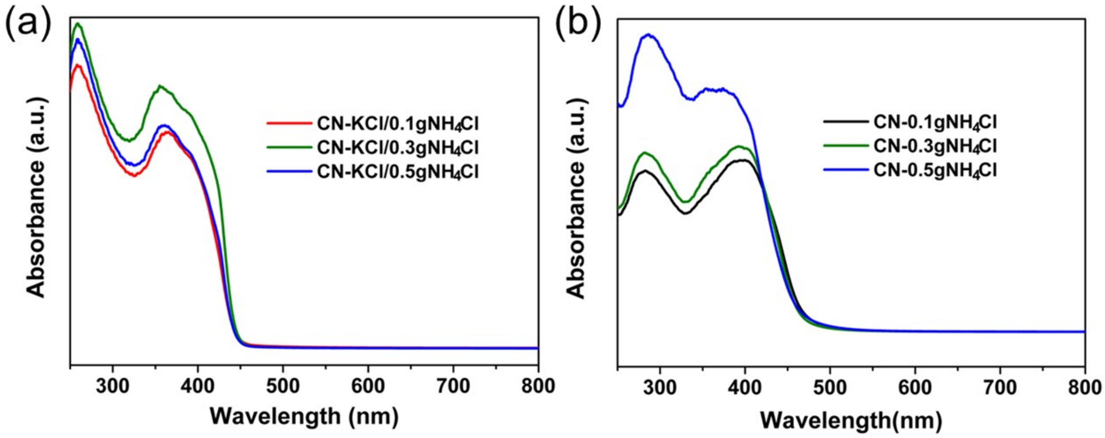

Figure S5. UV-visible absorption spectra of CN-KCl/xNH_4Cl (a) and CN-yNH_4Cl (b).

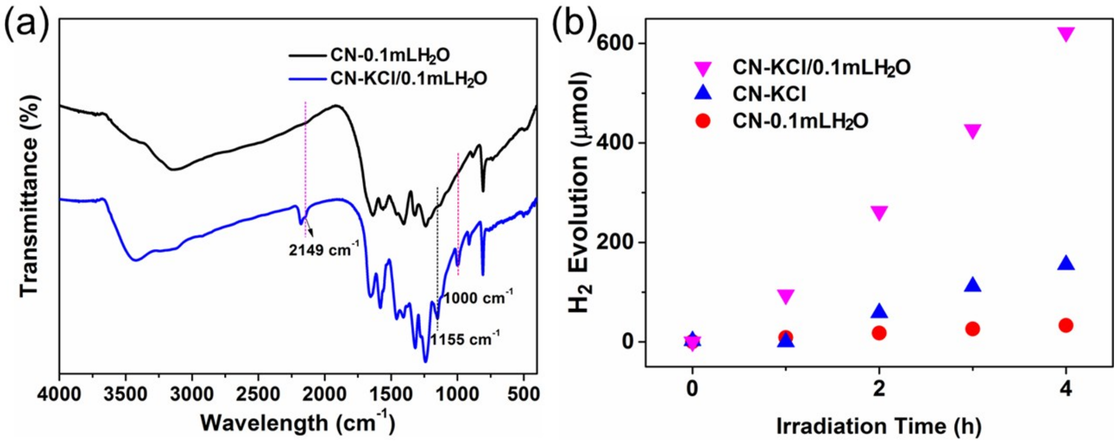

Figure S6. (a) FT-IR spectra of CN-0.1mLH_2O and CN-KCl/0.1mLH_2O. (b) Comparison of photocatalytic activities for H_2 evolution of CN-0.1mLH_2O and CN-KCl/0.1mLH_2O.

Table S1. Comparison of AQY for different modification g-C_3N_4

Samples	H_2 evolution rate (μmol/h)	AQY (%)	Notes
CN-KCl/0.1gNH_4Cl (this paper)	151 (λ > 400 nm)	5.7	100 mg, 2 wt% Pt λ = 420 nm, 6.4 mW cm^{-2}
HCNS[1]	574 (λ > 420 nm)	9.6	40 mg, 3 wt% Pt λ = 420.5 nm
Nanospherical g-C_3N_4[2]	224 (λ > 420 nm)	7.5	50 mg, 3 wt% Pt λ = 420.5 nm, 49.4 mW cm^{-2}
Intercalated g-C_3N_4[3]	502 (λ > 420 nm)	21.2	100 mg, 3 wt% Pt λ = 420 nm, 2 mW cm^{-2}

[1] J. Zhang, M. Zhang, C. Yang, and X. Wang, Adv. Mater. **26**, 4121 (2014).

[2] J. Sun, J. Zhang, M. Zhang, M. Antonietti, X. Fu, and X. Wang, Nature Communications, 1139 (2012).

[3] H. Gao, S. Yan, J. Wang, Y. A. Huang, P. Wang, Z. Li, and Z. Zou, Phys. Chem. Chem. Phys. **15**, 18077 (2013).

Table S2. Best fitted parameters of time-resolved PL spectra

Samples	Decay life times (ns)			Fractional contribution			Goodness of fit parameter (χ^2)
	τ_1	τ_2	τ_3	f_1	f_2	f_3	
CN	5.76	1.23	38.23	39.17	46.82	14.01	1.39
CN-0.1gNH_4Cl	5.02	0.99	33.22	36.39	49.87	13.74	1.37
CN-KCl	3.01	0.43	26.10	24.92	69.04	6.05	1.38
CN-KCl/0.1gNH_4Cl	2.53	0.32	27.25	18.34	75.82	5.34	1.22

Table S3. Comparison of H_2 evolution rate for different approaches to alkalinization over g-C_3N_4

Samples	Approach to alkalinization	H_2 evolution rate of the pristine samples (μmol/h)	H_2 evolution rate of the modified samples (μmol/h)	Notes
CN-KCl/0.1gNH$_4$Cl (this work)	In situ	10	151	100 mg, 2 wt% Pt (λ > 400 nm) Melamine (precursor)
CN–H1[5]	Soaking sample in alkalin solution	33	73	30 mg, 3 wt% Pt (λ > 420 nm) Urea
D52[4]	Tuning the pH of the photoreaction solution	~8 (pH=4.5)	47 (pH=13.3)	30 mg, 1 wt% Pt (λ > 400 nm) dicyandiamide

4. P. Wu, J. Wang, J. Zhao, L. Guo and F. E. Osterloh, *Chem. Commun.*, 2014, **50**, 15521-15524.
5. X. L. Wang, W. Q. Fang, H. F. Wang, H. Zhang, H. Zhao, Y. Yao and H. G. Yang, *Journal of Materials Chemistry A*, 2013, **1**, 14089.