Appendix A. Supplementary data

One-pot fabrication of 2D/2D HCa₂Nb₃O₁₀/g-C₃N₄ type II

heterojunction towards enhanced photocatalytic H_2

evolution under visible-light irradiation

Jinwen Shi, *^{a,b} Liuhao Mao,^a Chongze Cai,^a Gaosheng Li,^{a,d} Cheng Cheng,^a Botong Zheng,^a Yuchao Hu, *^{a,c} Zhesong Huang,^b Xiaowei Hu,^e and Gaweł Żyła^f

a. International Research Center for Renewable Energy (IRCRE), State Key Laboratory of Multiphase Flow in Power Engineering (MFPE), Xi'an Jiaotong University, Xi'an 710049, China. E-mail: jinwen_shi@mail.xjtu.edu.cn

- b. Department of New Energy Science and Engineering, School of Energy and Power Engineering, Xi'an Jiaotong University, Xi'an, 710049, China
- c. Center for Applied Chemistry, University of Electronic Science and Technology of China, Chengdu, 610000, China. E-mail: huyuchao@uestc.edu.cn
 - d. PetroChina West Pipeline Company, Urumqi, 830013, China
- e. School of Mechanical & Electrical Engineering, Shaanxi University of Science & Technology, Xi'an, 710021, China
 - f. Department of Experimental Physics, Rzeszow University of Technology, Rzeszow, 35-959, Poland

EXPERIMENTAL SECTION

Synthesis of HCNO-b

Bulk HCNO (HCNO-b) was prepared by solid-state and following ion-exchange methods as reported in previous literature.¹ Firstly, Nb₂O₅, K₂CO₃ and CaCO₃ powders were fully ground in a mortar, then the mixture were calcined in air at 800°C for 2 h in an alumina crucible. After cooling to room temperature naturally, the obtained white solids were ground for 30 min and then further calcined in air at 1150°C for 24 h. The sample thus obtained was designated as bulk KCa₂Nb₃O₁₀. Afterwards, the asprepared bulk KCa₂Nb₃O₁₀ (1 g) was dispersed in the mixed solution of deionized water (50 mL) and HNO₃ solution (5 mol L⁻¹, 50 g). Deionized water was then slowly added until the total volume of the solution reached 100 mL and then vigorous stirring was kept for 24 h. After centrifugation, washing and drying, HCNO-b was obtained.

Synthesis of HCNO

 $HCa_2Nb_3O_{10}$ nanosheets (HCNO) were prepared by an ultrasonic-assisted exfoliation method as previously reported.² The prepared HCNO-b (1 g) was dispersed in TBAOH aqueous solution (2 wt%, 50 mL) with stirring for 6 h followed by ultrasonic-assisted exfoliation for 30 min (1000 W, 50 Hz) to obtain a colloidal suspension of HCNO (0.02 g·mL⁻¹).

Sample	HCNO content ^[a] /wt%	Specific surface area ^[b] /m ² g ⁻¹	Pore volume ^[c] /cm ³ g ⁻¹
CN (0.0-HCNO/CN)	0.0	6.82	0.0516
0.5-HCNO/CN	0.5	11.02	0.0845
1.0-HCNO/CN	1.0	13.19	0.0917
5.0-HCNO/CN	5.0	18.99	0.0766
1.0-HCNO/CN-m	1.0	5.63	0.0373
1.0-TBAOH/CN	-	9.13	0.0734
HCNO	-	7.20	0.0233

Table S1. Specific surface areas and pore volumes of 1.0-TBAOH/CN, *x*-HCNO/CN with *x* equal to 0.0 0.5, 1.0 and 5.0, 1.0-HCNO/CN-m, and HCNO.

[a] Mass fraction of HCNO in the feed reagents, [b] calculated from N_2 adsorption-desorption isotherms by the Brunauer-Emmette-Teller (BET) method, [c] calculated from desorption branches of N_2 adsorption-desorption isotherms by the Barrett-Joyner-Halenda (BJH) method.



Fig. S1. XRD patterns of CN, HCNO-b, and *x*-HCNO/CN samples with *x* equal to 0.0 (i.e., CN) 0.5, 1.0 and 5.0.

Table S2. XPS binding energies (unit: eV) of C 1s and N 1s for CN and 1.0-HCNO/CN.

Sample		C 1s			N 1s		π excitation
CN	284.8	285.7	288.5	398.4	399.0	400.6	404.6
1.0-HCNO/CN	284.8	285.8	288.4	398.5	399.1	400.6	404.7



Fig. S2. TEM image of HCNO. Scale bar: 100 nm.



Fig. S3. Survey-scan XPS spectra of 1.0-HCNO/CN and CN.



Fig. S4. Determination of bandgaps of CN and 1.0-TBAOH/CN.



Fig. S5. Photocatalytic H₂-evolution activities of *x*-HCNO/CN samples with *x* equal to 0.0 (i.e., CN), 0.5, 1.0 and 5.0.

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- 2. Y. Hu and L. Guo, *ChemCatChem*, 2015, **7**, 584-587.