Electronic Supplementary Material (ESI) for Physical Chemistry Chemical Physics. This journal is © the Owner Societies 2018

> Electronic Supplementary Material (ESI) for **Physical Chemistry Chemical Physics**. This journal is © The Royal Society of Chemistry 2018

Supporting Information

Double Defects Modified Carbon Nitride Nanosheets with Enhanced Photocatalytic Hydrogen Evolution

Guilan Xu^{ab}, Jianchen Shen^{ab}, Shumei Chen^a, Yujie Gao^{ab}, Huabin Zhang^{*^c} and Jian Zhang^{*^b}



Figure S1 a)The image; b) The internal structure of Ultrasonic cell Disruption System .

Ultrasonic cell disruption system is a high strength ultrasonic instrument, which can use to exfoliate 2D material.



Figure S2 the XRD patterns of all samples.



Figure S3 N₂ adsorption/desorption isotherms for (a) the bulk g-C₃N₄, (b) CNNS (c) DCNNS-450 and pore size distribution curves for (d) bulk g-C₃N₄, (e) CNNS, (f) DCNNS-450.



Figure S4 Photocatalytic hydrogen production for all samples under visible light ($\lambda \ge 420$ nm)

irradiated.



Figure S5 Photocatalytic hydrogen production rate for all samples under visible light ($\lambda \ge 420$ nm) irradiated.



Figure S6 XRD patterns of DCNNS-450 beforen and after reaction.



Figure S7 TEM and HRTEM images (insets) of DCNNS-450 after reaction.



Figure S8 Color change of bulk g-C₃N₄, CNNS and DCNNS-T (T=200 °C, 300 °C , 400 °C, 450 °C, 500 °C, 530 °C).

Samples	Temperature	Heating	Time	Atmosphere	Instrument.
		rate			
Bulk-g-C ₃ N ₄	550	3 °C/min	4h	air	Muffle furnace
DCNNS-200	200	5 °C/min	4h	H ₂ (10%)/N ₂	Tube furnace
DCNNS-300	300	5 °C/min	4h	H ₂ (10%)/N ₂	Tube furnace
DCNNS-400	400	5 °C/min	4h	$H_2 (10\%)/N_2$	Tube furnace
DCNNS-450	450	5 °C/min	4h	$H_2 (10\%)/N_2$	Tube furnace
DCNNS-500	500	5 °C/min	4h	$H_2 (10\%)/N_2$	Tube furnace
DCNNS-530	530	5 °C/min	4h	$H_2 (10\%)/N_2$	Tube furnace

Table S1 The experimental conditions

Table S2 C, N, O, H elemental contents and C/N atomic ratios in DCNNS-T (T=200,

300, 400, 450, 500 and 530 °C)

Complea	С	N	0	TT	
Samples	C	IN	0	п	C/N
DCNNS-200	33.81	59.14	5.75	1.98	0.666
DCNNS-300	34.32	59.63	5.28	1.96	0.670
DCNNS-400	34.46	59.70	5.08	1.92	0.672
DCNNS-450	34.41	59.34	5.32	1.95	0.675
DCNNS-500	34.05	58.04	6.11	2.08	0.683
DCNNS-530	33.89	57.65	5.65	1.93	0.685

Note: The permissible error for the C and N elemental measurement is less than 1%. Moreover, the permissible error of oxygen elemental may be greater than that of C and N elemental. So, summation of values of N, C, O and H above- or below- 100, which is within the margin of error.

Table S3 Binding energies of O 1s core electrons of two kinds of O species recorded from the bulk $g-C_3N_4$, CNNS and DCNNS-450.

Samples	O1(eV)	O2(eV)
bulk <i>g</i> -C ₃ N ₄	531.84	532.63
CNNS	531.72	532.67
DCNNS-450	531.71	532.82

Table S4 Binding energies of C 1s core electrons of three kinds of C species recorded from the bulk $g-C_3N_4$, CNNS and DCNNS-450.

Samples	C1(eV)	C2(eV)	C3(eV)
bulk g -C ₃ N ₄	288.11	286.15	284.76
CNNS	288.15	286.16	284.77
DCNNS-450	288.19	286.24	284.80

Table S5 Binding energies of N 1s core electrons of three kinds of N species recorded from the bulk $g-C_3N_4$, CNNS and DCNNS-450.

Samples	Pyridinic N(eV)	Pyrrolic	Graphitic
		N(eV)	N(eV)
bulk g -C ₃ N ₄	398.58	399.54	401.18
CNNS	398.54	399.48	401.14
DCNNS-450	398.63	399.92	401.15

Table S6 Hydrogen evolution rate and standard Deviation for Bulk-g- C_3N_4 , CNNS and DCNNS-450.

Samples	hydrogen evolution	Standard Deviation	
Bulk-g-C ₃ N ₄	1.12×10^2	$1.65 \text{ x} 10^2$	26.86
CNNS	2.38×10^2	$2.56 ext{ x10}^2$	8.61
DCNNS-450	1.69×10^3	$1.44 \text{ x} 10^3$	122.04

Table S7 HER over bulk-g-C₃N₄, CNNS and DCNNS-450 at 420 nm, 450 nm and 500 nm.

Samples	420 nm	450 nm	500 nm
bulk-g-C ₃ N ₄	1.33×10^2	~	~
CNNS	3.81×10^2	$7.60 \mathrm{x} 10^{1}$	~
DCNNS-450	3.85×10^2	3.65×10^2	$1.60 \mathrm{x} 10^{1}$

Note: ~: No H₂ was detected.