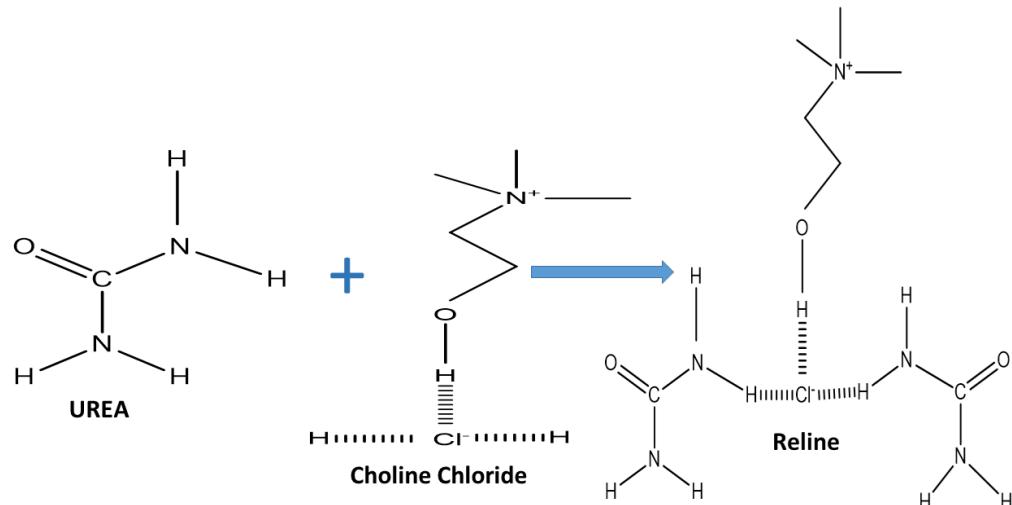


## Supporting Information on

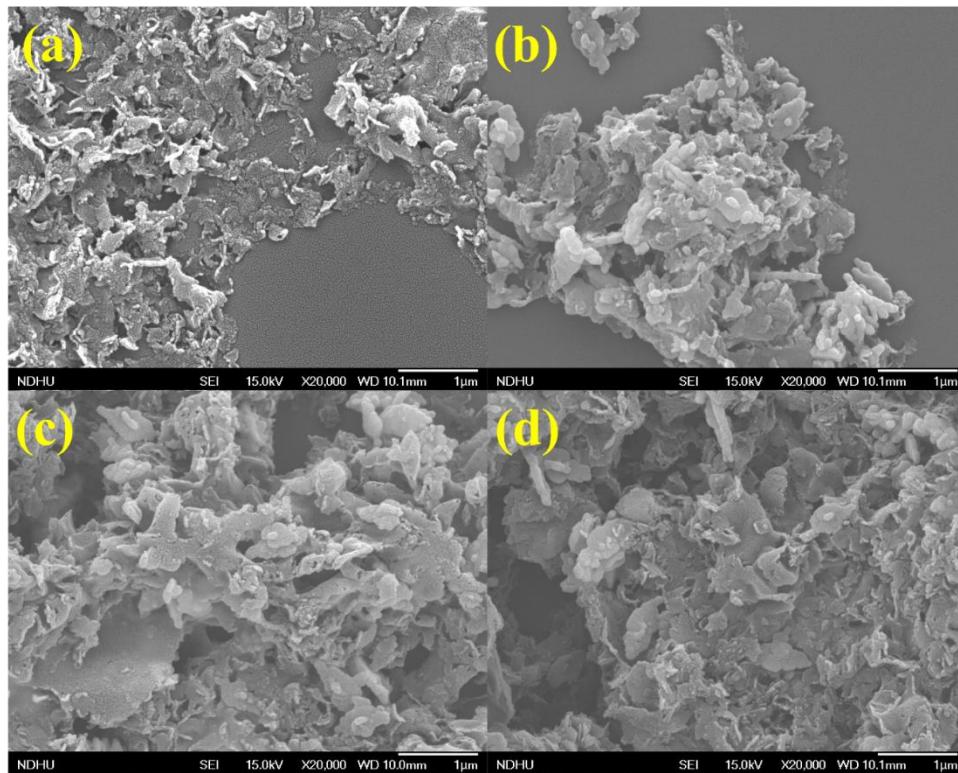
### Photocatalytic 4-nitrophenol degradation and oxygen evolution reaction in CuO/g-C<sub>3</sub>N<sub>4</sub> composites prepared by deep eutectic solvent assisted chlorine doping

Atul Verma<sup>‡</sup>, Dhayanantha prabu Jaihindh<sup>‡</sup>, and Yen-Pei Fu<sup>\*</sup>

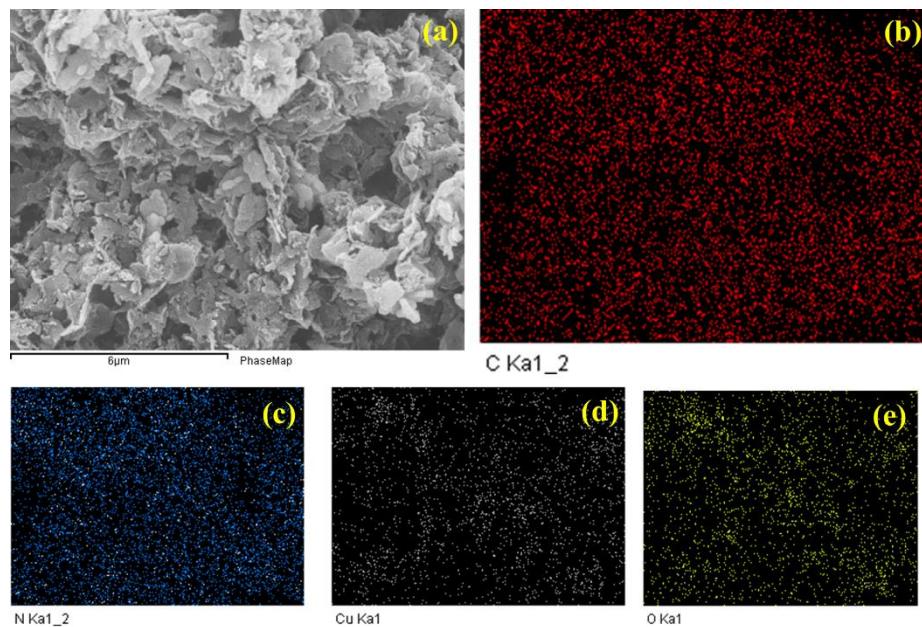
Department of Materials Science and Engineering, National Dong Hwa University, Shoufeng,  
Hualien-97401, Taiwan, R.O.C.



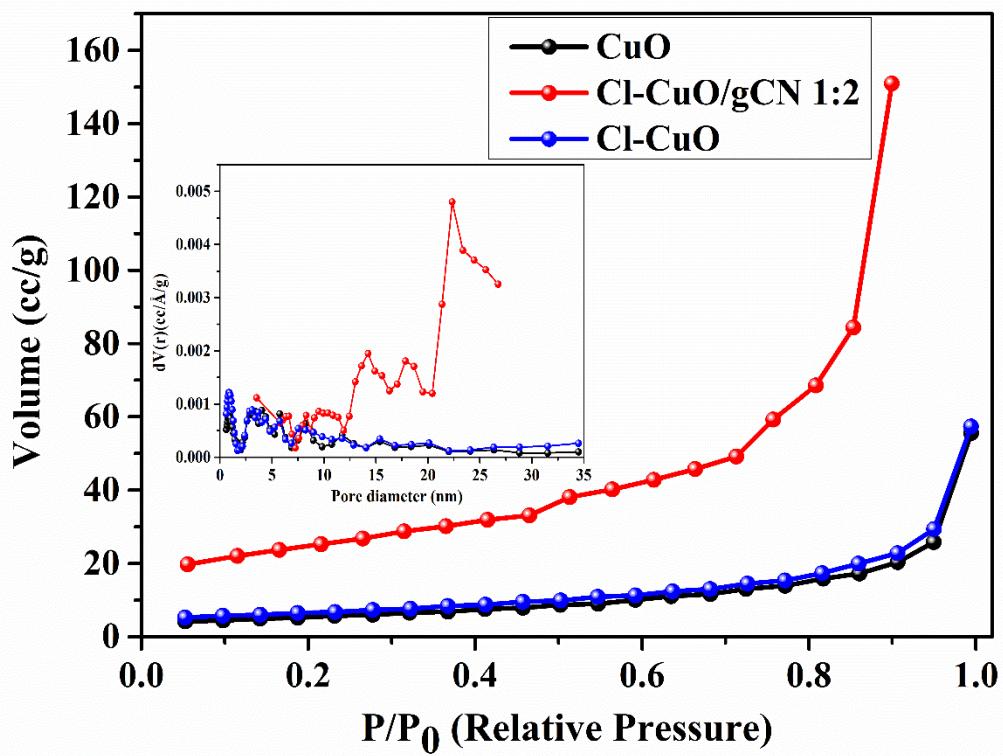
**Scheme S1.** Molecular structural formation of Reline



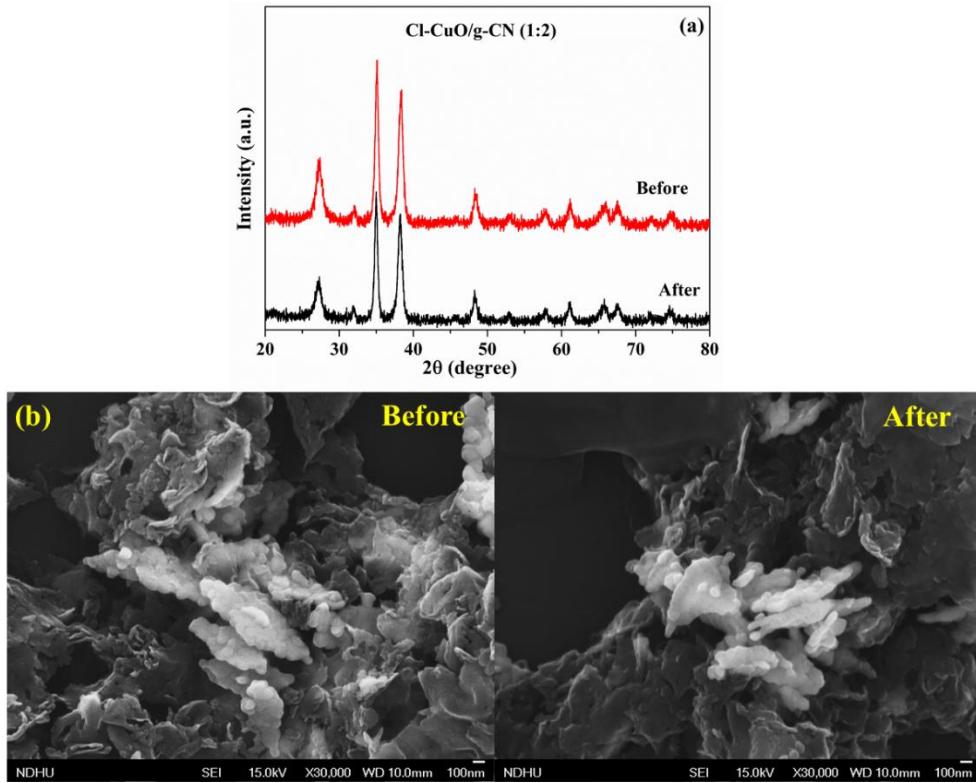
**Fig. S1.** FESEM images of (a)  $\text{g-C}_3\text{N}_4$ , (b)  $\text{CuO}/\text{gCN}$  2:1, (c)  $\text{CuO}/\text{gCN}$  1:1 and (d)  $\text{CuO}/\text{gCN}$  1:2.



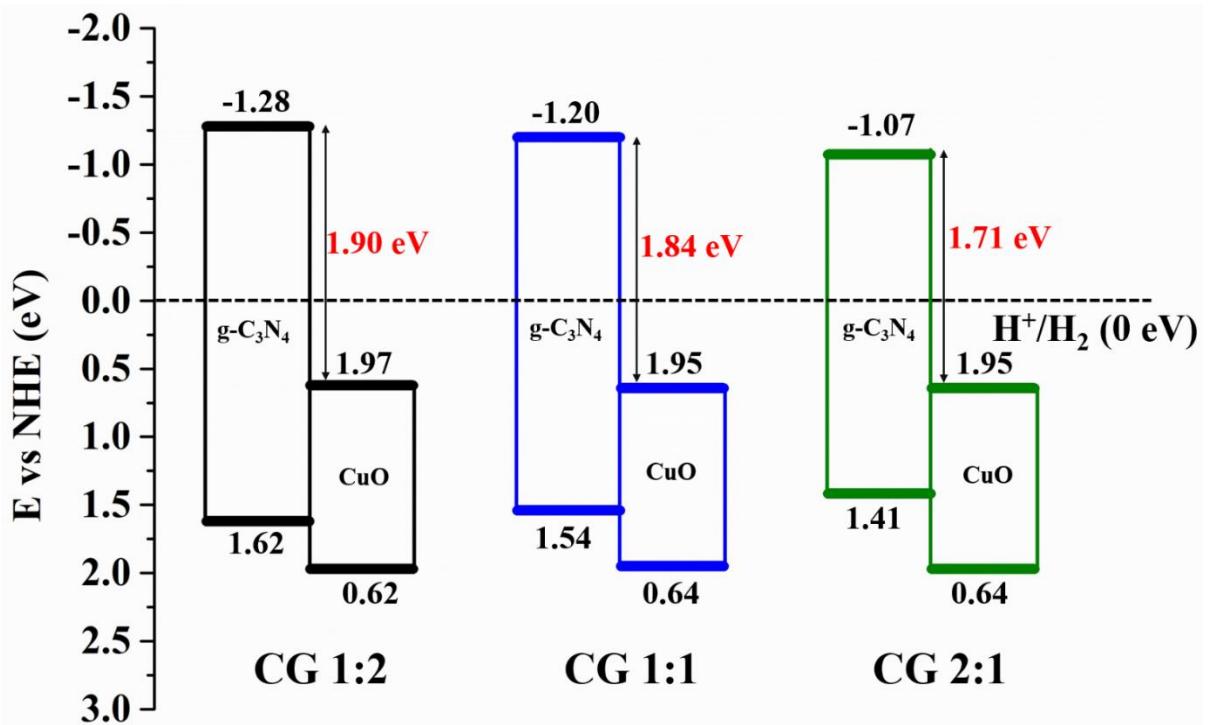
**Fig. S2.** FE-SEM elemental mapping  $\text{Cl-CuO}/\text{g-CN}(1:2)$ , (a) FE-SEM image, (b) C, (c) N, (d) Cu and (e) O



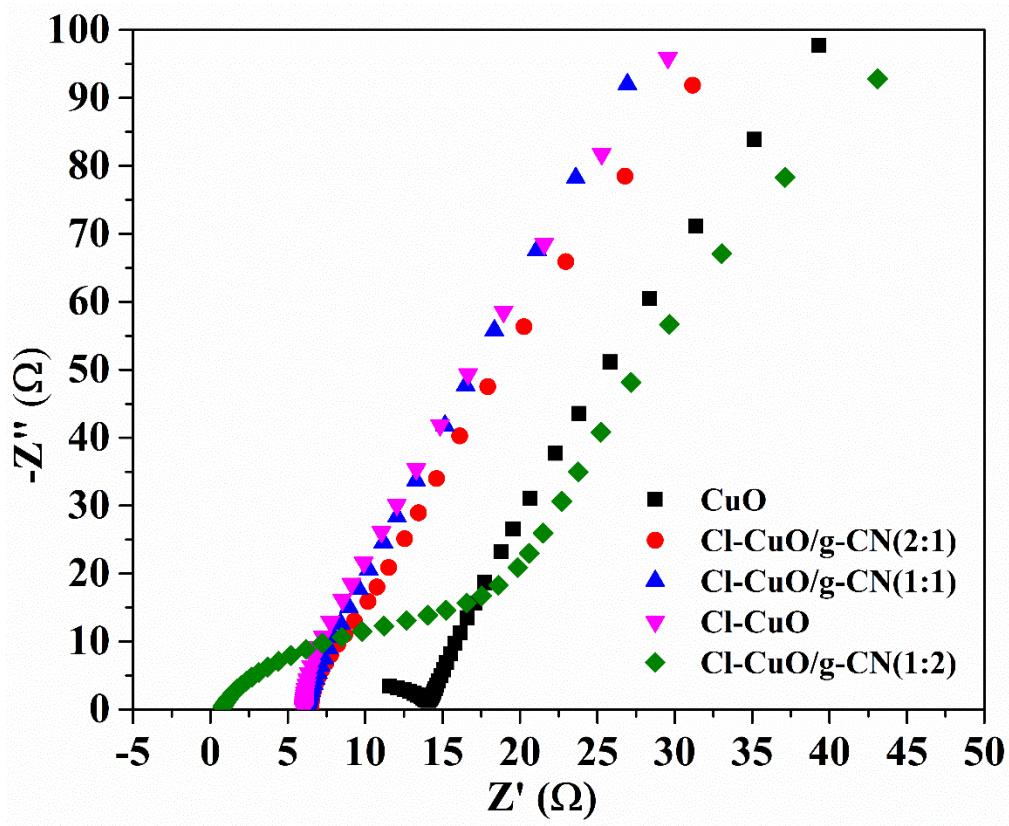
**Fig. S3.** BET and pore size distribution of CuO. Cl-CuO and Cl-CuO/g-CN (1:2)



**Fig. S4.** XRD spectra of Cl-CuO/g-CN (1:2) before and after recycle test of 4-NP degradation (a), FE-SEM images before and after recycle test (b)



**Fig. S5.** Comparison of band edge potentials (E vs NHE) among various ratio of Cl-CuO/g-C<sub>3</sub>N<sub>4</sub> composite (CG = Cl-CuO/g-CN)



**Fig. S6** Electrochemical impedance spectra of Cl-CuO/g-CN composites and CuO

**Table S1.** Band gap energy calculations of prepared composites

Photocatalysts	(αhv) <sup>2</sup> vs hv Band gap (eV)		(αhv) <sup>1/2</sup> vs hv Band gap (eV)		Absorbance vs wavelength Band gap (eV)	
	g-CN	Cl-CuO	g-CN	Cl-CuO	g-CN	Cl-CuO
Cl-CuO/g-CN (1:2)	2.90	1.35	2.24	0.85	2.57	1.16
Cl-CuO/g-CN (1:1)	2.74	1.31	-	0.73	2.33	1.15
Cl-CuO/g-CN (2:1)	2.49	1.31	-	0.77	-	1.15
Cl-CuO	-	1.30	-	0.66	-	1.14
g-C <sub>3</sub> N <sub>4</sub>	2.98	-	2.62	-	2.81	-

**Table S2.** Calculated relative contribution percentages of lifetimes t<sub>1</sub> and t<sub>2</sub> and their values.

Sample	A <sub>1</sub> (rel. %)	t <sub>1</sub> (ns)	A <sub>2</sub> (rel. %)	t <sub>2</sub> (ns)	t <sub>av</sub> (ns)
CuO/gCN 1:2	99.3	1.78	0.66	8.14	1.82
g-C <sub>3</sub> N <sub>4</sub>	97.9	3.28	2.0	16.38	3.54

**Table S3.** Calculated rate constant values of the prepared photocatalytic composites

Photocatalysts	Rate Constant k ( $\times 10^{-2}$ min <sup>-1</sup> )
Cl-CuO/g-CN (1:2)	2.74
Cl-CuO/g-CN (1:1)	1.76
Cl-CuO/g-CN (2:1)	1.65
Cl-CuO	1.74
g-C <sub>3</sub> N <sub>4</sub>	0.53
H <sub>2</sub> O <sub>2</sub> +light	0.53

**Table S4.** Calculated values of activation energy, enthalpy, entropy and Gibbs free energy by photodegradation at different temperatures

Temperature (°C)	E <sub>a</sub> (KJ mol <sup>-1</sup> )	ΔH (KJ mol <sup>-1</sup> )	ΔS (KJ mol <sup>-1</sup> K <sup>-1</sup> )	ΔG (KJ mol <sup>-1</sup> )
30	37.87	35.26	-0.236	106.8
40				109.1
50				111.5

**Table. S5** Comparison of photo-oxidation of 4-Nitrophenol with previous reports.

Photo Catalyst	Catalyst amount (mg)	4-NP concentration	Light source	Degradation % & Time (min)	Recycle test	Reference
GO/TiO <sub>2</sub>	100 mg	10 ppm	150 W Hg Lamp	59.5% & 120 min	---	<sup>1</sup>
TNTs: CoFe <sub>2</sub> O <sub>4</sub> /TCF	100 mg	10 ppm	4 Sylvania TLD 15 W/08 UV-A lamps	100% & 35 min	5 cycles	<sup>2</sup>
Ag–OMS-C	10 mg in 20 μL	0.10 mM in 2 mL	-----	96% & 120 s	5 cycles	<sup>3</sup>
Ag@TPHH-COF	4 mg	0.4 mM	----	100% & 150 s	6 cycles	<sup>4</sup>
MCNF@NiCo <sub>2</sub> O <sub>4</sub>	2 mg	0.01 M in 25 μL	----	100% & 45 min	20 cycles	<sup>5</sup>
Pt@Ag NPs	1.0 mg mL <sup>-1</sup>	0.10 mM)	----	100% & 8 min	5 cycles	<sup>6</sup>
Cl-CuO/g-C <sub>3</sub> N <sub>4</sub>	10 mg	20 ppm (50 ml)	35 W Xe	100 min & 92.8%	5 cycles	This work

**Table S6** Calculated values of Current density, onset potential and over potential

Photocatalysts	Onset potential (V)	Over potential (V)	Current density (mA cm <sup>-2</sup> )
Cl-CuO/g-CN (1:2)	2	0.77	33.11
Cl-CuO/g-CN (1:1)	2.20	0.97	24.29
Cl-CuO/g-CN (2:1)	2.22	0.99	19.29
Cl-CuO	2.12	0.89	30
CuO	2.28	1.05	9.7

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