

## Supporting Information

### Chemical Cleavage of Layered Carbon Nitride with Enhanced Photoluminescent Performances and Photoconduction

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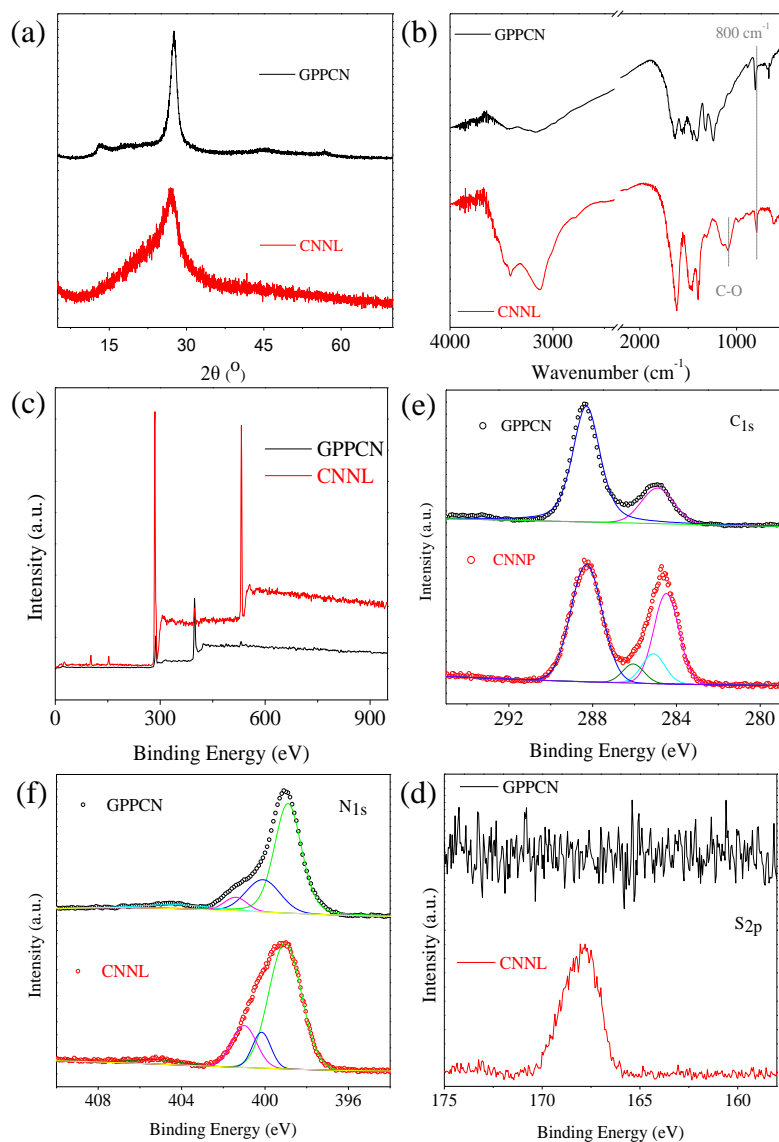
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**Figure S1.** Photo of bulk GPPCN (left) and GPPCN/H<sub>2</sub>SO<sub>4</sub> solution (50 mg/mL, right).



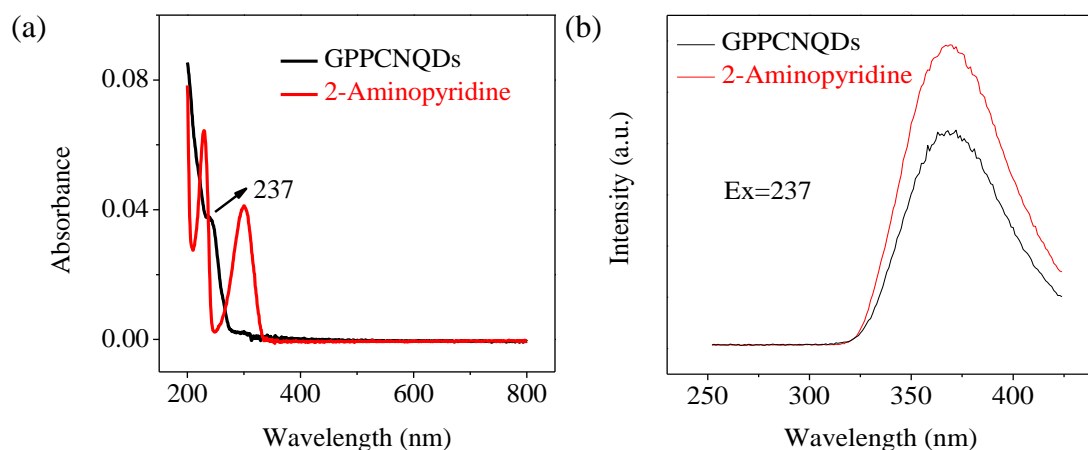
**Figure S2.** XRD pattern (a), FT-IR spectra (b), XPS survey spectra (c), XPS  $\text{C}_{1s}$  (d),  $\text{N}_{1s}$  (e) and  $\text{S}_{2p}$  (f) spectra of pristine bulk GPPCN and CNNL.

X-ray diffraction patterns (XRD) of as-prepared CNNLs presented one predominant (002) peak at ca.  $27^\circ$  similar with their bulk counterparts, while the low-angle reflection peak of bulk carbon nitride located at  $13.3^\circ$  disappeared for the CNNLs (Figure S2a). With respect to the (002) peak stemmed from the periodic stacking of layer structure, a slight increase of the peak width occurred, suggesting a similar intrinsic crystal structure as the bulk one but with an increased interlayer spacing due to the presence of functional groups on the surface of CNNLs.<sup>1</sup> The peak located at  $13.3^\circ$  was originated from the in-planar repeat period of tri-*s*-triazine units

with small tilt angularity in bulk carbon nitride, which was absent for CNNLs due to the structural defects and decreased planar size.<sup>1</sup>

As shown in Figure S2b, the characteristic IR spectrum of the CNNLs was largely reminiscent of that of the bulk GPPCN, both having a strong band at around 800 cm<sup>-1</sup> (tri-*s*-triazine ring sextant out of plane bending vibration), and 1200-1700 cm<sup>-1</sup> (C-N heterocycles stretches).<sup>2</sup> Nevertheless, the FTIR spectrum of CNNLs shows the disappearance of some stretching vibrations of connected units of C-N-C or C-NH-C located between 1200 and 1700 cm<sup>-1</sup>, which can be ascribed to partial break of triazine rings due to small sizes.<sup>1</sup> The appearance of new peaks ca. 3400 and 1110 cm<sup>-1</sup> can be attributed to the stretching vibrations of OH group and C-O, respectively.<sup>3</sup>

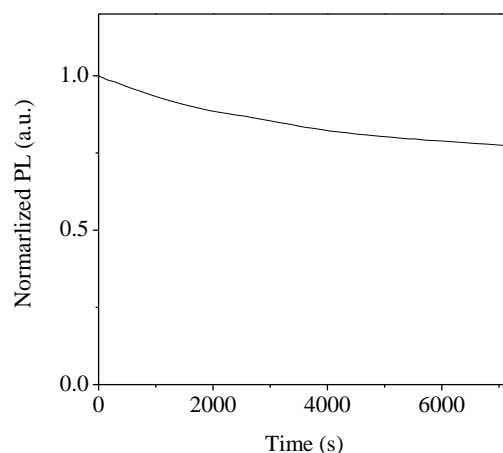
To further probe the chemical environment and bonding between the carbon and nitrogen atoms in the CNNLs, XPS measurements were conducted. As shown in Figure S2c, both carbon nitride and CNNLs shows the existence of C, N and, O, but a much higher O peak was observed for CNNLs, due to the introduction of OH group after hydrolysis reaction. As illustrated in Figure S2d, by deconvolution using Gaussian-Lorentzian fitting, one predominant C<sub>1s</sub> peak with a binding energy of ca. 288.3 eV (C1), corresponding to the typical aromatic C-N=C coordination in GPPCN framework, was observed for both carbon nitride samples.<sup>4</sup> However, the high resolution C<sub>1s</sub> XPS of the CNNLs shows the existence of C-OH (ca. 285.2 eV) and C-NH<sub>2</sub> (ca. 286.4 eV), due to partial break of triazine rings.<sup>1</sup> The high resolution N<sub>1s</sub> spectra of both carbon nitride samples (Figure S5e) can be deconvoluted into four different peaks at binding energies of 398.9 (N1, sp<sup>2</sup> N atoms involved in tri-*s*-triazine rings (C-N-C)), 400.1 (N2, bridging N atoms in N-(C)<sub>3</sub>) and 401.4 (N3, N bonded with H atoms (C-N-H)), respectively.<sup>5</sup> The intensity of N2 was decreased but N3 was increased, suggesting most of N-(C)<sub>3</sub> has been transferred into -OH or NH<sub>2</sub>. In addition, the high resolution S<sub>2p</sub> peak verified the absence of S-C and S-N bonds, suggesting there was covalent bonding between the H<sub>2</sub>SO<sub>4</sub> molecules and CNNLs. Evidently, the XRD, XPS and FTIR spectra all indicated that the CNNLs keep the same chemical structure as their parent bulk GPPCN with oxygen or amino functional group due to hydrolysis.



**Figure S3.** (a) UV-vis absorption spectra and (b) PL emission spectra of the standard 2-Aminopyridine (red) and CNQDs (black) with excitation at 237 nm, where the absorbance values of them are equal. The quantum yield of the CNQDs was calculated using the following equation, by referencing to that of 2-Aminopyridine.<sup>6-8</sup>

$$Q = Q_R \cdot \frac{I}{I_R} \cdot \frac{OD_R}{OD} \cdot \frac{n^2}{n_R^2}$$

where  $Q$  is the quantum yield,  $I$  is the measured integrated emission intensity,  $n$  is the refractive index, and  $OD$  is the optical density, which is measured on a UV-Vis spectrophotometer. The subscript  $R$  refers to the reference fluorophore of known quantum yield, i.e. 2-Aminopyridine used in present work. The 2-Aminopyridine (literature  $\Phi=0.60$ ) was dissolved in 0.05 M  $H_2SO_4$  ( $n=1.33$ ) and the CNQDs was dissolved in distilled water ( $n=1.33$ ). In order to minimize re-absorption effects, absorbencies in the 2 mm fluorescence cuvette were kept under 0.1. By the above equation, the PL QY of the CNQDs was measured to be 46%.



**Figure S4.** Photobleaching properties of the carbon nitride quantum dots under continuous excitation at 243 nm.

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